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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic properties of atmospheric aerosol particles over the Eastern Mediterranean: implications for regional direct radiative forcing under clean and polluted conditions" *by* M. Stock et al.

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Your statement: P25995, L16: You should reference the name of the manufacturer (PermaPure, LLC) rather than the local distributor.

Our response: Thank you for this hint, we replaced the former reference by Perma Pure LLC, Toms River, NJ, USA.

Your statement: P25997, L6: Replace "instable" with "unstable".

Our response: Thank you, this was corrected.

Your statement: P25997, L1-9: How accurately can you determine the RH at 90% (i.e., what is the uncertainty in this measurement)? I understand that you calibrate using salt solutions, but you can get the right answer for the wrong reasons. For example, the salts or water used might not be perfectly pure, and the RH sensor could be off, and yet the calibration might yield an answer that implies both are fine. (...)"

The Vaisala RH sensor has, according to our experience a measurement uncertainty of at least 2% (in RH) at RH $>$ 90%. (The manufacturer indicates 1%) An error of 2 % in RH would propagate into an error in GF of about 7%. If we consider all known measurement uncertainties the uncertainty associated with the derived GF is estimated to be 10%.

Your statement: P26005, L17-22: "However, we admit that uncertainties introduced by assumptions in the model - including spherical shape... were not taken into account (and) consideration of any particle morphology other than a homogeneous mixture is outside the scope of this work." I agree with this assertion. However, one simple test could have been performed that would have told the authors if non-spherical, externally-mixed particles were a major or minor contributor to the Aegean aerosol. This of course is some form of electron microscopy. Do you have archived samples that could be analyzed by SEM or TEM? (Also other remarks on EM).

Our response: In summertime, polluted air masses are associated with N/NE winds. Air masses will travel several hours above the seawater before reaching the station and thus the aerosol is always enriched with sea-water components. For instance, during continentally-pollution period a significant correlation between sulfate and methanesulfonic acid, an indicator of marine source, is always observed (e.g., Bardouki et al., 2003 or Kouvarakis et al., 2002). This observation supports our statement about sphericity.

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Your statement: P26009, L13-14: "The linear fitting was done to match zero." Rephrase.

Our response: Thank you, this was corrected.

Your statement: P26010, L16-19: The refractive index entries in Table 5 are transposed.

Our response: Thank you, this was corrected.

Your statement: P26011, L12-15: The choice of 80% RH for growth factors is not a good one if, as you state, much of the Finokalia aerosol is composed of ammonium sulfate. Ammonium sulfate deliquesces at 81% RH, and if you are off by only a little bit in your RH measurement you may see none, some, or all of the deliquescent growth of the salt at 80% RH. This may explain why you have higher growth factors than observed in the literature. I would do the analysis at 85% if you have an aerosol rich by ammonium sulfate... at least you know the salt has completely deliquesced at that point.

Our response: It probably needs to be pointed out that the aerosol sample was always humidified to more than 90% before the hygroscopic growth factors were measured. This ensures that the particles have a deliquescence history, i.e. they originate from the upper (wet) branch of the hysteresis. This picture also refers to the radiative transfer modelling, where the aerosol is assumed to have been in a state of high RH before the calculations are made. In addition, we have checked individual deliquescence measurements made with the H-TDMA at Finokalia. The deliquescence point of pure ammonium sulphate was just below 80%, deliquescence in ambient aerosols at Finokalia was not observed during these brief measurements above 30% at all, just a gradual hygroscopic growth along with RH.

Your statement: P26016, L1-10: Part of the effect is simply due to particle size and the increased forward scattering (i.e., decreased upscatter -> smaller negative forcing) of

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the larger marine aerosol particles.

Our response: Thank you, this is now mentioned in the text.

Your statement: P26025, T1: Should state explicitly that fsoot is derived from analysis of the SDI stages $<$ 1um. Some readers may think the MAAP was used to generate this.

Our response: Thank you, this was corrected.

Your statement: P26026-P26027: State clearly in the text that these are diameter growth factors. You also discuss scattering growth in the manuscript.

Our response: Thank you, this was corrected.

Your statement: P26037, F8: The lowest two points are essentially the same (i.e., well inside the envelope of their uncertainties). You really should have another point at a smaller particle size to confirm that you have found the peak of this number size distribution.

Our response: Thank you, this is now mentioned in the Figure caption.

Your statement: P26040, F11: Not explicitly stated, but I assume that the measured neph scattering was corrected for angular nonidealities. Is that correct? If so, what method was used?

The truncation error was corrected by a Mie calculation. Truncation error factors had been applied to each calculated scattering angles to simulate the nephelometer's output signals as done in Cheng et al. (2006). The truncation error factors were taken from Anderson et al (1998). This is now mentioned in the text.

Your statement: P26041, F12: Suggests a jump in the lower envelope of data points in the continental A4 aerosol at 80%. Refer to point above about 80% RH and ammonium sulfate.

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As stated above, the aerosol sample was always humidified to RH $>$ 90% before analysis, so deliquescence occurred before the measurement in any case. We are not certain about the statistical significance of the "jump", which you see "in the lower envelope". Mind that there is also a considerable scatterwidth in both data clouds. If this "jump" were due to any sulphate effects, it should be even more prominent in the marine case, where the aerosol consists of an even higher fraction of sulphate (Figure 6). In any case, the growth factors during period A4 (continentally mixed aerosol) are expected to be more complicated to predict than A1.

Your statement: P26042, F13: Can't see the letters (a) and (b) in the figures. May want to consider white letters on the dark background.

Thank you for this suggestion. The Figure will be modified accordingly.

Wolfram Birmili, on behalf of all co-authors.

References

Anderson, T. L., et al. (1996), Performance characteristics of a highsensitivity, three-wavelength, total scatter/back scatter nephelometer, J. Atmos. Oceanic Technol., 13, 967– 986.

Bardouki H., Liakakou H., Economou C., Smolik J., Zdimal V., Eleftheriadis K., Lazaridis M. and N. Mihalopoulos, Chemical Composition of Size Resolved Atmospheric Aerosols in the Eastern Mediterranean During Summer and Winter, Atmos. Environ., 37, 195-208, 2003.

Cheng, Y. F., et al. (2006), Mixing state of elemental carbon and non-light-absorbing aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta of China, J. Geophys. Res., 111, D20204, doi:10.1029/2005JD006929.

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Interactive Comment