**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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The authors thank Dr Riipinen for insightful comments on the manuscript. Dr Riipinen has provided several suggestions for improving the readability and quality of the manuscript. We have followed the suggestions in most cases, and our detailed response is outlined below.

Response to major comments:

1.) In the current setup, the model describes the evolution of gas species and particles in two compartments along calculated trajectories. The height of the trajectory is not directly used, as the model in principle only utilize the lat-long coordinates to move the

two layer box compartment to the receptor. It is however agreed that the height of the trajectory may bias the result due to the fact that transport at higher altitudes may be different from transport close to the ground. To address this issue, a test was performed to investigate the role of transport height as described by the trajectories. In this test we simplify the transport heights by dividing the data set in two groups depending on time spent in the mixing layer. The test is described under 3.3.4 in the revised MS:

"The simplified model set-up used in this study utilizes the coordinates of the trajectories to describe the movement of a quasi-1-D column consisting of a mixing layer (ML) and residual layer (RL) compartment. Thus, the model describes how the model compartments move along the latitude-longitude coordinates until the receptor station (in this case Hyytiälä) is reached. As transport path and speed may vary significantly with altitude, trajectories travelling at on average higher altitudes may not always yield a fair representation of experienced sources and transport speed of the air in the boundary layer above the receptor. On average during the simulations, the air-parcel spend 74 % (or 160 h of 216 h total transport) of the time within the mixing layer. In order to test the validity of our model setup we divide the model output into two groups, one of which the air spends more than 160 h in ML and one group that spends less than 160h in the ML. For this test we utilize only simulations along trajectories calculated for year 2000. For the trajectories spending more than 160h in the ML, the simulated average of the accumulation mode number concentrations was 480 cm-3, compared to measured average of 418 cm-3. Corresponding values for the Aitken mode was found to be 1185 cm-3 and 698 cm-3 for modeled and measured concentration, respectively. Modeled nuclei mode concentration was found to be 314 cm-3 compared to the measured average of 245 cm-3. In the case of less time spent in ML, the simulated average of the accumulation mode number concentration was 322 cm-3, compared to measured average of 368 cm-3. Corresponding values for the Aitken mode was found to be 1221 cm-3 and 837 cm-3 for modeled and measured concentration, respectively. Modeled nuclei mode concentration was found to be 468 cm-3 compared to the measured average of 326 cm-3. Thus, in the case of dominating ML transport, we slightly over predict the

accumulation mode concentration, and in the cases with less ML transport we under predict the accumulation mode number concentration. This result in, most likely due to the reduced condensation sink, more nuclei mode particles in the case of less ML transport as compared to cases dominated by ML transport. The differences between the two cases are typically small and both high ML and low ML transport conditions result in a fairly good agreement between modeled and measured number concentrations. In order to avoid this kind of bias, a more thorough description of the vertical structure and transport would be required, but this is unfortunately beyond the scope of this study and model framework."

Regarding the test of the meteorological parameters, we have in the original manuscript shown sensitivity tests for cloudiness and precipitation. Given the formulations used in the model setup, testing of other meteorological parameters than temperature would likely not contribute much to the paper. Changes in temperature will effect emissions of monoterpenes. This may mainly impact aerosol growth. However, we have in the original MS demonstrated the sensitivity to BVOC emissions, and concluded the model to be sensitive to these. Thus, additional detailed tests on temperature effect itself, although potentially interesting, seems less important at this stage.

Furthermore, we added a subplot to figure 7, showing the median altitude along each one of the trajectory clusters.

2.) The authors agree fully. In the revised MS we add a comparison of modeled and measured mass to figure 4. (The reviewer recommends changes to Figure 3, but most probably means Figure 4). Under section 3.2 we added:

"The total volume is further larger for the modeled data compared to the observed data as evident from second frame of Fig. 4. This also causes increased condensation sink, which may be part of the explanation to the slower growth of the modeled nuclei mode."

3.) This is clarified. At the end of section 2.3 we add: "The gas phase chemistry is further indirectly affected by cloudiness. When clouds are present, the photolysis con-

stants are adjusted accordingly, assuming a cloud optical depth of 20 (corresponding to reasonable cloudiness), modifying photolysis constants above (i.e. in the residual layer) and below the cloud column (i.e. in the mixing layer). " 4.) The initialization of the gas phase chemistry is in the current set-up admittedly quite crude. Thus it is, as pointed out by Dr Riipinen, important to demonstrate the sensitivity to different initial values of gas concentrations. This test is added as a new sub-section under "3.3 Sensitivity tests". We show here the evolution of OH radicals and NO2+NO during simulations initialized with 25 ppb O3 and 0.1 ppb NOx ozone and NOx, and compare with base case simulations (35 ppb O3 and 0.5 ppb NOx in base case). Following text was added under 3.3.1 together with a descriptive figure:

"The results from the runs with lower than base case concentration of ozone and NOx, the evolution of OH and NO2+NO as average along the trajectories of year 2000 is shown in Fig. 24. When choosing to initialize with lower NOx and ozone, one might expect at corresponding change in both ozone and OH concentration along the trajectories, and that this in turn will influence the oxidation potential and thus production of condensable species. In this test the initial concentration of O3 is 10ppb less than the concentration in the base case runs and the ozone recovers slowly during the length of the model run, and on average, at the end of the runs, the difference is less than 4 ppb (not shown). However, OH concentration and NO2+NO show a much more rapid recovery, and it is shown in Fig. 24 that NOx for the both types of simulation gets comparable after $\sim$80 h, and then follow each other until arrival at the receptor. When using the lower initial values of ozone and NOx OH requires slightly more time to recover to base case values, and gets comparable to the base case runs (at an average of $\sim$4*105 cm-3) after approx. 120 h. However, the change in final size distribution as a result hereof is very minor and not shown. This test shows, that the initialization of the model with proper gas phase concentrations is important to get an accurate description of the evolution of species such as ozone, but show at the same time that the final aerosol size distribution is largely unaffected by these moderate changes in ozone and NOx. This is due to comparable quick establishment of important oxidants and

precursors thereof."

5.) The number concentration in the smaller size range is larger for the simulated size distributions in May. However, we do not consider the difference to be very large as the modeled value still is within the 75th percentile of observations performed at Hyytiälä during the same month. It is hard to pinpoint the actual causes for this offset, but may be due to several reasons such as description of clouds, emissions, photochemistry etc. Indeed, the model seems to over-predict number in most cases, especially during winter months. The cause for the winter period discrepancy is addressed in a short discussion already included in the MS. Concerning the role of clouds; we have performed sensitivity tests for this under section 3.3. However, the role of ML height and transport within ML is discussed under point 1 above, where it was shown that transport predominantly taking place in ML is associated with overestimates of the accumulation mode concentration, while trajectories preferentially travel above the ML are associated with less accumulation mode particles.

6.) The figure 7 shows the cluster centroids of each separate cluster, i.e. the average trajectory. Although it is a good suggestion to add some bounds to these centroids, simply plotting bounds create a very blurry picture that is not easily interpreted. One solution would be to plot the trajectory clusters as density plots over the maps. This however requires several more figures to be added to the MS since the spread within each cluster still is quite large. Therefore we think that it is best to leave the trajectory figures as just the centroid of each cluster.

7.) This is a probable candidate for the overestimation of nuclei mode particles. The description of the production of low volatile (in principle non-volatile, sat. vap. of 3*1012 cm-3) is of course a simplification and this could indeed affect the nuclei mode particle concentration. In the text we add to the discussion:

"It is also clear that the model seems to overestimate the nuclei mode concentration while underestimating the growth. This could very well be a result of the way the

C8057

secondary organics are treated (i.e. treating first order products (15%) as very low volatile). This could result in higher concentration of small particles due to effective growth of the freshly formed particles to more or less stable sizes. This in turn could yield a larger condensation sink, which in turn hinders consecutive growth."

8.) The authors agree!

Minor comments:

9.) A subplot was added to figure 3 to show the evolution of the number size distribution as observed at Hyytiälä during the final day of simulation.

10.) This has been addressed under point 2 in this response letter by adding volume distribution comparison to the figure.

11.) This has been adjusted in the revised MS.

12.) Following text was added to the discussion of figure 7 & 8

"Based on the centroids, cluster 1, 2, 3, 7, and 8 are predominantly of marine origin and thus referred to as marine clusters , clusters 5, 6, and 10 are predominantly of continental origin (Continental clusters) and clusters 9 and 4 are considered to be of mixed marine-continental origin (Mixed clusters)." Same annotations are also given in the figure itself.

13.) I guess this comment actually refers to figure 8. If wrong, please correct. Y-scale changed to logarithmic.

14.) This is now adjusted. 15.) Table 2 referred to in first paragraph, section 3: "The model parameters of these initial size distributions are given in Table 2." Also, relevant information was added in the caption:

"Table 2: Modal parameters of the input size distributions. N(1-3) corresponds to number of particles in each mode (cm-3), GSD(1-3) correspond to the geometric standard deviation of each mode, and Dg(1-3) represents modal size in nm."

Table 3 is now referred to in section 3.2.1: "The number of trajectories belonging to each cluster is shown in table 3."

Technical comments:

16.) Changed to Trajectory-Driven 17.) The figure is for qualitative illustrative purpose only. Exact knowledge is not necessary, and we think it is better to try to reduce the number of figures in the MS. Solar radiation is not explicitly used. Instead, photolysis constants are extracted based on lat-long coordinates, day of year, time, and cloudiness. Only PAR is calculated and used in the calculation of isoprene emissions only. The rest of the technical comments are corrected following the suggestions of the referee.

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