

Interactive comment on “An analytical solution to calculate bulk mole fractions for any number of components in aerosol droplets after considering partitioning to a surface layer” by D. Topping

Dr Topping

david.topping@manchester.ac.uk

Received and published: 13 October 2010

1

I would like to thank referee1 for their constructive comments and suggestions. General and minor points are addressed in the following text with changes made in the original manuscript where required.

This manuscript describes an analytical treatment of the bulk to surface partitioning of surface active components in aerosol particles, effectively reducing the computational

C431

costs of including such a level of detail in the modelling of aerosol properties. The efficiency of the model is compared to other previously reported iterative approaches. The manuscript is clearly written and should be published once the author has had the opportunity to respond to the following comments. The approach, while applicable to multicomponent systems, has been applied to the bulk depletion of binary aerosol containing an organic component and water. The impact of the paper could be greatly improved by extending the application to multi-component aerosol.

Response: I apologise for the lack of transparency with regards to applicability to multicomponent systems. Whilst the original text did refer to the use of the stated equations for both binary and multicomponent mixtures, the revised manuscript specifically has an example illustrating how the bulk to surface ratio of components within a multicomponent mixture vary with wet droplet size. This is included within the revised section (3) which has two new subsections at the request of referee 2 (3.1 Binary mixtures, 3.2 Ternary and higher order mixtures).

Indeed, extending the predictions to a mixed inorganic/organic system would be helpful, demonstrating the application of the model to a system for which, for one of the components, the surface tension is set to increase with increasing concentration, rather than to decrease.

Response: In a previous study Topping et al (2007) assessed the applicability of the mixing rules from which the analytical solution has been presented for mixed inorganic-organic systems. Briefly it was found that measured surface tensions for ternary inorganic-organic systems were adequately captured for a range of concentrations, deviations increasing as the concentration of solutes increased. The successful application of the mixing rules presented in that study formed the basis of the equations presented in this manuscript. In the revised manuscript, in response to the other referees, I have used a specific test case focussed on mixtures of sodium chloride and sodium dodecyl sulfate (SDS) (an anionic surfactant used in many cleaning products). This highlights the capability of the model to replicate well known CCN characteristics

C432

of a common inorganic salt. It must be said however, that since the surface tension of inorganic solutions is quite close to that of pure water, dramatic impacts of partitioning are not seen in this instance.

It should perhaps be made clear that the model considers only the partitioning between bulk dissolution and surface enrichment. For atmospheric aerosol, the ubiquitous presence of inorganic components can significantly reduce the critical micelle concentration. Under these circumstances the importance of partitioning into micelles must be considered.

Response: Indeed, whilst I did try to present a clear description of the exact idealised phenomena captured with the presented equations, the revised manuscript will make reference to specific phenomena that even detailed model descriptions struggle to predict from first principles. Parameterised boundaries of micelle formation, similar to the inclusion of solubility based on empirical data, would be required to adequately capture the impact of micelle formation on, for example, the cloud activation behaviour of the aerosol. The assumptions required to derive a set of equations applicable to multicomponent solution are stated more clearly in the revised manuscript. Whilst the original manuscript did make reference to future developments including 'competing adsorption' at the surface layer, additional developments are suggested following the referees comments.

Discussion of the computational efficiency repeatedly states that the efficiency is less for the analytical treatment. Given that the analytic expression reduces the computational cost of a calculation, this would to me seem to indicate that the model is more efficient. This should be corrected throughout.

Response: Many apologies for the discrepancy. The manuscript has been updated accordingly.

The temperature T in equation 10 appear in the numerator where as it should

C433

appear in the denominator. This error is repeated in equations 11, 13, 18 and 19, although the correct expression is arrived at in equation 20. $\gamma_i B$ all appears in the subscript of the last derivative in equation 14 and this is not correct.

Response: Many apologies, this was a simple error in the latex document and has been corrected. The subscript has been removed.

At the bottom of page 1099, the bulk mole fractions with and without surface partitioning seem to be very similar (1.3512 and 1.3368×10^{-4}) although the text states that only 50 percent of the material remains in the bulk. I do not understand this statement.

Response: Again many apologies for this. The numbers have been removed and changed accordingly.

Powers of 10 should be written as powers of 10. The molar volume of water expressed in equation 29 seems to be incorrect.

Response: Yes, the equation for the molar volume of water has been expressed completely wrong. I'm not sure why that occurred but the error has been removed.

C434