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Atmospheric
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Interactive comment on “Effects of uncertainties in the thermodynamic properties of aerosol components in an air quality model – Part I: Treatment of inorganic electrolytes and organic compounds in the condensed phase” *by* S. L. Clegg et al.

Anonymous Referee #2

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This is a good paper. But I have a number of suggestions that I think will make the paper more easily comprehended.

Paragraph 1, Introduction: you state that inorganic models are well established. However, this gives the impression that differences among such models are small, while you show here that they are not. I would add a sentence towards the end of the paragraph which points this out. (See also recent comparison of results from different treatments of Feng and Penner, 2007, J.G.R.).

Page 3, 1st para. 1: you state that you focus on key elements of the activity coefficient treatment in this paper. But you actually calculate uncertainties due to activity coefficients, water content of aerosols associated with the choice of compounds treated, dissociation treatment, and vapor pressure uncertainties. And you partially evaluate uncertainties due to surrogate treatment of the aerosol compounds. You should state where you are going with this paper. Is the idea to evaluate all of these uncertainties and their effects in the model? Or are you going to state which uncertainties are important to reduce? And evaluate the relative importance of each?

Page 3, para. 2: State explicitly that the UCD-CACM model follows the scheme in Fig 1, with a few simplifications, explained below.

Page 4, 1st line para. 3: State that the 2D version is (time, height) or whatever the 2D refers to.

6th line: Are the gas/aerosol partitioning and total amounts of each species ?*at each time*? Given as input to AIM?

Line 12: here you admit that the inorganic components of the models are very different. Need to clarify statement referred to above, so reader understands.

Page 5, 1st full para: Do you mean that particles from different sources are not internally mixed, when you state that they are tracked separately?

Page 6, 1st para: when you say a "single bulk aerosol phase" is assumed do you mean that all sizes are lumped together and an internal mixture assumed?

2nd para: "models may not predict the same amounts of inorganic solids" – this differs from your introduction where you state the inorganic treatments are well-established (and hence well known).

Page 7, last para: Do the differences in calculation of Gibbs free energy differ significantly? You should state whether this is an issue to be explored or not.

Page 8: 1st full para: the UCD CACM model exchanges water dynamically. What about HNO3 and NH4? Are these treated dynamically? How about any of the other species?

Also clarify here again, that the differences in equilibrium vs dynamic behavior are not analyzed, because the focus is on a detailed comparison of a time period which is in equilibrium in the UCD CACM model.

2nd full para. Near end: what "additional terms" are needed? What do you mean by the "extra" liquid phase? The sub-cooled phase associated with hydrophobic components?

Page 8 last paragraph: Later on I was confused because in several places you state that the activity coefficients in both codes are calculated using the UNIFAC method. You should go through the paper, and when you state this, also state that the UDC-CACM model bases its calculations on the properties of single electrolytes, not aqueous ions.

End of page 9: at this point in my reading, I decided that the paper would benefit from a table describing each component calculation for each model, so it was clear what was different in each model. There are so many different components and different procedures, that it is hard to know what is important to retain.

Page 10: middle of page: here you mention that UNIFAC is used in both models for activity coefficients. Should also state that UCD CACM does not calculate these for ions.

Page 11, 3rd paragraph: does the AIM model somehow use UNIFAC to calculate activity coefficients independent of the ions and undissociated organics?

Page 12: 1st para in Section 2.4: Does paper II arrive at a "best" method for calculating vapor pressures?

Page 13: 1st para: "ignore interactions . . . between the TWO inorganic and organic (uncharged) species” – which two are you talking about here??

1^{st} para, section 3.1: It seems to me that the addition of NH3 starts at 13:00, not 18:00. There is a minimum at 18:00, and a max at 17:00 and 19:00.

End of para: How big is P8 compared to other compounds?

Page 14: 1^{st} line: what sizes are bins 6 – 15? Maybe include table?

3^{rd} para: RH is above 60% at times other than midnight to 8 or 9 am. i.e, at 9 am and at 10am. Also, you state that AIM predicts less water between about 11 and 1 pm, do you mean at 12p here? (the RH is 60% at 1:00p).

last para: which model is displayed in Fig 9?

Page 15, 1^{st} line: is Fig 9 the same for the other model?

2^{nd} para: Is Figure 10 at 8am?

PaGE 16, last line, 1^{st} full para: delete sentence.

1^{st} sentence next para: I would start: “in contrast to the simultations for AIM shown in Fig’s 7, 8 (and 9??), in these AIM calculations . . . ”

Last para: here you say that the 2 activity coefficient models result in different predictions of pHNO3 and pNH3 – state again that this is because UCD CACM does not allow dissociation in it’s calculations.

Page 17, 1^{st} full para: The amounts are the same after 9am in the normal AIM model.

Page 18, 1stfull para: “at low RH most of A1-5 would be in the gas phase” But Fig 14b shows that $>$ 80% of A5 and $>$ 95% of A1-4 are aqueous!? So certainly not “most”

Last sentence: need to state again: UCD CACM does not explicitly include calculation of the organic anion activity coefficient.

Page 20, 1^{st} para: here you say that the treatment of organic dissociation does not

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matter – but it was earlier said to be the major effect (page 18, bottom).

1st para, section 3.2.1: why use the model of Nannoolal? Was this determined to be "best " in paper II? Or later in para. you state that for P2 the fraction in aerosol phase is not reasonable. Is this because Nannoolal method is poor?

Page 21: Since the model result for P2 is not reasonable, are you stating here that the Robinson et al. work is not reasonable?

Page 22: last para: "based upon estimates such as those tabulated *here*. Here or paper II?

Page 23: "SOA concentrations roughly double" – can't see this in the figure, which uses different scales on the plots – also need to give the totals for the domain.

"species UR7 and UR17" – what are these?

Page 24, last para: The solution identified here only addresses the differences shown in Fig 2. Is this solution really needed? How do you know?

Page 26: the assigned lower limit for H2O is responsible for large differences, but here you state that this retention is seen in the atmosphere. So is the CACM approach better? "it would be preferabl to directly model metastable aqueous aerosols" – you haven't shown this. What is your reasoning?

Page 27: "than from improved predictive techniques for vapour pressure or activity coefficients" But you state above that the method used in CACM (Mydal and Yalkowsky) is poor.

Should say that paper II shows that "uncertainties in the vapor pressure are a factor of 2, and that this is larger than activity coefficient uncertainties based on calculating ions or undissociated molecules only" – paper 2 makes no assessment of which method for vapor pressure is best?

Page 29: table 4: What does the # column in Table 4 refer to?

Table 1 caption: state that these are in the aqueous and hydrophobic phases and at 8:00am.

Table 4: explain what # is. Group definitions of surrogate compounds and ??#.

Figure 4 caption: add that this is level 1 of UCD results.

Fig. 5 caption: add that this is UCD CACM model level 1 results.

Figur 9: which model is this?

Fig 10: Explain that AIM and UCD-CACM differ if not = 1. Is this at 8:00am?

Fig 21: The scales in the 2 figures need to be the same.

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