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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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We thank the referee for insightful comments on the MS. The questions raised by the referee have called for additional tests and clarifications. The main concerns relate to the chemistry and mixing within the box model framework. We have tried to approach these issues in a clear and through way in the revised MS. The changes and additions suggested by the referee have increased the quality and readability of the manuscript. In the following, we will address the different concerns and questions raised by the referee one by one.

Response to Major comments:

The first issue raised by the referee relates to the length of the trajectory. In the current study, we use 9-days trajectories to simulate the transport to a receptor (in this study Hyytiälä). It is well known that the accuracy of trajectory rapidly deteriorate with increasing distance from the final receptor. It is further agreed, that since trajectories describe single particles, and thus isolated entities of air, the methods relying on the use of single trajectories neglect exchange with the surrounding air. For shorter periods of time, however, the use of single particle trajectories is likely adequate to describe transport. In this study we simulate the transport to a receptor without exact prior knowledge of the initial conditions. This is different from other Lagrangian simulations where transport is evaluated between two specific measurement sites. In the latter cases, the initial conditions are generally better known. In order to initialize a model run one can usually chose from two different possibilities: either use climatological data with respect to properties of aerosols and trace gases, or allow the model to spin up and equilibrate to the environment where the simulations starts. We have in this study chosen a hybrid approach in which we first approximate the aerosol and trace gas concentration with more or less well known values, and combine this initial guess with a longer run time of each simulation. We believe that during the longer simulation time, we will better approach the actual conditions, lets say 5 days from arrival to the receptor. We believe that this allows the simulations to equilibrate to an environment closer to the receptor, thus producing a more accurate result compared to initializing the model runs with an anyway poorly known initial concentration of gases and particles. Prior submission, we have experimented with the use of trajectories of different length. We will add to section 3.3.4 tests to show the effect of trajectory run length. In these runs we have shortened the length of the trajectory to 120 hours, i.e. 5 days. The model runs along these trajectories were initialized with corresponding aerosol size distribution and initial gas phase concentrations.

Second major concern relates to BL transport. The referee question how long the trajectories actually spend in the boundary layer. This since if isolated particles as defined by the trajectories spend much time away from sources in the free troposphere,

they likely not represent boundary layer transport and processing very well. We agree that this may be the case. However, our modeled box consist of two compartments, i.e. mixing layer (ML) between surface and a calculated mixing height, and the residual layer (RL), defined by the volume of air between the top of the mixing layer and the maximum ML-height during the simulation. When performing the model runs, the trajectories describe the movement of this quasi 1-D column along the lat-long coordinates. Thus, we do not claim to simulate the actual small volume as described by the trajectory, but instead a portion of the lower atmosphere (ML and RL) that moves along the coordinates of the trajectory. Nevertheless, this may introduce problem since the movement of air at higher altitudes does necessarily not represent the movement in the lower atmosphere, a fact that may bias both transport time of and sources experienced by the modeled boxes. In the sensitivity test section we perform a test where we divide the simulations in two groups, one that spends above average time in the ML and one that spends less than average time in the ML. Thus, a new section (3.3.4) was added: "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, and this is unfortunately beyond the scope of this study and model framework."

To address the role of the crude gas phase initialization we also added to the MS results from a new run where we altered the initial concentrations of NOx and ozone to lower than base case concentrations. This is now discussed under section 3.2.2: "The results from the runs with lower than base case concentration 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 ∼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 ∼4*10-5 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. "

Specific comments:

Page 15199, line 16: Sentence changed to: "Box models are computational efficient since they since they omit the advection term from the continuity equation, and simulate processing in a flow relative framework. This gives the opportunity to investigate the usually computationally demanding aerosol dynamic processes with a higher level of detail than possible in large scale regional or global models."

Page 15200 line 27: " different transport sectors" changed to "air-masses of different origin".

Page 15201, line 8: "Single particle trajectories" changed to "back trajectories" for clarity. Page 15201, line 17/18: What controls the MLH in the model? Added: "The MLH is calculated by the HYSPLIT4 model along the trajectories and is defined as the height level at which the potential temperature is at least two degrees greater than the minimum potential temperature."

Page 15201, lines 24-26: There is no interaction with the air above the modeled layers. This is clarified in the revised MS: "There are no interactions with the air above the modeled layers in the current set-up."

Page 15202: FNL archive is clarified by adding: "The FNL data is a product of the

Global Data Assimilation System (GDAS), which uses the Global spectral Medium Range Forecast model (MRF) to assimilate multiple sources of measured data and forecast meteorology"

Page 15202, line 13/14: The origin of the cloud data is declared in section 2.2.

Page 15203, line 24: The statement refers to both terpenes and their products.

Page 15204, line 26: Albedo is a typo. Should be optical depth. This is change in revised MS. Page 15205, line 18: In fact, we do not use oxidation products from isoprene at all in the current setup. This is removed from MS. Page 15205, line 27-29: All anthropogenic NMVOC's are lumped as ethane, based on the mass of NMVOC. Rate constant for NMVOC-OH reaction is that of OH-Ethane. This likely has implications for the OH budget, but we have performed no tests in the current study.

Section 2.3: 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 constants 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). "

Page 15213, line 19-21: Poorer agreement expanded on following lines 19-21: "The sometimes poor agreement may be due to wrong description of cloudiness, inaccurate transport paths of simulated boxes or wrong representation of sources to mention a few possible causes." Page 15218, line 23: "With respect to the general shape and magnitude, the modeled and measured size distributions agree to a large degree" replaced with "Both modeled and observed aerosol number size distributions share similar shape and magnitude, both showing a high concentration of nuclei mode particles as well as similar concentration of accumulation mode particles." Page 15218, line 25: This could be due to many things: to small emissions, wrong representation of saturation vapor pressure, wrong concentration of oxidants, to mention a few possible causes. Further investigation is not within the scope of this study. Page 1219,

line 9: ". . .largely captures. . ." changed to "is often able to capture" Page 15220: The original idea was to show the average agreement between modeled and measured gas phase concentrations. We agree seasonality is more interesting to look at. However, in Figures 14-15, the interested reader may find information about the seasonality of the trace gases. Page 15221: Modeled concentration of monoterpenes is low: We add to this paragraph: "Furthermore, we cannot exclude the possibility of stronger sinks due to higher than actual abundance of OH radicals. " 15224, line 28: "without a doubt" replaced by "possibly" Page 15226, line 4: We agree. We rephrase according to: "This is the most probable parameter to be of importance during initialization of the model since gaseous components such as SO2 have relatively short lifetimes in relation to particles. We choose not to change the initial concentrations of gases like CO and CH4, although these compounds may affect the concentration of oxidants. However, our main interest is to study the effect of the initial size distribution representation." Page 15227, line 12: should be aerosol dynamic process and this is clarified in revised MS. Page 15227, line 20: We agree that this is not well formulated. Reformulated as: "The explanation for this remains open, but it may relate to a more complicated meteorology that is not captured by the trajectory model used, e.g. stratification of the lower atmosphere. It could also relate to a seasonality of the sources that is not well captured by the emission module." Page 15227, line 21: sentence rephrased as: "Measured and modeled accumulation mode number concentrations are similar with respect to both magnitude and seasonal trends." Page 15228, line 1: Changed to: "The model, however, seems to overestimate the Aitken mode number concentration." Page 15229, line 19: Sentence changed to: "The model is easily adoptable to different sites and locations" Table 4: This is corrected accordingly. Figure 1, 7, 16: This information is added as second frame to the figures. Figure 7 & 16: Although this is a good suggestion, simply plotting the sigma 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.

Technical comments:

All technical comments are adjusted according to the suggestion made by the referee.

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