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Atmospheric Chemistry and Physics Discussions

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

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We thank Reviewer 2 for his/her time and effort. Below are our responses to each of his/her comment.

(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.

We understand the point of the reviewer. We have changed Figure 2 (also in response to comment 4 of Reviewer 1), which now shows the evolution of the 1-h averaged size distributions together with time series of the number concentration of the particles in the nucleation, the Aitken and the accumulation mode. We have also removed the discussion on the nucleation events since those corresponded to days that the aircraft did not fly, as correctly pointed out by the reviewer.

Section 3.1.1 has been updated accordingly. The whole discussion about the size distributions now supports better the HTDMA and the airborne cToF-AMS measurements (cf. response to comment 4 of Reviewer 1 for the details of the updated text).

(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?

The referee implies that the choice of CE in some way affects the proportions of chemical components in the aerosol. This is not the case. The calculation, based on the method of Middlebrook et al. uses the bulk composition to establish the efficiency of collection. Middelbrook et al. also demonstrate how this is effective for a range of composition mixes typical of the ones observed in this study.

The referee also asks why PToF data were not used in this study. The noise in the PToF data is large and as a result statistically significant data delivered as a function of size is not possible from the flight data in any meaningful sense for this analysis. Hence only bulk data are used.

(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. 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 cannot accept your summary statement on lines 14-15 of page 5821; it is possible that it is correct, but you have not proven it.

We thank the reviewer for pointing out the importance of determining the hygroscopic parameters for the organics more accurately. Also in response to comment 10 of reviewer 1, we have revised our analysis to seek more representative values of the hygroscopic parameter and the density of the organic species of the particles that we observed. More specifically, we varied the values of κ_{org}, keeping the ρ_{org} at the lowest possible value (i.e., 1200 kg m^{-3} as indicate from the literature), to the point where we would still obtain an agreement with the HTDMA measurements within less than 5% (i.e., the accuracy of our instrumentation). Then we varied the values of ρ_{org} keeping the value of κ_{org} = 0 so that the agreement between predicted (by the AMS measurements and the ion pairing algorithm) and measured (obtained by the HTDMA) agreed

within 5%. From the ranges of κ_{org} and ρ_{org} values that were obtained in this way for each of the two flights, we used the mean values as representative. In this way we estimated κ_{org} = 0.03 and ρ_{org} = 1300 kg m^{-3} for the first flight, and κ_{org} = 0.1 and ρ_{org} = 1400 kg m^{-3} for the second.

To include the new analysis in the updated manuscript we have modified lines 16-28 in page 5820, and lines 1-15 in page 5821. The new paragraph now reads:

"*For the predicted growth factors shown in Fig. 6 we used the fixed κ and ρ values for the inorganic species shown in Table 1. The values of κ_{org} and ρ_{org} were determined as follows. By keeping ρ_{org} = 1200 kg m^{-3} (i.e., the lowest density of organic species as indicate from the literature; cf. Hallquist et al., 2009), we increase κ_{org} from 0 up to the value that the predicted hygroscopic growth factor agreed with the measured ones within 5%. In a similar manner, we increased ρ_{org} from 1200 kg m^{-3} by keeping κ_{org} = 0. From the resulting ranges of κ_{org} and ρ_{org} we used the mean values as the most representative for each day. Following this procedure we estimated κ_{org} = 0.03 and ρ_{org} =1300 kg m^{-3} for the first flight (1 September), and κ_{org} = 0.1 and ρ_{org} =1400 kg m^{-3} for the second flight (4 September). Evidently, the organic fraction of the particles observed during the closure on 4 September was more hygroscopic and slightly more dense compared to that on 1 September. In either case, the organic fraction of the particles was far less hygroscopic than the inorganic fraction, thereby inhibiting their water uptake.*"

Using the new values of κ_{org} and ρ_{org}, we have also updated the results shown in Figs. 5 and 9 of the manuscript. Regarding the text we also made the following changes:

1. lines 7-10 on page 5824 that originally read:

"*For these calculations we used the hygroscopic parameter and the density of the organic fraction of the particles derived from the closure study (i.e. κ_{org} = 0 and ρ_{org} = 1270 kg m-3; cf. Sect. 3.1.3 and Fig. 6), and assumed that all the samples were internally mixed.*"

are now changed to:

"*For these calculations we used the hygroscopic parameters and the densities of the organic fraction of the particles derived from the closure study (i.e.* κ_{org} *= 0.03,* ρ_{org} *= 1300 kg m-3 for the first flight, and* κ_{org} *= 0.1,* ρ_{org} *= 1400 kg m-3 for the second; cf. Sect. 3.1.3 and Fig. 6), and assumed that all the samples were internally mixed.*"

2. lines 10-11 on page 5824 that original read:

"*For the flight on 1 September (Fig. 9a) the hygroscopic parameter ranged from 0.17 to 1.03 with a median value of 0.28.*"

have been changed to:

"*For the flight on 1 September (Fig. 9a) the hygroscopic parameter ranged from 0.19 to 0.84, with a median value of 0.31.*"

3. lines 25-27 on page 5824 that originally read:

"*In this case, the calculated* κ_{mix} *values exhibited a variability that is smaller (from 0.15 to 0.93), and a median value (ca. 0.30) that is comparable to that calculated for the first flight.*"

have been changed to:

"*In this case, the calculated* κ_{mix} *values exhibited a variability from 0.22 to 0.80 (with a median value of 0.36) that is comparable to that calculated for the first flight.*"

4. lines 28,29 on page 5824 and line 1 on 5825:

"*At altitudes above 2000m (western leg of the flight) the particles exhibit a relatively high hygroscopicity with many points having* κ_{mix} *values greater than 0.8.*"

now read:

"*At altitudes above 2 km (western leg of the flight) the particles exhibit a relatively high hygroscopicity with many points having* κ_{mix} *values greater than 0.5.*"

5. lines 18-20 on page 5825 that originally read:

"*For the closure we assumed that the organic fraction of the particles was totally hydrophobic (i.e.* κ_{org} *= 0), having a density of 1270 kg m-3 that is representative of aged organic species.*"

have been changed to:

"*For the closure we used* κ_{org} *and* ρ_{org} *values determined from the analysis described in section 3.1.3; i.e.,* κ_{org} *= 0.03 and* ρ_{org} *= 1300 kg m*$^{-3}$ *for the first flight, and* κ_{org} *= 0.1 and* ρ_{org} *= 1400 kg m*$^{-3}$ *for the second.*"

6. lines 9-13 on page 5826:

"*Assuming that the organic fraction of the particles was hydrophobic having a density of 1270 kg m-3 (as indicated by the closure study performed when the aircraft flew in the vicinity of the ground station), the cTOF-AMS chemical composition measurements were used to estimate the aerosol single hygroscopic parameter* κ_{mix} *for the entire path of the flights.*"

have been changed to:

"*Using the particle parameters obtained from the closure study when the aircraft flew in the vicinity of the ground station, the cToF-AMS chemical composition measurements were used to estimate the aerosol single hygroscopic parameter* κ_{mix} *for the entire path of the flights.*"

7. lines 13-14 on page 5826 that originally read:

"*Although the median hygroscopic parameter was very similar for both flights (i.e. ca. 0.30), its spatial variability was higher during the first flight.*"

have been changed to:

"*Although the median hygroscopic parameter was not significantly different for the two*

flights (i.e. 0.30 and 0.36 on 1 and 4 September), its spatial variability was higher during the first flight."

9. The caption of Fig. 9 that originally read:

"*Fig. 9. Estimated hygroscopic parameters κmix of aerosol particles observed over the Aegean Sea on 1 (a) and 4 (b) of September 2011. The hygroscopic parameters are calculated using the κ-Köhler theory (Eq. 3) and the chemical composition measurements from the airborne cTOF-AMS. For the calculations we assumed that the particles were internally mixed, and that all the organic species were hydrophobic, i.e. κ_{org} = 0.*"

has been changed to:

"*Fig. 9. Estimated hygroscopic parameters κmix of aerosol particles observed over the Aegean Sea on 1 (a) and 4 (b) September 2011. The hygroscopic parameters are calculated using the κ-Köhler theory (Eq. 3) and the bulk chemical composition measurements from the airborne cToF-AMS. For the calculations we assumed that the particles were internally mixed, had spherical shape and that all the organic species had κ_{org} and ρ_{org} of 0.03 and 1300 kg m^{-3} for the first flight (1 September) and of 0.1 and 1400 kg m^{-3} in the second flight (4 September).*"

(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.

We thank the reviewer for pointing this out. The kappa value of 1.19 for H2SO4 were originally obtained by Petters and Kreidenweis (2007). Following the suggestion of the reviewer, we looked into the literature. It is worthwhile noting that reported kappa val-

ues for different substances differ depending on the instrumentation used. Although, kappa values from CCNCs are somewhat higher compared to those from HTDMAs, we found the opposite for H2SO4. The values reported by Shantz et al., (2008) correspond to CCNC measurements. More relevant (i.e., HTDMA) measurements of the hygroscopicity of H2SO4 particle are reported by Biskos et al. (2009). Using these measurements, the kappa value used in our work for H2SO4 has been changed to 0.97. The results shown in Figure 9 have been updated accordingly, while reference to the revised kappa value for H2SO4 is made in the update of Table 1 of the manuscript.

(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?

We agree with the reviewer that this data set offers a good opportunity to show some vertical profiles. However, we believe that the 3D plots have many advantages (including good depiction of the spatial position and evolution of the measurements) that the 2D plots cannot offer. To make the data easier for the readers to use and reference (as pointed out by the reviewer) we provide all the airborne measurements as supplementary material (cf. files flight_1_data.txt, and flight_2_data.txt). The data in the supplement include the volume fractions of each component as determined by the ion pairing algorithm, together with the predicted kappa values throughout both flights. The data for the ground-based SMPS and HTDMA measurements are also provided (files smps_data.txt and htdma_data.txt, respectively).

The reviewer also asks whether the vertical temperature profiles can induce organic matter from the sea can end up to the higher altitudes. This is well possible. To

address this point we have changed lines 4-8 in page 5823 that original read:

"*Considering that the air masses arriving over Lemnos and the central Aegean Sea during the respective missed approaches have passed over cities and rural areas and that their origin is similar (i.e. from eastern Europe), the organic fraction of the particles can be either biogenic or anthropogenic.*"

to

"*Considering that the air masses arriving over Lemnos and the central Aegean Sea during the respective missed approaches have passed over urban, rural and marine environments, and that their origin is similar (i.e. from eastern Europe), the organic fraction of the particles can be either biogenic or anthropogenic.*"

(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.

We thank the author for pointing out this detail. We have updated the sentence (page 5808 lines 2 to 5) that original read:

"*Scattering and absorption of atmospheric particles strongly depends on their chemical composition, which is often characterised by high variability as a result of the large diversity of their sources and the different physicochemical processes they are involved in during their lifetime (Hallquist et al., 2009).*"

to

"*Scattering and absorption of atmospheric particles strongly depends on their size and chemical composition, which are often characterised by high variability as a result of the large diversity of their sources and the different physicochemical processes they are involved in during their lifetime (Hallquist et al., 2009).*"

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

SMPS fitting algorithms are not suitable for fitting HTDMA data. This is because the first DMA together with the aerosol conditions (in our case the particles humidifier) significantly change the transfer function (i.e., the probability of a particle that enters the first DMA to be detected by the CPC downstream of the second DMA) of the system compared to transfer function of the SMPS configuration which includes only one DMA and the CPC. The algorithm we used (TDMAfit; Stolzenburg and McMurry,1988) has become a standard for inverting HTDMA data. We have further compared the output of the TDMAfit algorithm with that of the HTDMAinv (a newer algorithm for inverting HTDMA measurements) and obtained almost identical results (see response to comment 2 of reviewer 1).

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

The indication according to the suggestion of the reviewer has been made. The original sentence (pages 5808 line 29, and page 5809 lines 1-3) that read:

"*The polluted air masses blend with particles emitted from natural sources, e.g. biogenic marine and vegetation emissions, resulting in increased particle concentrations commonly observed in the region (Salisbury et al., 2003).*"

has been updated to:

"*The polluted air masses blend with natural primary and secondary particles, resulting in increased particle concentrations commonly observed in the region (Salisbury et al., 2003).*"

(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.

We thank the reviewer for this point, which helps make our introduction sharper. We have updated this paragraph to reflect more on the results of previous hygroscopicity measurements at Finokalia station, rather than just on the fact that there have been other studies in the region. The original paragraph that read:

"*Engelhart et al. (2011) performed measurements of the integrated hygroscopicity of particles in the submicron range using a Dry-Ambient Aerosol Size Spectrometer (DAASS), whereas Stock et al. (2011) carried out measurements using a Hygroscopic Tandem Differential Mobility Analyzer (HTDMA) system. Both studies were conducted at Finokalia, on the island of Crete, and reported particle hygroscopicities that correspond to mixtures of organic and inorganic compounds.*"

has been changed to:

"*Stock et al. (2011) carried out measurements using a Hygroscopic Tandem Differential Mobility Analyzer (HTDMA) system, at Finokalia, on the island of Crete. During that study the observed hygroscopic growth factors of particles having dry mobility diameters of 50, 80 and 150 nm ranged from 1.12 to 1.59 when exposed at 90% RH.*"

(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 $\pm$X%.

We have included the uncertainty limits between measured (HTDMA) and predicted growth factors in this sentence. Lines 21-23 in page 5809 that read:

"*Good closure between cTOF-AMS and HTDMA measurements was achieved when the aircraft flew at the vicinity of the ground station.*"

have been changed to:

"*Good closure between cToF-AMS and HTDMA measurements (agreement within $\pm$5% uncertainty) was achieved when the aircraft flew at the vicinity of the ground station.*"

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

Page 5810, lines 8-10 have been modified to provide more clear information about the number of flights conducted by the FAAM BAe-146 aircraft. The original text was:

"*The airborne measurements involved a total of three flights from Crete to Lemnos and back with the FAAM BAe146 aircraft (cf. Tombrou et al., 2012). Detailed paths of the flights performed on 1 and 4 September 2011 are shown in Fig. 1.*"

and in the updated manuscript reads:

"*The airborne measurements involved a total of three flights from Crete to Lemnos and back with the FAAM BAe146 aircraft (cf. Tombrou et al., 2012). The cToF-AMS was operational in only two (i.e., on 1 and on 4 September) of these flights. Detailed paths of these flights are shown in Fig. 1.*"

(12) Page 5810, line 16 – "halfway towards" what?

We have updated the text to avoid confusion here. The original sentence:

"*To capture the vertical variation of the chemical composition of the particles along this path, the aircraft performed two missed approaches: one halfway towards, and one over the island of Lemnos (Tombrou et al., 2012).*"

has now been changed to:

"*To capture the vertical variation of the chemical composition of the particles along this path, the aircraft performed two missed approaches: one over the central Aegean Sea, and one over the island of Lemnos (Tombrou et al., 2012).*"

(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?

The AMS sampled through a 1/4-inch stainless steel tube connected to a modified

Rosemount inlet (Foltescu et al., 1995). This inlet, which is a modified 102E Rosemount thermometer housing, samples aerosol through a forward facing aperture and decelerates it before sampling into a 3/8-inch stainless tube mounted at right angles to the flow. The Rosemount inlet operates sub-isokinetically and, thus, it can enhance aerosol measurements at larger diameters and higher densities. Particles with densities typical of pollution aerosol and less than 600 nm are transmitted with close to unit efficiency (cf. Trembath et al. 2012). To briefly elaborate on some details of the inlet we changed lines 1-2 on page 5811 that originally read:

"*In brief, air was sampled through a Rosemount inlet (Foltescu et al., 1995), mounted on the aircraft fuselage.*"

to:

"*In brief, air was sampled through a Rosemount inlet (a forward-facing, sub-isokinetic inlet with sampling efficiency close to unity for particles* $\leq$ *600 nm; cf. Foltescu et al., 1995 for more details), mounted on the aircraft fuselage.*"

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

Clarification has been made. Lines 7-8 in page 5811 that originally read:

"*The cTOF-AMS can measure particles having diameter in the range from 50 to 700nm (Liu et al., 2007) with a detection limit of ca. 50 ng m-*3."

have been updated to:

"*The cToF-AMS can measure particles having Vacuum Aerodynamic Diameters (VADs) in the range from 50 to 700 nm (Liu et al., 2007) with a detection limit of ca 50 ng* m^{-3}."

(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?

Differences in pressure and temperature are taken into account in the data inversion software (TSI 3034 operators manual, 2003; page 1-2, sect. Applications). The measuring range of the SMPS system we employed is according to the manufacturer 10-487 nm. The measuring range of 10.4 to 469.8 nm (page 5811, line 22) corresponds to the midpoint diameter of the first and the last size bin, respectively. We changed line 22 on page 5811 of the manuscript. The original sentence was:

"*The SMPS system measured the size distribution of the particles having diameters in the range from 10.4 to 469.8 nm, whereas the HTDMA measured the hygroscopicity of the particles having dry diameters from 50 to 170 nm.*"

and now reads:

"*The SMPS system measured the size distribution of the particles having diameters in the range from 10 to 487 nm, whereas the HTDMA measured the hygroscopicity of the particles having dry diameters from 50 to 170 nm.*"

(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.

We agree with the reviewer that growth factors smaller than 1 could be attributed either to uncertainties in the measurements or to intrinsic properties of particles. Regarding the uncertainties of the instrument, during calibration with ammonium sulfate and sodium chloride particles we observed that differences between the sizes of the particles selected by DMA-1 and those measured by DMA-2 were within less than 1%. Apart from that, growth factors < 1 are indicative of non-spherical particles (cf. Biskos et al. 2006) or volatile species. In our analysis we assume that all particles have spherical shape and that they do not contain any volatile species. As individual measurements of volatility or shape were not conducted in this campaign, we exclude all measurements indicating growth factors less than unity. In any case, including all the

samples with hygroscopic growth factors < 1 would affect the average growth factor by less than 1% which is negligible.

(17) Page 5816, line 23 – "in" rather than "at".

The correction has been made.

(18) Page 5819, line 24-25 – Should this read "1 September"? Fig 5a looks to have more NH4HSO4 than Fig 5b, and I don't see any significant H2SO4 in either plot.

Indeed there was a typo in the date. The correction has been made.

(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.

The sentence has been deleted.

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