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Atmospheric Chemistry and Physics Discussions

Interactive comment on "New trajectory driven aerosol and chemical process model: chemical and aerosol Lagrangian model (CALM)" *by* P. Tunved et al.

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This manuscript describes the development of a Lagrangian box model and its initial evaluation with atmospheric observations from Hyytiälä, Finland. The idea of the Lagrangian box model that can be used with trajectories is great, and model will potentially very useful in analyzing data observed at field stations. The manuscript is generally clearly written and fits well within the scope of ACP. The model is described well – the reader gets a good idea of what the model is doing. I do, however, have a few comments that I think could improve the manuscript and that I think the authors should address before the MS can be accepted to ACP.

Major comments:

1. It would be good if the authors could demonstrate how sensitive the model output is to the trajectories used – i.e. the information of the height and meteorological parameters?

2. The current version of the model is concentrating on reproducing the aerosol number size distributions. It would be good, however, if the authors could comment on how well the model is doing with the the mass size distributions? This could be done by e.g. adding a comparison of the observed and measured mass size distributions to Fig. 3

3. How is cloudiness related to radiation intensity and spectrum along the trajectory? This should be somehow described, and it would be naturally good, if these were somehow related in the calculations. Now I got the impression that they are not, as the clouds are generated randomly. It is also possible, of course, that I misunderstood the description. If this is the case, the authors should try to improve the text to make this connection clear to the reader.

4. How sensitive is the model output to the initial concentrations of trace gases? How long does the air parcel have to move to not be dependent on the initialization? I think it would be important to demonstrate this, in addition to the sensitivity tests that have already been presented.

5. Fig.5, and the related discussion: Why is the model overpredicting nucleation (?) so much in May? This should be discussed in the text. Also the reasons of the overpredictions in the wintertime should be discussed with this Fig. too, although some discussion is provided later in the MS. Generally the model seems to be systematically overpredicting the particle numbers rather than underpredicting them. It would also be important to demonstrate how sensitive the model is to meteorology (e.g. the mixing height), deposition and clouds along the trajectory, since these seem to be potential candidates for explaining the differences. This would also give useful information on which process descriptions and parameterizations to improve in the next stage.

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6. Fig. 7: It would be nice if the authors could somehow show, besides the cluster centroids, also the variation of trajectories in each cluster, i.e. some sort of “bounds” of the clusters. It would be useful to see the most “extreme” trajectories with respect to the centroid that are included in each of the clusters.

7. Figs. 9-13 and the related discussion: The authors note that the model overpredicts the nucleation mode number concentrations. Could it be that this overprediction is related to the assumption they make about organics (i.e. that 15 % of the monoterpene oxidation products condense as completely non-volatile components)? The 15 % yield is probably representative to the first order oxidation products, a large fraction of which probably has a non-negligible saturation vapor pressure. Treating these compounds as completely non-volatile sticks them to the particles permanently (i.e. will not let them evaporate). This might result in overpredicting the atmospheric lifetimes and thus the average concentrations of the nucleation mode particles. Could the authors comment on this? I think also the measurement-model comparison shown in Figs. 11 and 12 (particularly the stable accumulation mode) could indicate something like this. Based on Fig. 13 the authors conclude that the underprediction of the nucleation mode growth is related to underprediction of condensable vapors. However, at the same time the model is clearly overpredict the aerosol numbers. Could it be possible that the overprediction in particle numbers actually result in underpredicting their growth (even if the overall amount of condensable vapors was correct), as the condensable vapors need to be divided to larger number of particles? The authors also compare their organic condensable vapor concentrations to those estimated by Kulmala et al. (2002) and Spracklen e tal. (2008). It should be noted, however, that these authors used a similar approach as the authors are using, and thus correspond to the case where the vapors are effectively non-volatile.

8. As a comment that does not really require any action from the authors: I think result on the importance of FT nucleation is extremely interesting – although contradicting to the previous study by Merikanto et al. (2009). These results indicate strongly that

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further studies on this phenomenon are needed.

Minor comments:

9. What are the 76 species treated in the model? Same goes for the 56 chemical reactions.

10. Fig. 2: how do the particle number size distribution observations in Hyytiälä look like during the simulated period (or at least on the last day) – when was Hyytiälä in a similar airmass with the modeled air parcel? It would be useful to see this plot.

11. Fig. 3: I suggest using a logarithmic y-axis. Also, how does the large end of the size distribution look like/compare?

12. Fig. 5: Please give explanations to the curves in the caption or in a legend.

13. Fig. 8 and the related discussion: It might be a good idea to give each cluster and representative name, like “Atlantic”, “Central Europe”, “Eastern Europe” etc. and give these names and corresponding numbers in a table. That way the reader could better interpret the physical meaning of the results (e.g. comparisons to data) related to each of the clusters. Also, it would be nice if the authors would discuss the performance of the model and most importantly I think the authors should at least briefly touch the reasons for the overpredictions in e.g. clusters 1, 4, 5 and 10 (although this is discussed also related to Figs. 9-10). Can it be related to the anthropogenic influence, e.g. the emission inventories? This would be somewhat in line with the dsiscussion that the authors present as explanation to Figs. 9-10.

14. Fig. 9: I suggest using logarithmic scales in all y-axes. Please also add tick marks to the axes.

15. Reference to Fig. 12 should be before the reference to Fig. 13 in the text. Otherwise change the numbering of the figures.

16. Please refer to Tables 2 and 3 in the text. Currently I could not find references to

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these important tables in the text. Also, please explain parameters used in the first row of Table 2 in the caption.

Technical comments:

I have tried to list some of the small typos and grammatical errors. I suggest, however, that the authors read the MS through once more carefully to ensure that it reads well. Please also improve the figure quality and resolution. Some of the figures are difficult to read. Also consider using larger font size and line width in the plots and add tick marks where appropriate for helping the reader interpret the results.

17. Title: trajectory driven -> trajectory-driven?

18. Fig. 1: the y-axes are difficult to read. Maybe chop the figure in two vertical panels? Could the authors also give the solar radiation along the trajectory?

19. Fig. 8, legend, line 2 from the top: “Fig. AP3” -> “Fig. 7”

20. p. 15198, Line 1: have -> has

22. p. 15199, Line 16: computational -> computationally

22. p. 15202, line 22, p. 15204, line 24: “. . .a size range and resolution that we consider sufficient for this study.” and “For the purpose of this study we do not think that this assumption will affect the results in a major way.” Without aguments backing up these statements, they seem rather unnecessary.

23. p. 15207, line 6: Are clouds phenomena? Maybe think of another wording.

24. p. 15208, line 24: remove the second “cloud” after “in-cloud”

25. p. 15211, line 22: “Primary emissions” -> “Primary aerosol emissions”

26. p. 15213, line 5: “. . .particles remaining as individuals”. Can particles be individuals . . . somehow that makes me think of people. Think of a better wording for this sentence.

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Interactive Comment

27. p. 15213, line 27: “The population of activated CCN evaporates and leaving significantly larger . . .” rephrase to “The population of activated CCN evaporates and leaves behind significantly larger. . .” or “The population of activated CCN evaporates leaving behind significantly larger . . .”

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 15197, 2010.