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Interactive comment on "A pseudo-Lagrangian model study of the size distribution properties over Scandinavia: transport from Aspvreten to Värriö" *by* P. Tunved et al.

Anonymous Referee #1

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This is a potentially interesting paper that attempts to address important science questions pertaining to aerosol formation and growth, and is very much within the scope of ACP. The authors use an aerosol box model in Lagrangian frame of reference, which is a powerful tool for examining chemical and physical processes in greater detail than is normally possible with Eulerian models. However, this study makes too many critical assumptions in constraining and evaluating the box model, which I think seriously weaken the interpretation and conclusions.

First of all, the initialization of the model without any gas and aerosol chemical mea-

surements at Aspvreten is questionable. Using monthly average values for gas-phase species measured at Hyytiala is probably not a good assumption for episodic case studies.

Second, all the non-radical species are held constant in the model over the entire simulation period. This is especially problematic for SO2, which is concluded to be important in nucleation and secondary growth of aerosols. Also, OH loss due to reactions with isoprene, terpenes, and their oxidation products is ignored in the model.

Third, evaluation of model performance by just comparing size distributions at Varrio is not sufficient, especially since the critical model inputs were simply assumed. Also, because of the long transport times (˜2-3 days), comparison of model predictions with measurements is needed at some intermediate point (preferably aloft with an aircraft) along the trajectory for a convincing evaluation of the model and interpretation of the observations.

The authors admit in the first paragraph on page 7770 that model overestimates mass in most cases and underestimates in the rest. They attribute the shortcomings to miscalculated growth rates, which is of course obvious. However, it is very difficult to figure out the underlying causes due to lack of critical measurements to constrain the model. So the question is: What have the authors really learned from this exercise?

I think this study would have been significantly more useful and informative if the authors were to estimate the yield of low volatile terpene oxidation products required to explain the observed increase in mass at Varrio as opposed to assuming a constant value of 13%. The suggested analysis would result in a range of overall yield that may be expected from terpene oxidation, assuming that the terpene emissions, vertical mixing, and oxidation rates are treated reasonably well in the model. In the sensitivity analysis, the authors could turn the terpene emissions off and estimate the initial concentrations of SO2 needed to explain the observed increases in mass, and then see if those levels make any sense.

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Other specific comments/concerns: 1. The following two sentences are contradictory. Please revise or clarify. Page 7758, Line 19: “In order to explain the observed mass increase during transport, we conclude that a yield of Ĕ”

Page 7760, Line 26: We utilize a pseudo-Lagrangian box model approach to explain the observed decrease in particle mass and number concentrationĔ”

2. On Page 7760, line 24 it is stated that Aspvreten is located 70 km south of Stockholm, and on Page 7761, Line 14 it is stated that Aspvreten is located 70 km southwest of Stockholm. Which one is correct? It would also be useful to actually show Aspvreten, Stockholm, and Varrio on the map in Figure 2.

3. The authors need to do a more thorough literature search. Not a single reference is cited on Lagrangian modeling studies done in the past.

4. The mathematical construction of the pseudo-Lagrangian box model is not readily evident from the text. Why is it called “pseudo-Lagrangian” as opposed to “Lagrangian.” This may be obvious to some readers, but needs an explanation to be sure of what the authors mean. Please give the governing equations, and show how the box dimensions (base and height) are defined and how vertical and horizontal dilution is treated in the model? Also, what were the boundary layer heights in the selected cases, and how were they varied diurnally along the trajectories?

5. Page 7763, Line 9-10: Why is growth due to ammonia and water considered as numerical diffusion? Also, it is not clear how fixed size sections can eliminate numerical diffusion completely. Fixed sections will always introduce numerical diffusion when you try to re-map the new size sections on to the fixed size sections after condensation calculations.

6. It appears that only terpene emissions were varied diurnally in this study. What about emissions from Stockholm and other urban areas (if any) along the Lagrangian trajectories.

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7. How is growth of mixed layer treated when the air parcels are over water?

8. From Figure 2 it appears that most of the trajectories passed over water where there are no terpene emission. Was the terpene emission shutoff over water? If so, how was horizontal mixing of terpene emissions were handled due to directional wind shear in the vertical along the trajectory? For example, 200 m trajectory may be mostly over land while the 10 m trajectory is mostly over water. This can produce strong gradients in terpene concentrations, especially in the absence of strong vertical mixing over water.

9. What is the maximum height of the mixed layer over land? Is it same as the height of the residual layer which is assumed equal to 2000 m. If so, is it appropriate to use trajectories only up to 200 m?

10. Based on the sensitivity analysis, the authors conclude that dry deposition has relatively minor impact on aerosol mass, but still go on to say that it is important. How can it be important when it is of minor consequence (at least in the model)? Also, under what conditions would it actually become important?

11. It would be useful to show initial and final mass distributions in addition to the number distributions.

Editorial: The language is fairly precise, but needs a thorough revision by someone more fluent in English. Some sentences are rather awkward and hard to understand. Below is a list of editorial errors (probably incomplete) I was able to catch.

1. Page 7760, Line 10. Replace “was” with “were.”

2. Page 7760, Line 21. Suggest deleting “approach.”

3. Page 7762, Line 8. Replace “where” with “were” and reword that sentence.

4. Page 7766, Line 17. Change “decrease” to “decreases.”

5. Page 7766, Line 22. Change “increase” to “increases.”

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6. Page 7766, Line 25. Change “get” to “gets.”

7. Page 7766, Line 29. This is an awkward sentence. Try rewording it.

8. Page 7767, Line 11. Replace “changes” with “change.”

9. Page 7767, Line 20. Change “substantial” to “substantially.”

10. Page 7769, Lines 8 and 12. Avoid using the word “excellently.”

11. Page 7775, Line 4. Change “o” to “of.”

12. Page 7775, Line 13. This is an awkward sentence. Try rewording it.

13. Page 7775, Line 20. Replace “apart” with “different.”

14. Page 7776, Line 16. Replace “put down” with “made.”

15. Page 7777, Line 8. Change “no” to “not.”

16. Page 7777, Line 9. This is an awkward sentence. Try rewording it.

17. Page 7778, Line 7. Change “is” to “are.”

18. Page 7778, Line 14. Change “over all” to “overall.”

19. Page 7778, Line 23. Change “occur” to “occurs.”

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 7757, 2004.