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Comment

Interactive comment on “Particle concentration and flux dynamics in the atmospheric boundary layer as the indicator of formation mechanism” by J. Lauros et al.

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As Referee #1 mentioned the main conclusions of the manuscript are:

- 1) Based on the measurements and modeling of the vertical particle number concentration profiles it was illustrated that sulphuric acid nucleation alone cannot be responsible for the observed nucleation events in Hyytiälä but the organic-induced formation mechanism from monoterpene oxidation likely is.
- 2) It is important to capture the vertical mixing within the atmospheric boundary layer (ABL) and between the ABL and free atmosphere when quantifying new particle formation in the troposphere.

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The paper addresses relevant scientific questions concerning which mechanism(s) which is responsible for new particle formation in the troposphere. To answer these questions they using a new and interesting approach where hot-air balloon flight measurements is combined with detailed model simulations of the vertical particle concentration profiles. Therefore I recommend that the manuscript can be accepted for publication if the below given comments are considered by the authors.

Major comments

These are very interesting results and I fully agree with the second conclusions. However, although the comparison between the modeled and measured vertical profiles are convincing and support the hypothesis that the organic-induced formation mechanism is responsible for the particle formation in Hyytiälä it is still not a proof, and therefore I agree with Referee #1 that the conclusions about the nucleation mechanism should also be toned down and instead of as you write on line 2-4 on page 20019 “This result allows to conclude that the organic-induced formation mechanism is responsible for particle formation at a Boreal forest site.”, you could write e.g. This result indicate that organic-induced formation mechanism could be the responsible mechanism for particle formation at a Boreal forest site.

I miss the comparison between the modeled and measured O₃, CO, SO₂, NO and NO_x concentrations. It would be especially interesting to compare the modeled and measured O₃ and SO₂ concentrations since O₃ is a proxy for the oxidizing capacity of the atmosphere and the SO₂ concentration is strongly related to the H₂SO₄ concentration which affect the nucleation rate both for the sulfuric and organic nucleation mechanism which is tested. Where did you get the gas phase emissions from, which temporal and spatial resolution did the emissions have? On page 20013 line 18-20 you write that you set the initial particle concentration above the ABL to 0.2 of the observed concentration at the surface. How was this performed? Did you just multiply the whole particle number size distribution with 0.2? This would then give a significant smaller coagulation and condensation sink above the atmospheric boundary layer than

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within the boundary layer. Since the accumulation mode particles generally are relatively long-lived in the atmosphere I would expect to have an almost uniform vertical profile of the concentration of particles above 100 nm in diameter and not a drastic decreasing concentration above the boundary layer. These particles have a large influence on the condensation sink and coagulation sink, which I believe is important for the particle number concentration of particles below 25 nm in diameter. I suggest that you test to run the model with different assumption regarding the initial particle number size distribution above the ABL and evaluate the importance of the coagulation and condensation sink for the modeled particle number concentration at different altitudes. I also want to know which initial gas phase concentrations that you used. Did they vary with altitude? Is it OK to assume that the SO₂ concentration is constant within and above the ABL or should that also be scaled with 0.2 as the particle concentrations? If you don't have measurements of the SO₂ concentration from the hot-air balloon flight measurements I suggest that you do some sensitivity test by varying the initial SO₂ concentration within and above the ABL.

On page 20014 line 12-14 you write “on the selected days change of air masses by horizontal advection most likely prevented the appearance of clear “banana-plot” compared to the model”. I agree with you that this could be a possible explanation. I myself do similar model attempts as you, but then I want to compare the model results with measurements from one measurement site I use my model as a Lagrangian model along 7 days backward air mass trajectories. This way I get a representation of the long distance transported gas and particle properties which depend on the regions which the trajectory pass over before reaching the measurement site, and by applying the model along several backward trajectories which reach the measurement station with e.g. 3 hours in-between you can account for the change of air masses by horizontal advection. I think this is a methodology which would improve your model results. However, for the limited time period which you simulate (in total 3 days) it might be reasonable to assume that measurements in Hyytiälä are representative of the same ageing air mass. This you will need to illustrate with modeled air mass trajectories (e.g.

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HYSPLIT model) and I would also like to see a plot of the measured PM₁, PM_{2.5} or volume concentration in Hyytiälä for the three days. If the volume and mass show large variations with time it indicates that the air masses are of different origin.

On page 20007 lines 11-15 you write that the first order mixing parameterization (K-theory), have not succeeded to present turbulent mixing in all atmospheric layers and conditions, so higher order models have been developed, and on lines 26-29 you write “we have chosen a one-and-half order parameterization (Boy et al., 2010) which presents mixing better than first order models and is more proper for aerosol flux than the counter-gradient models mentioned above. Although I also believe that the more advanced one-and-half order parameterization often represents mixing better than the K-theory it does not always have to be this way. Therefore I suggest that you either referee to studies which clearly state that the K-theory does not work as accurately as the more advanced one-and-half order parameterization or that you perform this model comparison yourselves.

On page 20015 lines 12-13 you write “in the presented simulations (Fig. 2c and d) we have assumed that 5% of organic reaction products are able to condense on freshly nucleated particles. How many percent of the organic reaction products are able to condense on the Aitken and accumulation mode particles? Is it not the same as for the freshly nucleated particles? This seems to be a quite crude assumption to have a fixed fraction which does not vary with the temperature and total organic particle mass, but since it seems to give similar growth rates as detected with the DMPS I still think you could use this yield, especially if the temperature and organic particle mass show small temporal variation. Which compounds except organic oxidation products grow the particles e.g. NH₃, H₂SO₄, HCl and HNO₃?

Another question I have is how the organic reaction products which are able to condense or form nucleation clusters are formed? Are they assumed to form directly after the first oxidation step or do they need to be oxidized several steps? Most first oxidation step reaction products from monoterpenes will be too volatile to condensate and

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would require several oxidation steps before they are low volatile enough to form particles. Is this something you have considered? This will influence how far up into the atmosphere the organic compounds will be mixed before they can condense or form nucleation clusters, and therefore also influence the vertical particle number concentration profiles, especially when using the organic nucleation mechanism. I suggest that you do some sensitivity tests by varying the number of oxidation steps required before the organic compounds can condense or form nucleation clusters.

The text and message is usually easy to follow, however the language need to be improved. I give a few examples as minor comments

Minor comments

All figures are easy to understand and have describing legends and labels. The abstract gives a good summary of the paper.

I do not think that C in equation 4 is defined in the text.

Page 20006 line 5, missing “the”. “We show that kinetic nucleation of sulphuric acid cannot be responsible for new particle formation alone as the vertical profiles of “the” particle number size distribution does not correspond to “the” observations” Similar changes need to be made on many more places in the manuscript.

Page 20011 line 20-21. “Nucleation is followed by growth according to nano-Köhler theory having sulphuric acid and reaction products of organics with OH as condensing vapour.” I understand this sentence but it is not correct English. Instead of writing “reaction products of organics with OH” you can write, reaction products from organic compounds oxidized by OH.

What is the temporal and spatial resolution of the monoterpene emissions calculated with MEGAN? Can you specify the temporal and spatial resolution? How detailed is the forest species composition described?

On page 20011 line 14-15 you write that the constant P value used when calculating

the organic nucleation rates varies between 1×10^{-4} and $2 \times 10^{-4} \text{ cm}^3$ depending on the organic reaction products which are participating nucleation. What do you mean by that? And how do you assign realistic values of P which is different for different organic reaction products?

On page 20012-20013 you describe the new advanced deposition parameterization with several levels inside the canopy. This seems to be an important improvement of the model however you never test the new model against earlier model versions which described the deposition to canopy with a bulk parameterization. Although this is not a requirement from me this would be a useful and good test to do.

Do you consider any primary particle emissions?

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 20005, 2010.

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