

***Interactive comment on* “Composition and properties of atmospheric particles in the eastern Atlantic and impacts on gas phase uptake rates” by J. D. Allan et al.**

J. D. Allan et al.

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Received and published: 17 November 2009

Reviewer: The abstract states “Marine aerosol composition continues to represent a large source of uncertainty in the study of climate and atmospheric chemistry”, and some of these issues are discussed in the Introduction, but these questions are then never explicitly addressed in the manuscript’s Discussion. This really leaves me wondering what was learned from this dataset, and how does it address some of the issues presented in the Introduction? As currently presented I don’t feel this paper will significantly contribute to the field’s understanding of marine aerosol chemistry. For example, the importance of marine sources of organic aerosol are discussed in the Introduction,

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but then barely mentioned in the Discussion. There are several other similar disconnects. In general, the paper could be significantly improved if novel and significant results obtained from this study were more clearly stated and presented, and then summarized in the Conclusions.

Response: As recognised elsewhere in the review, much of this data feeds into other studies as part of the wider RHAMBLE campaign. However, we feel there are a number of aspects of this paper that are novel in themselves. This is the first time that the ability of the ADDEM model to reconcile aerosol composition and subsaturated growth factors and composition has been demonstrated with MBL data. In addition, while the physical size of the modes in the uptake distribution have been characterised in previous studies, this is the first time that they have had compositions assigned through experimental work. The revised text stresses these points more in the introduction and conclusions and cuts back on aspects of the introduction that are not dealt with in this paper.

Reviewer: The final section, regarding the feedbacks between aerosol hygroscopicity and multiphase chemistry had the most potential, but in the end did not seem very well developed or interesting. Why was the uptake of HOI the only compound that was studied using this analysis?

Response: HOI was used for the sake of comparison with the previously published MBL works cited. A much larger work activity is being undertaken as part of RHAMBLE that uses the ambient data presented and treats many other systems other than HOI uptake. However, given the scale of the work, this is seen as outside the scope of this paper. This is mentioned in the revised text as follows: "Other processes can and will be modelled as part of the halogen-aerosol interaction studies within RHAMBLE and will be the subject of future publications."

Reviewer: Some of the findings presented here have been observed in other marine studies. In particular the influence of mineral dust on the heterogeneous chemistry

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and chloride displacement of sea salt (pg. 18349) was observed during the ACE-Asia campaign in a polluted marine air mass (Bates et al., 2004; Sullivan et al., 2007; Tang et al., 2004).

Response: Agreed; the ACE-Asia work is already cited, albeit using a different reference (Topping et al., 2004).

Reviewer: Page 18333, line 17: Marine sources of organics – extensively studied by Kawamura's group, e.g. (Kawamura and Gagosian, 1987; Mochida et al., 2003; Mochida et al., 2007).

Response: References added.

Reviewer: Page 18334, line 15: Particle phase also important for determining reaction pathways and kinetics. Page 18334, line 22: Particle must deliquesce before undergoing hygroscopic growth. Should be some discussion of phase transitions in this section.

Response: Agreed, but it is unlikely that any effloresced particles will ever occur in the MBL, given the high ambient humidity and mixed sulphate/organic composition. This is reflected in the contiguous humidograms reported. These are mentioned in the revised introduction and results sections.

Reviewer: Page 18334, line 24: This muddles the differing roles of solubility and hygroscopicity on a particle's hygroscopic growth, see for example (Hori et al., 2003; Petters and Kreidenweis, 2008; Sullivan et al., 2009).

Response: This section has been reworded as follows: "This describes the effect of two competing factors on the equilibrium water content of a particle; the Raoult (solute) term, which accounts for bulk intermolecular interactions on the water activity, or 'effective' concentration of water and the Kelvin (curvature) term, which accounts for the increase in vapour pressure of water above a curved surface, the dependency on composition manifest in the surface tension of the mixed solution. For subsaturated conditions, the dependency of the growth factor on particle composition is mainly cap-

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tered in the Raoult term, the impact of surface tension becoming important only at higher relative humidities (RH>95 %).”

Reviewer: Page 18342, line 11: Calcite has a very low solubility. You can use this to estimate what mass of total calcium was present in the aerosol based on the fraction of soluble calcium that is measured, for the extreme forms of insoluble calcite, and soluble calcium nitrate.

Response: This calculation would require many arbitrary assumptions be made regarding the exact composition of the dust (in particular, the source mineralogy and the distribution of secondary nitrate between the sea salt and dust particles), the extraction methods used during analysis and the amount of soluble calcite that could be released from the dust matrix during extraction. Given these inherent uncertainties, we feel that this calculation is of questionable merit. The text has been modified slightly to convey this as follows: “but as the impactor analyses capture an indeterminate fraction of the calcium content (dependent on the exact composition and extraction procedure), this can only be taken as a qualitative indicator.”

Response: Page 18342, line 12: Calcium sulfate is not soluble, it is as insoluble as calcium carbonate.

Response: Corrected.

Reviewer: Page 18342, line 22: What about uptake of nitrate (and chloride, and SOA) by mineral dust when it is present?

Response: This is currently mentioned on page 18349, but the text now is modified to say, “the nitrate appears to be associated with coarse particles, which is consistent with sea salt or dust particles taking up nitric acid from the gas phase.” If chloride or SOA were being taken up by dust, this would not be resolvable by any of the instruments deployed here. A mention to this is now given in the discussion on page 18349.

Reviewer: Page 18343, line 25: The mass absorption efficiency of 6.67 m²/g is a bit

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low, Bond & Bergstrom (2006) recommend 7.5 m²/g at 550 nm for uncoated soot.

Response: The appropriate absorption efficiency to use in a given environment is a contentious topic and not one we intend to deal with in this paper. The MAAP reports EBC using its own internal value of 6.67 m²/g and we decided it better to give the values as reported by the instrument and state the value used if a reader wished to modify accordingly. Given that its data is not used for anything more than a qualitative indicator, we did not see the absolute values as critical.

Reviewer: Page 18349, Line 1: Could the greater chloride depletion in submicron sea salt also be attributed to the larger surface area to volume ratio of smaller particles? (Song and Carmichael, 1999) As you are sampling directly above the ocean I would not expect the sea salt particles to have a widely different residence time versus particle size when sampled.

Response: This line of discussion is added in the text.

Reviewer: Page 18349, Line 6: Compare the influence of dust on chloride displacement from sea salt with ACE-Asia, and possibly other dust-influenced marine studies.

Response: We made that particular comment with ACE-Asia in mind. References to this have been added.

Reviewer: Table 1: “(2300)” is repeated.

Response: This mistake does not appear in our source text so must have occurred during production; we will check to make sure it does not appear in the proofs of the revised version.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 18331, 2009.

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