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Aerosol climatology and discrimination of different types over Athens, Greece, based on MODIS data

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Abstract

A long-term (2000–2005) monitoring of aerosol data from the moderate resolution imaging spectroradiometer (MODIS) is analyzed focusing on the Greater Athens Area (GAA) in the Eastern Mediterranean region. The MODIS aerosol optical depth standard product (AOD at 550 nm) and its respective ratio attributed to fine-mode (FM) particles are employed to evaluate the seasonal variability of the aerosol properties over Athens. The climatological trend of both parameters in the period 2000–2005 is nearly absent, while remarkable year-to-year variability can be observed. The seasonal analysis reveals a significant AOD variability over Athens, with minimum values in winter (AOD_{550}~0.2), and maximum in summer (AOD_{550}~0.45). Regarding the FM fraction, maximum values are present in spring and minimum in summer, thus revealing the dominance of FM and coarse-mode particles, respectively. For the whole data set, a method is implemented to distinguish the main aerosol types (urban/industrial (hereafter UI), clean maritime (hereafter CM type) and desert dust (hereafter DD) over Athens, based on both AOD and FM values. Because of the mixing processes in the atmosphere the majority of the cases (46.6%) belong to a mixed (hereafter MT) aerosol type. The UI aerosols are more frequent in spring (41.2%) and less in winter (9.1%), while the coarse particles, probably DD, more frequent in summer (35.8%) and less in winter (3.5%). In contrast, the clean atmospheric conditions are more frequent in winter (23.9%), when the mixing processes are also well established (63.5%). For each aerosol type, the mean AOD_{550} and FM values are also computed. Their seasonal variability exhibits a clear summer maximum for UI, CM and MT aerosols, while the DD exhibits maximum in spring. As regards the FM values of the different aerosol types they exhibit a rather constant variation with small fluctuations from season to season.

Keywords: Aerosol optical depth; Fine-mode fraction; MODIS; Aerosol types; Athens; Greece

1. Introduction

It is known that optical and physical properties of atmospheric aerosols, such as optical depth, particle size, single scattering albedo, etc., play an important role in the Earth's climate and radiation budget

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(IPCC, 2001). Furthermore, Greece is one of the most interesting regions for aerosol studies, because soil dust from the neighboring North African desert areas, and the emission of local aerosols, such as sulfuric, nitric and carbonaceous particles, are increasing due to rapid urbanization. Therefore, the location of Athens offers a unique opportunity to monitor aerosols from different sources. The satellite-derived aerosol information is useful for indicating air quality on a global scale. The combination of radiometric aerosol information and the surface-level particulate mass is also useful for studying air quality and aerosol properties. Previous studies (Hutchison, 2003) have shown that moderate resolution imaging spectroradiometer (MODIS) data and products can detect and track the migration of pollutants. Moreover, they analyzed some cases to show the usefulness of remote sensing in monitoring air quality in urban areas.

Till now there are still significant uncertainties related to the aerosol physical and optical properties and to their response to global climate, due to their large spatio-temporal variability and the complex interactions between them, solar radiation and clouds (direct and indirect effects), in terms of their physical and chemical properties (IPCC, 2001). In order to improve the scientific knowledge about these issues, a worldwide effort has been undertaken in the last two decades to produce a global aerosol climatology by combining satellite-based observations (such as TOMS, MODIS, MISR, POLDER, SPOT, SCIAMACHY) and measurements from ground-based monitoring networks (AERONET, EARLINET, BSRN, GAW). These data sets are complemented with those of field campaigns (ground-based and airborne), for calibration and validation of satellite data (Chu et al., 2002; Remer et al., 2002) or associated with model (GOCART) simulations (Chin et al., 2004).

Due to the variety of the regions surrounding the Mediterranean, different types of particles can be found throughout the basin, having both strong temporal and spatial variability (Antoine and Nobileau, 2006; Barnaba and Gobbi, 2004): desert dust (hereafter DD), originated from Sahara, polluted aerosols produced mainly in industrialized areas of Europe, aerosols formed over Mediterranean Sea or transported from the North Atlantic and smoke from biomass burning often produced from seasonal forest fires. The various aerosol types display different optical and physico-chemical characteristics. As a consequence, the Mediterranean Sea has received recently a great scientific interest regarding the aerosol monitoring; many experimental and modeling studies have taken place (Formenti et al., 2001; Gerasopoulos et al., 2003; Kalivitis et al., 2007; Lelieveld et al., 2002; Meloni et al., 2007; Papayannis et al., 1998, 2005; Pace et al., 2006; Zerefos et al., 2006). The Eastern Mediterranean and the Aegean Sea is a crossroad where aerosols from different sources and mixtures of different kinds of particles converge (urban pollution, maritime aerosols, DD particles and seasonal biomass burning). The significant aerosol loads in the Eastern Mediterranean strongly influence the local radiation budget, and hence the climate of the region, especially during summer (Hatzianastassiou et al., 2004). The majority of the studies are based on observations carried out in the coastal Mediterranean regions (Formenti et al., 2001; Israelevich et al., 2003; Perrone et al., 2005), as well as in coastal Greece (Amiridis et al., 2005; Balis et al., 2003; Gerasopoulos et al., 2003; Kalivitis et al., 2007; Kaskaoutis and Kambezidis, 2006b; Papayannis et al., 2005; Fotiadi et al., 2006). Analyses based on satellite observations were also carried out to identify the transport of Saharan dust (Antoine and Nobileau, 2006; Dulac et al., 1992; Israelevich et al., 2002; Moulin et al., 1998).

In the present study aerosol measurements from the EOS-Terra-MODIS sensor are analyzed to characterize the aerosol properties over the Greater Athens Area (GAA). The MODIS aerosol data include aerosol optical depth at 550 nm (AOD_{550}) and the fine-mode (FM) fraction, covering the 6-year period 2000–2005, allowing the annual aerosol cycle to be studied. The main goal of this article is the analysis of the seasonal variation of aerosol optical properties and the discrimination of different aerosol types, which dominate the Athens atmosphere in specific seasons and under the influence of favorable meteorological conditions. This work is among the first long-term aerosol climatology studies over Athens. A previous interesting effort for the investigation of the dust transport over Athens and its vertical distribution has been conducted by Papayannis et al. (2005) in the frame of the EARLINET project covering the period 2000–2002. The present study also focuses on additional aerosol types (like urban/industrial (hereafter UI) or maritime particles) further investigating their properties and seasonality over the Athens basin.

2. Climatology of the area

The GAA is located in an oblong basin that is characterized by a complex topography and constitutes an area of $\sim$450 km^2. The basin is surrounded by mountains at the three sides and by the Saronikos Gulf in the South (see topography map in Kaskaoutis and Kambezidis, 2006a). This complex topography gives a result of poor air mass mixing within the basin (Kambezidis et al., 1995). Approximately 3.5 million inhabitants (census 2001) reside within the GAA, which represents about one-third of the country's population. High levels of air pollutant concentrations have been reported over the last two decades within the Athens basin (Kambezidis et al., 1995). Therefore, Athens' air quality has been of great concern for the Greek Ministry of Environment.

The synoptic meteorological pattern over the region during winter is affected by: (a) the Siberian anticyclone causing polar continental air flow from Northeastern Europe, (b) an anticyclone over Middle and Eastern Europe and (c) the low-pressure systems in the Western Mediterranean and Atlantic Ocean, moving from West to East. These regimes, in conjunction with the higher precipitation in this season, result in lower AODs and more transparent atmospheric conditions. During summer Greece, and generally the Eastern Mediterranean basin, is under the influence of two pressure systems that drive the synoptic circulation pattern: the quasi-permanent Azores anticyclone and the Indian thermal low, which extents to Middle East and South of Cyprus (Metaxas and Bartzokas, 1994; Katsoulis et al., 1998). Thus, a strong pressure gradient is created over the Aegean Sea, resulting in Northerly "Etesian" winds. It was found that under the influence of the Etesian winds the turbidity levels over Athens decrease rapidly (Adamopoulos et al., 2000). During late spring and early summer calms or presence of the sea-breeze circulation when the synoptic flow is weak is frequently associated with high levels of anthropogenic aerosols and air pollution. The sea breeze and calms favor the accumulation of pollutants in the Athens atmosphere and the formation of photochemical smog, a brown cloud over Athens called "nephos" (Kambezidis et al., 1995).

3. Data set

The MODIS was launched in December 1999 on the polar orbiting NASA-EOS-Terra spacecraft. Terra's sun-synchronous orbit has a dayside equatorial 10:30 am local crossing time. Since February 2000, MODIS data are acquired in 36 spectral bands from visible to thermal infrared (29 spectral bands with 1-km, 5 spectral bands with 500-m and 2 with 250-m nadir pixel dimensions). Aerosol retrievals from MODIS data are performed over land and ocean surfaces by means of two separate algorithms thoroughly described in Kaufman and Tanrè (1998). Aerosol products are stored in MODIS Level 2 (MOD04_L2) files, each corresponding to 5-minute acquisition along the satellite orbit. The two aerosol products employed in the present study are: (1) the optical thickness at 550 nm (AOD_{550}) and (2) the FM fraction. The latter is derived as the ratio of optical depth of small mode versus effective optical depth at 550 nm. Both products are given at a spatial resolution of 10×10 km ($1^\circ \times 1^\circ$). The MODIS aerosol products are only performed over cloud-free regions. Both AOD_{550} and FM data correspond to collection 5 (c005) data, where much of the high bias is removed (at least on a global average; R. Levy, personal communication). However, in dust aerosol regimes, the retrieved AOD_{550} will have greater error due to the non-spherical particles shape (Remer et al., 2002). The FM fraction was initially given over oceans. Nevertheless, it can also be obtained over coastal areas. Over land, the retrieval is made by combining radiances from two aerosol "models" that each contains two or more "modes". For example, over Athens, the algorithm chooses a UI model for the "fine-dominated" mode and a dust model for the "coarse-dominated" mode (R. Levy, personal communication).

Chu et al. (2003) showed that separation of FM and coarse-mode particles is also possible over land by comparing the path radiances at 660 and 470 nm. As discussed in detail in the MODIS aerosol products validation studies (Chu et al., 2002; Remer et al., 2002), a different accuracy is associated to the MODIS AOD_{550} retrievals over land ($\pm 0.05 \pm 0.2$AOD) and ocean ($\pm 0.03 \pm 0.05$AOD). Over land, even larger errors can be found in coastal zones (such as Athens) due to subpixel contamination. This effect tends to produce AOD overestimation (Chu et al., 2003). Similarly, a significant watercolor contribution can reduce the ocean AOD retrieval quality in coastal areas. With tests performed on Mediterranean sites, Remer et al. (2002) also indicated the particle size-dependent parameters (such as FM) to be retrieved with an accuracy within ±25%.

The MODIS aerosol standard products (AOD_{550} and FM) are provided in a spatial resolution of 10×10 km. In order to reduce the aforementioned uncertainties, the MODIS products used in the present study refer to the mean AOD_{550} and FM values obtained from four pixels covering the area (36.5°−38.5°N and 22.5°−24.5°E) completely including the GAA. A total of 1804 daily data were collected from 26 February 2000 to 31 December 2005. The winter, spring, summer and autumn days represent 20.8%, 25.7%, 28.4% and 25.1%, respectively, of the whole data set (Table 1). Thanks to the long-term period (2000–2005) the MODIS data are employed to evaluate the seasonal variability of the aerosols over the region, since the retrieval uncertainties are strongly reduced. There are fewer winter daily values (376 data) because of extent cloud cover, in contrast to the clear-sky conditions during summer (510 data). Also, there are some gaps in the measurement period, mainly due to the presence of clouds or to instrumental problems. The longer data gap is 15 days, lasting from the 15th to the end of June 2001. Other long-term gaps are observed in the periods 17–26/12/2003, 19–28/3/2002 and 6–18/8/2000.

4. Methodology

The method proposed by Barnaba and Gobbi (2004) is implemented here to distinguish the main aerosol types (UI, maritime and DD). Based on the combination of AOD_{550} and FM data, we separate the presence of the three aerosol types over Athens on seasonal basis. Fig. 1 shows the correlation of AOD_{550} versus FM in the period February 2000–December 2005. The correlation of the AOD with the size distribution allows, in general, the discrimination of the different aerosol types. This method has been used in a large number of studies and is based on the sensitivity of the two parameters to different, somewhat independent, microphysical aerosol properties; the FM fraction depends on the size of the particles, while the AOD_{550} depends mainly on the aerosol column density. Therefore, the AOD_{550}–FM plot qualitatively indicates the amount and dimension of the observed aerosols. Similar discrimination has been performed by Barnaba and Gobbi (2004) using the MODIS data too. The following considerations were taken into account to define the three-region limits shown in Fig. 1, corresponding to maritime, UI and coarse-mode, probably DD aerosols, respectively.

Table 1
Number of occurrences and mean AOD_{550} and FM values for each aerosol type

Aerosol type	Whole period	Winter	Spring	Summer	Autumn
Whole data set					
Number of occurrences	1804	376 (20.8%)	466 (25.7%)	510 (28.4%)	452 (25.1%)
AOD_{550}	0.35 ± 0.18	0.21 ± 0.10	0.41 ± 0.18	0.44 ± 0.18	0.25 ± 0.11
FM	0.69 ± 0.21	0.72 ± 0.15	0.77 ± 0.11	0.63 ± 0.12	0.68 ± 0.18
Urban/industrial (UI)					
Number of occurrences	442 (24.5%)	34 (9.1%)	192 (41.2%)	117 (22.9%)	99 (21.9%)
AOD_{550}	0.38 ± 0.13	0.31 ± 0.10	0.41 ± 0.15	0.45 ± 0.16	0.37 ± 0.11
FM	0.90 ± 0.06	0.92 ± 0.06	0.91 ± 0.06	0.88 ± 0.06	0.90 ± 0.06
Clean maritime (CM)					
Number of occurrences	183 (10.2%)	90 (23.9%)	15 (3.2%)	14 (2.7%)	64 (14.2%)
AOD_{550}	0.15 ± 0.03	0.13 ± 0.04	0.15 ± 0.03	0.17 ± 0.03	0.14 ± 0.04
FM	0.49 ± 0.13	0.47 ± 0.14	0.58 ± 0.10	0.42 ± 0.14	0.50 ± 0.16
Desert dust (DD)					
Numberof occurrences	337 (18.7%)	13 (3.5%)	40 (8.6%)	182 (35.8%)	102 (22.5%)
AOD_{550}	0.52 ± 0.19	0.57 ± 0.31	0.60 ± 0.24	0.46 ± 0.12	0.45 ± 0.10
FM	0.44 ± 0.12	0.43 ± 0.14	0.46 ± 0.11	0.43 ± 0.13	0.43 ± 0.12
Mixed type (MT)					
Number of occurrences	842 (46.6%)	239 (63.5%)	219 (47.0%)	197 (38.6%)	187 (41.4%)
AOD_{550}	0.31 ± 0.14	0.19 ± 0.10	0.36 ± 0.17	0.40 ± 0.15	0.29 ± 0.15
FM	0.70 ± 0.14	0.78 ± 0.13	0.72 ± 0.10	0.66 ± 0.14	0.65 ± 0.18

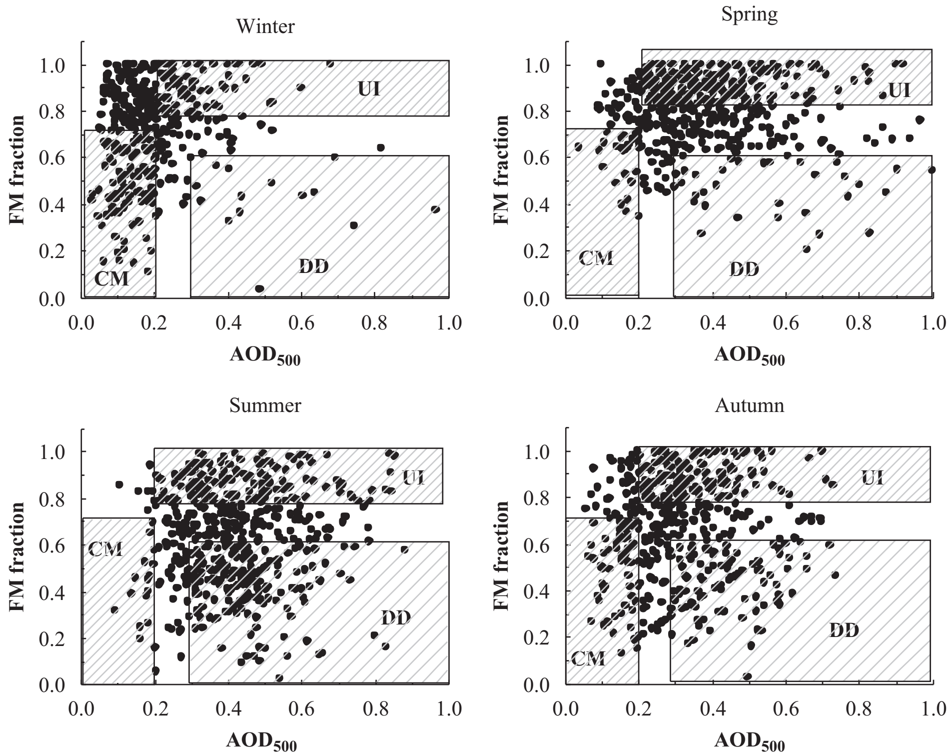

Fig. 1. Correlation of AOD_{550} versus FM fraction for the discrimination of the different aerosol types in each season. The three areas correspond to urban/industrial (UI), clean maritime (CM) and desert dust (DD). The cases outside these areas correspond to mixed or undetermined aerosol type (MT).

In contrast to remote oceanic regions, it is quite difficult to define "pure maritime conditions" in closed seas such as the Mediterranean and the Aegean. In the Mediterranean the clean background conditions ($AOD_{500}<0.1$) are rather rare, and "standard" atmospheres with a mean AOD_{500} in the range 0.2–0.3 (Smirnov et al., 2002) are most common. A mean AOD of 0.29 ± 0.22 was found when averaging the 11 AOD values referring to the Mediterranean Sea and collected by Smirnov et al. (2002) on the basis of 30 years of published data. This average value reduces to 0.18 ± 0.11 when AODs higher than 0.5 are not considered since they do not correspond to clear maritime (hereafter CM type) conditions. Taking into account the over-estimation of MODIS for AODs in land–ocean subpixels, 0.2 was chosen as the upper limit for maritime AOD_{550}, also corresponding to clear conditions. Due to the variability of both meteorological conditions (relative humidity, wind speed) and contamination sources, a significant variability is observed regarding the relative contribution of FM and coarse-mode particles in marine environments (Smirnov et al., 2002). Over the Atlantic and Pacific Oceans the fine-aerosol contribution to the total AOD was found to be as high as 70% (Kaufman et al., 2002). The present study maintains this threshold for the FM fraction for maritime aerosols. For the identification of UI aerosols, representative of light, moderate or severe pollution, the AOD_{550} values above 0.2 were associated with significant FM contribution to the total AOD (above 80%). The dust particles presented over whole Mediterranean are usually attributed to significant dust outbreaks having high AOD values (Pace et al., 2006; Papayannis et al., 2005; Tafuro et al., 2006). Also, these particles are associated with small fine-to-coarse ratio. Therefore, the threshold values for the DD aerosols were selected to be $AOD_{550}>0.3$ and $FM<0.6$.

Between the three portions some gaps reveal where the aerosols are difficult to be discriminated and they are taken as well-mixed type. This fact constitutes the main difference from Barnaba and Gobbi (2004) discrimination criteria. An additional "mixed (hereafter MT) aerosol type" was chosen bearing in mind the considerable effects of the various aerosol-mixing processes in the atmosphere (e.g. coagulation, condensation, humidification, gas-to-particle conversion). Therefore, these thresholds designate the areas in Fig. 1 which are classified as mixed type, but prevent from overpredicting the presence of either UI or DD particles over Athens. Because of its simplicity, Fig. 1 is associated to some arbitrariness (e.g. incorrect aerosol type interpretation), particularly at the borderlines of the three regions. A sensitivity analysis, which has been performed, showed that the definition of FM value of 0.7 instead of 0.8 for the UI aerosol type would increase its presence in spring about 15%. This is the largest difference regarding any change in the threshold values.

The scatterplot of AOD_{550} against FM is shown in Fig. 1 for each season. The wide range of FM values involves various types of particles, such as natural sulfate maritime aerosols, mineral dust or DD, soot, anthropogenic particles or photochemical pollution. The MT is also very common especially in winter. The absolute as well as the relative frequencies of each aerosol type for both the whole data set and each individual season are summarized in Table 1. The main findings from this table can be underlined to that in both whole data set and each individual season the MT dominates, exhibiting its maximum frequency of occurrence in winter (63.5%). The UI aerosol type is more frequent in spring (41.2%) due to most favorable atmospheric conditions (sea breeze and calms) dominated in late spring, while the DD type is more frequent in summer (35.8%). It was found (not presented here) that the UI aerosols are associated with Northern wind directions, carrying polluted air masses from Central, Eastern Europe and Balkan countries or local stagnant air masses enriched with industrial emissions. Both types (UI and DD) exhibit the least frequency of occurrence in winter (9.1% and 3.5%), respectively, while in this season the CM type presents maximum (23.9%).

5. Results and discussion

This section is divided into two parts: the first presents the 6-year aerosol climatology over Athens, and the second analyzes further the aerosol types. To our knowledge this is the first study utilizing such a long-term aerosol data set over Athens and discriminating the main aerosol types.

5.1. *Aerosol climatology*

In Fig. 2 the monthly mean AOD_{550} (lower part) and the FM (upper part) values are shown for the period February 2000–December 2005, associated with the standard deviations expressed by vertical bars. Both parameters exhibit a significant annual variability, but no remarkable trend is detected. The trend lines are also shown in the figure, which remain nearly constant in the entire period. As a consequence, both intercepts are similar to the mean AOD_{550} and FM values, 0.35 ± 0.18 and 0.69 ± 0.21, respectively (Table 1) of the 6-year period. In contrast to the climatology trend, there is a rather significant year-to-year variability in aerosol properties, AOD_{550} and FM. Despite the year-to-year variations it is obvious that the AOD_{550} annual pattern is rather bimodal with two peaks, the first in spring (March to May) and the second in early summer (July or August). The FM annual variation seems to be more complicated, not presenting an organized pattern. Despite this, a spring maximum and summer minimum may be identified.

The annual distribution of the monthly averages of AOD_{550} and FM fraction with their standard deviation (1σ) is shown in Fig. 3(a, b) for the examined period. The monthly AOD_{550} has a distinct annual cycle, with minima below 0.2 in winter, and spring/summer peaks between 0.4 and 0.5. These AOD_{550} values are generally higher than the respective data published for Athens in various periods (Giavis et al., 2005; Kaskaoutis and Kambezidis, 2006a). Nevertheless, in a variety of studies there has been established that MODIS overestimates the AODs derived by sun photometers (Ichoku et al., 2002; Chin et al., 2004). Moreover, the mean AOD_{550} values over Athens (0.35 ± 0.18) are higher than those in Northern Greece, 0.23 (Gerasopoulos et al., 2003), and FORTH-CRETE AERONET station, 0.21 (Fotiadi et al., 2006). This is attributed to the more polluted Athens urban atmosphere in conjunction with the slighter overestimation of the AODs obtained by MODIS. In contrast, the mean AOD_{550} values in Athens are comparable with those at Lampedusa, 0.35 (Meloni et al., 2007), mainly due to its proximity to the African coast. The AOD_{550} varies from 0.05 to 0.98 on daily basis and within the range 0.18–0.48 on a monthly basis. The evolution of FM fraction does not seem to have such a depicted annual variation, except for the lower summer

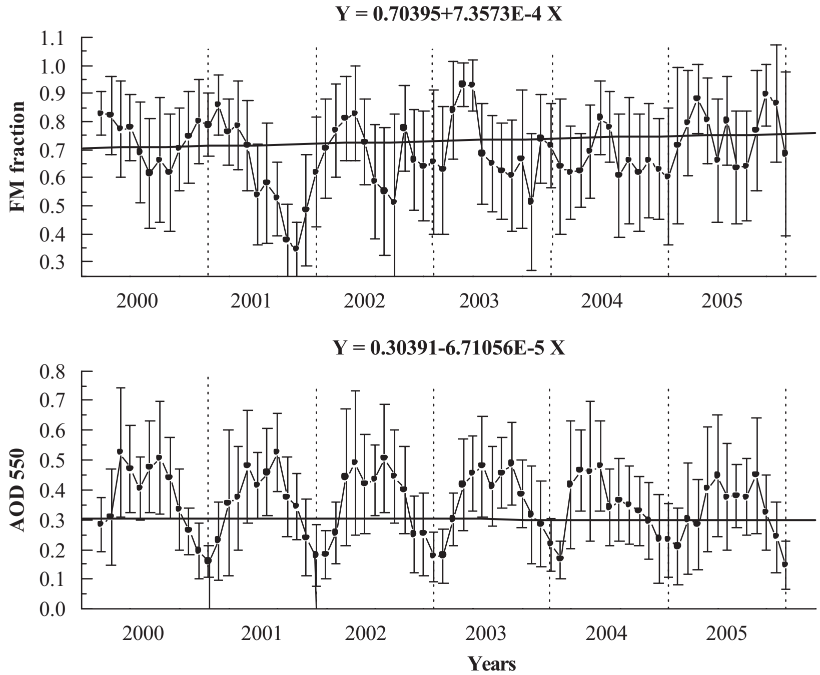

Fig. 2. Inter-annual variation of monthly mean AOD_{550} (lower part) and FM fraction (upper part) values derived from Terra-MODIS in the period February 2000–December 2005 over GAA. The vertical bars express one standard deviation from the monthly mean value.

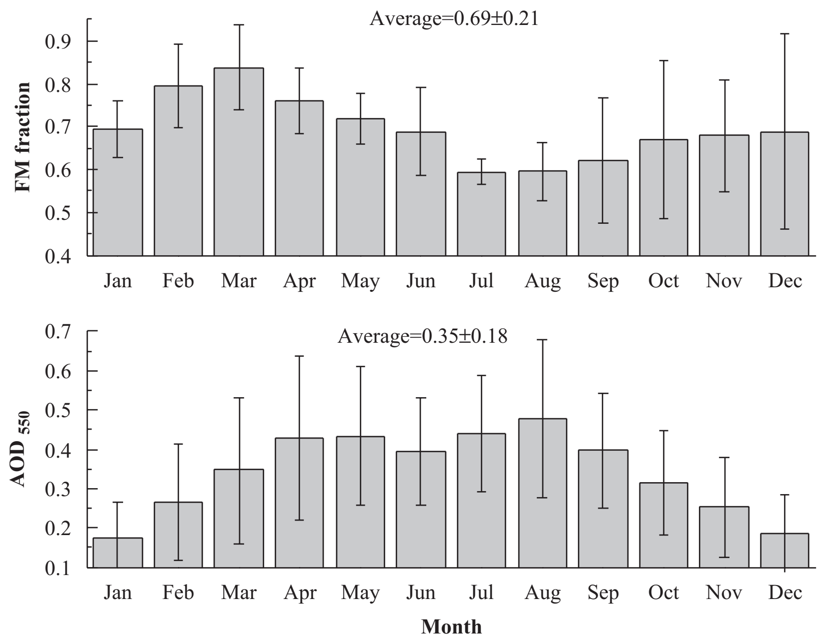

Fig. 3. Monthly mean variability of AOD_{550} (lower part) and FM fraction (upper part) values in the period February 2000–December 2005. The vertical bars express one standard deviation from the monthly mean value.

values, possibly related to the strong flow of the North "Etesian" winds, which uplifts mineral dust, favored by the dry soil conditions, and sea-spray particles. Moreover, the FM fraction can take values from low (0.61) to high (0.82) on monthly basis. Different types of particles are observed over

Athens, as suggested by the large variability of the aerosol particles, since the daily FM fraction values are comprised between 0.2 and almost 1.

The annual distribution of the monthly mean AOD_{550} values seems to be bimodal, with the absolute maximum (0.48 ± 0.20) in August and the secondary in April–May period. In contrast, June exhibits lower AOD_{550} values as well as lower standard deviations. Local minimum AOD values in June have also been reported for the FORTH-CRETE AERONET station. This may be attributed to the synoptic systems developed in this season in Eastern Mediterranean (Fotiadi et al., 2006). The high AOD_{550} values in late summer and early fall in conjunction with small values of FM fraction indicate the predominance of coarse particles, probably dust transported from African deserts or sea spray lifted by the strong "Etesian" winds. In late spring, large AOD_{550} values, together with relatively large FM fraction, are associated with fine aerosols produced mainly from local activities.

The low monthly mean AOD_{550} values in winter, below 0.2, together with FM fraction values about 0.7, indicate near-background aerosol conditions dominated in a great fraction by maritime aerosols. This is in agreement with measurements performed during winter in Crete (Fotiadi et al., 2006) and in the whole Mediterranean (Antoine and Nobileau, 2006; Barnaba and Gobbi, 2004) that indicate a significant contribution of sea salt compared to dust. The presence of anthropogenic FM aerosols over Athens, especially in spring, is supported by the corresponding monthly means FM fraction values of 0.77 ± 0.06. In late spring, the sea-breeze circulation and calms favor the accumulation of local anthropogenic aerosols over the region. The plateau of high AOD_{550} values in the warm period (April–September) is probably related to the stable atmospheric conditions in the Eastern Mediterranean, which favor the accumulation of aerosol particles advected by long-range transport processes mainly from the North (Fotiadi et al., 2006). The maximum AOD_{550} values in spring and summer can also be associated with maximum production of maritime (sea-spray and sulfate) aerosols in the Eastern Mediterranean and Aegean Sea (Antoine and Nobileau, 2006), also mixed with dust particles.

The standard deviations of AOD_{550} are found to be largest during spring and late summer, when AOD_{550} is maximum, indicating strong day-to-day variability in aerosol load over Athens, also associated with frequent dust events. The large standard deviations of AOD_{550} can also be related to the wet and dry deposition processes, associated with local meteorological conditions. Furthermore, the standard deviations of FM fraction exhibit larger values in autumn probably due to the combined effects of dust events, wet removal processes and local anthropogenic activities.

From the literature, the high spring and late summer AOD values in the Eastern Mediterranean are associated with strong dust episodes taking place in these seasons (Antoine and Nobileau, 2006; Barnaba and Gobbi, 2004; Moulin et al., 1998; Fotiadi et al., 2006), when dust particles are transported from African deserts. This is also indicated by lower values of the Ångström exponent during spring and late summer. Nevertheless, these seasonal features are not depicted clearly over Athens using the MODIS AOD_{550} and FM fraction data set, since the AOD_{550} depicts a plateau of high values from April to September, while the FM fraction presents a remarked decrease during summer months. Therefore, it is believed that the aerosol climatology over Athens is driven not only from the Sahara dust outbreaks and the Mediterranean circulation patterns, but also from the local emissions and the industrial activities in the urban environment. In addition, it is worth mentioning a further factor playing a major role in determining the AOD's seasonal pattern. This factor is the precipitation, which is the most efficient removal process of atmospheric particulates. Therefore, the observed seasonal patterns of the AOD_{550} and FM fraction should also be interpreted in connection to the seasonal precipitation above Athens. In particular, even considering the mentioned increase of photochemical and convective activity in spring–summer period, the higher mean AOD_{550} values in this period constitute also a consequence of a minimum aerosol scavenging by precipitation. In contrast, the higher precipitation rate over Athens in the fall–winter period tends to reduce the residence time of aerosol particles in the atmosphere and also the mean AOD_{550}.

The relative frequency distribution of both AOD_{550} and FM fraction values is presented in Fig. 4(a, b), respectively. The frequency histograms of AOD_{550} are quite broad with values occurring even at above 0.8, especially in spring (see Fig. 1). Here for a better representation the AOD_{550} interval is limited to 0.8. The winter presents the narrowest AOD_{550} frequency distribution, with a maximum of

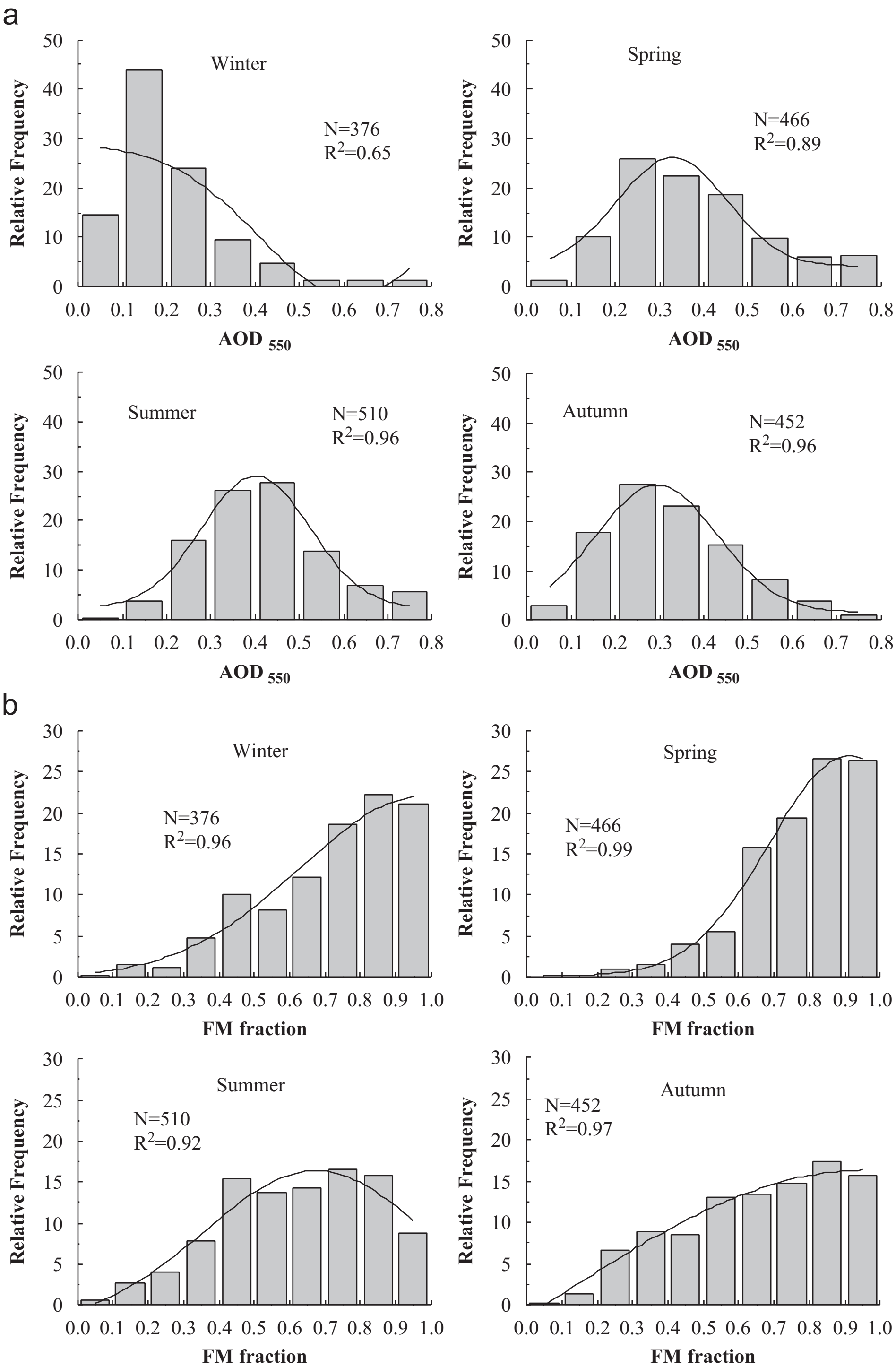

Fig. 4. Seasonal relative frequency distribution of the number of occurrences of (a) AOD_{550} and (b) FM fraction. The coefficient of determination (R^2) of the Gaussian curve fit and the number of data in each season (N) are also given in the graphs.

about 45% in the range 0.1–0.2 corresponding to background conditions. The frequency distribution is highly skewed towards lower values, far away from the Gaussian. In spring and summer, a large fraction (~70−80%) of the daily AOD_{550} values are almost equally distributed in the range 0.2–0.5, while values above 0.7 correspond either to strong dust outbreaks or to heavy pollution events. In spring, the frequency distribution broadens out to larger AOD_{550} values, directly affected by the intense dust events and the frequent occurrence of pollution episodes. A significant portion of spring daily values (23%) have $AOD_{550}>0.5$, and ~10% has $AOD_{550}>0.7$. In summer, the frequency of occurrence of AOD_{550} exhibits a broad peak within the range 0.3–0.5, yielding 57% of the total observations in this season, whereas another 7% has $AOD_{550}>0.7$. Note also the absence of AOD_{550} values below 0.1. In autumn the frequency distribution is broader exhibiting a slightly skewed curve at the lower AOD_{550} values. The peak is centered in the 0.2–0.3 interval, with steadily decreasing values at larger optical depths, quite similar to the spring distribution.

In all seasons the frequency distribution of the daily FM fraction values is broad, covering the range 0.1–1.0 due to the great variety of aerosol types (UI, biomass burning, maritime, DD). The frequency distribution of FM fraction in winter is skewed towards large values, with 45% of the cases having $FM>0.8$. Similar feature is also presented in spring, when the portion of cases with $FM>0.8$ increases to 54%. It is worth noticing that in this season the shortened FM fraction distribution is presented, highly skewed towards larger values. In contrast, similarities are revealed from the frequency distribution patterns in summer and autumn, when the broadest distributions of the daily FM values occur. Therefore, in both seasons there is almost equal frequency of occurrence in small $FM<0.5$ and large $FM>0.8$, revealing simultaneous occurrence of coarse-mode and FM aerosols. In summer, the frequency distribution of FM is rather uniform with values equally distributed in size bins from 0.5 to 0.9. In autumn, the frequency distribution reveals the predominance of the UI fine aerosols against the dust or maritime coarse particles, since the frequency of occurrences is slightly skewed towards larger FM values. All the FM distributions are limited by a cutoff threshold value of 1, thus representing only the left part of the Gaussian distribution. A different pattern is obvious in summer when the frequency of large (>0.8) FM values drops significantly.

5.2. Investigation on the different aerosol types

The previous analysis established that both AOD_{550} and FM fractions varied widely in the measurement period. This behavior indicates a great variety of aerosol types over the study region, such as UI and biomass-burning aerosols, DD and maritime particles as well as MT aerosol types. The mean AOD_{550} and FM fraction values for all aerosol types, both in the whole period and in each season, are shown in Fig. 5(a, b), respectively. Values of AOD_{550} and FM averaged over the whole measuring period are 0.38 ± 0.13 and 0.90 ± 0.06 for urban pollution, 0.15 ± 0.05 and 0.49 ± 0.13 for CM conditions and 0.52 ± 0.19 and 0.44 ± 0.12 for DD aerosols (Table 1). This distribution is essentially produced by the annual evolution of the meteorological synoptic patterns over the Eastern Mediterranean, with strong Northwestern and Western winds in winter as a consequence of the

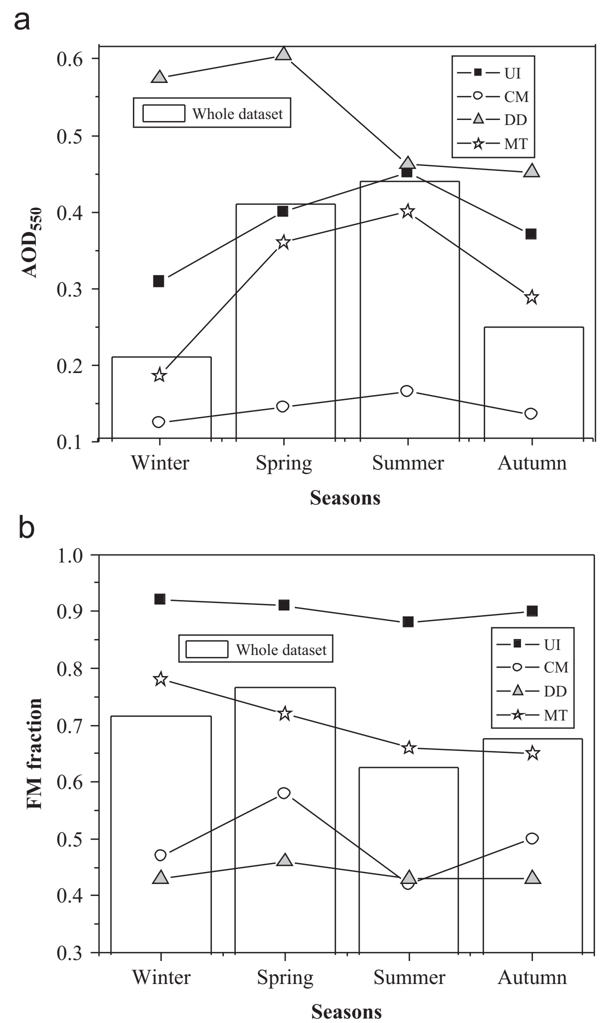

Fig. 5. Mean seasonal (a) AOD_{550} and (b) FM fraction values for the whole data set as well as for each individual aerosol type.

cyclonic circulation patterns, the ''Etesian'' winds in late summer and the Southwestern flow in late spring and summer.

The optical depth for the three main aerosol types (UI, CM and DD) exhibits a distinct seasonal pattern, having in general larger values in summer with the exception of desert aerosols, which present higher AOD_{550} values in spring, when the dust events are more intense. It should be noted that the term DD does not correspond to dust particles only but generally to coarse-mode aerosols. This is the reason why the lower summer values are mainly attributed to air masses from the Northern sector possibly carrying soil particles or sea spray during their trip above North Aegean Sea. It was found that only a fraction (28%) of these coarse-mode aerosols is associated with Saharan dust events in summer. On the other hand, in winter, a significant fraction of the coarse-mode particles (~45%) is associated with Saharan dust events, thus presenting high (0.57 ± 0.31) AOD_{550} values.

Over Athens the lowest anthropogenic AOD_{550} values are observed in winter and maximum in spring–summer. The latter is possibly related to the increase of both photochemical and convective activity registered during the warm months of the year. The modeling study of Duncan and Bey (2004) also highlights the important role of convection, as a pathway to the export of European pollution to the middle troposphere in summer. The summer maximum is also presented for the CM and MT mainly due to more stagnant air masses and the absence of wet removal processes. It was also established (Antoine and Nobileau, 2006) that the production of maritime sulfate aerosols, also mixed with dust, is maximum in summer in the Eastern Mediterranean. The small winter AOD_{550} are also consistent with aerosol-mixing processes, involving low-concentration maritime aerosols from sea spray, whose production increases in winter due to strong winds (Fotiadi et al., 2006).

Regarding the seasonal variation of the FM fraction values, no remarkable variability is found for all aerosol types. Particularly, the UI type exhibits similar values throughout the year since the anthropogenic local emissions are continuous. Low FM fraction values are observed in winter for DD and CM aerosols, while their lowest values for all aerosol types in summer may be associated with a slight mixture with dust. Nevertheless, there are remarkable differences between our findings and the results given for Northern Greece by Gerasopoulos et al. (2003), comprising large α-Ångström values in summer months, which are mainly linked to fine-aerosol particles transported from Eastern Europe. In this season, large and extensive fires take place in the Northern Balkan countries and Ukraine; thus, the air masses, which are mainly from this sector in summer, transport FM soot aerosols over Greece, a finding also reported by Balis et al. (2003).

The seasonal discrimination and classification of aerosols was further investigated based on AOD_{550} and FM fraction values (Fig. 6(a, b)). At first, a classification of aerosol types was performed based on AOD_{550} values (Fig. 6(a)). In all seasons, except spring, it was found that the larger fraction of the daily AOD_{550} values greater than 0.5 corresponds to dust cases. AOD_{550} values higher than 0.4 in winter are mainly related to some Sahara dust events. The larger fraction of CM aerosols is also observed in winter, while the MT dominates especially for low AOD_{550} values. In summer, a portion of 30% of daily values with $AOD_{550} > 0.5$ corresponds to fine particles (UI type), especially under intense urban pollution, while the coarse particles are dominant. In spring, this fraction is significantly enhanced reaching up to 50%, while the DD type exhibits significant fraction for $AOD_{550} > 0.6$. Also, in both seasons (spring and summer) the MT has significant fraction in all AOD_{550} intervals. In fall, the situation is clearer, with the CM and MT aerosols to dominate at lower AOD_{550} values, while the DD at higher.

The classification of aerosol types based on FM fraction values instead of AOD_{500} is given in Fig. 6(b). Due to threshold values a clear discrimination of fine- and coarse-aerosol types can be depicted. Thus, for $FM > 0.8$ the UI type is dominant, while the fraction of MT is significant only in winter. In this season, the CM type dominates at low FM values, while in the other seasons the DD type. In general, the DD type exhibits its higher frequency in the interval 0.4–0.5 or 0.5–0.6. The larger frequency of the UI type is for $FM > 0.9$ in winter and spring, shifting towards lower values (0.8–0.9) in summer and fall. In the FM values interval (0.7–0.8) only the MT is present exhibiting similar frequency in all seasons, while in intervals < 0.6 and > 0.8 a negligible frequency is observed in spring and summer. Since the fractions of the seasonal occurrence of each aerosol type are given in Table 1, more discussion about Fig. (6a, b) is not needed. The seasonal contribution of each aerosol type to the total AOD_{550} was analyzed from a 1-year (2001) data set

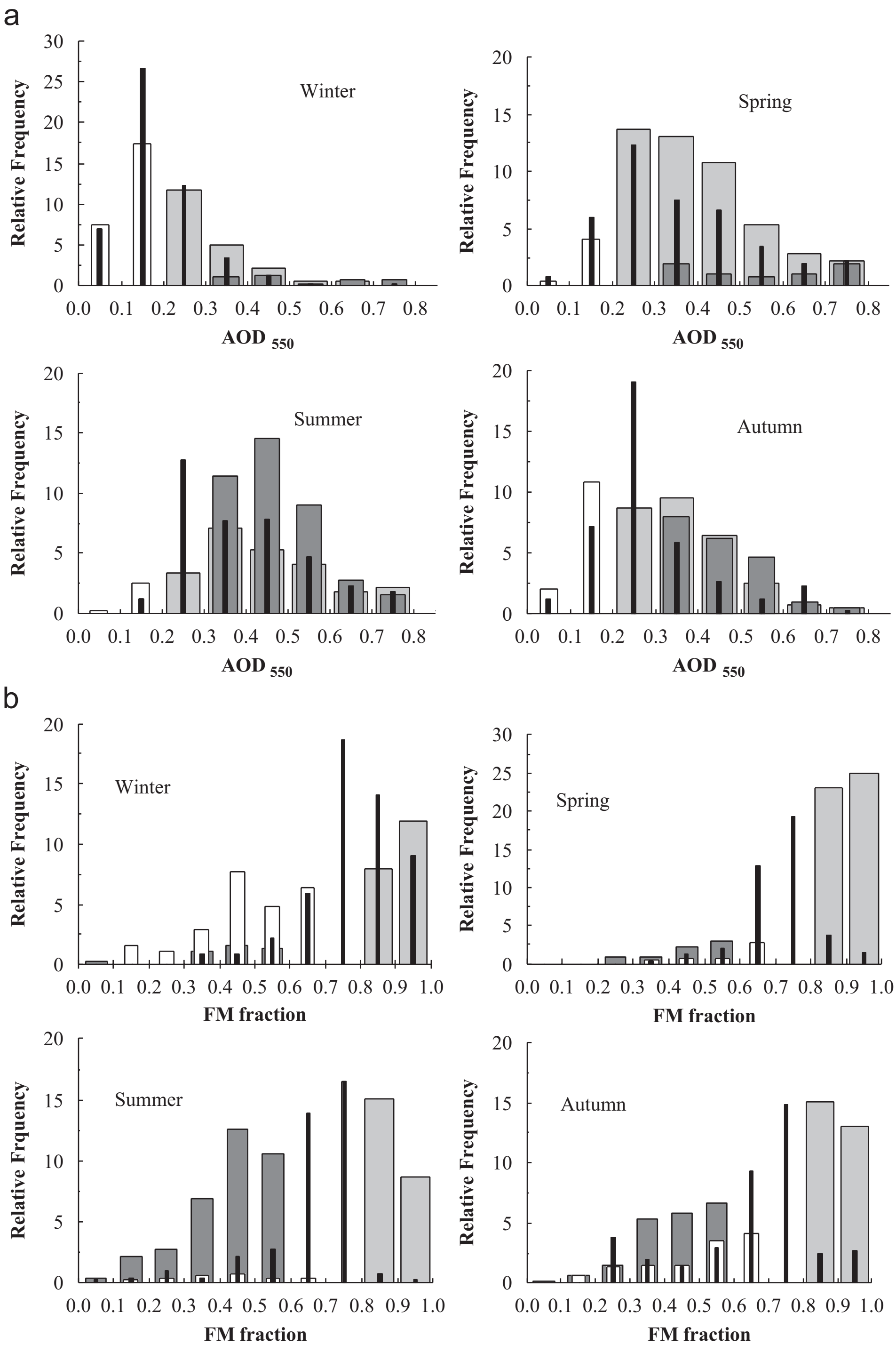

Fig. 6. Seasonal relative frequency of occurrence of different aerosol types based on (a) AOD_{550} and (b) FM fraction values. UI in light gray, CM in white, DD in gray and MT in black.

from MODIS (Barnaba and Gobbi, 2004) over the whole Mediterranean. As regards the Mediterranean subregions close to Athens (e.g. Aegean Sea), the contribution of the three aerosol types (UI, CM and DD) to the total AOD_{550} seems to be strongly seasonal and spatial dependent.

6. Conclusions

A 6-year (2000–2005) data set of AOD_{550} and FM fraction from the MODIS instrument onboard NASA-Terra satellite was analyzed in order to evaluate the aerosol seasonal variability and to discriminate various aerosol types over GAA. The results highlighted an evident AOD_{550} seasonal cycle with minimum and maximum values in winter and late spring/summer, respectively. The FM fraction seasonal cycle presented maximum values in spring and minimum in late summer. In the period examined, no trend was observed for both parameters, despite the significant annual variations from year to year.

The seasonal distribution of the meteorological patterns over Athens, the efficiency of the aerosol production mechanisms and aerosol residence time produced a distinct seasonal cycle of AOD and FM fraction. Therefore, strong Northeastern winds lead to ventilation of the Athens basin, while Southwest sea breezes and calms favor the accumulation of pollutants and the formation of second generation aerosols and photochemical smog. Also, the mixing height as well as the city circulation patterns play a very important role in the aerosol properties over Athens complex terrain. Therefore, the combination of satellite data, ground-based instruments, meteorological parameters, air mass trajectories and air pollutant concentrations seems to be necessary for the air pollution monitoring and aerosol characterization in GAA.

The data set was classified according to the aerosol optical properties; four types were identified: urban/industrial (UI), clean maritime (CM), desert dust (DD) and mixed type (MT). Values of AOD_{550} and FM fraction over the whole observation period are 0.38 and 0.90 for UI, 0.15 and 0.49 for CM, 0.52 and 0.44 for DD and 0.31 and 0.70 for MT aerosols. These values also exhibit significant seasonal variation, especially of the AOD_{550} for the UI, DD and MT aerosols and the FM for the CM aerosols. All the aerosol types, except DD, indicated an obvious maximum AOD_{550} in summer and minimum in winter, while the coarse-mode aerosols seemed to be dominant in the Athens atmosphere during summer months. This is probably attributed to dry conditions favoring the erosion of dust into the atmosphere as well as to the rare precipitation, thus enhancing their residence time. The spring maximum of AOD_{550} for DD aerosols suggests an increase in the total number of particles and a relatively more intense transport of dust. The anthropogenic aerosols are dominant in spring due to local emissions and favorable (sea breeze and calms) meteorological conditions, while the CM aerosols are frequent in winter, when the atmospheric conditions are more transparent. The results of our study confirm the large variability of aerosol optical properties.

The direct comparison of our results with relevant studies conducted in the Mediterranean does not constitute an easy task. The differences in instrumentation (ground-based sun photometers, satellites, lidar), differences in data collection, in the period of measurements, in the used methodology for the classification of aerosols and in the location where each study was conducted, are among the main difficulties. However, satellite and ground-based data are available over extended periods, and a comparison of our results with these climatological values may provide useful indications.

This study provided a quantitative overview of the aerosol seasonal variability over Athens, well representing the impact of the different aerosol types over this urban environment. The AOD_{550} and FM fraction data are thought to be important for providing a basis of comparison for radiative-transport models. With the use of aerosol vertical profiles, ground-based aerosol measurements and high-spatial-resolution satellites (such as SPOT HRVIR), the aerosol and air pollution monitoring in the Athens basin can be highly efficient. Given that spectral information on aerosol properties is being increasingly available all over the globe (through both ground- and space-based sophisticated instruments, e.g. AERONET or MODIS) it is very hopeful that our knowledge on globally distributed aerosols will be enriched soon.

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