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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.

Anonymous Referee #2

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This paper presents a description of a new Lagrangian model of aerosol microphysics and gas-phase photochemistry, its evaluation against surface observations from a Scandinavian forest observatory, and model-based sensitivity studies to quantify processes contributing to size-resolved aerosol abundances at the site. The development of a Lagrangian tool to simulate aerosol processing is a worthy undertaking and will likely be of great interest to the community. The application of the Lagrangian model framework to aerosol processing in the troposphere is also somewhat novel, and is certainly a suitable topic for ACP. However, there are some shortcomings in the formulation of the model and its application which I feel must be addressed before the paper is accepted for publication in ACP. In particular, the treatment of the gas-phase chemistry

and assumptions regarding mixing (or lack of) and initial conditions are not adequate and may have significant impacts on model results. The paper is very detailed and thorough, but contains several overly qualitative and subjective statements and some unqualified speculation regarding impacts of different processes and assumptions on model performance.

Below I first outline my scientific concerns, followed by a detailed list of recommended typographical / technical corrections.

Major comments

For trajectories of 4-5 days in length, Lagrangian models are generally able to reproduce tracer structure in the troposphere, despite the fact that each air mass trajectory remains isolated. As trajectories become longer, the assumption that air masses remain isolated from others surrounding them as they are advected becomes less applicable, and unrealistically strong gradients are retained in the modelled tracer time series at the trajectory arrival location. The use of 9-day tajectories in this study is likely too long to neglect exchange between the simulated air masses and surrounding air. It is certainly not obvious that 9-day trajectories are justified for all transport events encountered over during a year at the study site. In addition, isolation of the air masses can produce a strong sensitivity to initial conditions used. This is particularly the case for longer-lived tracers. The inclusion of emissions in the model does allow species to be affected by processes external to the trajectory, and in the BL is likely sufficient for modelling the evolution of the shorter lived species (e.g. BVOCs). However, away from the surface and for longer lived tracers the modelled evolution is likely to be unrealistic without mixing processes (see e.g. Lagrangian modelling of CO in Real et al., 2007). Similarly, away from emissions, model concentrations of some constituents may become unrealistically small over 9 days without exchange with surrounding air. Have the authors experimented with different trajectory lengths to demonstrate that the 9 day advection does not produce such problems?

- It is not clear from the paper to what extent the modelled air masses are advected within the BL during their transport to the site, or how much of their time is spent in the free troposphere. If much of the time is spent aloft before descent to the surface, the lack of mixing and assumptions regarding initial conditions will have strong impacts on the modelled concentrations, again due to possibly unrealistic isolation of the air masses. It would be useful to show the time/height profiles of e.g. the cluster trajectories in addition to the maps.

- The tests of sensitivity to initial aerosol distributions suggest that initial conditions may bias the model results significantly. Since most of the gas-phase species are initialised as constant values, their assumed initial concentrations are also likely to impact the final model results, since they control the oxidants important for aerosol processing. Assuming a single ozone and single NOx concentration throughout the year and across all trajectory origins seems inadequate. Again, air masses e.g. descending from the mid/upper troposphere will likely contain far more than 35ppbv ozone, with implications for OH, H2O2 etc.

The authors must do a better job to demonstrate that the above problems do not bias their model results, and to better to account for these issues if the model does demonstrate strong sensitivity to them. This could perhaps be investigated in an extension to the existing sensitivity tests part of the manuscript.

Specific comments

Page 15199, Line 16: Box models are efficient since they omit the advection term from the continuity equation, and simulate processing in a flow-relative framework. They often don't consider dispersion or diffusion, although some do diffuse with assumed surroundings.

Page 15199, Line 27: 'different transport sectors' may be confusing. Could say 'abundance of particles associated with different air mass origins'.

Page 15201, line 8: The word 'particle' used to refer to Lagrangian modelled air mass may be confusing, since particle may also refer to aerosol particle. Consider re-phrasing this.

Page 15201, Lines 17/18 - What controls the mixing height variation in the model?

Page 15201, Lines 24/-26. Is there no interaction with the large-scale free troposphere above the model layers? e.g. exchange with 'background' concentrations of long-lived species or background aerosol? If this is the case it should be made clear here. This is also related to my major point on mixing above.

Page 15202, Line 1. Please define 'FNL'.

Page 15202, Line 13/14. Please describe where cloudiness data is taken from (e.g. satellite, model climatology?).

Page 15203,line 24: Does this refer to deposition of the terpenes themselves or their oxidation products (e.g. peroxides, carbonyls etc)?

Page 15204, Line 26: 'cloud albedo of 20'. This should presumably be 20% or 0.2? Albedo cannot be greater than 1.

Page 15205, line 18: 0.2%(?) yield for condensables from isoprene oxidation. Please give references for this. Is this value correct?

Page 15205, lines 27-29: Treatment of VOCs. How are the different emitted VOCs lumped into the ethane species - is there any mass adjustment to account for different reactivities? Does the whole NMHC burden of the air mass react with OH with the ethane rate constant? What are the implications of this for the radical budget and OH in the air mass? Does it mean the OH sinks for the non biogenic (emitted) VOCs are likely smaller than is realistic?

Section 2.3 - Clouds. Do the clouds impact photochemical processes in the model? Please make this clear.

Page 15213, Lines 19-21: Model / obs agreement in other cases. It is stated that agreement in this case is good but less good in others. Can this be expanded on, or possible reasons for particularly good agreement here be suggested?

Page 15218, line 23: ".. agree to a large degree." This is subjective. Please quantify the agreement (e.g. model mean bias).

Page 15218, Line 25: Slower Aitken mode growth / possible slow production of condensing gases. Could this be related to a problem with the model oxidants? Could this be investigated further?

Page 15219, Line 6: ".. largely captures.." is again subjective. Please be more quantitative.

Page 15220: Evaluation of trace gas concentrations. What is the justification of comparing annual average obs/model concentrations at the site? Does this a give a useful comparison? The concentrations are likely to show a large degree of seasonal variation and variations between trajectory transport clusters. Can they instead be compared e.g. seasonally for each trajectory cluster? This will likely yield more useful information about periods modelled well and those not so well, and what may control this.

Page 15221, Line 1: Modelled concentrations of terpenes. These are found to be low compared with observations. Does this suggest a possible OH bias in the model? Could this be related to the lumped VOC treatment (see earlier comment)?

Page 15224, line 28: "This is without a doubt..." Please do not use this phrase unless you demonstrate this level of certainty in the analysis (it doesn't appear to be shown).

Page 15226, line 4: "gaseous components have relatively short lifetimes in relation to particles". This is not the case for many gases important for oxidant abundances in the model. e.g. CO and ethane lifetimes are several weeks at typical tropospheric OH concentrations.

Page 15227, line 12: "role of dynamic processes" Should this be "aerosol dynamic processes" to avoid confusion with dynamic processes due to meteorology?

Page 15227, Line 20: Poor model performance due to more complicated meteorology. Isn't the model meteorology driven by the trajectory? Why would this period be more difficult for the model, since the meteorology is always taken offline from the trajectory input? Does this comment refer to periods when the meteorology may not be well represented by the trajectory? If so, please expand on this.

Page 15227, Line 21: "generally quite a good agreement" Please replace this phrase with something more quantitative.

Page 15228, Line 1: "fairly good agreement" Please replace this phrase with something more quantitative.

Page 15229, line 19: "The model is associated with a high level of transparency..." It is not clear what this means. Please rephrase.

Table 4: Caption states that median and 25-75 percentile ranges are shown, however data in table does not show this.

Figures 1, 7, 16: Please also show altitude profile of trajectories.

Figures 7 and 16: It would be more informative to show some spread around the trajectories for each cluster to demonstrate variability in transport within each cluster. Please clarify what is plotted. Do trajectories show mean paths of advection in each cluster, or simply a representative single trajectory from each cluster? Plotting the former would then allow spread to be plotted as +/- 1-sigma from the mean trajectory points.

Typographical /technical corrections:

Page 15198, lines 21/22: Change 'while' to 'during' and omit 'takes place'.

Page 15199, line 6: Omit 'better'.

Page 15199, line 16: 'computational' should be 'computationally'.

Page 15199, line 28: Change 'holds today' to 'has'.

Page 15200, line 28: change 'advection' to 'sources'.

Page 15201, line 3: Change 'Besides on' to 'Aside from'.

Page 15203, line 1: Omit first word 'The'

Page 15205, lines 2/3: Move parenthesis before 'Andersson' to before '2001'.

Page 15211, line 5: 'estimates adopt' should be 'estimate adopts'

Page 15213, line 9: 'events is' should be 'events are'

Throughout: consider changing the empirical coefficient for nucleation 'A' to italic typeface to improve readability of text.

References

E. Real, et al., (2007), Processes influencing ozone levels in Alaskan forest fire plumes during long-range transport over the North Atlantic, J. Geophys. Res., 112, D10S41, doi:10.1029/2006JD007576.

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