RESPONSE TO REVIEWERS

RESPONSES TO POINTS RAISED BY REVIEWER #1

We thank the reviewer for his time and effort. His/her comments have been very valuable for improving the quality of our manuscript. Below are our responses to all the points raised.

Point 1: Which is the time resolution and air flow used for the AE51?

The time resolution is 1 second and the air flow is 0.2 liters per minute. This information has now been added in the manuscript.

Point 2: The sun photometer inversion is valid for AOD equal to 0.1?

According to bibliography (Dubovik et al. 2000, Dubovik et al. 2001) retrieval of the particle volume size distribution was demonstrated to be adequate in practically all situations with AOT > 0.05. For low aerosol loading significant errors are induced in the retrieval of the single scattering albedo, the real and imaginary part of the refractive index. This information has been added in Section 2.6 of the manuscript.

Point 3: S1 Which is the original time resolution of the OPC data?

The time resolution of the OPC data is 1 second. This information has been added in Section 2.4 of the updated version of the manuscript.

Point 4: p 2 1 3: instrumental -> fundamental

The change has been applied.

Point 5: p 817: define PBL here

PBL is now defined in the manuscript.

Point 6: Figure S1 why so different panels graphic?

We understand the point of the reviewer here. As a response we have improved Figure S1 in the updated version.

RESPONSES TO POINTS RAISED BY REVIEWER #2

We thank the reviewer for his time and effort. His/her comments have been very valuable for improving the quality of our manuscript. Below are our responses to all the points raised.

Point 1: I would like to see word "derived" in paper title.

The title has been changed to:

"Vertical profiles of aerosol mass concentrations <u>derived</u> by unmanned airborne in-situ and remote sensing instruments during dust events."

Point 2: Article structure seems to be unsuitable for this journal, in my opinion the part related to description of individual cases is a little too long. The paper should rather be about technique then case studies.

The point raised by the reviewer is well taken. The methodology-technique part of the paper was extended by including parts that were previously included in the supplementary material, namely the sections:

- 1. Particle Mass Concentration Calculation from the OPC_a Measurements
- 2. POLIPHON Method Error Estimation

Point 3: Authors show AOT and Angstrom Exponent time-series for two locations. Is it crucial for the presented methods or is it placed just to prove that they compare the same air masses?

We thank the reviewer for this point. Indeed we show this graph to prove that we compare the same air masses. To avoid confusion we added a clarification in an existing sentence in Section 3.1. The updated sentences reads as follows:

"This good correlation was further enhanced during the dust event cases (e.g. on 15 April 2016) when the relative contribution of the aerosol fine mode was minimized, <u>which supports that a comparison of</u> <u>aerosol measurements at these locations is meaningful.</u>"

Point 4: I suggest fitting bimodal distribution to OPC data with boundary condition applied to CIMEL size distribution with zeros at the ends. Maybe it could help.

We understand the point of the reviewer here. We fitted lognormal distributions to the volume size distribution data of the OPC_a and, based on these curves, calculated the mass concentration. The results showed that in Case Study 1 the mass concentration calculated by the lognormal fits is in very good agreement (\pm 5%) with the mass concentration calculated after correction based on the sun-photometer data (as described in the manuscript). However, in Case Study 2 the use of the lognormal fits did not improve the results and the differences between the two methods (lognormal fits and sun-photometer correction) reached up to 30 %.

Point 5: Authors discussed uncertainties caused by refraction index of material used for calibration of OPC and index of real aerosol. Could they discuss uncertainties caused by aerosol shape? I can suppose that non-spherical aerosol may give different signals than spherical one used for calibration.

We thank the reviewer for this comment. Yes, particle morphology can affect the sizing of the particles by OPCs as shown by Osborne et al. (2008). The following text was added in the updated version of the manuscript (see Section 3.2.3) in order to address this point:

"Regarding particle shape, the effect of non-sphericity on the particle sizing by light-scattering instruments having similar scattering angle range that of OPC_a (90°±60°) is within less than 20%, with a tendency towards undersizing (Osborne et al., 2008)."

Point 6: Another question, in the manuscript is stated that authors used constant density (page 4, line 7) whilst in Appendix, section S1 they mentioned dust particle density and non-dust particle density. How was it really calculated, one density for the whole spectrum or different densities for different modes?

We understand the confusion. We used different densities for the fine and coarse modes. However, since we do not show the comparison between the fine mode measurements, this information is redundant and confusing. Thus, we removed the sentence referring to the density of the fine mode in the Section 2.4 that was added in the main text as a response to point 1 of the reviewer (see above).

Point 7: I understand that retrieval of mass concentration from POLIPHON method bases on assumption that coarse mode is dominated by large depolarizing particles. What happens when coarse mode is a mixture of polarizing and non-polarizing particles. In example mixture of dust and maritime aerosols? This is the case in second episode (page 13, line 4). Maybe, in case of second episode, authors should not apply correction to OPC measurements but to POLIPHON retrievals because assumptions of POLIPHON methods seems to be not fulfilled. [... from another comment] The same for Figure 9, trajectories proving that it is dust start at the ground level. What about anthropogenic aerosol from Cairo or Alexandria?

We thank the reviewer for his very good points. His comments made us revisit Case Study 2 and modified the text in Section 3.2.2 accordingly:

"These relatively low δ_{532} values indicate a mixture of Saharan dust with spherical continental/pollution particles. This is supported by the paths that the air mass follow between 1-2 km which originated from north-eastern Africa close to Cairo and Alexandria. The lidar ratio of 40 ± 7 Sr, measured during the previous night (at a height where the signal is mostly free of noise; i.e. 1.2-1.4 km), agrees with the findings of Schuster et al. (2012) and Nisantzi et al. (2015) who reported respectively that $S_{532} = 40 \pm$ 5 Sr and $S_{532} = 47$ Sr for dust originating from eastern Sahara."

Point 8: In general Figures containing LIDAR signals, LIDAR quicklooks and figures containing mass concentrations have different vertical scales. Unification of scales will help in quick comparison of results. I would also like to see time window of both LIDAR measurement, for LIDAR ratio and mass concentration estimation, as well as for UAV flight.

Case Study I: The vertical scales of the quicklooks and the lidar retrievals are now the same (maximum altitude 8 km) and time windows were added to the lidar quicklook indicating the time spans of the Raman retrievals and the UAV flight (Figure 5 of the revised manuscript).

Case Study II: The plots were updated in a similar manner as in the previous case but with a maximum altitude of 3 km (Figure 8 of the revised manuscript).

We prefer to keep different vertical scales for the two case studies as we think that in that way it is more clear for the reader to grasp the atmospheric conditions during our measurements.

For the mass concentration profiles we used the maximum altitude that the UAVs flew (Figure 11).

Point 9: What are colors in Figure 6. I can suppose that are altitudes of trajectory endpoints, it is not defined.

The different colors are indicative of the altitudes of the different back-trajectories starting points. A legend was added to the plot for clarification.

Point 10: Authors estimated LIDAR ratio (LR) around midnight. It is OK for Raman measurements. However during LR measurements and UAV flight different depolarization ratios are observed? Is it really the same aerosol? Increase of depolarization ratio (Figure 5) may suggest some changes. Could you please comment that.

We thank the reviewer for this insightful comment. Indeed Fig. 5 shows that the depolarization ratio increases from 25% to 30% between the time of the Raman measurements and the time of the UAV flight (red rectangles). This implies that the dust layer initially was not pure but slightly mixed with another aerosol type, which explains why our Raman LR retrieval was 47 sr. The back-trajectory analysis showed that during both measurements the air masses arriving over our site, originated from the same region. Thus, we can safely assume that the dust particles originated from the same source.

Consequently, it is possible that the mean dust lidar ratio that was measured during night (Sd = 47 ± 10 Sr) is valid and representative also for the day-time observations. However, former studies of Saharan dust in the region around Cyrpus (e.g. Nisantzi et al., 2015, Mamouri et al., 2016) report Saharan dust lidar ratios of Sd = 53 ± 6 Sr. Taking into account the lidar ratio uncertainties (±10 Sr), the Raman measured value of 47 Sr can therefore be safely used in the POLIPHON retrievals.

Point 11: A few comments regarding trajectory analysis. I suppose that Figure 6 should prove that air mass originates from northern Africa. However, it passes over southern Italy, Greece and Turkey. Could you please comment possible influence of anthropogenic aerosol on your results. Height of trajectory may be large enough not to capture anthropogenic aerosols. However, it is not clear from the figure and text.

The point of the reviewer is very well taken. Indeed, the altitude of the trajectory is too high (above the PBL) to have any direct influences from ground sources over Italy, Greece and Turkey. To make this clear we have clarified the discussion regarding the backtrajectories in Section 3.2.1:

"Backtrajectory analysis (cf. Figure 9) corroborated that this layer resulted from a Saharan dust event that originated in Algeria and traveled over Italy, Greece and Turkey before reaching Cyprus. Despite passing over polluted areas, the core of the dust layer remained pure (2.5-4 km, see analysis below) due to it high elevation (>2 km) throughout its path."

Point 12: I would like to see discussion of uncertainties induced by shape of particles (OPC) and contribution of only polarizing particles to coarse mode (POLIPHON). It is mentioned but not discussed.

These two issues have been addressed in our responses of points 5 and 7 of the reviewer (see above).

Point 13: Corrections of OPC mass concentration by POLIPHON mass concentration and integrated volume size distributions makes sense when authors are sure that POLIPHON works well. In my opinion in second case study POLIPHON assumption is not fulfilled. That's why OPC correction is so large. I would rather extrapolate somehow (for example by fitting bimodal function) OPC size distribution and then compare OPC results with POLIPHON one.

This point has already been addressed above (see responses to points 4 and 7).

Point 14: Statements that values are within error bars and provision of pretty large correlation coefficients looks great but could you give more sophisticated statistical analysis? In my opinion it is necessary especially in case of measurements taken in different locations. Could you provide tests for mean values or for distribution of mass concentration. It may be done for whole population or for different altitude ranges.

The reviewer has a good point here. We tested the differences between the means with a student t-test which in both cases showed that the difference between the in-situ and the remote sensing measurements is not statistically significant. The results of the tests are shown below. In order for our hypothesis (that the means of the two measurements are not statistically significant) to be valid the p values should be higher than the significance level 0.05. The results of the tests were added in the text in Section 3.2.3:

'Further statistical analysis between the lidar and the corrected OPC_a measurements showed that our hypothesis that the two observations refer to the same aerosol population is valid. To be more specific, the two-tailed T-test yielded a P-value of 0.70 (assuming equal variances), indicating that the differences between the mean values of the two types of observations are not statistically significant."

"Also in this case, the two-tailed T-test (assuming equal variances) yielded a P-value of 0.05 indicating marginal statistically insignificant differences between the means of the two types of measurements."

15/04/2016	in-situ	lidar
Mean	61.6794732	63.1364067
Variance	667.09617	347.776601
Observations	34	181
Pooled Variance Hypothesized Mean	397.248647	
Difference	0	
df	213	
t Stat	-0.3910822	
P(T<=t) one-tail	0.34806371	
t Critical one-tail	1.65203888	
P(T<=t) two-tail	0.69612742(>0.05)	
t Critical two-tail	1.97116389	

t-Test: Two-Sample Assuming Equal Variances

The differences between the means are not statistically significant

t-Test: Two-Sample Assuming Equal Variances

22/04/2016	in-situ	lidar
Mean	20.1101707	24.95662895
Variance	135.155273	100.7737154
Observations	19	167
Pooled Variance	104.137129	
Hypothesized Mean Difference	0	
df	184	
t Stat	-1.9615539	
P(T<=t) one-tail	0.02566152	
t Critical one-tail	1.65317709	
P(T<=t) two-tail	0.05132305 (>0.05)	
t Critical two-tail	1.97294054	

The differences between the means are not statistically significant

References

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Vertical profiles of aerosol mass concentrations observed during dust events concentration derived by unmanned airborne in-situ and remote sensing instruments <u>during dust events</u>.

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Abstract. In-situ measurements using Unmanned Aerial Vehicles (UAVs) and remote sensing observations can independently provide dense vertically-resolved measurements of atmospheric aerosols; information which is highly required in climate models. In both cases, inverting the recorded signals to useful information requires assumptions and constraints, and this can make the comparison of the results difficult. Here we compare, for the first time, vertical profiles of the aerosol mass concentration

- 5 derived from Light Detection And Ranging (iidarLIDAR) observations and in-situ measurements using an Optical Particle Counter (OPC) onboard a UAV during moderate and weak Saharan dust episodes. Agreement between the two measurement methods was within experimental uncertainty for the <u>coarse-mode coarse mode</u> (i.e., particles having radii > 0.5 μ m) where the properties of dust particles can be assumed with good accuracy. This result proves that the two techniques can be used interchangeably for determining the vertical profiles of the aerosol concentrations, bringing them a step closer towards their
- 10 systematic exploitation in climate models.

1 Introduction

Aerosol particles affect the atmospheric energy balance directly by interacting with solar radiation, and indirectly through the formation of clouds (Lohmann and Feichter, 2004). Determining the radiative forcing of the atmospheric aerosol particles is

highly uncertain partly because of the significant spatial (both vertically and horizontally) and temporal variability of their concentration, size, and chemical composition (IPCC, 2013). The vertical variability in the properties of the atmospheric aerosol can be independently determined by modern in-situ measurements using airborne platforms and remote-sensing observations. Comparison of the measurements obtained by these two types of techniques, however, is <u>instrumental fundamental</u> for improv-

5 ing the accuracy of the resulting observational data for use in climate models.

Light Detection and And Ranging (LIDAR) instruments are among the most powerful tools for probing vertically-resolved properties of the atmospheric aerosol. A number of retrieval algorithms that have been developed over the years can be used to obtain aerosol optical parameters from the lidar LIDAR raw signals, including the aerosol backscatter coefficient β_{aer}

- 10 (Klett, 1981; Fernald, 1984), the aerosol extinction coefficient α_{aer} (Ansmann et al., 1990, 1992), and the particle depolarization ratio δ^p (Freudenthaler et al., 2009). Under certain assumptions, recently developed algorithms can now be used to retrieve other vertically resolved aerosol properties such as particle absorption and mass concentration using the synergy of lidar and sunphotometer (Ansmann et al., 2011; Lopatin et al., 2013; Chaikovsky et al., 2016)LIDAR and sunphotometer (Ansmann et al., 2011; Lopatin et al., 2013; Chaikovsky et al., 2016). To check the validity of these assumptions and to as-
- 15 sure the quality of the final data, certain aerosol properties retrieved from lidar_LIDAR observations have been compared with vertical in-situ observations using research aircraft (Feingold and Morley, 2003; Weinzierl et al., 2011; Bravo-Aranda et al., 2015; Granados-Muñoz et al., 2016; Rosati et al., 2016; Kokkalis et al., 2017; Tsekeri et al., 2017).

Airborne in-situ measurements using research aircraft are complex and costly, and therefore their availability is scarce and time-restricted, limiting comparability with remote sensing observations. What is more, manned aircraft cannot cover the lowermost part of the atmosphere due to safety restrictions, posing another major limitation. Recent efforts in aerosol instrumentation have provided lightweight and miniaturized instruments that can measure the size and concentration of aerosol particles onboard UAVs (Altstädter et al., 2015; Brady et al., 2016; Renard et al., 2016)

25 a much simpler and cost-effective manner. As a result, vertical profiling of key aerosol parameters can now be performed over long periods of time on a routine basis, and at much lower altitudes compared to measurements with manned research aircraft. Considering, however, that these advantages come in many cases at the expense of the quality of the recorded data, measurements of aerosol properties using miniaturized instruments onboard UAVs need to be validated before using them to bridge the long-lasting gap between in-situ measurements and remote sensing observations.

(Altstädter et al., 2015; Bezantakos et al., 2015; Barmpounis et al., 2016; Brady et al., 2016; Renard et al., 2016; Surawski et al., 2017; Be

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Here we compare, for the first time to our knowledge, vertical profiles of the aerosol mass concentration, derived from <u>lidar_LIDAR</u> measurements using the POlarization <u>LIdar_LIDAR</u> PHOtometer Networking technique (POLIPHON), and insitu measurements with an OPC onboard a UAV (hereafter referred to as OPC_a). It should be noted that the two techniques do not measure the mass concentration directly, but this is estimated from the recorded signals of the two instruments. The measurements were recorded during the BACCHUS-INUIT-ACTRIS (Impact of Biogenic Versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding; Ice Nuclei Research Unit; European Research Infrastructure for the observation of Aerosol, Clouds and Trace gases Research InfraSctructure network) campaign that took place in Cyprus during April 2016.

5 2 Instrumentation and Methods

2.1 Site Description

Cyprus is located in the Eastern Mediterranean (cf. Figure 1 inset), receiving air masses from Europe, the Middle East and North Africa (Lelieveld et al., 2002). Therefore, it represents an ideal location for characterizing different aerosol types and investigating the role of particles in various atmospheric processes.

- 10 The measurements reported here were conducted at three different locations. Aerial measurements, using a UAV, were carried out at Orounda (35°09' N; 33°07' E; 310 m above sea level; a.s.l.), providing highly-resolved spatially and temporally distributed data up to ca. 2 km above ground level (a.g.l.). Ground-based in-situ aerosol measurements, were performed at the Cyprus Atmospheric Observatory (CAO) at Agia Marina-Xyliatou (35°04' N; 33°06' E; 535 m a.s.l.), located 6.5 km south of Orounda. A Polly^{XT} Raman lidar-LIDAR was located at the suburbs of Nicosia (35°14' N; 33°38' E; 190 m a.s.l.), ca.
- 15 35 km east of Orounda, providing round-the-clock measurements of the atmospheric conditions up to 12 km a.g.l.. The exact locations of the measuring points are shown in Figure 1 and detailed descriptions of the instruments are given below.



Figure 1. Map of of Cyprus showing the locations of the observation sites used for the measurements reported in this paper. The inset in the upper-left corner shows the greater area of South Europe, North Africa, and the Middle East, with the white square showing indicating the location of Cyprus. The maps were generated with by Google Earth Pro version 7.1.7.2606 (https://www.google.com/earth/download/gep/agree.html).

2.2 Unmanned Aerial Vehicle (UAV)

The UAV employed during the campaign (cf. Figure 2) has a fixed wingspan of 3.8 m, and is powered by a two-stroke internal combustion engine. It has a take-off weight of 35 kg that results in a payload capacity of approximately 12 kg. The payload bay is $1.3 \text{ m} \times 0.23 \text{ m} \times 0.34 \text{ m}$ (length-width-height), and can fit multiple instruments. When loaded, the UAV can fly for up to 4 hours with an air speed velocity of $25 \pm 10 \text{ m s}^{-1}$ and can reach altitudes up to 4 km a.g.l. (due to airspace limitations, however, only flights up to 2 km were permitted). An autopilot system allowed predetermined flight plans that involved spiral rectangular-shaped ascending and descending patterns (cf. Figure S1 of the Supplementary Material) preventing contamination

5 of the sampling system from the engine's exhaust. For consistency, the results shown in the rest of the paper correspond to measurements during ascends.



Figure 2. Photograph of the UAV of the Cyprus Institute used for the measurements reported in this work.

2.3 UAV-based Optical Particle Counter Measurements

Vertical profiles of the particle size distributions of the atmospheric aerosol were measured using an OPC (i.e., the OPC_a; MetOne, Model 212-2) onboard the UAV, which reported particle size distributions, ranging from 0.15 to 5 μ m in radius, in 8

- 10 <u>size bins</u>. The sampled aerosol was dried to below 50% relative humidity (RH) by gently heating the sampling tube of $OPC_{a\bar{j}}$ which reported particle size distributions, ranging from 0.15 to 5 μ m in radius, in 8 size bins. Assuming spherical shape and constant mass density for the particles, the size distributions were converted into aerosol mass concentrations (see Section S1 in the Supplementary Information2.4). In addition to OPC_a , a single wavelength aethalometer (AethLabs - Model AE51) with a sampling time resolution of 1 second and a flow rate of 0.2 lpm, was onboard the UAV to verify that no contamination of the
- 15 sampled air by the engine exhaust took place. Each instrument was equipped with an individual sampling inlet that extended 5 cm from the UAV nose to ensure representative sampling.

2.4 Lidar Particle Mass Concentration Calculation from the OPC_a Measurements

The mass concentration profiles of the coarse particles were calculated from the size distribution measurements recorded by

- 20 OPC_a. Before converting the aerosol number concentrations to mass concentrations, the OPC measurements were averaged over 30 s (original time resolution 1 s). This was found to be optimal for suppressing a high frequency noise of the OPC raw data and at the same time maintaining a relatively high spatial resolution of ~ 80 m in the vertical direction. The number concentration (dN) of each size bin was converted to volume concentration according to $dV(r) = dN(r)\frac{4}{3}\pi r^3$, where r is the mean radius of each size bin in the recorded measurements. The volume concentration of particles with radii larger that 0.5 µm were summed and multiplied by ρ_d , yielding the coarse mode mass concentration. The variability in the number size
- 5 distributions averaged every 30 s propagated an uncertainty of the order of 10% in the estimated volume size distributions and the mass concentrations of the particles (Taylor, 1997).

2.5 LIDAR Measurements

A depolarization Raman lidar LIDAR Polly^{XT} (Althausen et al., 2009; Engelmann et al., 2016) was used in the measurements reported here. This lidar LIDAR emits laser pulses simultaneously at three wavelengths: 1064, 532 and 355 nm. The laser beam
interacts with the atmospheric molecules and particles, and a part of it (backscattered light) is collected by the receiver unit that consists of two telescopes (near-field and far-field). The elastically backscattered signals are used as input to the Fernald-Klett algorithm (Klett, 1981; Fernald, 1984; Böckmann et al., 2004) to retrieve the vertical profile of the particle backscatter coefficient β_{aer}. This method assumes a linear relationship between the aerosol extinction-to-backscatter ratio constant (i.e., the lidar LIDAR ratio S) throughout the entire atmospheric column; a critical assumption that can induce uncertainties up to 20-30% of the retrieved property from statistical and systematic errors (Bösenberg and Brassington, 1997; Comerón et al.,

2004; Rocadenbosch et al., 2010).

In addition to the elastically backscattered signal, $Polly^{XT}$ receives the nitrogen Raman-shifted signal at wavelengths 387 and 607 nm, and the water vapor Raman signal at 407 nm wavelength. The Raman technique (Ansmann et al., 1992; White-

- 20 man et al., 1992) utilizes the elastic and inelastic signals to retrieve the particle extinction α_{aer} and scattering β_{aer} profiles independently, without any critical assumptions. The range-resolved aerosol lidar LIDAR ratio can then be directly estimated as the ratio α_{aer}/β_{aer} . In our analysis, we used the Raman technique to retrieve the α_{aer} and β_{aer} profiles during night-time, and the Fernald-Klett method during day-time when the Raman signal is highly affected from the background noise induced by the scattered sunlight. The Polly^{XT} system also provides information on volume depolarization ratio δ^v from which the
- 25 particle depolarization ratio δ^p can be estimated (Murayama et al., 1999; Sakai et al., 2000; Shimizu et al., 2004; Sugimoto and Lee, 2006; Freudenthaler et al., 2009). This allows discrimination between spherical particles (e.g., water droplets) and non-spherical particles such as dust.

2.6 Sun/sky Photometer Measurements

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- 30 A lunar/sun sky photometer of the Aerosol Robotic Network (AERONET; Holben et al. 1998) was collocated with the lidar LIDAR at Nicosia, whereas an additional sunphotometer was situated at CAO. Both instruments provided measurements of the Aerosol Optical Thickness (AOT) at seven wavelengths (i.e., 340, 380, 440, 500, 675, 871 and 1020 nm). The AERONET products include, among others, parameters corresponding to the total atmospheric column such as the Ångström exponent Å (at several wavelength pairs), the particle volume size distributions in the size range 0.05 to 15 μ m (particle radius), the fine- and coarse-mode coarse mode AOT (τ_f and τ_c , respectively) at 440, 675, 871, 1020 nm (O'Neill et al., 2003) and the fine- and coarse-mode coarse mode volume concentrations (v_f and v_c , respectively; Dubovik et al., 2000a, 2006). According
- 5 to Dubovik et al. (2000b) and Dubovik et al. (2002) the retrieval of the particle volume size distribution was demonstrated to be adequate in practically all situations with AOT > 0.05, which was also the case for the observations reported here. Cloud screened and quality assured level 2.0 data products were used in this work. The uncertainties of the AOT were < 0.02 for UV wavelengths and < 0.01 for wavelengths above 440 nm (Eck et al., 1999).</p>

10 2.7 Particle Mass Concentration Profiles - The Derived by the LIDAR Measurements and the POLIPHON Method

The mass concentration profiles from the lidar LIDAR measurements were calculated using the POLIPHON method (Ansmann et al., 2011) as stated above. In the first step of the method, the contribution of the fine-mode and coarse-mode coarse mode particles to the total backscatter coefficient (β_t) is calculated based on depolarization measurements (Tesche et al., 2009a). Here we assumed an externally-mixed aerosol consisting of a fine component with low depolarization (5 ± 1%; Ansmann et al., 2011), and a coarse component that induces light depolarization of 31 ± 4% (Freudenthaler et al., 2009), corresponding to dust particles. The dust-related backscatter coefficient was determined as:

$$\beta_d = \beta_t \frac{(\delta_t - \delta_{nd})(1 + \delta_d)}{(\delta_d - \delta_{nd})(1 + \delta_t)} \quad , \tag{1}$$

where δ_t, δ_{nd} and δ_d are respectively the observed total depolarization ratio, the assumed non-dust depolarization ratio and the measured depolarization ratio of dust particles. Once β_d was determined, the non-dust backscatter coefficient was calculated
by β_{nd} = β_t - β_d. In the calculations presented here we used β and δ^p values corresponding to 532 nm wavelength.

In the second step of the method, the mass concentrations of the fine (non-dust; m_{nd}) and coarse (dust; m_d) aerosol fractions are calculated according to (Ansmann et al., 2011):

$$m_d = \rho_d \left(\overline{v_c / \tau_c} \right) \beta_d S_d \tag{2}$$

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$$m_{nd} = \rho_{nd} \left(\overline{v_f / \tau_f} \right) \beta_{nd} S_{nd}$$
, (3)

where ρ is the mass density, whereas the product of the backscattering coefficient and the lidar LIDAR ratio $\beta \cdot S$ is the extinction coefficient of the particles, with subscripts d and nd denoting dust (coarse) and non-dust (fine) particles. It should be noted that the factors $\overline{v_c/\tau_c}$ and $\overline{v_f/\tau_f}$ are used to convert the extinction measurements to particle volume concentration for the coarse and the fine faction, respectively. In this work these factors were determined from the daily mean data of the sunphotometer that was collocated with the lidar LIDAR. The volume concentrations v_f and v_c were obtained from the AERONET data, whereas the fine and coarse mode AOTs, τ_f and τ_c , at 532 nm wavelength, were calculated using Å (determined in the 440-675 wavelength range) according to:

5
$$\tau_{f,c(532)} = \tau_{f,c(440)} \times \left(\frac{440}{532}\right) \overset{\text{A}}{\underbrace{}_{f,c(675-440)}} \overset{\text{A}}{\underbrace{}_{f,c(440-675)}}$$
(4)

Another assumption we made was that the lidar-derived-LIDAR-derived dust and non-dust fractions are identical to the photometer-derived coarse and fine particle fractions. The inflection point of the AERONET data was adopted as the limit between the fine and the coarse-mode coarse mode particles. As a result, the fine mode ranged between 0.05-0.5 μ m (particle radius) and the coarse-mode coarse mode between 0.5-15 μ m as shown in Figure 3. The calculated values of $\overline{v_f/\tau_f}$ and $\overline{v_c/\tau_c}$ (cf. Table 1) are in line with the conversion factors mentioned by Mamouri and Ansmann (2016, 2017) who performed an

10 (cf. Table 1) are in line with the conversion factors mentioned by Mamouri and Ansmann (2016, 2017) who performed a extensive analysis of the conversion factors of dust over Cyprus.

Apart from v/τ , the other parameters required for determining the aerosol mass concentration from the <u>lidar_LIDAR</u> measurements are ρ , β and S. Regarding ρ , we used a density of $\frac{1.5 \pm 0.3 \text{ g cm}^{-3}}{1.5 \pm 0.3 \text{ g cm}^{-3}}$ (Hess et al., 1998) for the fine-mode particles

- 15 and 2.6 ± 0.6 g cm⁻³ for the coarse-mode coarse mode particles (corresponding to dust according to Gasteiger et al., 2011). Chemical analysis of filter samples collected during the measurements showed that the dust density assumed here is valid (data not shown). Values for S_{nd} (60 ± 10 sr) were taken from the literature (Mamouri and Ansmann, 2014), and actual measurements were used for S_d. More specifically, the S_d value d measurements were only possible during night-time when the Raman channels were operating. We measured the same S_d values (47 ± 10 sr) was estimated by night-time Raman measurements
- 20 when pure and dense Saharan dust layers occurred over Nicosia on 15 April 2016 for both events analyzed here (cf. Figure S2 of the Supplementary Material). The analysis of the lidar data for the estimation of the S_d along with the error calculations for equations (1) to (3) are given in the Supplementary Information (cf. Sections S2 and S3 in the Supporting Information). The mean uncertainties of β_d and β_{nd} were in the Supporting Information). All the values of the parameters that are required as input for the calculations are summarized in Table 1.

2.8 POLIPHON Method - Error Estimation

5 The uncertainties of β_d and β_{nd} in Equation (1) were calculated using the Monte-Carlo method (Bevington et al., 1993). For each input parameter, we generated 100 normally distributed random numbers. The values provided in Table 1 were used as the mean parameter and the standard deviation of the normal distributions. Then, 100 β_d and β_{nd} values were calculated for each point in the atmospheric column and from these the mean values and the standard deviations (errors) of β_d and β_{nd} were estimated to be 22% and 28%, respectively. All the values of the parameters that are required as input for the calculations are

10 summarized in Table 1-For equations (2) and (3) the uncertainties were calculated analytically using the error propagation law.



Figure 3. Column-integrated volume size distribution measured with the sunphotometer over Nicosia at 06:57 UTC on 15 April 2016. The ranges of particle sizes measured by AERONET sunphotometers, and by the OPC_a are also indicated in the figure.

Table 1. Values of the input parameters used in the POLIPHON algorithm.

Parameter	Symbol	Values
Dust depolarization ratio	δ_d	$31\pm4\%$
Non-dust depolarization ratio	δ_{nd}	$5\pm1\%$
Dust lidar LIDAR ratio	\mathbf{S}_d	$47\pm10~\mathrm{sr}$
Non-dust lidar_LIDAR ratio	\mathbf{S}_{nd}	$60\pm10~{ m sr}$
Dust particle density	$ ho_d$	$2.6 \pm 0.6 \ g \ cm^{-3}$
Non-dust particle density ρ_{nd} 1.5 \pm 0.3 g cm ⁻³ (Hess et al., 1998) Dust conversion factor (15.04.2016)	$\overline{v_c/\tau_c}$	$0.67\pm0.05\times10^{-1}$
Non-dust conversion factor (15.04.2016)	$\overline{v_f/ au_f}$	$0.24\pm0.018\times10$
Dust conversion factor (22.04.2016)	$\overline{v_c/\tau_c}$	$0.81\pm0.04\times10^{-1}$
Non-dust conversion factor (22.04.2016)	$\overline{v_f/ au_f}$	$0.14\pm0.019 imes10$

3 Results and Discussion

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3.1 Homogeneity of Aerosol Measurements Properties over the Measurement Sites

Given the proximity (6.5 km) of the ground (at CAO) and the airborne in-situ observations (at Orounda), as well as the absence of any strong pollution sources in the region, the measurements were considered to correspond to the same air parcel in terms of atmospheric composition. The third measurement location (Nicosia) was situated 35 km away from the airfield. As suggested

from by the comparison of sunphotometer measurements at Nicosia and CAO, however, all locations were influenced affected by the same air masses with minor local influence which was influence from local emissions that were mostly trapped within the PBLPlanetary Boundary Layer (PBL).

Figure 4 shows the AOT₅₀₀ and the $Å_{440-870}$ measured by the sunphotometers in Nicosia and at CAO from 13 to 24 April 2016 when concurrent measurements were performed at the two locations. Overall, the temporal variability of these two parameters observed at Nicosia was very similar with the respective measurements at CAO, exhibiting correlations coefficients of 0.89 and 0.87 for AOT₅₀₀ and $Å_{440-870}$, respectively. This good correlation was further enhanced during the dust event cases (e.g., on 15 April 2016), when the relative contribution of the aerosol fine mode was minimized, supporting that a comparison of aerosol measurements at these locations is meaningful. In terms of absolute values, AOT₅₀₀ was 15-50% higher at Nicosia compared to CAO, even during the cases with the dust events, when coarse particles dominated. These higher values at Nicosia are mainly due to the altitudinal difference between the sites (Nicosia is at an altitude of 190 m whereas CAO at 535 m above

<u>sea level</u>) and the contribution of the local aerosol sources to the total aerosol burden. This was further justified by the higher $\mathring{A}_{440-870}$ measurements at Nicosia which signify the presence of small aerosol particles from anthropogenic sources.

3.2 Comparison of the Mass Concentration Measurements

A total of 6 UAV flights with OPC_a onboard were performed during the entire campaign. However, only 2 fulfilled all the necessary requirements for comparison with the <u>lidar_LIDAR</u> observations. Those requirements are that 1) there are simultaneous measurements of <u>lidar_LIDAR</u> and OPC_a , 2) there are cloud-free conditions or clouds are above 7-8 km altitude so that the <u>lidar_LIDAR</u> retrievals can be made, 3) there is enough dust loading, 4) there is availability of AERONET data, and 5) the airborne in-situ measurements were performed before the full development of the PBL. All these requirements were fulfilled during the measurements on 15 April 2016 and 22 April 2016, which are analyzed below.

3.2.1 Case Study I: 15 April 2016

5 The atmospheric situation over South-Eastern Europe on 15 April 2016 was dominated by a high-pressure system resulting in mostly cloud-free conditions over Cyprus. A dust event of moderate intensity was observed, resulting in an average AOT_{500} value of 0.4 over Nisosia and CAO (cf. discussion in section Section 3.1 and Figure 4). Figure 5 shows the lidar-LIDAR



Figure 4. (a) AOT₅₀₀ and (b) $Å_{440-870}$ as measured with the sun-photometers at CAO (blue circles) and Nicosia (red circles) from 13 to 24 April 2016.

time-height display during that day, with :- the upper panel showing the range-corrected signal of the 1064-nm channel, which provides information about the aerosol loading and the presence of clouds, and the lower panel the linear volume depolarization

- 10 ratio δ^v at 532 nmthat can be, which is used to discriminate particles of different shapes that can be indicative of different sources. Throughout the day, high concentrations of aerosol particles were observed even up to ca. 7 km altitude (Figure 5a), with a persistent aerosol layer extending from 2.5 to ca. 7 km. Backtrajectory analysis (cf. Figure 6) corroborated that this layer resulted from a Saharan dust event that originated in Algeria and traveled over Italy, Greece and Turkey before reaching Cyprus. Despite passing over polluted areas, the core of the dust layer remained pure (see analysis below) due to its high
- 15 <u>elevation (>2 km) throughout the path.</u> The δ^v plot (Figure 5b) also shows the temporal evolution of this dust layer. From 00:00-03:00 UTC the dust extends from 2 to 7 km altitude, but later (until 14:00 UTC) it becomes shallower. From the early morning hours (07:00 UTC) to early afternoon (14:00 UTC) when the boundary layer develops, the dust layer is confined above it, reaching an altitude of up to 5 km altitude. After the collapse of the boundary layer, the dust layer starts to descend and finally reaches the ground at 18:00 UTC.
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The cloud-free and time homogeneous atmospheric scene between 07:00 and 07:50 UTC, which overlapped with the time window of the UAV flight, was selected for calculating the parameters of the atmospheric aerosol using the POLIPHON method. The lidar As mentioned earlier, the S_d values used as input in POLIPHON were not measured at the same time window (07:00 and 07:50 UTC), but they were derived from night-time measurements. However, back-trajectory analysis verified that the LIDAR was measuring the same air mass type during these time spans (Fig. 6 and Fig. S3). The LIDAR

profiles of β (355 nm, 532 nm, 1064 nm; retrieved with the Fernald-Klett method; Section 2.5) and δ^p (355 nm, 532 nm), that

were used as input in POLIPHON (532 nm only), are shown in Figure 7a-b. The backscatter signal increased gradually from $1 \text{ Mm}^{-1}\text{sr}^{-1}$ at 1 km (532 nm), reaching a maximum of ca. 2.3 Mm⁻¹sr⁻¹ at 3 km where the dust layer core was. The pure dust layer spanned from ca. 2.5 to 3.8 km ($\frac{\delta^p}{\delta_{-}} \approx \delta_{-}^p \approx 30\pm 2\%$) while below 2 km, the dust was mixed with almost spherical

- 30 particles, probably from the residual layer, as indicated by the relatively low δ^p values ranging between 12% and 30%. Figure 7c shows the POLIPHON-derived dust and non-dust related backscatter coefficients β_d and β_{nd} derived by from Equation (1), and respective uncertainties determined by Monte-Carlo calculations (cf. Section S2 in the Supplementary Information for details 2.8). The backscatter coefficient of the fine-mode particles β_{nd} decreased with altitude, while the dust particles were present even down to 0.7 km. As discussed in 2.5, the lidar LIDAR ratio value used in the Fernald-Klett retrieval and the the
- 35 **lidar_LIDAR** ratio corresponding to the dust particles S_d that is required as input in the POLIPHON algorithm, were estimated from Raman lidar measurements that were LIDAR measurements performed between 00:00-01:40 UTC (UTC+3 local time), just before sunrise. It should be noted here that Raman measurements can only be performed during the night because are only possible at night as during the day the scattered sunlight induces high background noise signal. The fact that the dust layer observed during the Raman measurements had the same origin and followed the same atmospheric path before reaching
- 5 the measurement site between 07:00-07:50 UTC was confirmed by back-trajectory analysis (data not shownFig. 6 and Fig. S3).

Vertical profiles of the RH measured with the UAV and predicted by the WRF-ARW atmospheric model (Skamarock and Klemp, 2008) showed that the atmosphere was dry enough (RH ≤ 50%), at the ground level and up to 4 km altitude (Figure 7d). As a result we could safely assume that the aerosol particles were dry and thus changes in the mass density and backscatter
10 coefficient due to water uptake were negligible.

3.2.2 Case Study II: 22 April 2016

Contrary to ease Case Study I, a low intensity dust event (AOT₅₀₀ = 0.1) was recorder recorded over Cyprus on 22 April 2016. The evolution of the boundary layer dominating the atmospheric situation that day is depicted in the lidar LIDAR time-height plots shown in Figure 8. From 00:00 to 10:00 UTC a sparse dust layer extended between 1 and 2 km a.g.l. while after the PBL decay a shallower dust plume was observed between 1 and 1.5 km altitude. According to the back-trajectory analysis (Figure 9) the dust air mass at 1.5 km originated from Egypt at the ground level, then it was elevated and passed over Libya, the Mediterranean and Turkey before reaching Cyprus.

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The UAV flight on that day was performed between 04:22 and 05:16 UTC. The atmospheric scene between 04:20-05:00 UTC (Figure 8) was chosen selected for the comparison due to its stable conditions above 0.8 km. Also in this case, the same procedure as in case I — was followed to retrieve the lidar_LIDAR profiles that were used as input in the POLIPHON algorithm. The backscatter coefficient, the particle depolarization ratio, the POLIPHON-derived dust and non-dust related backscatter coefficients as well as the RH profiles of this atmospheric scene are shown in Figure 10. In contrast to the esti-

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Figure 5. Range-corrected <u>lidar_LIDAR</u> signal at 1064 nm (a) and Volume Linear Depolarization ratio (b) reflecting the atmospheric conditions over Nicosia on 15 April 2016. Blue color indicates weak backscattering, yellow-red colors in the range corrected <u>lidar_LIDAR</u> signal (a) indicate backscattering mainly from fine aerosols and dust, whereas the dotted line shows the <u>Planetary Boundary Layer (PBL</u>) top. The <u>lidar_LIDAR</u> observations used for the comparison with the UAV measurements were those recorded between 07:00-07:50 UTC.

mated δ^p values determined from the measurements on 15 April, here δ_{532}^p decreases gradually with height from 0.8 to 2 km having values between 10-17%. The combination of these These relatively low δ_{532}^p values with the estimated S₅₃₂ value of ≈ 30 Sr (estimated from Raman retrievals between 01:00-03:00 UTC) indicate a mixture of non-spherical dust particles with almost spherical acrosols, possibly of marine origin (Amiridis et al., 2005; Tesche et al., 2009b). Saharan dust with spherical continental/pollution particles. This is supported by the paths that the air mass follow between 1-2 km which originated from north-eastern Africa close to Cairo and Alexandria. The LIDAR ratio of 40 ± 7 Sr, measured during the previous night (at

5 a height where the signal is mostly free of noise; i.e. 1.2-1.4 km), agrees with the findings of Schuster et al. (2012) and Nisantzi et al. (2015) who reported respectively that $S_{532} = 40 \pm 5$ Sr and $S_{532} = 47$ Sr for dust originating from eastern Sahara.

3.2.3 Particle Mass Concentration Profiles



Figure 6. Back-trajectories of the air masses arriving at several altitudes over Cyprus on April 15, 07:00 (UTC). The back-trajectories were calculated for a duration of 5 days using the HYSPLIT transport and dispersion model (Rolph, 2003; Stein et al., 2015) with GDAS 1° meteorological data through the Real-time Environmental Applications and Display sYstem (READY; http://ready.arl.noaa.gov/index.php).



Figure 7. Daytime Day-time profiles of β (355-, 532-, and 1064-nm wavelength) (a) δ^p (355- and 532-nm wavelength) (b) β_d and β_{nd} (c) determined by POLIPHON, as well as RH profiles from in-situ measurements onboard the UAV and from WRF–ARW model simulations over Nicosia at 08:00 UTC (d).

Figure 11Figures 11a and 11c shows the mass concentration profiles for the coarse particles (particles larger than 0.5 μ m in radius) as derived by the lidar LIDAR observations using POLIPHON method for inversion, and the OPC_a measurements. The lidar LIDAR profiles, were calculated by Equations (2) and (3) using the measured β_d and β_{nd} , profiles and the dust density



Figure 8. Range-corrected <u>lidar_LIDAR</u> signal at 1064 nm (a) and Volume Linear Depolarization ratio (b) reflecting the atmospheric conditions over Nicosia on 22 April 2016. <u>In (a), blue Blue</u> color indicates weak backscattering, yellow-red colors in the range corrected LIDAR signal (a) indicate backscattering mainly from fine aerosols and dust. <u>The</u>, whereas the dotted line shows the <u>Planetary Boundary Layer</u> (PBL) top. The <u>lidar_LIDAR</u> observations used for the comparison with the UAV measurements were those recorded between 04:22-05:00 UTC.

values from the literature (cf. Table 1). The respective OPC_a profiles were determined by the recorded particle number size distributions assuming the same dust particle density (cf. Section 2.3-2.4 for details). To ensure that the lider LIDAR observations are representative of the atmospheric aerosol over Orounda and over CAO we compare the data for altitudes higher than 0.8 km a.s.l. during morning hours when the PBL was shallow.

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The mass concentration profiles from the lidar LIDAR and the OPC_a observed on 15 April 2016 (Figure 11a), show a good correlation, with $R^2 = 0.8 = 0.9$. In terms of absolute values, the mass concentrations measured by the OPC_a (red curve) lie within the uncertainty limits (38%) of the lidar LIDAR observations, with the former being equal or lower for the entire range of altitudes, exhibiting a mean bias of -11μ bias ranging from -23.0 to -2.4μ g m⁻³ with a mean of -12.0μ g m⁻³ (Figure 11b). The discrepancies between the two methods can be partly attributed to the assumptions used in POLIPHON: 1) constant S throughout the atmospheric column, 2) contribution in the coarse-mode coarse mode only from depolarizing particles, and

15 3) the assumption of an externally-mixed aerosol. Assumptions used for the manipulation of the OPC measurements that can



Figure 9. Back-trajectories of the air masses arriving at 500 m, 1000 m and 1500 m over Cyprus on April 22, at 04:00 (UTC). on April 22. The back-trajectories were calculated for a duration of 6 days; the black circles indicate the locations where the air-mass was below 100 m altitude.



Figure 10. Daytime Day-time profiles of β (532-, and 1064-nm wavelength) (a) δ^p (355- and 532-nm wavelength) (b) β_d and β_{nd} (c) determined by POLIPHON, as well as RH profiles from WRF–ARW model simulations over Nicosia at 04:00 UTC (d). The 355 nm channel of the lidar LIDAR was discarded due to misalignment.

explain differences between the two methods are mainly related to the refractive index and the shape of the particles. The



Figure 11. Aerosol mass concentration profiles for case study I and II (a, c) and the respective biases (b, d). The In plots (a) and (c), the blue solid lines show represent the mass concentration derived by the POLIPHON. The mass concentration measured by the OPC_a is plotted in red with the red shaded area representing the uncertainties of the in-situ measurement. The green lines show the mass concentration from the OPC_a corrected for the particles losses due to cut-offs. In plots (b) and (d), the red dots show the biases between the values measured by the OPC_a and the LIDAR ($OPC_a - LIDAR$) before applying the corrections, while the green dots are the biases after corrections. The red and green solid lines show the mean biases before and after correction, respectively.

refractive index can notably influence the size distribution measured by the OPC, inducing sizing uncertainties of up to 30% (Rosenberg et al., 2012; Granados-Muñoz et al., 2016). The refractive index used for calibrating OPC_a , however, has a value of for n = 1.59, which is very close to literature values for Saharan dust (n = 1.56; Petzold et al., 2009). The difference between

- 20 the refractive index values used for the calibration of $OPCa_OPC_a$ and that used to for the retreival for the retrieval of the LIDAR measurements is estimated to introduce a bias of 2% to the calculated mass concentration values. Regarding particle shape, the effect of non-sphericity the particle sizing by light-scattering instruments having similar scattering angle range that of OPC_a (90° ± 60°) is within less than 20%, with a tendency towards undersizing (Osborne et al., 2008).
- Another source of the discrepancy between the mass concentrations determined by OPC_a and the <u>lidar LIDAR</u> is the limitation of the former to measure particles larger than a few tens of microns due to aerodynamic inlet loses (sedimentation and inertial deposition), resulting in an underestimation of 20% of the <u>coarse mode coarse mode</u> volume concentration (cf. yellow-green shaded hatched area in Figure 3). To account for that, we corrected the OPC_a measurements using the formula: $m_{OPC} = m_{POLIPHON} \frac{\int_{OPC_a} dV/d\ln r}{\int_{POLIPHON} dV/d\ln r}$. This correction significantly improved the agreement between the OPC_a (green curve in Figure 11a) and the lidar measurements, decreasing the mean bias to -1μ LIDAR measurements, constraining the bias range
- 30 to -11.1 and 8.8 μ g m⁻³ , which results in a decreased mean bias of -1.1μ g m⁻³. Further statistical analysis between the LIDAR and the corrected OPC_a measurements showed that our hypothesis that the two observations refer to the same aerosol population is valid. To be more specific, the two-tailed T-test yielded a P-value of 0.70 (assuming equal variances), indicating that the differences between the mean values of the two types of observations are not statistically significant.
- The mass concentration profiles determined by the lidar LIDAR and the OPC_a measurements on 22 April 2016 (Figure 11bc) also show a good correlation, with R² = 0.72 = 0.9. In terms of absolute values, the mass concentrations determined by the OPC_a measurements (green line) are lower compared to those determined by the lidar LIDAR observations for the entire range of altitudes, exhibiting a mean bias of -11 μbiases in the range from -14.7 to 0.6 μg m⁻³ with a mean value of -8.7 μg m⁻³ (Figure 11d). The integrated volume size distribution measured by the sunphotomer in Nicosia (cf. Figure \$3.54) showed that in this case the OPC_a underestimates the coarse volume fraction by 48%. Upon correction, the mean bias decreases to
- -5μ -1.6μ g m⁻³ and, with the exception of one point at 1.8 km altitude, the mass concentration values from the OPC_a lie within the calculated uncertainty resulting from the POLIPHON algorithm used to invert the lidar LIDAR data (32%). At higher altitudes the mass concentration decreases drastically and OPC_a measurements drop below the POLIPHON uncertainty limits. Also in this case, the two-tailed T-test (assuming equal variances) yielded a P-value of 0.05 indicating marginal statistically
- 10 insignificant differences between the means of the two types of measurements

Overall, the airborne in-situ and lidar LIDAR observations are in good agreement both during the observation of a dense as well as of a weak dust event, after the necessary corrections for the OPC_a measurements. In the case of the moderate dust event the volume concentration fraction that is not captured by the OPC range is small and so is the corresponding correctionis small. In contrast, during the weak dust event, the OPC misses almost 50% of the volume size distribution which introduces

15 small. In contrast, during the weak dust event, the OPC misses almost 50% of the volume size distribution which introduces large measurement ambiguities.

4 Summary and Conclusions

In this study we compare, for the first time to our knowledge, vertical profiles of the aerosol mass concentrations determined independently by an OPC on board onboard a UAV and by remote sensing observations using data from a LIDAR and a sun-

20 pohotometer. The measurements were performed during two cases of dust events that occurred in the region of the Eastern Mediterranean on 15 and 22 April 2016. During those days, the UAV flew up to ca. 2 km altitude with the OPC measuring the size distributions of sampled aerosol particles having radii in the range 0.15-5 μ m, from which the aerosol mass concentrations were-concentration was calculated. The same information was retrieved by concurrent lidar the concurrent LIDAR and sunphotometer measurements that were inverted using the POLIPHON method.

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During the measurements on 15 April 2016 the dense dust layer extended from 2 to 4 km, while a mixture of dust and almost spherical particles was observed below 2 km. The mass concentration of the coarse-mode coarse mode particles increased from 30 μ g m⁻³, at ca. 0.8 km, to ca. 70 μ g m⁻³, at ca. 1.8 km. Agreement between the in-situ measurements and the lidar LIDAR observations retrieved with the POLIPHON method was very good (R² = 0.8 = 0.9), with the in-situ measurements lying within the POLIPHON uncertainty limits (38%), exhibiting a mean bias of $-11 \mu - 12.0 \mu$ g m⁻³ that can be mainly attributed to the difference in the cut-off diameters measured by the two techniques. Corrections applied to account for this difference in

the cut-off diameters further enhanced the agreement, decreasing the mean bias to $-1 \mu - 1.1 \mu g m^{-3}$.

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In the measurements carried out on 22 April 2016, a sparse dust layer was observed between 0.8-2 km altitude during the morning hours. Information from the lidar measurements LIDAR measurements and the backtrajectory analysis suggests that this layer was a mixture of desert dust particles with particles from another source with continental/pollution particles. Despite that, however, agreement between the airborne in-situ and remote sensing measurements in this case was also very good (R = 0.720.9). In terms of absolute values, the corrected mass concentrations measured by the OPC_a airborne OPC were equal or lower than those derived from the lidar LIDAR measurements for the entire range of altitudes and exhibited a mean bias of $-5 \mu - 1.6 \mu \text{g m}^{-3}$. The concentrations measured by the airborne OPC were within the calculated uncertainty of POLIPHON.

The measurements reported here indicate that unmanned airborne OPC measurements and lidar-LIDAR observations can provide reliable ways to determine coarse-mode coarse mode aerosol mass concentration profiles in the atmospheric column, thereby bridging the gap between in-situ and remote sensing observations. Considering that both methods can provide dense datasets in a cost-effective manner and on a regular basis, this finding paves the way towards their systematic exploitation in climate models.

Author contributions statement

15 J.S., A.A. conceived the experiment, E.M., M.P., M.K., C.K., R.E. conducted the experiment(s), D.M. analysed the data, prepared the figures and wrote the manuscript. H.B. applied corrections to lidar LIDAR profiles. G.B. supervised the work and

contributed to the writing of the manuscript. P.K., I.B., A.T., E.M. and V.A. supported the data analysis procedure. All authors reviewed the manuscript.

Data availability. The sun photometer datasets used in this study are available at https://aeronet.gsfc.nasa.gov

20 The OPC datasets analysed during the current study are available from the co-author M.Pikridas (m.pikridas@cyi.ac.cy) on reasonable request.

The Polly^{XT} LIDAR (http://polly.tropos.de) datasets analysed during the current study are available from the co-authors A. Ansmann (albert@tropos.de) and E.Marinou (elmarinou@noa.gr).

Competing interests. The authors declare no competing interests.

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