Atmos. Chem. Phys. Discuss., 1, S181–S184, 2001
www.atmos-chem-phys.org/acpd/1/S181/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Field measurements of hygroscopic properties and state of mixing of nucleation mode particles” *by* M. Väkevä et al.

Anonymous Referee #2

Received and published: 18 January 2002

General Comments This paper summarizes the measurements done by the Helsinki group on the hygroscopic properties of nucleation mode particles. At least two of these data sets have been published elsewhere (Hyytiälä, Mace Head), so the only new data presented here are the urban/suburban (Helsinki, Luukki) studies. Nevertheless, I appreciate the effort made by the authors to compile and summarize their present knowledge of the hygroscopic properties of nucleation mode particles. As far as I know, there are no similar data sets available.

It is interesting to note that freshly nucleated particles at the coastal site were hydrophobic in clean marine air masses, and hygroscopic in polluted continental air. At the forest site, freshly nucleated particles were slightly hygroscopic, similar to that of terpene oxidation products. This shows that the UTDMA can be used to draw conclusions regarding the chemical composition of these particles. Since the particles formed by homogeneous nucleation are ˜1 nm, the UTDMA can not reveal the nature of the nucleating gases, only the compounds that condense on the newly formed particles. Nevertheless, the UTDMA provides insight into the dynamics of tropospheric new particle production.

Even though the UTDMA does not provide the exact chemical composition, the instrument can measure with high time resolution, and therefore serve as a very important complement to other types of single-particle analysis, such as TEM (Transmission Electron Microscopy) and EELS (Electro Energy Loss Spectrometry).

I recommend this paper for publication in ACP.

Specific Comments

The term Şultrafine particlesŤ should be used with some caution, since for instance the epidemiological scientists tend to define ultrafine particles as those with diameters $<$ 100 nm. To be on the safe side, the term should be defined.

Inversion algorithm I recommend that the authors spend some time on acquiring an inversion algorithm for the raw TDMA data, so that all spectra can be inverted properly. The routine by Stratmann appears to be cumbersome to apply in practice, since the authors only used it for a limited number of spectra. What is then the use of such an inversion routine? There are easy-to-use (non-commercial) software products available.

Concentration In many of the tables and figures, the concentrations of particles after DMA2 are given. I assume that this concentration was estimated using the simplified inversion procedure described in section 2.2.

Normally, concentrations at a specific size are given as dN/dlogDp (cm-3) or similar. The important thing is that the number of particles within the narrow size interval selected by a DMA is normalized according to the width of the interval. When this normalization is done, it is then possible to compare directly number concentrations (as a function of size) between instruments using different widths. This is normal practice for

Full Screen / Esc

Print Version

Interactive Discussion

Original Paper

all size distribution measurements.

The concentrations given in this paper donŠt really tell us anything about the concentrations (dN/dlogDp, cm-3) in the atmosphere during the actual measurements. It only provides a picture of the relative variations in concentration during the experiments.

I suggest that the authors do either one of the following:

(1) Express the proportion between the various hygroscopic modes only by giving their relative fractions (as already presented in many of the figures). Use DMPS size distribution measurements to show particle concentrations.

(2) Recalculate the concentrations as dN/dlogDp (cm-3), and plot these instead.

The latter should be fairly easy to do, since the quasi-monodisperse size interval of DMA1 is known, and also the fraction of particles in each hygroscopic mode (taken from the simplified inversion procedure). The particles can be assumed to be singly charged, with a charging probability given by ordinary bipolar charging theory. The efficiency of the CPC 3010 should also be known as a function of particle size. The outcome of this calculation can be validated against DMPS size distribution measurements, which were obviously carried out in parallel (at least at Hyytiälä and Mace Head).

Corrections of field data First, the authors state that Şlaboratory measurements and theoretical calculations do not give exactly the same growth factors for ultrafine ammonium sulphate particles.Ť Fair enough. Then they give the impression that this is the reason why they have to apply a correction factor given by equation (4). In this equation, I fail to see how the theoretical calculations play a role, since only measured growth factor (laboratory and field) data are used. This correction could have been made even without any theory.

I also fail to understand the logic behind the correction factor defined in equation (4). Let us take a simple example. If the ammonium sulphate (AS) growth factor measured in the laboratory (GFlab) is 1.7, and the AS growth factor measured in the field (GFfield) is 1.6, then the correction factor corr defined in equation (4) is 0.7/0.6=1.167. If then this correction factor is multiplied with the observed field growth factor, which for pure a pure AS particle was observed to be 1.6 in our example, then the corrected growth factor would be 1.167Œ1.6=1.867, and not 1.7. All other observed growth factors for atmospheric aerosols would also be Şover-correctedŤ in the same way. Was this the intention? Did I misunderstand the application of this formula?

Hygroscopic modes From the growth factor data presented in the figures, it is sometimes difficult to see the distinction between the various modes of hygroscopic behaviour described in the text (hydrophobic, less-hygroscopic, more hygroscopic, sea salt). For instance for the Helsinki May 1999 data, the growth factors seem to slide up and down in a more or less continuous way. The definition of the modes is likely to be somewhat dependent on the measurement site, and the exact factors (1.05, 1.2, 1.4) separating the various modes should not be taken to strictly.

Figure 5 Ű Hyytiälä Autumn 1999 20 nm data In Figure 5, the concentrations of 20 nm particles appear extremely constant most of the time at 1 cm-3. My own experience is that one rarely finds such stable conditions in atmospheric aerosols. Any comments?

Interactive comment on Atmos. Chem. Phys. Discuss., 1, 379, 2001.

Full Screen / Esc

Print Version

Interactive Discussion

Original Paper