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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Treatment of non-ideality in the multiphase model SPACCIM-Part2: Impacts on the multiphase chemical processing in deliquesced aerosol particles" *by* Ahmad J. Rusumdar et al.

Anonymous Referee #1

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The study presented in this paper aims to quantify the activity coefficients of species dissolved in deliquescent atmospheric particles. The approach is based on the modeling of two continental scenarios (urban, remote) simulating the chemical evolution in various phases (gas – particle - cloud droplets). The study examines the activity coefficients of inorganic (HOx, SOx, H2O2, metals) and organic (C1-C3) species. The effect of the non-ideal behavior of the species on the dynamic evolution of the system is evaluated by comparing the results obtained when the ideal behavior in the aqueous phase is imposed or not. For the scenarios tested, the budget of key species (S(VI),

H2O2, organic acids ...) is substantially modified when the non-ideal behavior of the solution is considered. The results presented are relevant to this journal. The paper is well organized but the writing could likely be more concise. I have a few comments (see below) that the authors might consider for submitting a revised article to ACP.

1. The description of the two scenarios considered in this study is inadequate. The authors refer to an article by Ervens et al. (2003) (page 7, line 24), but I did not find more detailed information in the article cited. Additional information is necessary to characterize the simulations carried out with the box model (initial conditions, emissions, speciation of organic species, deposition, NOx regime, NH3 level, etc.) and to weigh the simulated behavior of the species (e.g. the accumulation of S(VI) presented in fig.3, page 16 and reaching 90 μg/m3). The details of the simulation conditions could for example be added to the additional material.

2. The paper examines the effect of the non-ideality of the solution on the condensed phase reactivity but does not discuss its effect on the gas/particle partitioning, as suggested for example page 12, line 4. No result of the non-ideal behavior is presented on the simulated aerosol mass concentrations, their compositions (e.g. inorganic vs organic), their liquid water content ... Adding a section presenting the effects on the overall aerosol properties (mass concentration, composition, water content in particular) would be useful to evaluate the sensitivity of this non-ideal behavior in models dealing with aerosols formation.

3. The iron activity decreases notably when the non-ideal behavior of the species is considered. The authors show that this decrease in activity is one of the main causes explaining the simulated effects. However, the authors state that the model ignores the "middle range interaction" for Fe and Mg ions, leading to uncertainties about the activity coefficients calculated for these species. This limitation is reminded very regularly in the paper, including in the conclusion of the article (e.g. line 15, page 34). Although clearly mentioned, this uncertainty considerably weakens the conclusions of this study. I think that a test would be necessary to examine the sensitivity of the simulations to

uncertainties about the activity of iron. An educated guess could perhaps be made to estimate the effect of the middle range interactions, e.g. using another TMI as a proxy.

4. Minor comments: The term "flux" is used systematically by the authors to define the rate of processes. I think that using "flux" is inappropriate here and should be replaced by "rate" everywhere in the text and in figures 5, 8, 10 and 12.

The writing of the chemical formula are often difficult to read for organic molecules. For example in Table 4, OH2CHCH2OH would be more easy to read as CH(OH)2CH2OH to represent hydrated glycolaldehyde. Similarly, C2H4COOH2 would be more easy to read as C(O)OHCH2CH2C(O)OH to represent succinic acid, etc ... Furthermore, the writing is sometime misleading, like in figure 8 for OHCCHCHCHO (which I guess is for butenedial CHOCH=CHCHO) or, in figure 10, C(OH)2O2COOH which I cannot identify (a typo?) but understand to be glyoxylic acid from the text. I suggest to use only “conventional” formula in the text, figures and tables.

Page 2, line 20: the word "including" in the context of the sentence is unclear. Change to "leading to"?

Page3, line 16: the paper use both ALWC and ALW. Duplication of 2 similar acronyms could be avoided (ALWC is used only 2 times in paper).

Page 10, line 26-27: One of the 2 "finally" should be removed.

Page 11, line 22: pyruvic acid is not a simple carboxylic acid (i.e. without another functional group), as suggested in the context of the sentence.

Page 12, line 18-20. The sentence seems useless in the context of the paragraph and could be removed.

Page 13, line 8. I don’t understand the meaning of the word "obstruct" in the context of the sentence.

Page 14, line 5-6: the sentence is obvious in the context of the paragraph and could

be removed.

Page 15, line 14-16: The sentence is obvious in the context of the paragraph and could be removed.

Page 19, line 1: the second processing in "processing of Fe(II) processing aqueous" could be removed.

Page 19, line 17-31: The reading of this paragraph is tedious. A figure similar to 5, 8, 10 or 12 could be useful.

Page 21, line 7: units are missing for OH concentration. Number of significant figures should be consistent for the 2 numbers.

Page 24, line 25: I don’t understand "formation-substituted" in the sentence.

Page 26, line 10-32: The reading of this paragraph is tedious. A figure similar to 5, 8, 10 or 12 could be useful.

Page 31, line 8: Typo in "sSect."

Page 31, line 14: it is stated that keto malonic acid is the final C3 oxidation product. Figure 12 (page 32) show however a loss process which is not discussed in the text (and not "explained" from the legend in the figure).

Page 32, figure 12: Only the first 3 processes (dark blue, light blue and pink) can be discerned in the figure. The other processes could be merged in either the "other source" or "other sinks" boxes.

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