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Aerosol physical and optical properties in the Eastern Mediterranean Basin, Crete, from Aerosol Robotic Network Data

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Abstract

In this study, we investigate the aerosol optical properties, namely aerosol optical thickness (AOT), Angström parameter ($\alpha_{440-870}$) and size distribution parameters over the Eastern Mediterranean Basin, using spectral measurements from the recently established FORTH (Foundation for Research and Technology-Hellas) AERONET station in Crete, for the two-year period 2003–2004. The location of the FORTH-AERONET station offers a unique opportunity to monitor aerosols from different sources. The AOT is maximum during spring, because of the high dust load transported mainly from African deserts, and minimum in winter. There are secondary maxima in AOT at 870 and 1020 nm in October, attributed to dust transport events occurring in autumn. Large values of AOT at 340 and 500 nm persisting during summer are associated with transport of fine aerosols of urban/industrial and biomass burning origin. The dust events are characterised by a drastic increase in AOT at all wavelengths accompanied by a drastic decrease in Angström parameter to values below 0.3. The mean annual values of AOT_{340}, AOT_{500}, AOT_{870} and $\alpha_{440-870}$, are equal to 0.34±0.14, 0.21±0.11, 0.11±0.09 and 1.17±0.53 respectively. The scatterplots of Angström parameter versus aerosol optical thickness indicate a great variety of aerosol types over the study region including dust, urban-industrial/biomass burning, maritime, as well as mixed aerosol types. This is supported by back-trajectory analyses, and agrees with the measurements of experimental campaigns that took place in Crete during summer. The aerosol volume-size distributions are bimodal over all seasons, with a fine and a coarse mode having effective mean radius of 0.13 μm and 2.12 μm, respectively, and columnar volume concentrations of about 0.038 and 0.061 $\mu m^3/\mu m^2$. There is a general dominance of coarse to fine mode in terms of aerosol volume, in agreement with other maritime locations persisting through the year except for summer. Our analysis shows that the highest values of AOT are related to wind directions from the east, southeast and south, as well as from northwest. Northwestern winds are associated with maximum fine aerosol loads from industrial areas, while eastern, southeastern and southern winds

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are related to maximum coarse aerosol loads, namely sea salt and desert dust.

1 Introduction

Aerosol particles affect the Earth's climate by influencing its radiation energy budget in two ways: i) directly (aerosol direct effect) by scattering and absorbing solar radiation, and ii) indirectly (aerosol indirect effect) by modifying the cloud microphysical properties since they act as cloud condensation nuclei, affecting thus either cloud optical and radiative properties (cloud albedo and optical thickness, first indirect effect) or cloud amount, lifetime and precipitation efficiency (second indirect effect). Given the importance of aerosols, climate models consider today the aerosol direct and indirect radiative forcings. However, despite significant progress in understanding aerosol effects on climate, there are still significant uncertainties due to the large spatial-temporal variability and heterogeneity of aerosols associated with their short atmospheric lifetime and to the complex interactions between aerosols and clouds, in terms of their physical and chemical properties (IPCC, 2001; Haywood and Boucher, 2000; Lohmann and Feichter, 2005; Chen and Penner, 2005). In order to improve both the aerosol parameterization schemes in climate models and the accuracy of aerosol radiative forcings, monitoring of the spatial and temporal distributions of aerosol physical, chemical, optical and radiative properties is required both on global and regional scales. To this aim, a worldwide effort has been undertaken since the early 1990s, to produce a global aerosol climatology by combining satellite-based observations (such as Total Ozone Mapping Spectrometer, TOMS, Moderate Resolution Imaging Spectro-Radiometer, MODIS, Multiangle Imaging Spectroradiometer, MISR, Polarization and Directionality of the Earth's Reflectance, POLDER) and measurements from ground-based monitoring networks (Aerosol Robotic Network, AERONET). These data sets are complemented with those of field campaigns (ground based and airborne), for calibration and validation of satellite data.

A region that has received much interest with regard to aerosol effects is the clima-

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tologically sensitive region of Mediterranean Basin, in particular its eastern part, where several experimental studies have taken place (Luria et al., 1996; Pinker et al., 1997; Mihalopoulos et al., 1997; Paronis et al., 1998; Papayiannis et al., 1998; Ichoku et al., 1999; Formenti et al., 2001a, b, 2002a, b; Andreae et al., 2002; Lelieveld et al., 2002; Kouvarakis et al., 2002; Zerefos et al., 2002; Gerasopoulos et al., 2003; Balis et al., 2004a, b; Papayiannis et al., 2005). The Eastern Mediterranean is a crossroad where aerosols from different sources are superimposed and mixtures of different kinds of particles converge, e.g. maritime aerosols from Mediterranean sea spray, seasonal biomass burning aerosols (Formenti et al., 2002a, b), anthropogenic aerosols transported by polluted air masses from urban and industrial areas (Lelieveld et al., 2002; Zerefos et al., 2000; Formenti 2001b; Eisinger and Burrows 1998), and mineral dust aerosols mainly from north African deserts, and to a lesser extent from the Middle East. Indeed, concentrations of trace gases and aerosols over the Mediterranean region are higher by factors 2 to 10 than over the North Pacific Ocean, which is the least polluted environment at northern latitudes (Lelieveld et al., 2002).

The strong climatic effect of aerosols in the Eastern Mediterranean Basin, especially in summer, is due to cloud-free conditions and high solar radiation intensity (Millan et al., 1997; Kouvarakis et al., 2002), amongst other factors. These conditions, in combination with the significant aerosol loadings, the different aerosol types, and the practically absent wet removal, make the Eastern Mediterranean a very interesting climate study region. Several model predictions indicate that direct forcing by sulfate aerosols reach an absolute maximum over this region (Charlson et al., 1991; Kiehl and Rodhe, 1995; Boucher and Anderson, 1995). Moreover, recent campaigns (e.g. Lelieveld et al., 2002) and modeling studies (e.g. Hatzianastassiou et al., 2004) have shown that the direct forcing from all aerosols obtains large values in the Eastern Mediterranean. More specifically, Lelieveld et al. (2002) based on measurements taken during the Mediterranean Intensive Oxidation Study (MINOS) conducted in the summer of 2001 in Crete, reported a large daily mean surface radiative forcing induced by all aerosol species, of about $-18\,\mathrm{Wm}^{-2}$, with a corresponding forcing of $-6.6\,\mathrm{Wm}^{-2}$ at the top-of-

atmosphere. Note that such experimental estimates are limited in temporal and spatial coverage. Hatzianastassiou et al. (2004) in a recent modeling study using mean climatological aerosol data from the Global Aerosol Data Set (GADS), computed a monthly mean aerosol surface radiative forcing of $-9\,\mathrm{Wm}^{-2}$.

In order to improve our scientific knowledge of the role of aerosols for such a climatologically sensitive region and the accuracy of relevant model computations, more and better data on aerosol properties and budgets are required. Site-specific ground-based aerosol data from a global network such as AERONET, provide continuous time series with a very high temporal resolution.

The globally distributed AERONET (Holben et al., 1998; 2001), which is an automated robotic Sun- and sky scanning measurement program, was established in the early 1990s and is growing rapidly in the number of stations. Since January 2003, a new AERONET station (FORTH), was installed in Crete, additional to others in the Eastern Mediterranean (Sede Boker, Ness Ziona, Dead Sea, Erdemli). The location of the FORTH-AERONET station is unique, located in a relatively isolated island in the central part of the Eastern Mediterranean Basin.

In this work, aerosol measurements from the FORTH-AERONET station in Crete are analysed for the first time, to characterise aerosol properties over the Eastern Mediterranean Basin. The aerosol data include aerosol optical thickness (AOT) at 340, 500, 870, and 1020 nm, Angström parameter ($\alpha_{440-870}$), and aerosol volume-size distribution, and cover the complete 2-year period 2003–2004, allowing the annual aerosol cycle to be studied. A brief description of the data and the retrieval procedure is given in Sect. 2. In Sect. 3, the temporal variation of aerosol optical properties is discussed, with emphasis on the advection of dust aerosols from Africa. In addition, the relationship between the Angström exponent and the aerosol optical thickness, and the features of the aerosol volume-size distribution are also examined. Finally, we investigate the relationship between aerosol properties, air masses, and wind direction and speed.

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2 The data used

The aerosol data used in this work were derived from measurements carried out by the AERONET station, named FORTH-CRETE, which is located on the northern coast of Crete (35°19′58″ N, 25°16′55″ E, 20 m altitude, see Fig. 1). The station is located 15 km east from Heraklion, which is the largest city in Crete island with 200 000 inhabitants. The measured aerosol data cover a complete 2-year period from January 2003 to December 2004. The measurements were made with the CIMEL sunphotometer (CE-318) an automatic sun-sky scanning spectral radiometer. The instrumentation is set up on the roof of the building of National Center for Marine Research in Crete, which is 20 m high and is found 100 m from the coast. The instrumentation, data acquisition, retrieval algorithms and calibration procedure are conform to the standards of the AERONET global network, and are described in detail, along with the uncertainty of final released products, by Holben et al. (1998), Dubovik and King (2000), Dubovik et al. (1998, 2000, 2002) and Kaufman et al. (2002). In this study, level 2.0 AERONET data are used; more specifically, we analyse daily mean values of AOT, Angström exponent and volume size distribution of aerosol particles. For AOT, we have chosen to use spectral values at 340, 500, 870 and 1020 nm to cover the solar spectrum to which aerosol properties are most relevant.

Analytical air-mass back trajectories of 4-days obtained with the Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT), are also used in this study to identify the pathway of air-masses over Crete. The back trajectories are computed for 3 distinct arrival levels, namely 500, 1500 and 3000 m at 12:00 UTC, on a day-by-day basis.

Surface meteorological observations are ensured by an automatic meteorological station operating at the AERONET FORTH-CRETE site. Air-temperature, wind speed and direction and relative humidity are routinely recorded as 15 min averages. The total (direct and diffuse) shortwave and longwave solar radiation measurements were performed by the Kipp & Zonen CG4 and CM11 radiometers respectively. The meteo-

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rological data are available since 20 June 2003, when the meteorological station was set up, i.e. 6 months latter than the beginning of the AERONET station.

3 Results and discussion

3.1 Temporal variation of aerosol optical properties

The seasonal variation of monthly mean AOT values at four wavelengths (340, 500, 870 and 1020 nm) as well as that of monthly mean Angström parameter ($\alpha_{440-870}$) for the FORTH-AERONET station in Crete are given in Fig. 2, where the corresponding time series over the 2-year period 2003–2004 are shown. Table 1 summarizes the seasonal variation of AOT and $\alpha_{440-870}$, as well as annual average values for the entire study period (2003–2004). The monthly mean values were computed from corresponding daily means. The annual cycle of AOT in the ultraviolet and visible has maximum values in spring (March–May), late summer and autumn (August and October) and minimum values in winter and early summer (June). At the near-infrared wavelengths of 870 and 1020 nm, there are clear maximum AOT values in spring, but also in autumn (not so strong at 340 and 500 nm) and minimum values in winter. The spring and autumn maximum AOT values are associated with strong dust episodes taking place in these seasons (Dayan et al., 1991; Marticorena and Bergametti, 1996; Moulin et al., 1998; Karyampudi et al., 1999; Israelevich et al., 2002, 2003; Kubilay et al., 2003) when dust particles are transported from North African deserts. This underlines the importance of synoptic systems to aerosol loads in the Eastern Mediterranean Basin, but also in the rest of the basin. This is also supported by other features of AOT, such as the local minimum values in June when cyclones that develop in west Sahara are blocked by high-pressure systems over Libya, and thus cannot reach the eastern Mediterranean, traveling therefore northward and transporting dust to the central and western Mediterranean (Moulin et al., 1998; Israelevich et al., 2002).

The seasonal dust aerosol activity in the Mediterranean has been investigated by

several workers (Moulin et al., 1997, 1998; Marticorena and Bergametti, 1996; Prospero et al., 2002; Israelevich et al., 2002, 2003; Kubilay et al., 1995, 2003; Masmoudi et al., 2003a; Perrone et al., 2005; Esposito et al., 2004; Lyamani et al., 2004). In summary, during spring the main aerosol activity takes place in the eastern Mediterranean, where dust is transported by thermal lows (Sharav cyclones) created over the North Africa deserts then moving along the African coast from west to east (Alpert and Ziv, 1989; Dayan et al., 1991; Moulin et al., 1998; Karyampudi et al., 1999; Barkan et al., 2005). In summer, the maximum of dust activity occurs in the central Mediterranean, and can be detected even in central Europe (Dulac et al., 1992; Moulin et al., 1998; Collaud Coen et al., 2003). By late summer – early autumn, under the influence of low pressures near the Balearic Islands, the maximum dust activity is found in Western Mediterranean (Moulin et al., 1998). In general, the maximum dust activity appears in spring and summer. Israelevich et al. (2002) have shown that during these periods desert dust is always present in the region's atmosphere. Dust is transported over the Mediterranean basin whenever meteorological conditions are appropriate and most time dust intrusions occur under the form of "storms". Overall, the aerosol distribution over the Mediterranean is largely controlled by synoptic conditions.

The maximum AOT values in spring and autumn (Fig. 2) can also be associated with maritime aerosols, produced by sea spray, mixed with dust aerosols. Several studies have shown that unique aerosol types never occur in the atmosphere, but aerosol populations are rather mixtures of various aerosol types (Pruppacher and Klett, 1997); this was shown to be valid for Crete (Lelieveld et al., 2002).

The AOT values at 340, 500, 870 and 1020 nm indicate a significant spectral dependence of aerosol optical thickness, with decreasing annual mean values (Table 1) from 0.34±0.14 at 340 nm down to 0.11±0.09 at 1020 nm, implying an AOT reduction of 68% from the near-UV to near-IR wavelengths. Comparison between Figs. 2a, b, c and d, reveals a spectral difference in the seasonal cycle of AOT. More specifically, apart from the omnipresent spring maximum and winter minimum, there are more distinct secondary maxima in autumn along with summer minima at 870 and 1020 nm,

than in the near-UV and visible wavelengths. The maximum AOT values in spring and autumn at all wavelengths along with the minimum Angström parameter values during these seasons are due to episodes that advect dust from North African deserts towards the Mediterranean. It should be noted however that we performed a systematic 4-day back-trajectory analysis which revealed that mineral dust particles can also be advected to Crete from the Middle-East and Arabian Peninsula, but this case is quite rare compared to the transport from African deserts. Note that the annual variation of AOT in the UV-visible wavelengths is larger than in the near-IR (see Table 1). The high AOT values in summer at the smaller wavelengths are related to the prevailing meteorological conditions. Thus, the stable atmospheric conditions during summer in the Mediterranean favour the accumulation of aerosol particles, especially those of fine dimensions, advected by long-range transport processes mainly from the north. Moreover, during this period the aerosol wet removal is practically absent over the southeastern Mediterranean (Bartzokas et al., 2003). The presence of fine-mode aerosols in summer over Crete is supported by corresponding mean monthly Angström parameter ($\alpha_{440-870}$) values of 1.6±0.4 (Fig. 2e), while large sizes of aerosol particles, such as dust, during spring (April-May) and autumn (October) are related to monthly mean values of $\alpha_{440-870}$<1 (see Fig. 2e and Table 1). The spectral differences in the annual cycle of AOT shown in Fig. 2 and Table 1, indicate the presence of aerosols of different size distributions throughout the year over Crete, having also different chemical composition. The aerosol population over Crete is heterogeneous, and based on campaigns, it has been found to be composed by mineral dust, marine biogenic particles and anthropogenic aerosols at different layers (Lelieveld et al., 2002; Kouvarakis et al., 2002; Bardouki et al., 2003; Sciare et al., 2003). The standard deviations of AOT are maximum during spring i.e. when AOT is maximum, are associated with frequent dust events and suggest strong day-to-day variations in aerosol load over Crete. Large standard deviations of AOT are also related to variability in wet deposition processes, associated with local meteorological conditions. Note the relatively smaller number of winter daily values from which the monthly means were computed, because of cloud

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screening, in contrast to complete number of days in a month during summer, i.e. when sky is almost completely clear in Crete.

Up to now, only few studies have investigated the annual variation of aerosol properties in this region, since most of them are based on measurements taken only during summer. Kubilay et al. (2003) using AERONET observations from the Erdemli-AERONET station, located on the southern Turkish coast, covering the period from January 2000 to July 2001, found that AOT_{440} and AOT_{870} show the same seasonal pattern as in our study, with primary and secondary maximum values of ~0.4–0.5 in April and ~0.2–0.3 in August, and minimum values in winter. Similarly, the annual variation of AOT_{500} and AOT_{1020} in Sede-Boker-AERONET station in Israel has its primary maximum values in April-May, equal to ~0.25–0.27 for AOT_{500} for the years 1996, 1998–1999 (Holben et al., 2001) and ~0.15 for AOT_{1020} for 1996–2001 (Israelevich et al., 2003), whereas it has secondary maxima of ~0.22-0.23 and ~0.13, respectively, in late summer-early autumn (July–October). Our values for the FORTH site are slightly lower than those of Erdemli, and very close to the values of Sede-Boker. Therefore, it seems that the features of the annual cycle and magnitude of AOT in Crete are very similar to those reported for Erdemli (South Turkey) and Sede-Boker (Israel), located further east. Gerasopoulos et al. (2003) using ground-based aerosol measurements at two sites in Northern Greece, one rural (Chalkidiki) and the other urban (Thessaloniki), found that AOT_{500} at the rural site exhibits a distinct annual cycle with a summer maximum in August (0.3) and a minimum in December (0.09), while no clear annual cycle was observed at the urban site due to the permanent influence of local pollution sources. The differences in the annual cycle and magnitudes of AOT between this work and ours can be explained by the different location of the sites. Crete is an island in the central part of the Eastern Mediterranean Basin, in relatively close proximity to North Africa, while Northern Greece is far enough from dust sources, allowing the deposition processes to be more effective and the aerosol population to be depleted. Thus, Papayannis et al. (2005) in the framework of the project EARLINET (A European Aerosol Research Lidar Network) reported more than 50 dust events in Athens versus

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17 over Thessaloniki during the period 2000–2002. The large AOT values in August reported by Gerasopoulos et al. (2003), also found in our study, are mainly linked with fine aerosol particles transported from Eastern Europe, and to a lesser extent with dust aerosols. This is supported by the synoptic conditions during this season, and the quite large values of Angström parameter. The slightly larger AOT in Northern Greece than Crete during summer are due to the fact that Northern Greece is closer to the pollution sources in Central and Eastern Europe than Crete.

A more detailed temporal variation of spectral AOT and $\alpha_{440-870}$ values, within the 2-year study period 2003–2004, is depicted in Fig. 3. The annual cycle of AOT is more distinct at λ=340 and 500 nm. Apart from the features of annual cycles of AOT and $\alpha_{440-870}$, also seen in Fig. 2, strong temporal variability of AOT and Angström parameter during spring, late-summer and autumn period is also found. Peak values of AOT_{500}, as high as 0.91 in March 2004, 0.5 in September 2003, and 0.46 in May 2004 are found, with even larger values at 340 nm, and smaller ones at 1020 nm. These peak values are attributed to short-lived dust storm episodes, as indicated by the corresponding small values of the Angström parameter, decreasing down to 0.13 in March 2004 and 0.10 in May 2004, while it has values of 0.29 in September 2003. Peak AOT and minimum $\alpha_{440-870}$ values are attributed to dust events, as supported also by the results of our back-trajectory analysis for the specific days, which indicate dust particles transport by air masses originating mostly from North Africa. Only one single case of dust transport from the Middle-East has been identified (11 May 2003). Of course, apart from dust, the maximum AOT values in Figs. 2a, b, c and 3a, b, c are also related to the contribution of large-size coarse sea-salt particles, involving values of AOT up to 0.62 at 340 nm and 0.45 at 1020 nm, with corresponding values of Angström parameter between 0.09 and 0.8. In spite of the general pattern, according to which AOT maximum and $\alpha_{440-870}$minimum values occur simultaneously, there are some days when both AOT and $\alpha_{440-870}$ exhibit maximum values (e.g. 30–31 March 2003). This behavior involving high values of the Angström parameter (1.43–2.05) is seen at near-UV wavelengths only (e.g. 340 nm), while the corresponding near-IR AOT

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values at 1020 nm are very low, varying from 0.03 to 0.14. This behavior indicates the presence of fine urban-industrial aerosols either from local sources or transported from continental polluted air masses. Indeed, the back-trajectory and meteorological analyses revealed that these cases are characterised by northern wind directions, and they are associated with air masses originating from central Europe and the Balkan peninsula, carrying pollution from areas such as Athens, Eastern Europe, and western Turkey.

During summer, the day-to-day variability of AOT_{340} and AOT_{500}, as well as that of the Angström parameter $\alpha_{440-870}$, is relatively high. More specifically, values of AOT_{340} range between 0.2 and 0.65, those of AOT_{500} between 0.1 and 0.5, and values of $\alpha_{440-870}$ vary from 1 to 2.2. On the other hand, during summer the variability of AOT_{870} and AOT_{1020} is much smaller, except for the identified dust events, with values varying within the range 0.03–0.16. In this period, large values of AOT and $\alpha_{440-870}$, are attributed to fine urban-industrial aerosols, which are mainly transported from Central and Eastern Europe, and secondarily from Western and North-Western Europe. During summer, the broader Greek region and generally the Eastern Mediterranean Basin, are under the influence of two pressure systems: the quasi-permanent Azores anticyclone and the Pakistan thermal low-pressure system, which extends through to the Middle-East up to the South Anatolian plateau (Metaxas and Bartzokas, 1994; Palutikof et al., 1996; Katsoulis et al., 1998). Thus, a strong pressure-gradient is created over the Aegean Sea, resulting in a northerly flow known as etesian winds, which carries anthropogenic aerosols from Central and Eastern Europe, Balkans, and the polluted city of Athens to Crete. Note that the stable atmospheric and dry-weather conditions prevailing over the Mediterranean in summer favour the accumulation of continental anthropogenic aerosols, explaining thus the large AOT values in UV and visible wavelengths, along with large values of Angström parameter (even larger than 2). Large values of $\alpha_{440-870}$ in summer, associated with fine aerosol particles have been also reported by Gerasopoulos et al. (2003) for northern Greece (background AOT_{500} values of 0.3–0.5 up to 0.7–0.8 at specific cases, and $\alpha_{440-870}$ values gener-

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ally larger than 1.4). Balis et al. (2003) reported values of AOT_{500} and $\alpha_{440-870}$ as large as 0.77 and 1.78 respectively, in August 2001, at Thessaloniki (North Greece), which is the second largest city in Greece, with a population of about 1 million. They stated that these smoke plumes were transported from different European regions (Balkans, Northern Black-Sea, Danube region, Southern Italy). Our corresponding AOT_{500} values are slightly smaller (not exceeding 0.5) than those of Gerasopoulos et al. (2003) and Balis et al. (2003) because FORTH-AERONET station is located in a maritime environment and maritime aerosol concentrations are generally smaller than continental ones (Jaenicke, 1993). The sporadically increased daily mean $\alpha_{440-870}$ and AOT values in summer are due to biomass burning aerosols, largely transported by smoke plumes from the European continent to the South Mediterranean. Our Angström parameter values however are similar to those computed by Balis et al. (2003). Note that, Eck et al. (1999; 2001) have found that biomass burning particles are characterised by large values of $\alpha_{440-870}$, higher than 1.5, whereas Dubovik et al. (2002) reported worldwide AOT_{440} and $\alpha_{440-870}$ values within the range 0.4–0.8 and 1.2–2.2, respectively, from AERONET measurements.

It is interesting that the temporal variability in AOT at near-IR wavelengths (AOT_{870}, AOT_{1020}) and $\alpha_{440-870}$ values seems to be greater in summer 2004 than summer 2003 (Figs. 3c, d). The larger AOT variability and magnitudes in summer 2004 is associated with more peaks in AOT due to the stronger dust transport in summer 2004 than 2003, which is well depicted in the lower values of $\alpha_{440-870}$ in summer of 2004 (see Fig. 3e). Note also that summer mean values of AOT_{340} and $\alpha_{440-870}$ are slightly lower in 2004 (0.37 and 1.41, respectively) than 2003 (0.39 and 1.62, respectively). This indicates the presence of more fine aerosol particles in summer 2003 than 2004, which can be related to the fact that summer 2003 has been the hottest one in Europe since 1500 (Luterbacher et al., 2004), with a persistent high-pressure system established over western Europe during July and August 2003, resulting in extremely hot and dry weather conditions. Such conditions favoured the development of numerous forest fires (huge forest fires took place in Portugal, Southern France, Italy

and Balkans, as illustrated well by satellite images; e.g. http://visibleearth.nasa.gov, http://landqa2.nascom.nasa.gov/), inducing strong emissions of fine-mode aerosols that accumulated in the atmosphere under stable conditions. Therefore, the prevailing synoptic conditions and the northerly flow over Crete advected a lot of anthropogenic and biomass burning aerosols.

The low monthly mean AOT values during winter, down to 0.15 for AOT_{340} and 0.1 for the other wavelengths, together with $\alpha_{440-870}$ values mostly within the range 0.7–1, indicate near background conditions most probably dominated by the presence of marine aerosols. Although this is based on relatively few AERONET data, it is in agreement with measurements performed during winter (Bardouki et al., 2003) indicating significant contribution from sea-salt compared to dust. Time-series of AOT and $\alpha_{440-870}$ along with back-trajectory analyses have shown that dust outbreaks can also take place in winter, but they are rather rare and weak. This is normal given the less favourable conditions for dust-transport to occur. The synoptic pattern over the region during winter is characterised by: (1) the Siberian anticyclone causing polar continental air to flow from eastern Europe, (2) an anticyclone developed over middle and eastern Europe, and (3) the low-pressure systems in the central Mediterranean, moving usually from west to east. Basically, these regimes do not favour the transport of dust from Africa or Middle-East. On the contrary, they are rather favourable for transport of fine anthropogenic aerosols, resulting in smaller values of AOT during this season than others. Such small AOT values are also consistent with aerosol mixing processes, involving low-concentration maritime aerosols from sea-spray (whose production increases in winter due to strong winds), as well as efficient wet-removal aerosol processes, i.e. a lot of clouds and precipitation.

3.2 Frequency of occurrence of aerosol optical thickness and Angström parameter

The distribution of percent frequency of occurrence of daily averages of spectral AOT values and Angström parameter ($\alpha_{440-870}$) are shown in Fig. 4, whereas Fig. 5 shows

the corresponding seasonal distributions.

3.2.1 Ultraviolet optical thickness

The histogram of AOT_{340} is quite broad with values occurring even at ~1.2 (Fig. 4a). The 63% of the daily averaged AOT_{340} values are almost equally distributed within the range 0.2–0.45, while only a few cases, i.e. 1.3%, have AOT_{340} values greater than 0.7; these cases correspond either to strong dust outbreaks or to heavy pollution events. Overall, the average AOT_{340} value for the entire period is found to be equal to 0.34. The winter season presents the narrowest AOT_{340} frequency distribution (Fig. 5i-a) with a maximum of about 30% of total values at 0.125 corresponding to background conditions. Thus, 70% of total AOT_{340} daily observations have values between 0.1 and 0.25. In spring the probability distribution broadens out to larger near-UV optical depths (Fig. 5i-b) mainly due to the frequent occurrence of dust episodes in this season (see 3.1). Therefore the maximum probability of AOT_{340} shifts towards greater values (0.375). A significant portion of all cases (21%) have AOT_{340}>0.45 and 7% of total cases have AOT_{340} >0.6. The frequency of occurrence of AOT_{340} in summer (Fig. 5i-c) exhibits a broad peak within the range of values 0.25–0.45, yielding 62% of total observations in this season, whereas another 25% have AOT_{340} values up to 0.6. These values indicate the predominance of continental anthropogenic aerosols and probably maritime ones. A portion of 20% of total daily values with $AOT_{340}\geq 0.525$ correspond to fine particles, especially smoke aerosols from biomass burning. The autumn frequency distribution is broader, showing AOT_{340} peaks near 0.2–0.3 and 0.35–0.45.

3.2.2 Visible optical thickness

The frequency distribution of AOT_{500} (Fig. 4b) is broad, having values up to ~0.9. It is however narrower than the distribution of AOT_{340}, due to the smaller values of AOT_{500} at the right side of the distribution. Such a narrowing, even stronger, is apparent when moving gradually to larger wavelengths, i.e. AOT_{870} (Fig. 4c) and AOT_{1020} (Fig. 4d),

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due to the inverse wavelength dependence of AOT for most aerosol types such as water-soluble, soot and sulfate (Koepke et al., 1997). The maximum frequencies of occurrence of AOT_{500} (20%) are centered around 0.1–0.2, with steadily decreasing values at larger optical depths. The probability distribution of AOT_{500} for each season is quite similar to that of AOT_{340} (Figs. 5ii). The seasonal mean values of AOT_{500} are equal to 0.14, 0.25, 0.22 and 0.21 for winter, spring, summer and autumn, with the larger values occurring in spring (see also 3.1). The winter distribution is narrower compared to the others, whereas the spring distribution is the broadest one. Note that in spring about 25% of total AOT_{500} data have values beyond the dominant range 0.1–0.3.

3.2.3 Near-infrared optical thickness

The frequency of occurrence of AOT at near-IR wavelengths (870 and 1020 nm, Figs. 4c and d, respectively) is almost identical, and this is also valid for the corresponding seasonal distributions (Figs. 5iii and iv). They are both narrower than frequency distributions at near-UV and visible wavelengths. The maximum frequencies (≈50%) are found, both on a year and seasonal basis, around 0.075. The secondary maximum frequency (≈20%) occurs at 0.125. In spring, about 25% of total days have AOT_{1020}>0.2, and are characterised by dust transport. This is also the case in autumn, though to a smaller extent, with about 10% of total days with AOT_{1020}>0.2.

3.2.4 Angström parameter

The frequency of occurrence of the daily mean Angström parameter ($\alpha_{440-870}$) values (Fig. 4e) is wide, covering the range 0.05–2.15 owing to the great variety of aerosol types (such as urban/industrial, dust, maritime or mixtures) and sizes in the atmosphere over Crete. Given the location of the AERONET monitoring site in the central part of the Eastern Mediterranean Basin, with many and different emission sources all around unique aerosol types do not occur; in contrast aerosol populations are rather

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consisted of numerous types of particles, frequently distributed in different layers in the atmosphere as shown by experimental analyses (e.g. MINOS, Lelieveld et al., 2002). The intermediate $\alpha_{440-870}$ values in Fig. 4e correspond to aerosol background conditions, whereas $\alpha_{440-870}$ values <0.5 are related with coarse mode particles (dust or maritime ones) and days with large $\alpha_{440-870}$ values include significant contribution from fine mode particles (industrial or biomass burning). Approximately 16% of all days have $\alpha_{440-870}$<0.5, resulting mainly from dust transport. Note that values of $\alpha_{440-870}$ do not exceed 2.2 in contrast to continental polluted sites (Holben et al., 2001). This is due to the relatively remote marine environment of FORTH-AERONET station, far from polluted areas. Aerosols from polluted areas are mixed with maritime aerosols, resulting in reduced $\alpha_{440-870}$ values, so that less than 5% of the cases have values of $\alpha_{440-870}$ above 2.0. The histogram of $\alpha_{440-870}$ seems to be bimodal, with the larger-amplitude mode peaked at 1.65, and the lower amplitude mode peaked at 0.45; the first one is associated with fine mode particles. The computed annual average value of $\alpha_{440-870}$ is equal to 1.17. Seasonal differences are revealed from the diagrams of Fig. 5v. Thus, the atmosphere over Crete has quite small $\alpha_{440-870}$ values in winter (Fig. 5v-a), with 70% of total cases with $\alpha_{440-870}$<1.0. This, in combination with the small winter AOT values (Figs. 5i, ii, iii, iv) implies that aerosol populations in winter are dominated by low-concentration sea-salt aerosols. In contrast, the frequency distribution of $\alpha_{440-870}$ in summer is skewed towards large $\alpha_{440-870}$ values, with 88% of total cases having $\alpha_{440-870}$>1.0. Thus, the aerosol populations are dominated by fine mode particles during summer. During the transient seasons the situation is totally different. In spring, the frequency distribution of $\alpha_{440-870}$ is rather uniform with $\alpha_{440-870}$ values equally distributed in size bins from 0 to 1.7. More than 25% of total cases have $\alpha_{440-870}$<0.5, associated with dust particles. In autumn, the probability distribution of $\alpha_{440-870}$ has a bimodal structure with two distinct modes near 0.5 and 1.5 related with dust/maritime coarse particles, and continental urban/industrial fine aerosols, respectively.

3.3 Relationship between Angström parameter and aerosol optical thickness

The Angström parameter $\alpha_{440-870}$ versus AOT_{870} is shown in Fig. 6. The combined information from these two physical parameters allows a rough characterisation of aerosols. Different modes of aerosols can be identified. More specifically, small values of $\alpha_{440-870}$ (<0.5) and large values of AOT_{870} (>0.20) are related to coarse particles, associated with dust outbreaks in North African deserts during spring and late summer-autumn. Points corresponding to small values of $\alpha_{440-870}$ (<0.5) and AOT_{870} (≈0.10–0.15) whether correspond to transported dust during winter (involving frequent and efficient wet removal processes) or are associated with transport of maritime aerosols (sea-spray). There is a wide range of $\alpha_{440-870}$ values (~0.10–2.0) for AOT_{870}<0.2, which is due to the presence of various types of aerosol for background conditions, arising from transport and mixing processes. Such mixtures can involve background sulfate maritime aerosols, along with mineral dust, soot, and other continental natural or anthropogenic particles. The contribution of each of these particles varies with time, but coexistence of many types as internal or/and external mixture, is often possible. This was illustrated by back-trajectory analyses that were performed for identified dust events (with AOT_{870}~0.15 and $\alpha_{440-870}$~0.5) showing that air masses either originated from the Atlantic Ocean traveled over western Mediterranean and North Africa to reach Crete, or originated from North Africa deserts traveled across central Mediterranean or Libyan Sea before arriving over Crete. Both pathways suggest active mixing processes of aerosols. There is another group of points with values of AOT_{870}>0.15 and $\alpha_{440-870}$ between 0.5 and 1.5, indicating mixed aerosol types as well, and dominated either by relatively coarse aerosols (if $\alpha_{440-870}$<1.0) or fine particles (if $\alpha_{440-870}$>1.0). Back-trajectory analysis for these cases revealed that they usually correspond to mixtures of dust with urban/industrial aerosols, transported either from Western Europe (Atlantic Ocean, Spain, France, Italy) or from Central and Eastern Europe. In addition, there are cases for which air masses come from the NW or NE, and pass over Athens, thus advecting pollution to Crete. The predominance of fine aerosols with anthropogenic

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and biomass-burning origin is suggested by values of AOT_{870}<0.15 and $\alpha_{440-870}$>1.5. Similar patterns to ours have been also reported by Kubilay et al. (2003) for the Erdemli site located in the Eastern Mediterranean, and by Pace et al. (2005) for the island of Lampedusa in the Central Mediterranean.

3.4 Aerosol size distribution

Figure 7 shows the seasonal variation of aerosol columnar[1] volume size distribution[2] at the FORTH-AERONET monitoring site, in Crete, derived from sky radiances. The distributions were averaged from daily distributions for each season, for the period 2003–2004. The AERONET daily aerosol volume size distributions are retrieved using the inversion procedure proposed by Dubovik and King (2000). The FORTH-AERONET site shows an average bimodal aerosol volume size distribution with two distinct modes: a fine mode (composed by smaller particles) of principally anthropogenic origin, with radius between 0.05 and 0.5 μm, and a coarse mode of larger particles of natural origin, with radius ranging from 0.5 up to 15 μm. It is clear that both fine and coarse particles exist over Crete over all seasons, with sizes that do not change drastically from one season to another. However, the ratio between the two modes changes drastically, indicating variations in the relative contribution of different particles that constitute the total aerosol column. Thus, in winter and spring, the fine-mode peak is at 0.11 μm, and shifts towards 0.15 μm in summer and autumn. In the coarse mode, the major part of aerosol volume is contributed by particles with radius from 1.0 to 5.0 μm. It is worth noting that in the coarse mode, two pseudo-modes can be distinguished,

[1] Note that henceforth (based on AERONET retrievals) V refers to the integrated volume of aerosols over the whole atmospheric column.

[2] $\frac{dV}{d\ln r} = \frac{V}{\sqrt{2\pi}\cdot\ln\sigma}\exp\left[-\frac{1}{2}\left(\frac{\ln(r/r_v)}{\ln\sigma}\right)^2\right]$, where dV/dlnr is the aerosol volume size distribution, V is the total volume of particles, r is the particle radius, r_v is the volume geometric mean radius and σ is the geometric standard deviation.

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centered around 1.7 and 3.9 μm; these two pseudo-modes are attributed to desert dust and sea-salt maritime aerosols, as also suggested by Chiapello et al. (1999) and Tanré et al. (2001) for other AERONET sites. These two pseudo-modes are more distinct in spring, autumn and winter, in contrast to summer. Note that during winter the mode that peaks at 1.7 μm becomes dominant, whereas in summer the dominant mode peaks at 3.9 μm. Our bimodal structure of aerosol volume size distribution (Fig. 7) is in agreement with the results of Kouvarakis et al. (2002) and Bardouki et al. (2003) derived from experimental campaigns conducted in Crete in May 1999, July 2000 and January 2001. Their analysis of the chemical and physical aerosol properties revealed a bimodal size distribution both in summer and winter, with peaks of both anions and cations at 0.13 μm for the accumulation mode, and at 2.20 μm for the coarse mode. The analysed size distributions of different individual chemical species suggested that anthropogenic compounds such as non-sea-salt sulfate (nss-SO_4^{2-}), and biogenic-origin species such as NH_4^+ and MSA have their maximum in the fine mode (at about 0.13 μm, in terms of radius). On the other hand, anthropogenic nitrate (NO_3^-), sea-salt (Na^+) and dust particles (nss-Ca^{2+}) exhibit their maximum in the coarse mode (within the range 1.7–3.4 μm, in terms of radius, depending on the species). Note that the results of Kouvarakis et al. (2002) from the PAUR II (Photochemical Activity and Solar Ultraviolet Radiation) project suggest the peak radius of sea-salt and dust particles in May to be similar, while the results of Bardouki et al. (2003), from an experimental campaign in July 2000, show the peak of dust particles to be shifted to slightly larger wavelengths compared to that for sea-salt particles. According to these findings, our coarse pseudo-mode centered around 3.9 μm should be attributed to dust aerosols, while that at 1.7 μm to maritime aerosols. Based on this, during summer the volume of dust particles dominates that of maritime aerosols, while in winter the situation is reversed. Certainly, more detailed analyses are required to draw definite conclusions about particle concentrations, since the total volume of an aerosol population involves the number density and the size of particles. In any case, it is very interesting to see that over all seasons, the coarse-mode aerosol populations over Crete consist of both

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dust and maritime aerosols, with an almost equal contribution. The results of our study are in line with the results from the experimental campaigns in summer and winter, but provide additional insights for the other seasons.

The bimodal structure of aerosol spectra in Crete is in agreement with the results of Smirnov et al. (2002, 2003) who studied the optical and physical properties of maritime aerosols in five key island locations of AERONET network in tropical and subtropical Pacific and Atlantic oceans over 2–5 years. They found that in the presence of pure maritime or mixed with dust, smoke or urban/industrial aerosols, the atmospheric column aerosol is characterised by a bimodal lognormal size distribution with a fine mode at effective radius, R_{eff}[3], at ~0.11–0.14 μm, and a coarse mode at R_{eff}~1.9–2.3 μm. There is a remarkable agreement with our fine mode at 0.11 μm and the first pseudo-mode of coarse mode at 1.7 μm. Nevertheless, the aerosol size distributions in Crete are different to those in Pacific and Atlantic oceans in terms of the second coarse pseudo-mode at 3.9 μm. This is associated with a strong contribution of dust in Crete, originating from desert areas in Africa. Note that bimodal aerosol size distributions in maritime environments are common and it has been suggested that their coarse mode arises from efficient processing by numerous consecutive cloud cycles (Hoppel et al., 1990; Pruppacher and Klett, 1997). There is another difference between the FORTH-Crete size distribution and those over the Atlantic and Pacific Oceans. In Crete fine and coarse modes contribute equally, while at the oceanic sites, especially for the Pacific Ocean, the coarse mode is more significant than the fine one. This can be attributed to the less efficient cloud processing of aerosols over Crete than over remote oceanic areas due to the proximity of continental areas surrounding the Mediterranean.

Table 2 presents computed parameters of the bimodal lognormal aerosol volume size distribution in Crete, shown in Fig. 7, such as the effective radius (R_{eff}) and the

[3]The effective radius is defined and computed as: $R_{\text{eff}} = \frac{\int_{r_{\min}}^{r_{\max}} r^3 \frac{dV}{d\ln r} d\ln r}{\int_{r_{\min}}^{r_{\max}} r^2 \frac{dV}{d\ln r} d\ln r}$, where r_{min} is the minimum and r_{max} the maximum radius of the aerosol spectra

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columnar volume of particles per unit cross-section of atmospheric column (C_v)[4] for each mode. It should be noted that the number (343) of retrieved daily mean aerosol volume size distributions, and hence of associated parameters, is much lower than the number of retrieved AOT and $\alpha_{440-870}$ (533), though still meaningful. The effective radius for fine mode varies around 0.13 throughout the year, while that for the coarse mode varies slightly, i.e. within the range 2.00–2.32. It should be noted that R_{eff} of the coarse mode is slightly larger in summer than in the other seasons; this could arise from the larger relative contribution of the second (dust) pseudo-mode with respect to other seasons (see Fig. 7), due to the absence of wet removal processes of dust. The columnar volume concentration of aerosols, C_v, ranges from 0.025 to 0.048 for the fine mode and from 0.046 to 0.075 for the coarse mode. The modal volume of coarse particles in Crete dominates that of the fine mode, except for summer when they are equivalent. Additionally, note that the maximum columnar volume of fine mode aerosols occur in summer, owing to the significant contribution of anthropogenic and natural fine pollution particles (see Sects. 3.1 and 3.2). The coarse mode has maximum columnar volume in spring and secondarily in autumn, i.e. when there is strong transport of dust. In spring and autumn, when the frequency of occurrence of dust episodes is highest, the C_v of the coarse mode is larger than that of the fine mode by up to 1.8 times (Table 2). The dominance, in terms of aerosol volume, of the coarse mode has also been reported by Smirvov et al. (2002) for Atlantic and Pacific AERONET sites. The very large coarse-to-fine ratio of columnar volume concentration in winter (2.3) is attributed to strong processing of aerosols by winter clouds, leading to broadening of aerosol spectra towards larger sizes and also to the presence of mineral dust and maritime particles, as suggested by back-trajectory analyses. Nevertheless, note that only 12 cases were available for analysis from FORTH-AERONET during this season. The maximum total C_v in Crete occurs in spring (0.117 $\mu m^3/\mu m^2$, Table 2), while the minimum (0.082 $\mu m^3/\mu m^2$) occurs in winter.

[4] C_v was computed as: $C_v = \int_{r_{min}}^{r_{max}} \frac{dV}{d \ln r} d \ln r$

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3.5 Relationships between aerosol optical thickness, Angström parameter and wind conditions

In this section, the aerosol optical properties are analysed in relation to the prevailing wind conditions at the FORTH-AERONET station in Crete. The wind measurements at the FORTH-AERONET station over the study period, indicate two prevailing directions: northwest and south. The prevailing winds are of southerly from November to February, whereas they become northwesterly from March to October. Daily mean wind speed as function of wind direction exhibit maximum values for northern and southern directions, equal to 5.2 and 4.4 $m\,s^{-1}$, respectively. Figure 8 shows AOT_{340}, AOT_{870} and Angström parameter $\alpha_{440-870}$, as function of wind direction, on a daily basis, over the study period. The corresponding diagrams for AOT_{500} and AOT_{1020} were found to be similar. It is evident that the maximum values of AOT occur for wind directions from easterly to southerly (90°–180°) and from the northwest. Nevertheless, note that NW wind directions are absent from the AOT_{870} diagram (Fig. 8b), but they exist for AOT_{340} (Fig. 8a). This indicates that NW winds are associated with maximum loads of fine aerosol particles, while eastern, southern, and southeastern (SE) wind directions are related to maximum loads of coarse aerosols (sea-salt and particularly dust). These conclusions are strongly supported by Fig. 8c indicating maximum values of $\alpha_{440-870}$ (up to 1.5) for NW directions, and minimum values for SE winds. The coarse aerosol particles from southern, southeastern and eastern directions are essentially dust (advected mainly from North Africa deserts, and much less from the Middle East) and also background maritime aerosols, without excluding mixtures of both aerosol types. Our analysis shows that only 1% of the total number of days with coarse aerosols are characterised by eastern wind directions. This implies that most of dust aerosols are transported from Africa under cyclonic situations centered southwards from Crete, especially in North Africa. This conclusion is also supported by back-trajectory analyses for the relevant days, which have shown air-masses originating from Africa. Nevertheless, care must be taken in attempting to analyse wind measurements in combina-

tion with back-trajectories; wind measurements are instantaneous at the surface layer, while back-trajectories go back to a few days and are obtained at different levels in the atmosphere. The fine particles associated with NW winds, are anthropogenic pollution transported from industrialised northwestern European countries also including the Athens area. Note also that relatively large values of AOT_{340} (~0.35), along with low AOT_{870} values for northeastern (NE) to north wind directions, suggest fine pollution aerosol particles originating from the Balkans, former Soviet Union countries around the Black-Sea and Turkey. Cleaner conditions (i.e. smaller aerosol loads) are found for S-SW and N-NE wind directions.

4 Summary and conclusions

In this paper we have characterised the aerosol physical and optical properties over the eastern Mediterranean basin using, for the first time, a complete series of two-year (2003–2004) measurements from the recently established FORTH-AERONET station in Crete. This has been achieved by examining aerosol spectral optical thickness (AOT), Angström parameter $\alpha_{440-870}$, and properties of aerosol spectra such as volume size distribution and effective radius.

The amount and properties of aerosols in the eastern Mediterranean basin/Crete were found to be strongly determined by the marine environment involving low concentrations of sea-salt aerosols produced by sea-spray, which constitute the background conditions. This results in relatively small AOT values, equal to 0.34±0.14, 0.21±0.11 and 0.12±0.09 at 340, 500 and 870 nm, respectively, on a yearly basis. The AOT values are also influenced by the prevailing synoptic conditions implying transport of dust aerosols mainly from African deserts, and secondarily from the Middle-East and Anatolian plateau, and transport of pollution fine aerosols from central and east Europe, Turkey and west Europe.

Aerosol optical thickness varies seasonally with maximum values in spring (March-May), mainly due to frequent dust transport from Africa, and minimum values in winter,

with a secondary maximum in late summer and autumn (August and October). The spring and autumn maximum in AOT are due to dust episodes, as supported by corresponding Angström parameter values less than 0.5. The seasonal variation of AOT is more pronounced in the UV-visible wavelengths than in the near-infrared. The relatively large AOT_{340} and AOT_{500} values during summer, associated with large values of $\alpha_{440-870}$ (>1.6) indicate the presence of urban/industrial and biomass burning fine aerosols transported and accumulated in the atmosphere of Crete due to stable meteorological conditions and the absence of wet removal processes.

The measured spectral values of AOT vary within the range 0.02–1.17, whereas the values of $\alpha_{440-870}$ range from 0.05 to 2.18, indicating a large variability in aerosol content and particle size. The strong short-term temporal variability of AOT and Angström parameter is attributed to short-lived dust storm episodes occurring primarily in spring and secondarily in autumn, and to the transport of continental pollution and biomass burning aerosols during summer. The largest values of $\alpha_{440-870}$, up to 2.2, in summer 2003 were due to forest fires that took place in southern European countries, having produced fine smoke particles that were transported over the eastern Mediterranean. A large variety of aerosol types have been identified over Crete, as indicated by broad frequency distributions of spectral AOT and $\alpha_{440-870}$ values, resulting from aerosol transport from European, African and Asian countries.

The Angström parameter $\alpha_{440-870}$ versus AOT_{870} shows significant scatter, involving values of $\alpha_{440-870}$ from about zero up to 2.2, and AOT from 0.02 up to 0.7. The points with large AOT_{870} and $\alpha_{440-870}$ <0.5 are associated with coarse aerosols, namely dust or maritime ones, or mixtures of them, while those with $\alpha_{440-870}$>2.0 correspond to fine particles either anthropogenic or biomass burning. Values of $0.5<\alpha_{440-870}<2.0$ correspond to mixtures of fine and coarse particles that coexist within the atmospheric column either in different layers or in within the same layer. These conclusions, derived solely from optical aerosol properties have been supplemented by back-trajectory analyses identifying the source areas and pathways of air masses carrying aerosols to Crete. Thus, the origins of air masses were found to be the Atlantic Ocean, north Africa,

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and Europe. The trajectories were found to be quite long, and sometimes complicated, involving also close pollution sources such as Athens.

The volume size distribution of aerosols is bimodal, typical of maritime environments, and indicate the presence of both fine and coarse aerosols over Crete. The fine mode peaks at 0.11–0.15 μm depending on season, with an effective radius of about 0.13 μm, while the columnar volume of aerosols, C_v, ranges from 0.025 in winter to 0.048 in summer. The coarse mode has two pseudo-modes centered at 1.7 and 3.9 μm. From our analysis, and the results of experimental campaigns, the first mode (at 1.7 μm) is attributed to maritime aerosols and the second mode (at 3.9 μm) to dust aerosols. The first pseudo-mode becomes dominant in winter, in contrast to summer, when the dominant mode peaks at 3.9 μm, whereas in the transient seasons the two pseudo-modes are about equal. The ratios of C_v for fine and coarse particles, indicate that coarse aerosols dominate the total columnar volume of particles during all seasons except for summer, when they are equal. The columnar volume of fine aerosols obtains maximum values in summer because of accumulated anthropogenic and natural fine pollution particles, whereas for coarse particles it is maximum in spring, and secondarily in autumn, i.e. when there is strong transport of dust.

It was found that maximum values of AOT occur for eastern, southeastern and southern wind directions, as well as for northwest winds. The maximum aerosol loads corresponding to NW winds are associated with maximum loads of fine aerosol particles, while maximum loads for eastern, southeastern, and southern winds are related to coarse (basically dust) particles. These conclusions are supported by maximum values of $\alpha_{440-870}$ (up to 1.5) for NW wind directions, and minimum ones for SE winds. Back-trajectory analysis has shown that dust particles are mainly advected from northern African deserts, and much less from Middle East, whereas the fine particles associated with NW winds, are anthropogenic pollution transported from western Europe, but also from Athens. They were also found relatively large values of AOT_{340} (~0.35), along with low AOT_{870} and large $\alpha_{440-870}$ values for NE to N wind directions, suggesting transport of fine pollution aerosols from the Balkans, former Soviet Union countries

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near the Black-Sea and Turkey.

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Table 1. Monthly mean values of aerosol optical thickness (AOT) at 340, 500, 870 and 1020 nm, and Angström parameter $\alpha_{440-870}$ at the FORTH-AERONET station in Crete, averaged over the 2-year period 2003–2004. The monthly means were computed from mean daily values, and are accompanied by corresponding standard deviations. In the last row are given the corresponding mean annual values for the entire study-period, whereas the numbers in parentheses correspond to minimum and maximum daily value for each parameter.

Month	AOT_{340}	AOT_{500}	AOT_{870}	AOT_{1020}	$\alpha_{440-870}$
January	0.15±0.05	0.10±0.03	0.07±0.02	0.07±0.02	0.68±0.45
February	0.22±0.09	0.15±0.08	0.10±0.07	0.10±0.07	0.87±0.51
March	0.42±0.23	0.29±0.20	0.17±0.18	0.16±0.17	1.16±0.51
April	0.33±0.14	0.22±0.11	0.14±0.10	0.13±0.10	0.99±0.47
May	0.35±0.12	0.24±0.12	0.17±0.12	0.16±0.12	0.93±0.52
June	0.35±0.12	0.20±0.08	0.10±0.06	0.09±0.06	1.51±0.40
July	0.38±0.13	0.22±0.08	0.10±0.05	0.09±0.05	1.48±0.43
August	0.41±0.11	0.23±0.08	0.10±0.06	0.09±0.06	1.55±0.39
September	0.37±0.11	0.22±0.08	0.12±0.07	0.11±0.07	1.26±0.47
October	0.33±0.12	0.22±0.10	0.14±0.09	0.14±0.09	0.93±0.55
November	0.29±0.17	0.19±0.12	0.11±0.07	0.10±0.06	1.08±0.38
December	0.23±0.09	0.16±0.08	0.11±0.08	0.11±0.07	0.79±0.35
Year	0.34±0.14 (0.06–1.17)	0.21±0.11 (0.05–0.91)	0.12±0.09 (0.02–0.84)	0.11±0.09 (0.02–0.80)	1.17±0.53 (0.05–2.18)

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Table 2. Seasonal averages of aerosol volume size distribution parameters at the FORTH-AERONET station in Crete; R_{eff} is the effective radius (in μm) and C_v is the columnar volume of particles per unit cross section of atmospheric column ($\mu m^3/\mu m^2$).

	Fine mode		Coarse mode		Fine + Coarse
	R_{eff} (μm)	C_v ($\mu m^3/\mu m^2$)	R_{eff} (μm)	C_v ($\mu m^3/\mu m^2$)	C_v ($\mu m^3/\mu m^2$)
Winter	0.131	0.025	2.004	0.057	0.082
Spring	0.128	0.042	2.026	0.075	0.117
Summer	0.132	0.048	2.322	0.046	0.094
Autumn	0.135	0.037	2.141	0.066	0.103
Year	0.132	0.038	2.123	0.061	0.099

Fig. 1. The Eastern Mediterranean basin with the FORTH_CRETE AERONET station located in Crete-island.

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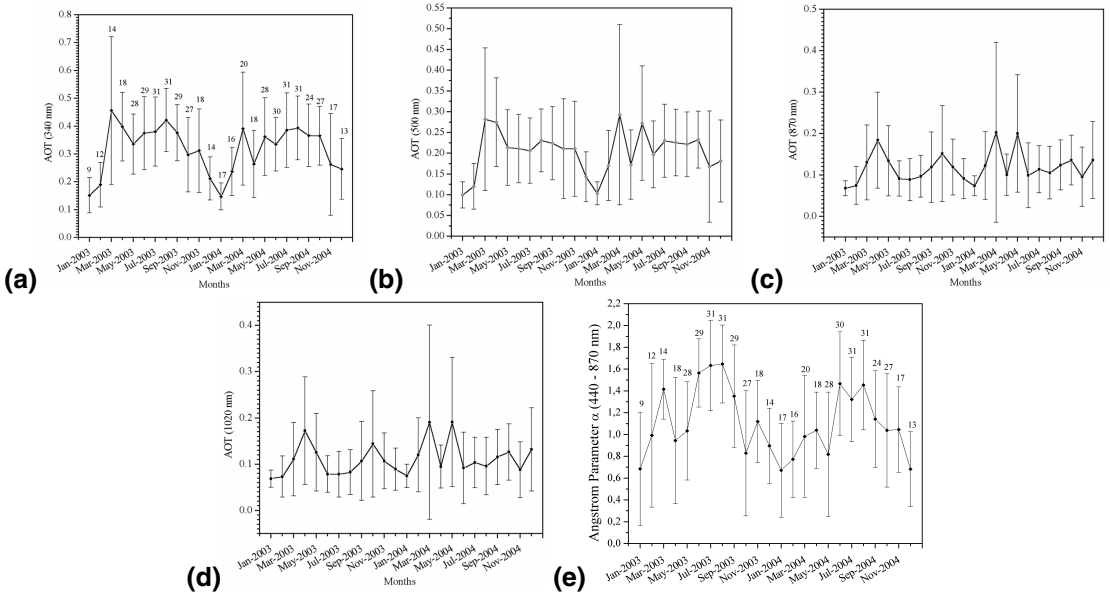

Fig. 2. Time-series of computed monthly mean aerosol optical thickness (AOT) at **(a)** 340 nm, **(b)** 500 nm, **(c)** 870 nm, and **(d)** 1020 nm, and of the Angström parameter, $\alpha_{440-870}$, derived from AOT measurements at 440 and 870 nm **(e)**, for the two-year period 2003–2004 at the FORTH-AERONET station in Crete. Standard deviation ($\pm\sigma$) values are also given (vertical bars) along with the number of days over which the monthly means are computed.

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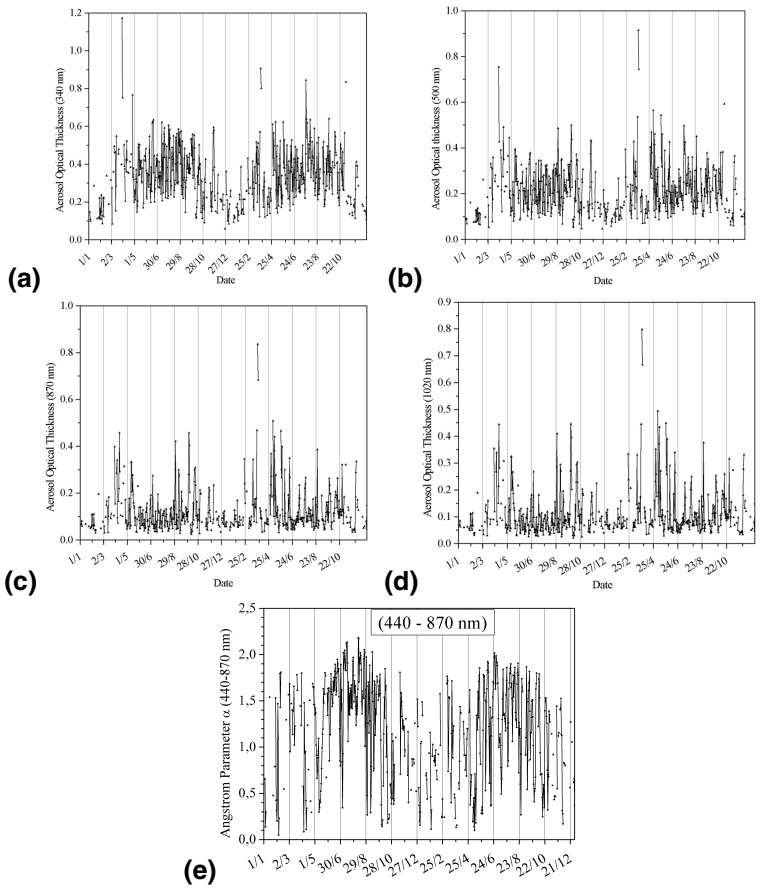

Fig. 3. Time-series of daily mean AOT values at **(a)** 340 nm, **(b)** 500 nm, **(c)** 870 nm, and **(d)** 1020 nm and of Angström parameter, $\alpha_{440-870}$, derived from AOT at 440 and 870 nm **(e)**, for the two-year period 2003–2004 at the FORTH-AERONET station in Crete.

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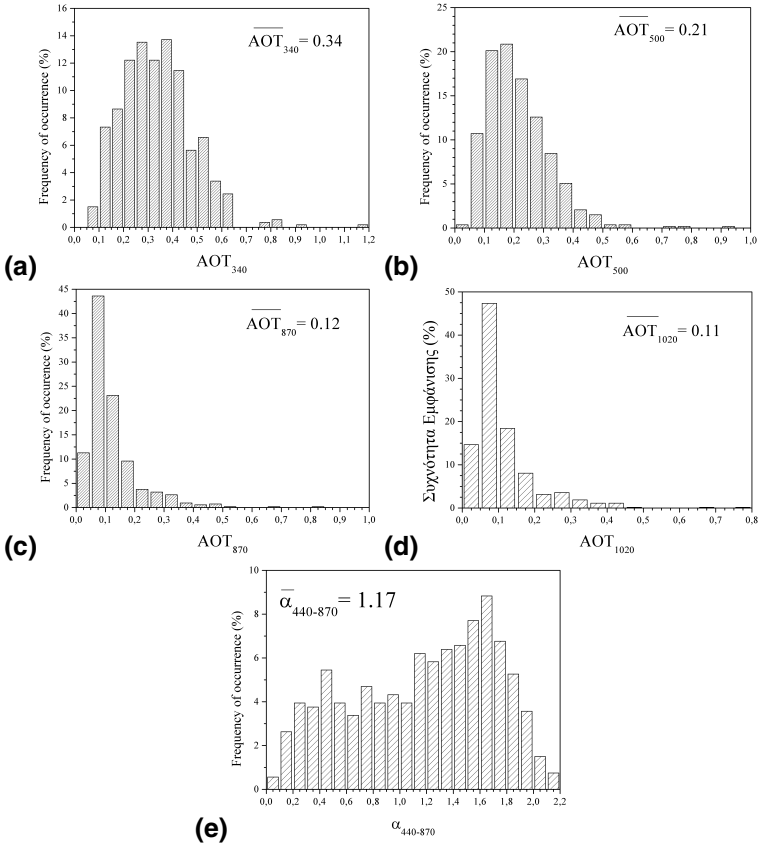

Fig. 4. Percent frequency of occurrence (%) of daily mean aerosol optical thickness (AOT) at **(a)** 340 nm, **(b)** 500 nm, **(c)** 870 nm, **(d)** 1020 nm, and **(e)** Angström parameter ($\alpha_{440-870}$), measured at the FORTH-AERONET station in Crete, for the 2-year period 2003–2004.

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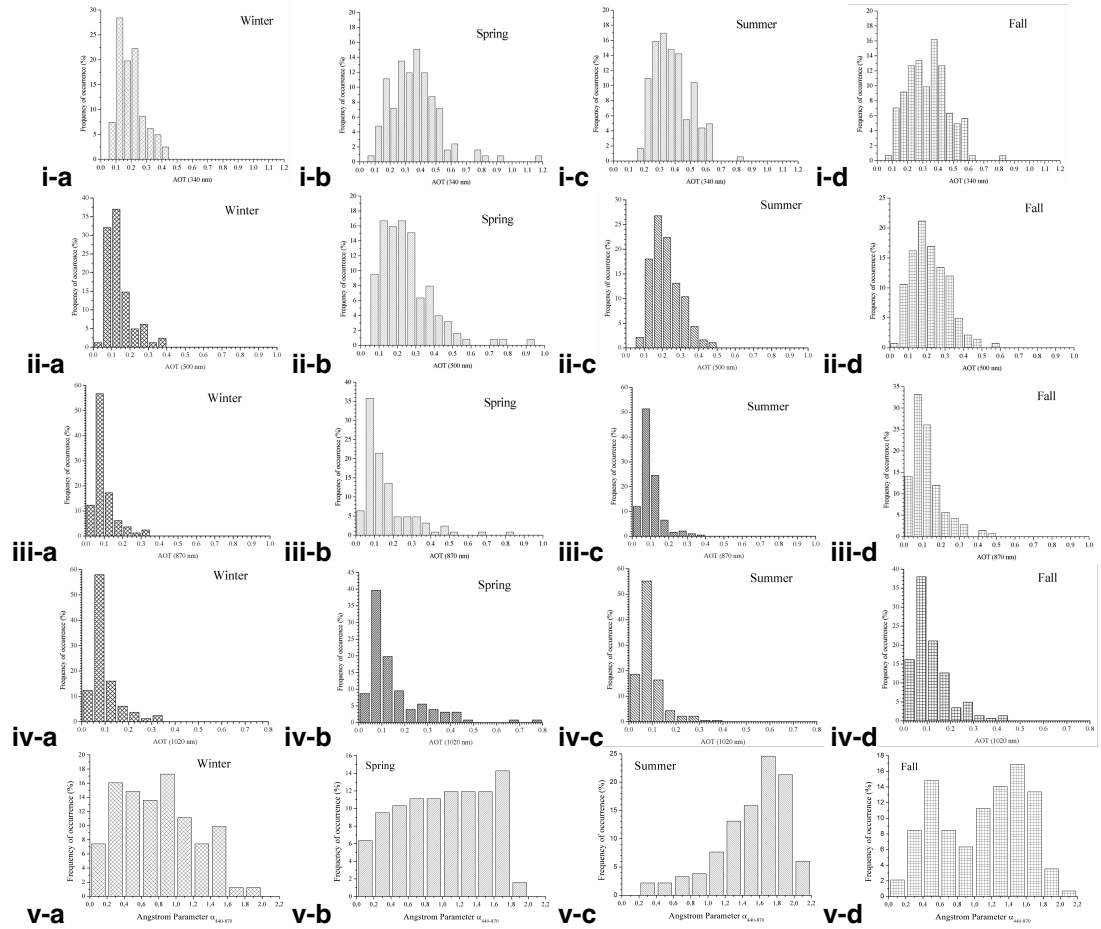

Fig. 5.

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Fig. 5. Seasonal frequency of occurrence (%) of daily mean aerosol optical thickness (AOT) at **(i)** 340 nm, **(ii)** 500 nm, **(iii)** 870 nm and **(iv)** 1020 nm, and **(v)** of Angström parameter ($\alpha_{440-870}$) measured at the FORTH-AERONET station in Crete, for the 2-year period 2003–2004 during **(a)** winter (December–January–February), **(b)** spring (March–April–May), **(c)** summer (June–July–August) and **(d)** autumn (September–October–November).

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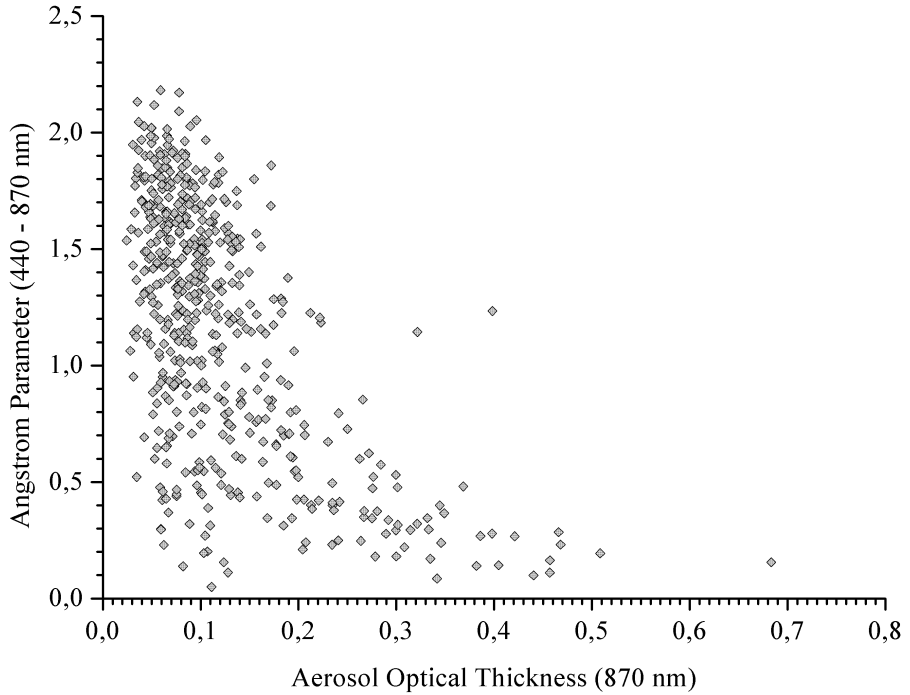

Fig. 6. Scatterplot of Angström parameter $\alpha_{440-870}$ versus aerosol optical thickness at 870 nm (AOT_{870}).

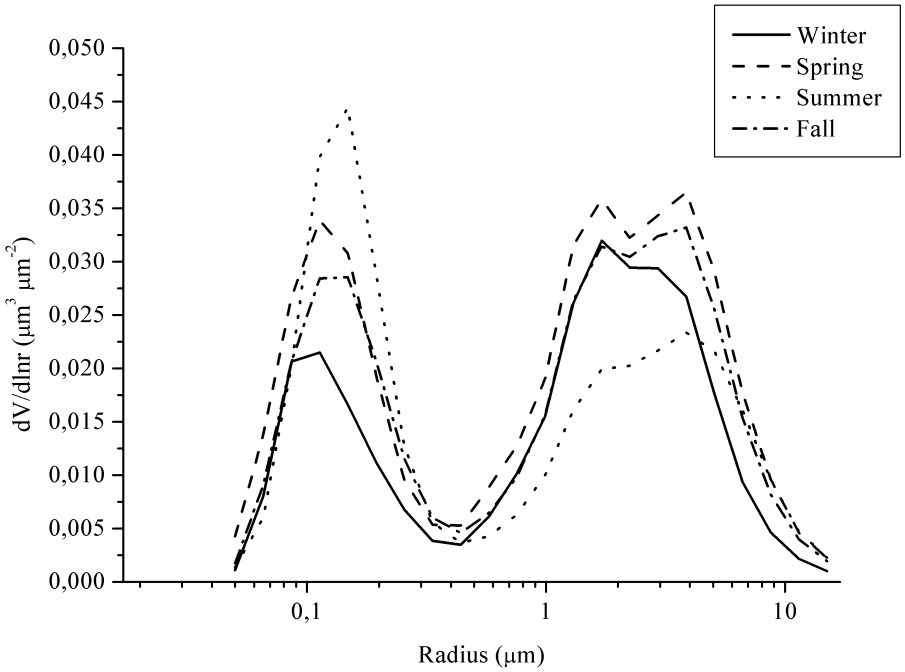

Fig. 7. Seasonal variation of aerosol columnar volume size distribution at the FORTH-AERONET station in Crete.

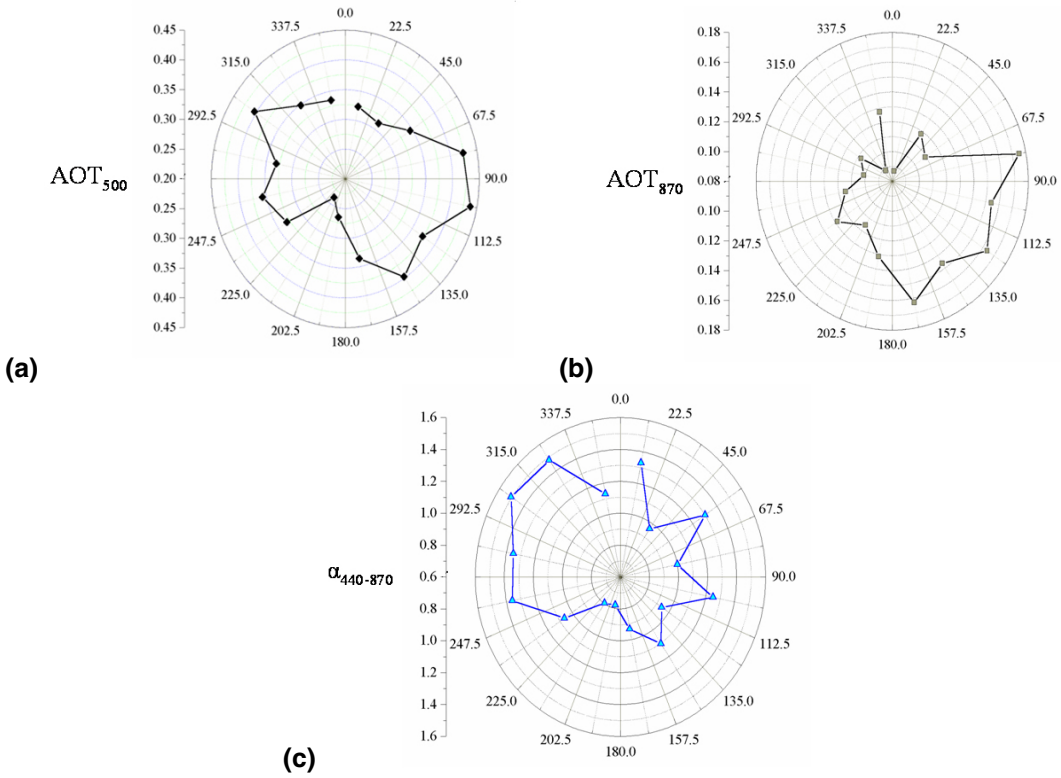

Fig. 8. Rose diagram of daily mean **(a)** aerosol optical thickness at 340 nm (AOT_{340}), **(b)** at 870 nm (AOT_{870}), and **(c)** of Angström parameter, $\alpha_{440-870}$, as function of wind direction at the FORTH-AERONET station in Crete.

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