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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric Cluster Dynamics Code: a flexible method for solution of the birth-death equations" *by* M. J. McGrath et al.

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Referee 3 has requested clarification on several sentences, and we hope that it will make the manuscript easier to understand.

Section 2.2 contains a detailed discussion of uncertainties concerning ionic-cluster formation but in the result section no results (e.g. sensitivity test of different cluster formation enhancement factors) are presented. Either you should include such results or you should remove the detailed discussion of the model uncertainties concerning ionic-cluster formation.

As mentioned in the responses to Referees 1 and 2, we prefer to retain this section to make future citations simpler, but we have removed references to preliminary results,

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instead just mentioning the current capabilities of the code.

Since this study rely on the results from the Vehkamäki et al., 2011 study this manuscript should at least be submitted before the manuscript can be accepted.

This manuscript has been submitted to the Journal of Chemical Physics.

To me it is not clear how you determined the free energy of formation of clusters. You write that with ACDC you can calculate the free energy with any method, but exactly how do you do it in this study and what are the uncertainties?

All of the free energies used in the results section of this work are taken from the data set of Ortega et al. (2011). The methods are described in detail there, but briefly, they are based on a series of quantum mechanical frequency and energy calculations which are able to reproduce the values of very expensive methods but at a much more reasonable computational cost, allowing them to be used for the larger clusters studied here. Since there seemed to be some confusion over this issue, we have tried to clarify this in the first paragraph of the Results section.

"When running ACDC, the first choice that needs to be made is the source of the thermodynamic data used to calculate the evaporation coefficients by Eq. (3). We have decided to perform all calculations here on the neutral acid/base (where the base can be either dimethylamine (DMA) or ammonia) clusters explored by Ortega et al. (2011). We wanted to include in our model only clusters for which we have commensurable high level quantum chemical data, thus restricting ourselves to clusters with a maximum of four acid and four base molecules. The free energies of formation were taken direction from this manuscript (supplementary information), which computes them using quantum mechanical methods that reproduce high level results to within an average of about 1 kcal/mol while being inexpensive enough to allow their use on large clusters. In the future, we will be able to add larger clusters when quantum chemical data becomes available with increased computer power, or through the use of liquid drop model properties for larger clusters. It should be noted that Nadykto et al. (2011) have

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recently published results on a similar system, which could also be used for this test; however, Ortega et al. (2011) also report results for larger clusters, which reduces the boundary effects. Kurten (2011) has also pointed out that the exchange/correlation density functional used by Nadykto et al. (2011) can significantly underestimate the stability of DMA/sulfuric acid clusters."

I don't exactly understand what you want to say with the sentence which starts on line 14 in the abstract:

"In particular, the temperature and sticking probabilities both have a large impact on all clusters, while the boundary effects (allowing clusters to grow to sizes beyond the largest cluster that the code keeps track of, or forbidding such processes), coagulation sink terms, non-monomer collisions, and monomer concentrations can all have significant effects."

This sentence has received a slight modification already due to comments from referee 2, and we have further modified it to be:

"In particular, changing the temperature had a significant impact on the steady-state concentrations of all clusters, while the boundary effects (allowing clusters to grow to sizes beyond the largest cluster that the code keeps track of, or forbidding such processes), coagulation sink terms, non-monomer collisions, sticking probabilities and monomer concentrations did not show as large of effects under the conditions studied."

Section 2, page 6, line 12-14: What do you mean with the sentence: "However, given that the generation of the equations in ACDC is done by a Perl script, there is not a lot of extra effort required to include these clusters."

Do you mean that it is not a lot of extra effort for the person which uses the code or that the computer time is not significantly altered? I expect that you mean that it is no large effort for the user. Please specify.

We have modified the sentence to be, "However, given that the generation of the equations in ACDC is done by a Perl script, there was only a small amount of programming

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effort required to include these terms, and it requires no special effort for the user."

You mention at several places in the text that you use a simulation time of 50 000 s. Do you then mean model time or actual computer time. I assume that you talk about the model time. Please specify.

This is correct. We have modified the manuscript on page 25276 to reflect this (one instance on page 25277 already included "simulation time").

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