Atmos. Chem. Phys. Discuss., 2, S30–S31, 2002
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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.

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Received and published: 18 March 2002

The topic of the Williams et al. paper is certainly very interesting to a wide audience as are the results they have obtained. At the same time, the paper remains somewhat speculative. One of the reasons for this is that instead of obtaining the lifetime-variability relationship directly from an ensemble of model runs they simply assume it to hold. They make a limited number of model runs, and make conclusions about the underlying reasons (related to different physical processes) of the observed variability vs. particle diameter relationships based on the lifetimes of particles in the different size bins of the model.

The efforts of the authors to connect obervations and model results are admirable, and

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the paper could be accepted to ACP although the results are not yet strictly proven. However, the conclusions are dependent not only on assumed lifetime-variabiliy relationship, but also of the processes included in the model and the way those processes are described. I agree with Referee#1 that under normal circumstances condensation is a more effective growth mechanism for Aitken mode particles than coagulation. Especially in boundary layer air the concentrations of condensable gases can be expected to be clearly higher than those in the model of Williams et al. Observations of growth rates of particles growing after nucleation events in clean air indicate that concentrations of condensable gases can reach levels on the order of 10ˆ8 molecules per cc (see e.g. Kulmala et al, Tellus 50B:449, 1998), and presumably even higher levels can be reached in polluted air. Note that the (diffusional) condensational growth law predicts that the diameter growth rate is independent of particle diameter itself, and therefore smaller particles will move to the next size bin much more rapidly than larger particles, which could well explain the observations (see end of p. 49 of Williams et al.) Before acceptance of the paper, the authors should perform additional model runs with higher levels of condensable molecules. The exact nature of the condensable species is not important, it can e.g. be described using properties of sulfuric acid. However, it is not necessary to make the production of the condensing molecules dependent on OH concentration since many low vapor pressure species are formed in ozone reactions.

As a minor issue, if coagulation really turns out to be a dominant process for small particle growth, isn't the integration procedure described on p. 60 flawed (because in this case it cannot be assumed that the particle "visits all sizes")?

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

ACPD

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