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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Application of the variability-size relationship to atmospheric aerosol studies: estimating aerosol lifetimes and ages" *by* J. Williams et al.

J. Williams et al.

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We are grateful for the helpful comments of both reviewers which have stimulated thought and discussion amongst the authors. To address the points raised clearly we divide them into 4 main issues.

1) The importance of condensation in this region

The main concern raised by both reviewers was that this work underestimates the importance of condensation to particle growth. In this paper we argue that geometrically coherent features in the variability data indicates a common underlying physical process. We have sequentially added coagulation, condensation and nucleation processes to an aerosol model and we note from a comparison of model and measurement, that we do not need condensation in order to reproduce the geometry between

20 and 160 nm in Figure 3a. In order to investigate further the potential effect of condensation we have performed two extra sets of model simulations where the amount of condensable vapours (H2SO4) were fixed between 1x106 to 1x1010 molecules per cm3 throughout the simulation. In one set of simulations no new particles were allowed, in the other set of simulations the new particle production was prescribed at 144 particles per second. These particles where introduced into the smallest size bins between 1 and 2.5 nm diameter. Hence, there are no feedbacks between condensable vapours, new particles or the pre-existing size distribution. The results from these show that when the condensable vapours are increased to a point where the accumulation mode is significantly influenced, this also affects the smaller particles in the Aitken-mode. The conclusion is that simply increasing the amount of condensable vapours and/or changing the temporal variation of the same can not make the calculations fit the observations better. When considering the importance of condensation, the regional environmental conditions must be considered. The paper of Kulmala 1998 was referenced in the review of Laaksonen to exemplify a case where high concentrations of condensable gases prevail. However, our study was made over the remote northern Indian Ocean (ca. 5°N) several hundred kilometres from source regions. Over the forests of Finland there is a large flux of biogenic, reactive and condensable gases, such as terpenes. Such species are no longer present in air that has been transported over the Indian ocean for several days, and as a result the relative importance of condensation for Aitken mode particles in this region can be expected to be less. The above results and discussion will be included in the revised version of the manuscript.

2) Use of more complex models

While reviewer Laaksonen suggests using an ensemble of model runs, we argue that geometrically coherent features in the variability data indicates a common underlying process, and this will hold whether we use one distribution with time or whether we study many distributions. Use of the ensemble approach merely introduces further large uncertainties such as inital conditions, integration time, sources and sinks as a

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function of time and space. Instead of our simplified modelling approach it is in principle feasible to perform this exercise using a 3D model, making a large number of longer simulations to emulate the sampling from aircraft, but this would necessitate a large number of assumptions to be made and is beyond the scope of this study. Reviewer 1 states that our nucleation model is inadequate but gives no indication why making it difficult to address. In general, this paper presents a new way of analysing aerosol measurements with model data in a case study. It is hoped that other groups with more advanced models and datasets from other regions will be interested to examine these effects further.

3) Limits of the integration technique to obtain particle age.

As pointed out by Laakenson, the derived ages must be viewed with caution especially in the boundary layer where there is a high concentration of larger particles and hence surface area for condensation. Newly formed particles in the boundary layer may well be condensing directly onto the available larger particles rather than growing through the particle sizes by coagulation which is assumed in the age derivation. The surprisingly high ages derived for 3 nm particles in the boundary layer indicate that this is indeed probably occuring to some extent. The derived ages can be extected to be more reliable in the free troposphere where the concentrations of large particles are low.

4) Technical corrections

a) Correction of the spelling of Mäkalä has been made. b) 10-3 and 103 should be 10-3 and 103 on page 57, line 13. c) Figures 3a, 3b and 3c will be enlarged in the final version for clarity. (This must be done in collaboration with the EGS for the revised version for ACP).

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 43, 2002.