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Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Cloud condensation nuclei measurements in the eastern Mediterranean marine boundary layer: CCN closure and droplet growth kinetics” *by* A. Bougiatioti et al.

Anonymous Referee #1

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The authors present an interesting data set of aerosol size distribution, chemical composition, and CCN concentrations at supersaturations ranging from 0.22% to 0.78% at a remote marine site (Finokalia Station). The aerosols from different air masses were observed and were mostly aged. The major finding of the study is that for aged aerosols, aerosol CCN concentrations can often be efficiently predicted using simplified chemical composition and mixing state (e.g. insoluble organics and internal mixing). This finding reinforces results from previous studies (Dusek et al., 2006; Andreae and Rosenfeld, 2008; Rose et al., 2008a; Wang et al., 2008, Andreae, 2009; Gunthe et al., 2009. Please refer to the references listed in Uli Pöschl’ comment). The authors also show that the aged organic species observed do not significantly affect droplet growth

kinetics.

Overall the presented data are important and the study is nice. The topic is well suited for Atmospheric Chemistry and Physics and I recommend its publication after my comments below have been addressed.

General comments:

The authors show that the measurements can be divided into five periods based on the origin of air masses sampled. The aerosols from different air masses exhibit quite different size distributions (Fig. 6) and chemical compositions (Fig. 7). I would suggest comparing the predicted and measured CCN concentration for each air mass in addition to supersaturation. Can CCN concentrations be efficiently predicted with limited chemical composition for all air masses? Will the closure agreement be better for "cleaner" air masses? It may be useful to compare closure agreements of different air masses to previous studies.

I echo Uli Pöschl’s comment on calibration of CCN counter using NaCl. Due to the shape of NaCl particles, the mobility equivalent diameter can substantially deviate from volume equivalent diameter, which is needed for calculation using Kohler theory. A discussion of the assumption on particle shape, and its impact on calibrated supersaturation will be very helpful.

Specific comments:

Page 10310, line 15: Here the authors describe that both PM1 and PM2.5 were measured. Later on page 10312, it is mentioned that PM10, PM1.3, and PM1.3-10 for PTFE and PM1 and PM10 for quartz filter samples were analyzed. Are these different measurements?

Page 10312, line 15-21: At Qs= 1 Lminˆ-1 and sheath-to-aerosol flow ratio of 10:1, the size range measured by TSI SMPS 3080 is somewhat limited, and is likely much smaller than 20-460 nm. (e.g. the classifying voltage of DMA will be close to 17000 volts for particles of 460 nm).

Page 10313, line 12-13: Similar CCN concentrations at 0.60, 0.76, and 0.87% do not necessarily suggest that most particles activate at 0.8%. The high activation fraction at high supersaturation is more convincing.

Page 10314, line 20-23: Please also show size distribution and values for period D.

Page 10315, line 1-2: Similar PM1/PM2.5 ratios for organics and sulfate do not necessarily mean internally mixed aerosols. Maybe it is more appropriate to change "suggest" to "are consistent with".

Page 10315, line 21: Please clarify what "gregs" stands for.

Page 10315, line 23: Ammonium sulfate accounted for 75% of the total inorganic mass concentration (page 10314, line 26). Therefore the other inorganic components accounted for about 25% of the total mass concentration, and might not be negligible. Does it improve the closure agreement by taking other inorganic species into consideration?

Page 10316, line 6: Please review the number of significant figures.

Page 10316, line 19: Should Vs be Vo?

Page 10317, first paragraph: what is the value of Vo?

Page 10317, line 17: PM10 or PM2.5? For spherical particles with density of $\sim$ 2000 kg/m^3, A mobility diameter of 1.7 micron corresponds to an aerodynamic diameter of $\sim$ 2.5 micron.

Page 10317, line 13-25: The authors suggest that the underprediction at low supersaturation is most likely due to neglecting the soluble organics instead of different composition at Dp50 ($\sim$100 nm at 0.2% supersaturation) from bulk measurement. However, the evidences presented are not very strong. Similar compositions for PM1 and PM10 do not necessarily suggest same composition at 100 nm, as 100 nm particles might

only contribute a small fraction of total mass. Also Fig. 6 shows substantial contributions at 100 nm from both lognormal modes (Sept 21 and Sept 25), which likely had different chemical compositions. The deviation of particle composition at Dp50 from bulk likely contributed (at least partially) to the underprediction. In addition, the deviation of particle chemical composition at small sizes (30-100nm) from that of larger ($\sim$100 nm and above) is suggested as the reason for the overprediction at higher supersaturations.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 10303, 2009.