

Interactive comment on “Chemical composition and hygroscopic properties of aerosol particles over the Aegean Sea” by S. Bezantakos et al.

Anonymous Referee #2

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This paper discusses aerosol measurements at a ground location on the island of Lemnos and from the two flights of the FAAM BAe-146 over the Aegean Sea between Crete and Lemnos. Particle size distribution and particle hygroscopic growth factors (HGF) were measured at Lemnos using a SMPS and a HTDMA, respectively. Particle composition was measured only on the BAe-146 using an Aerodyne CtofAMS. The measured HGF factors are compared with HGF estimated from the CtofAMS composition measurements during two periods when the 146 was flying relatively close to the Lemnos measurement site. The results of those comparisons suggest that the CtofAMS measurements can be used to estimate the particle HGF in the region. Subsequently the CtofAMS data were processed to produce such HGF for the complete flights. In my opinion, the data analysis does not match the sophistication of the observations.

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Major comments:

1) What is the reason for reducing the size distribution data to modal descriptions, and how does the data shown in Figure 2 contribute to the HGF discussion? I do not see any evidence of how section 3.1.1 contributes to the theme of the paper. There is a brief discussion of apparent nucleation on three days, but those are days when the aircraft was not present. It is insufficient to only present these observations; there needs to be some discussion of their importance. Time series of the unprocessed N70, N90 and N150 concentrations would provide some rough indication of the importance of the measured HGFs, but I do not see any value added to the HGF discussion by Figure 2.

2) Can the application of the Middlebrook et al method to “correct” for the CE of the CtofAMS affect the HGF estimates? In other words, are you increasing one chemical component relative to another by adopting that approach? You need to indicate how the applied method is appropriate for your “mix” of aerosol. The Ptof measurements from the CtofAMS are appropriate to this HGF closure. Why are they not used or discussed?

3) Page 5808, line 10-12 – It can be argued that existing knowledge does allow for reasonable predictions of hygroscopicity. We know kappa values for the major inorganic species, and, as you point out, most organics lie in the 0-0.2 range. If hygroscopicity is “one of the greatest uncertainties” in predicting the role of atmospheric aerosols on climate (I do not believe it is), then it is because we don’t know enough about the hygroscopicity of the organic components. However, you effectively dismiss the organic kappa by setting it to zero in combination with a density that fits; a density at the lower end of the range you quote. In other words, you conclude nothing other than the inorganic components dominate the HGF, a point that has been established many times when organics and inorganics are in comparable fractions. A considerable amount of work has been done in the past four years to document the organic kappa. You could add to that by including as a comparison of a test using an empirical parameterization

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of organic kappa as a function of the level of oxygenation of the aerosol, which can be estimated from the CtofAMS m/z44. Sensitivity to the density estimate is needed as well. I also suggest that you consider recent publications from the Petters group (NCSU) that offer a more fundamental approach to organic kappa. I can not accept your summary statement on lines 14-15 of page 5821; it is possible that it is correct, but you have not proven it.

4) Your kappa value of 1.19 for H₂SO₄ (Table 1) is too high. The value for H₂SO₄ is in the range of 0.68-0.74; see page 5882 of Shantz et al., *Atmos. Chem. Phys.*, 8, 5869–5887, 2008 www.atmos-chem-phys.net/8/5869/2008. Although little H₂SO₄ is evident in your Figure 5, you quote upper limits to your kappa values of 0.93 and 1.03 in your abstract. Also, on page 5823 and in the conclusions you refer to significantly higher H₂SO₄ and acidity overall with increasing altitude.

5) You have the opportunity to show some very useful vertical profile data of chemical components (from the AMS) and derived kappa values. Instead you choose 3D plots that are weak in quantification and are not easy for others to reference. Simple 2D plots of the profile periods will show more clearly the important vertical variation you discuss at the top of page 5823. Also related to the discussion at the top of page 5823, does the temperature structure from the vertical profiles indicate that the sea can be a source of OM higher up in the profile, or is the sea influence contained to lower levels?

Minor comments:

6) Page 5808, line 2-3 – “Scattering and absorption of light by atmospheric particles depends on their size and composition...”. The scattering depends more strongly on size as opposed to composition. The statement may be more defensible for absorption, but strong absorbers in smaller particles will not be efficient.

7) Why do you use a fit for the HTDMA data different from the one used for the SMPS data?

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8) Page 5809, line 1 – indicate the emission is of both primary and secondary particles from natural sources.

9) Page 5809, lines 6-13 – I find no point to this paragraph. There is no summary of the results of the work done by others. It is the results of their work that are important and relevant, not that they did work.

10) Page 5809, line 21 – Needs quantification; something like “Agreement between the HGF measurements from the HTDMA and the HGF estimated from the CtofAMS was within the uncertainty limits of +/-X%.”

11) Page 5810, line 8 – clarify the number of flights.

12) Page 5810, line 16 – “halfway towards” what?

13) Page 5811, line 1 – The “Rosemount” inlet needs some discussion. Is it diffusing? Does its attempt to reduce turbulent flow? Is it forward facing? Is it isokinetic?

14) Page 5811, line 8 – Clarify that the range of diameters is VAD (Vacuum Aerodynamic Diameter).

15) Page 5811, line 22 – how confident are you in your decimal place? Is the SMPS that accurate? Did you adjust for the pressure level of the site?

16) Page 5814, line 17-18 – Growth factors <1 have meaning. They indicate uncertainty in the measurement as well as particles with low hygroscopicity. By removing the 3%, you bias your HGF values and presumably underestimate your uncertainty.

17) Page 5816, line 23 – “in” rather than “at”.

18) Page 5819, line 24-25 – Should this read “1 September”? Fig 5a looks to have more NH₄H₂SO₄ than Fig 5b, and I don’t see any significant H₂SO₄ in either plot.

19) Page 5821, lines 17-18 – This first sentence is unnecessary; it is a repetitious statement of something that has been well known for many years.

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