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## Seasonal variation of columnar aerosol optical properties over Athens, Greece, based on MODIS data

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### Abstract

A long-term (2000–2005) data set of aerosol optical properties obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS) is analyzed focusing on the Greater Athens Area in the Eastern Mediterranean region. The MODIS aerosol optical depth standard product (AOD at 550 nm) and its respective ratio attributed to fine-mode particles (FM) are employed to evaluate the inter-annual and seasonal variability of the aerosol properties over Athens. Based on AOD<sub>550</sub> and FM values three specific aerosol types are discriminated corresponding to different aerosol load and optical properties. The aerosol types considered correspond to urban/industrial aerosols, coarse-mode particles and clean maritime conditions. This study focuses on the seasonal and year-to-year fluctuation of the number of occurrences as well as the AOD<sub>550</sub> and FM values of each aerosol type. The coarse-mode particles are observed mainly in the summer, while spring is the most favorable season for the occurrence of urban/industrial aerosols. On the other hand, clean maritime conditions occur mainly in the winter. The AOD<sub>550</sub> values for the coarse-mode particles are higher in spring, while the urban/industrial and clean maritime aerosols exhibit slightly higher values in the summer. The seasonal distribution of the aerosol properties is related to anthropogenic and dust emissions in the spring/summer period, but is modified by atmospheric dispersion and precipitation in late autumn/winter. The main conclusion of the study is that the coarse-mode particles exhibit much stronger inter-annual and seasonal variability compared to the urban/industrial aerosols. Finally, three cases corresponding to each aerosol type are analyzed with the aid of synoptic weather maps, air mass trajectories and MODIS data.

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### 1. Introduction

In the last two decades aerosols have been recognized as a major factor in determining global climatic change (IPCC, 2001), since they play a crucial role in the solar and thermal radiative transfer in the atmosphere. Through their direct and indirect effects aerosols strongly modify the radiation budget at the surface of the earth as well as the cloud microphysical properties, precipitation rate and hydrological cycle (Haywood & Boucher, 2000; Lohmann & Feichter, 2005). The climatic

effect of aerosols is closely related to their optical properties and size, surface albedo and their relative position in respect to that of clouds in the atmosphere (above or below them) (Kinne & Poeschel, 2001). As a consequence, the climate response to the different types of aerosols varies significantly from negative (cooling) to positive (heating) still having great uncertainties (see the review study by Satheesh & Krishna Moorthy, 2005 and references therein). In order to reduce these uncertainties through specific measurements, various networks have been established (e.g., Aerosol Robotic Network (AERONET), Global Atmosphere Watch (GAW), European Aerosol Lidar Network (EARLINET)), which aim to improve the scientific knowledge on aerosol properties and their impact on climate. To this respect, extensive field campaigns (e.g., ACE-1, ACE-2, MINOS, PRIDE, TARFOX, SAFARI) have been conducted at

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various locations over the globe in order to quantify and investigate the optical properties of various aerosol types, both natural and anthropogenic. Nevertheless, a better understanding of the uncertainties in the impact of aerosols on the surface air interactions, global surface air temperatures, hydrological cycle, photochemistry and ecosystems requires a holistic approach in terms of platforms (e.g., ground-based networks, satellites, ships and aircrafts), sensors (e.g., spectroradiometers, sun photometers, lidars, satellite sensors) and techniques (e.g., in-situ measurements, remote sensing and computer modeling) (Heintzenberg et al., 1996).

The ground-based instruments are readily calibrated and, therefore, the least imprecise, thus constituting the base for the calibration of satellite algorithms (Chu et al., 2002; Remer et al., 2002). However, their use is representative for the area only of their operation. On the other hand, satellite remote sensing has been increasingly used in the last decade to quantify aerosols over the globe (King et al., 1999). It constitutes a recent, but powerful tool, for assessing aerosol spatial distribution and properties due to its major benefit of providing complete and synoptic mapping of large areas in single snap-shots. Satellite sensors provide global images of the Earth and allow retrieving the spatio-temporal aerosol distribution, which results from the spatial inhomogeneities and the short aerosol lifetime (Remer et al., 2005; Santese et al., 2007). Thus the aerosol remote sensing from long-term operational satellites provides a unique opportunity to achieve a global and seasonal monitoring of aerosol load and properties. Kaufman et al. (2002) underlined the important role of satellite sensors in providing the much needed aerosol information for global climate studies. A variety of satellite sensors (e.g., AVHRR, TOMS, SCIAMACHY, POLDER, MODIS, MERIS, MISR) and techniques have been employed for aerosol and pollution monitoring at regional and global scales with spatial resolution varying from low to high. On the other hand, the disadvantages in the use of satellite sensor techniques are the low temporal resolution, the effects of surface albedo and the presence of clouds. Extensive analysis and comparison of satellite retrievals over central Europe has recently been published by Kokhanovsky et al., (2007).

In the Mediterranean basin aerosols have attracted a great scientific interest due to the variety of sources, natural and anthropogenic, which influence their optical properties (Pace et al., 2006; Zerefos et al., 2006). With the increasing urbanization and industrialization, especially at coastal Mediterranean areas, the aerosol load, particularly in the lower troposphere, increases continuously (Papayannis et al., 2005) leading to high aerosol extinction values (Barnaba & Gobbi, 2004) and large reductions in the solar radiation at the surface (Markowicz et al., 2002). The aerosols over the Mediterranean can be desert dust particles, directly transported from the North African desert regions, anthropogenic aerosols from the industrialized areas in Europe, maritime particles produced over the Mediterranean or transported from the Atlantic and smoke particles from seasonal forest fires (Barnaba & Gobbi, 2004). However, due to the variety of sources, the long-range transport and the mixing processes in the atmosphere, the aerosols in this area are difficult to be distinguished in a single type; the most likely scenario is a

mixed aerosol type, especially over regions that are influenced by local sources. This fact seems to be the case for Athens as has been clearly established by recent studies (Kaskaoutis & Kambezidis, 2006; Kaskaoutis et al., 2006). On the other hand, more recently, Kaskaoutis et al. (in press), using a long-term dataset from MODIS sensor on board the EOS-Terra satellite, discriminated three main aerosol types over Athens (urban/industrial aerosols, maritime component and coarse-mode dust particles). Despite this, the most common situation, especially in winter, was an “undetermined” (or blended) aerosol type as a result of the mixing processes in the atmosphere.

The present study focuses mainly on the inter-annual and seasonal variation of the three “specific” aerosol types based on satellite data over the Greater Athens Area (GAA). This work is among the first aerosol climatology studies over Athens based on long-term satellite data. The main results regarding the climatology of the different aerosol types over GAA were presented and discussed in a previous study (Kaskaoutis et al., in press). The present study presents, via Tables and Figures, the diachronic fluctuation (2000–2005) of the aerosol properties, in order to reveal the variability of each examined parameter (number of occurrences, AOD<sub>550</sub> and FM values). It is hoped that the results from both Figures and Tables will constitute the base for further research over the area using satellite sensors and/or ground-based instruments.

## 2. Description of the study region

Athens is a Mediterranean city of about 3.5 millions of people (census of 2001) located in an oblong basin with an area of 450 km<sup>2</sup>. The main axis of the basin lies in the NE–SW direction. High mountains on the three sides and the Saronikos Gulf on the south surround Athens. The mountains that act as physical barriers, the warm and dry climate in the warm period of the year and the concentrated pollution sources are responsible for the deterioration of air quality within GAA. Thus, under favorable meteorological conditions (sea breeze and calms), the concentration of air pollutants may exceed the air quality standards of the European Union and the World Health Organization (Chaloulakou et al., 2003; Kambezidis et al., 2001).

The major influence of the anthropogenic aerosol and air pollutants within the Athens basin is the level of their concentration in the air very much affected by the local meteorological conditions. Two opposite wind regimes are identified in the summer; calms or sea-breeze circulation on the one hand that appear when the synoptic flow is weak, frequently associated with high levels of air pollution, and, on the other, the strong North-easterly winds, called “Etesians”, mainly driven by the combined effect of the thermal low over the Eastern Mediterranean and the Azores anticyclone. These two opposite wind regimes lead to different aerosol load, size and optical properties (Kambezidis et al., 2001; Adamopoulos et al., in press). The synoptic meteorological pattern during winter is affected by: a) the Siberian anticyclone causing polar continental air flow from North-Eastern Europe, b) an anticyclone over Middle East 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. The prevailing meteorological conditions, the photochemical reactions, the local emissions, the long-range transport and the precipitation rates are the major factors influencing the annual and seasonal fluctuation of the aerosol types and properties over GAA. In Table 1 the main meteorological parameters are given for each month in the Athens area.

### 3. Data set and methodology

The data set used in this study concerns 6 years of observations over GAA of the standard aerosol products (AOD<sub>550</sub> and FM) derived from MODIS and corresponding to collection C005 data fully described in Kaskaoutis et al. (in press). Therefore, a brief description will be given here. Since February 2000, the 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 resolution, nadir pixel dimensions), having equatorial overpass time at approximately 10:30 UTC. Numerous parameters describing various properties over land and ocean surfaces as well as in the atmosphere are retrieved operationally from MODIS data at different spatial and temporal resolutions (daily, weekly, monthly). The land algorithm deduces ground reflectivity from the measured radiance at 2.13  $\mu\text{m}$ . Numerous studies (e.g. Kaufman et al., 2002) have demonstrated that, for certain vegetated surfaces over the globe, the surface reflectance at 0.47  $\mu\text{m}$  and 0.66  $\mu\text{m}$  can be derived from the mean radiance at 2.13  $\mu\text{m}$  by empirical relationships. The ocean inversion algorithm is based on Lookup Tables that consists of four fine modes and five coarse modes (Remer et al., 2005). The two aerosol products employed in the present study are: 1) the aerosol optical depth at 550 nm (AOD<sub>550</sub>) and 2) the fine-mode aerosol fraction (FM). The latter is derived as the ratio of the optical depth of fine mode versus total optical depth at 550 nm. To assess the quality of these parameters, a substantial part of the MODIS aerosol products acquired in 2000 have been validated globally and regionally, as reported by Chu et al., (2002), Ichoku et al., (2002) and Remer et al. (2002). The

uncertainties in determining these parameters are different for each algorithm (Remer et al., 2005) and are mainly attributed to non-spherical particles, the different algorithms used over land and ocean and the sub-pixel water contamination. These uncertainties usually lead to overestimation of the AOD (Chin et al., 2004). The MODIS aerosol retrieval is calculated on a 10 km  $\times$  10 km resolution (known as Level 2 data), and is directly retrieved from the radiance data observed by MODIS (Level 1B data). Since the radiance/reflectance data are observed at 500 m (for most channels), this means that the 500-m data are aggregated into these 10-km boxes (i.e., a 20  $\times$  20 box of radiance data). Clouds are screened within the 20  $\times$  20 box (Levy et al., 2007) and the aerosol retrievals are performed if there are sufficient numbers (approximately 10% remaining) of non-cloudy or otherwise not masked) pixels. Therefore, the 10-km (Level 2) products may be valid even when the box is  $\sim$ 90% cloudy. Depending on the “quality” of the retrieval (and the number of the remaining valid pixels), the 10-km retrieval is assigned a “quality assurance (QA)” value. These 10-km retrievals are aggregated to a 1-degree box (known as Level 3), and are weighted by their QA. Thus, a Level 3-aerosol retrieval may be reported even when the grid is very cloudy. Via the new developed MODIS algorithm (Levy et al., 2007), the AOD<sub>550</sub> and FM values are quite accurate after extensive comparison with the AERONET data. In this study the Collection 5 (C005) Level 3 QA-weighted products are used by Giovanni website (<http://giovanni.gsfc.nasa.gov/>).

In order to reduce the aforementioned uncertainties, the MODIS products used in the present study refer to the mean AOD<sub>550</sub> and FM values obtained from four pixels covering the area with coordinates 36.5°–38.5°N and 22.5°–24.5°E that includes the entire GAA. A total of 1804 daily data were collected from 26 February 2000 to 31 December 2005. The winter (January, February, December), spring (March, April, May), summer (June, July, August) and autumn (September, October, November) days represent 20.8%, 25.7%, 28.4% and 25.1%, respectively, of the whole dataset (Kaskaoutis et al., in press). Because of this long-term period the MODIS data can be used to evaluate the seasonal variability of aerosols over the region since the retrieval uncertainties are now very much

Table 1  
Mean values of the main meteorological parameters in Athens for the period 2000–2005

Month	Mean air temperature (°C)	Max air temperature (°C)	Min air temperature (°C)	Relative humidity	Mean total precipitation (mm)	Mean number of rainy days
January	9.76	13.14	7.17	72.37	63.58	6.60
February	10.20	14.32	7.09	68.83	33.30	4.60
March	13.04	17.65	9.63	65.86	19.28	3.00
April	15.98	20.78	12.33	64.77	33.50	4.40
May	21.80	27.39	17.45	54.14	6.58	1.20
June	26.99	32.66	22.33	47.87	6.54	0.80
July	29.75	35.53	25.16	49.15	20.94	0.40
August	29.01	34.74	24.66	47.86	11.70	0.60
September	24.34	29.86	20.57	59.91	47.68	2.80
October	20.40	25.40	16.93	65.14	32.36	2.60
November	15.75	19.74	12.91	74.75	107.82	5.80
December	10.93	13.97	8.59	74.77	126.96	8.80

The data obtained at the National Observatory of Athens.

Table 2  
Number of occurrences and mean AOD<sub>550</sub> and FM values for each individual year and season for the UI aerosol type

UI type		Winter		Spring		Summer		Autumn		Year	
Frequency of occurrences	2000	0 (0%)		39 (50%)		24 (31%)		15 (19%)		78 (18%)	
	2001	3 (8%)		23 (62%)		7 (19%)		4 (11%)		37 (8%)	
	2002	11 (15%)		32 (44%)		16 (22%)		14 (19%)		73 (17%)	
	2003	9 (9%)		46 (48%)		21 (22%)		19 (20%)		95 (22%)	
	2004	2 (2%)		26 (41%)		21 (33%)		15 (24%)		64 (14%)	
	2005	9 (9%)		26 (27%)		28 (29%)		32 (34%)		95 (22%)	
Whole		34 (7%)		192 (44%)		117 (27%)		99 (22%)		442	
		AOD <sub>550</sub>	FM								
Aerosol properties	2000			0.484	0.887	0.566	0.881	0.427	0.877	0.469	0.661
	2001	0.419	0.918	0.446	0.858	0.501	0.880	0.443	0.822	0.452	0.870
	2002	0.352	0.898	0.471	0.906	0.478	0.863	0.436	0.893	0.434	0.890
	2003	0.363	0.934	0.451	0.913	0.468	0.876	0.382	0.864	0.416	0.897
	2004	0.293	0.979	0.458	0.888	0.396	0.881	0.370	0.872	0.379	0.910
	2005	0.446	0.974	0.460	0.936	0.479	0.896	0.439	0.939	0.456	0.936
Average		0.31±	0.92±	0.41±	0.91±	0.45±	0.88±	0.37±	0.90±	0.38±	0.90±
		0.10	0.06	0.15	0.06	0.16	0.06	0.11	0.06	0.13	0.06

reduced. Recently, MODIS AOD<sub>550</sub> and FM values have been used in conjunction with lidar and AERONET data to monitor the aerosol optical properties in East Asia (Kim et al., 2007). The same parameters are also correlated with those derived from AERONET in South Italy with promising results using different spatial and temporal resolutions (Santese et al., 2007).

In the literature, there are some techniques to distinguish between major aerosol types, like the combined use of AOD and Ångström exponent (Pace et al., 2006; Kaskaoutis et al., 2007) or the combined use of AOD and FM values (Barnaba & Gobbi, 2004; Kaskaoutis et al., in press). The correlation of the aerosol optical depth 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, since the fine-mode fraction depends mainly on the particles size, while AOD<sub>550</sub> on

the aerosol load. Following the aerosol mask scheme of Barnaba and Gobbi (2004), Kaskaoutis et al., (in press) discriminated three aerosol types over GAA using the same MODIS data set and appropriate threshold values. Thus, urban/industrial (UI) aerosols correspond to AOD<sub>550</sub>>0.2 and FM>0.8, clean maritime conditions (CM) are associated with AOD<sub>550</sub><0.2 and FM<0.7, and coarse-mode, probably desert dust (DD) aerosols, have AOD<sub>550</sub>>0.3 and FM<0.6. Between these areas some gaps are presented, where the data are not included in a specific aerosol type, corresponding to an additional “undetermined” type, with a percentage of 46.6% of the whole dataset (Kaskaoutis et al., in press). The “undetermined”, rather mixed aerosol type, was introduced in order to quantify the mixing processes in the atmosphere and also prevent from over predicting the presence of either UI or DD aerosols. Because of its simplicity, this method is associated with some arbitrariness (e.g., incorrect aerosol type interpretation), particularly at the borderlines of the three aerosol

Table 3  
As in Table 2, but for the CM aerosol type

CM type		Winter		Spring		Summer		Autumn		Year	
Frequency of occurrences	2000	2 (22%)		2 (22%)		0 (0%)		5 (56%)		9 (5%)	
	2001	11 (44%)		2 (8%)		1 (4%)		11 (44%)		25 (14%)	
	2002	20 (54%)		1 (3%)		1 (3%)		15 (41%)		37 (20%)	
	2003	14 (50%)		2 (7%)		0 (0%)		12 (43%)		28 (15%)	
	2004	21 (42%)		5 (10%)		6 (12%)		18 (36%)		50 (27%)	
	2005	22 (65%)		3 (9%)		6 (18%)		3 (9%)		34 (19%)	
Whole		90 (49%)		15 (8%)		14 (8%)		64 (35%)		183	
		AOD <sub>550</sub>	FM								
Aerosol properties	2000	0.169	0.529	0.165	0.528			0.162	0.633	0.164	0.423
	2001	0.132	0.517	0.116	0.560	0.167	0.462	0.153	0.474	0.142	0.503
	2002	0.134	0.493	0.155	0.637	0.161	0.197	0.149	0.531	0.150	0.465
	2003	0.137	0.460	0.172	0.645			0.142	0.439	0.143	0.386
	2004	0.126	0.461	0.161	0.601	0.189	0.529	0.148	0.537	0.156	0.532
	2005	0.110	0.472	0.150	0.526	0.157	0.400	0.155	0.408	0.143	0.452
Average		0.13±	0.47±	0.15±	0.58±	0.17±	0.42±	0.14±	0.50±	0.15±	0.49±
		0.04	0.14	0.03	0.10	0.03	0.14	0.04	0.16	0.03	0.13

Table 4  
As in Table 2, but for the DD aerosol type

DD		Winter	Spring	Summer	Autumn	Year					
Frequency of occurrences	2000	0 (0%)	6 (12%)	26 (51%)	19 (36%)	51 (15%)					
	2001	1 (1%)	9 (11%)	42 (50%)	32 (38%)	84 (25%)					
	2002	1 (2%)	8 (14%)	39 (70%)	8 (14%)	56 (17%)					
	2003	2 (3%)	4 (6%)	36 (57%)	21 (33%)	63 (19%)					
	2004	4 (9%)	3 (7%)	23 (51%)	15 (33%)	45 (13%)					
	2005	5 (13%)	12 (32%)	14 (37%)	7 (18%)	28 (11%)					
Whole		13 (4%)	40 (12%)	182 (53%)	102 (30%)	337					
		AOD <sub>550</sub>	FM	AOD <sub>550</sub>	FM	AOD <sub>550</sub>	FM	AOD <sub>550</sub>	FM	AOD <sub>550</sub>	FM
Aerosol properties	2000			0.673	0.541	0.430	0.470	0.455	0.448	0.590	0.365
	2001	0.636	0.448	0.628	0.487	0.607	0.489	0.435	0.352	0.577	0.444
	2002	0.315	0.476	0.787	0.510	0.499	0.363	0.464	0.459	0.516	0.452
	2003	0.328	0.413	0.645	0.466	0.458	0.418	0.450	0.404	0.470	0.425
	2004	0.603	0.391	0.501	0.467	0.432	0.433	0.445	0.432	0.495	0.431
	2005	0.511	0.351	0.600	0.599	0.410	0.465	0.404	0.457	0.481	0.468
Average		0.57±	0.43±	0.60±	0.46±	0.46±	0.43±	0.45±	0.43±	0.52±	0.44±
		0.31	0.14	0.24	0.11	0.12	0.13	0.10	0.12	0.19	0.12

types (Barnaba & Gobbi, 2004). Despite this, the proposed method is quite effective and the three types are representative in the region. It was found that 81% of the UI cases corresponds to European air masses, 73% of the CM to Atlantic air masses and 50% of the DD to African air masses. Since these three directions are representative for the origin of specific aerosol types, the above percentages give accuracy to the classification scheme used. The frequency of occurrences, as well as the optical properties for each aerosol type, are given in Tables 2–4 for UI, CM and DD types, respectively.

## 4. Results and discussion

### 4.1. Number of occurrences

Firstly, the number of occurrences for each specific aerosol type is analyzed, since significant differences occur on annual or seasonal basis. These differences are mainly attributed to the meteorological conditions favoring the presence of a specific aerosol type, to local emissions and to long-range transport. Another significant parameter that affects the aerosol load and properties is precipitation, which constitutes the most efficient removal process in the atmosphere.

Regarding the UI type, it is obvious from Fig. 1 and Table 2 that its presence is favored in spring (44%), followed by summer (27%) and autumn (22%), while the occurrence of this aerosol type in winter is rather rare (7%). The meteorological and atmospheric conditions favoring the presence of this aerosol type and the accumulation of locally-emitted air pollutants are due to the presence of an anticyclone above the Eastern Mediterranean and Greece that favors stable atmospheric conditions and trapping of local emissions. The sea-breeze circulation over GAA in the warm season constitutes an additional condition favoring the accumulation of urban aerosols (Adamopoulos et al., 2007). Out of the total number of days analyzed, 442 are dominated by this aerosol type, which constitutes the most common situation in such an urban environment. Papayannis et al. (2005) showed that a

thick layer of urban aerosols is always present within the boundary layer of GAA, even though long-range transport occurs in the upper atmosphere. The inter-annual variability of this aerosol type is rather significant, having its higher frequency in 2003 and 2005 (22%), while in 2001 only 8% of this aerosol type was observed. The spring maximum in the occurrence of UI type is observed in all years with the exception of 2005, when the autumn presents the higher fraction (Fig. 1). Another important highlight from this Figure is the steep increase in the number of occurrences from winter to spring.

Clean maritime conditions, dominated by optical properties of marine aerosols, are rather rare over Athens urban environment (183 out of 1804 days), despite its proximity to the sea. The intense local emissions and the anthropogenic activities within GAA are continuous sources of aerosols, thus increasing AOD and reducing visibility. These sources, favored by the local meteorological conditions in the warm period of the year (e.g., low wind speed and stable atmospheric conditions), conclude to increased aerosol load within the basin. As a consequence, CM conditions in spring and summer are very rare

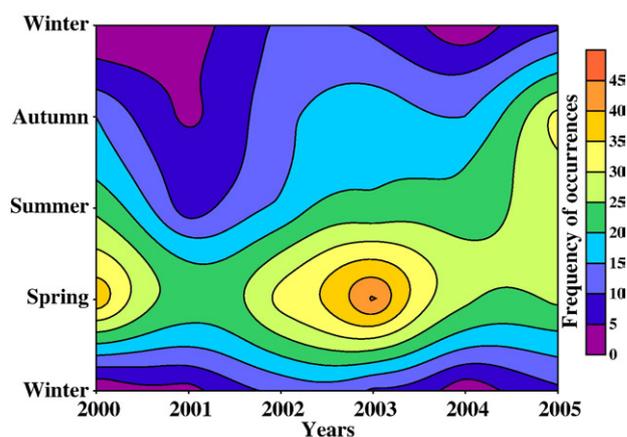


Fig. 1. Year-to-year variation of the frequency of occurrence for the UI aerosol type in each season.

over Athens, exhibiting a maximum of 6 individual days in summer of 2004 and 2005, (Table 3). It is worth to be noticed that no clean maritime day is observed in the summer of 2000 and 2003. On the other hand, clean maritime conditions are frequently observed in the cold period of the year (winter, 49%, autumn 35%). This is also in agreement with previous studies conducted over Athens (Kambezidis et al., 2001) and, in general, over Mediterranean (Barnaba & Gobbi, 2004). The strong winds in winter, in combination with the wet removal processes, are the main factors for cleansing the Athens atmosphere. On annual basis, the majority of the CM conditions are observed in 2004, while the low occurrence in 2000 is attributed to the shorter dataset for this season and not necessarily to more turbid atmospheric conditions. Note that the MODIS data set begins on 26 February 2000. Fig. 2 is very characteristic for the frequency of occurrence of the CM aerosol type presenting a deep “valley” in spring and summer and a steep increase towards autumn and winter.

The frequency of occurrence for the coarse-mode aerosols over GAA (Table 4) presents a clear maximum in summer (53%) followed by that of autumn (30%). On the other hand, this aerosol type is rather rare in the winter (4%), when the atmospheric conditions are clearer, and in the spring (12%), where the UI type dominates. Regarding the whole period, 337 cases correspond to this aerosol type, which should be underlined that does not correspond only to DD aerosols, but includes a general category of coarse-mode particles (e.g., desert dust, soil particles, sand, sea spray, etc). Therefore, its higher frequency in the summer can be justified by the dry atmospheric conditions favoring the erosion of soil dust and its presence in the atmosphere for several days, since in this season the precipitation is rather absent. It was found, that only 23% of the coarse-mode aerosols corresponds to Sahara dust events over Athens, depicted mainly in the April–May period and in July. Comparing Tables 2 and 4, it is established that at least on an annual basis, the occurrence of the DD type presents maximum when the respective UI is minimum and vice versa. This is strongly verified from the 2001 and 2005 data. Thus, the highest frequency of DD type in 2001 is mainly attributed to its higher frequency in summer and autumn, when the frequency of UI type is small. In contrast, the lowest frequency of DD type in

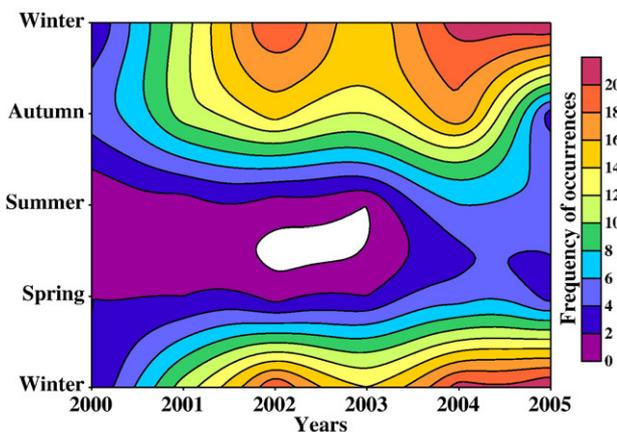


Fig. 2. Same as in Fig. 1, but for the CM aerosol type.

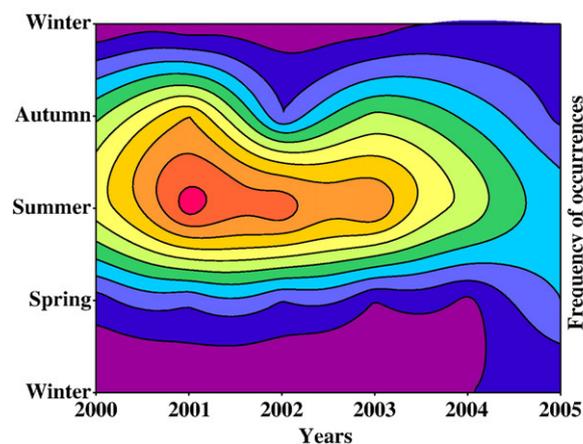


Fig. 3. Same as in Fig. 1, but for the DD aerosol type.

2005 is mainly attributed to the low frequency in summer and autumn, when the respective frequency of UI type is maximum. The enhanced frequency of the DD type in the spring of 2005 is attributed to frequent Sahara dust events in that period. Fig. 3 is quite characteristic regarding the frequency of occurrence for the DD type, highlighting the summer peak, which continuously decreases towards 2005.

#### 4.2. Annual and seasonal variability of AOD<sub>550</sub> and FM

Fig. 4 shows the seasonal variability of AOD<sub>550</sub> for the whole data set. In a previous study (Kaskaoutis et al., in press), no significant trend in the AOD<sub>550</sub> values was observed, while the mean AOD<sub>550</sub> for the whole period is  $0.35 \pm 0.18$ . Nevertheless, the AOD<sub>550</sub> values exhibit significant annual variability, with lower values in winter ( $0.21 \pm 0.10$ ) and higher in spring ( $0.41 \pm 0.18$ ) and summer ( $0.44 \pm 0.18$ ). In contrast to the climatology trend (not existed), there is a significant year-to-year variability in AOD<sub>550</sub>, which is highlighted in Fig. 4. The AOD<sub>550</sub> annual variation exhibits a plateau of high values in the spring and summer months. This AOD<sub>550</sub> maximum is attributed to the relative contribution of various processes such as stagnant synoptic meteorological fields, secondary photochemical aerosol

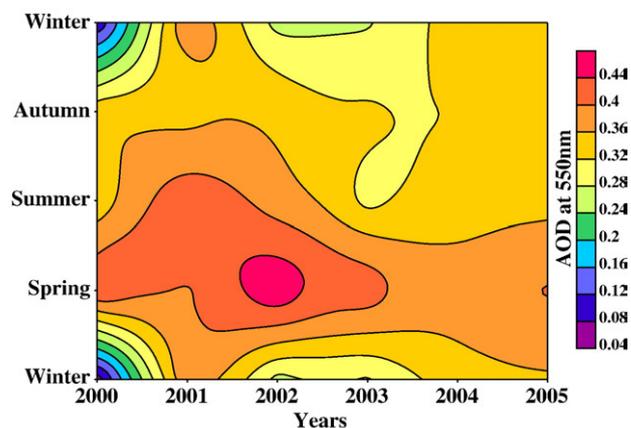


Fig. 4. Year-to-year variation of the mean seasonal AOD<sub>550</sub> values over GAA for all aerosol types (1804 individual days).

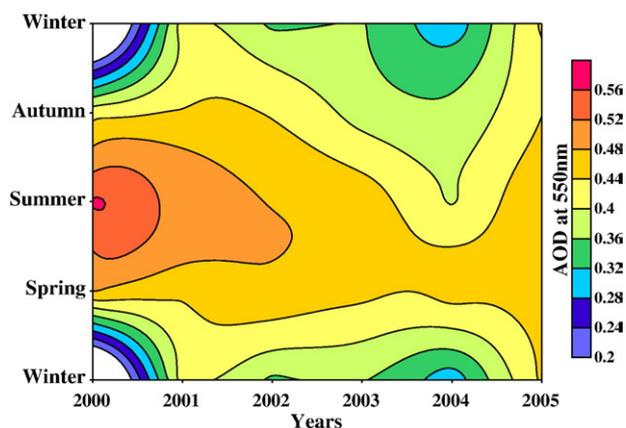


Fig. 5. Same as in Fig. 4, but for the UI aerosol type (442 individual days). The white gaps correspond to the lack of data.

formation, hygroscopic growth of water-soluble industrial aerosols and biomass smoke from seasonal forest fires. Maximum  $AOD_{550}$  values were observed in the spring of 2002. It was found that slightly lower  $AOD_{550}$  variability and magnitudes occurred in the summer 2004, which are probably attributed to the rare dust events and air masses from Africa in that year. The same feature was also observed in FORTH-CRETE AERONET station (Fotiadi et al., 2006) for the same years. Another important characteristic in Fig. 4 is the high winter  $AOD_{550}$  in 2001 and the lows in 2002 and 2004. The  $AOD_{550}$  variability in this season is mainly driven by the Sahara dust events (Kaskaoutis et al., in press). In contrast, the  $AOD_{550}$  values in autumn do not present a significant year-to-year variability.

Regarding the UI aerosol type, the seasonal and year-to-year variability of  $AOD_{550}$  seems to be insignificant, Table 2, Fig. 5. Despite the large range (0.2–1.0) of the daily  $AOD_{550}$  values for this aerosol type (Kaskaoutis et al., in press), the mean seasonal and annual values are rather constant. This indicates the continuous and nearly constant local emissions throughout the year. The  $AOD_{550}$  values increase from winter to summer and then reduce in the autumn. The summer increase is mainly attributed to the more stable atmospheric conditions in this season, which favor the accumulation of aerosols in the atmosphere, and not to additional local emissions, since, as reported by Chaloulakou et al., (2003), the  $PM_{10}$  concentrations are slightly lower in the summer compared to those in the winter. In addition, the photochemical reactions associated by the nearly absence of precipitation play a significant role in the production of secondary aerosols in the warm period of the year. Moreover, the difference between the mean summer and winter  $AOD_{550}$  values (0.14) of the UI type is significantly lower than the respective (0.23) of the whole dataset. This fact indicates the remarkable contribution of the UI type to the winter  $AOD_{550}$  values, since the winter UI mean  $AOD_{550}$  ( $0.31 \pm 0.10$ ) is significantly higher than the respective ( $0.21 \pm 0.10$ ) for the whole data set. A bit higher annual  $AOD_{550}$  for the UI type is observed in the first years of the measuring period, while the mean minimum annual value was observed in 2004. In spring, when the frequency of the UI type occurrence is maximum, the  $AOD_{550}$  values exhibit the least inter-annual variability. This fact highlights the rather constant and significant

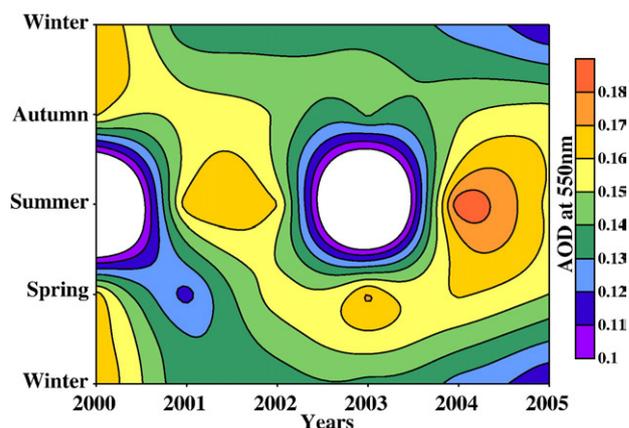


Fig. 6. Same as in Fig. 4, but for the CM aerosol type (183 individual days). The white gaps correspond to the lack of data.

emission rates in this season. In contrast, the largest year-to-year variability of the  $AOD_{550}$  values is presented in the winter, mainly attributed to the longer duration and more intense rate of precipitation, a parameter which constitutes the major aerosol removal process. Nevertheless, note that  $AOD_{550}$  values lower than 0.2 in Fig. 5 are absent, since this was the defined cut-off threshold value for the occurrence of the UI aerosol type (Kaskaoutis et al., in press). Thus the white gaps in winter 2000 (Fig. 5) correspond to lack of data.

As regards the CM aerosols, it is observed from Table 3 and Fig. 6 that the  $AOD_{550}$  variability is not significant. Nevertheless, the higher values in the summer months are obvious, except for the years of 2000 and 2003 when this aerosol type was absent, thus the white gaps. The little higher  $AOD_{550}$  values in the summer are attributed to the same reasons presented above (stagnant air masses, photochemical aerosol production, and absence of precipitation). Moreover, the relatively high values in winter 2000 are not representative for the whole period, since the data set includes December 2000 only. However, considering the very limited range of the  $AOD_{550}$  values of this aerosol type ( $AOD_{550} < 0.2$ ), the inter-annual and inter-season variability is more pronounced than the respective for the UI type. The CM aerosol type dominated on days with low pollutant

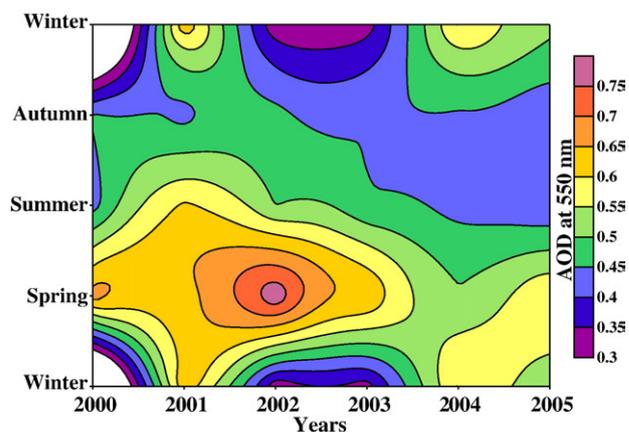


Fig. 7. Same as in Fig. 4, but for the coarse-mode (DD) aerosol type (337 individual days). The white gaps correspond to the lack of data.

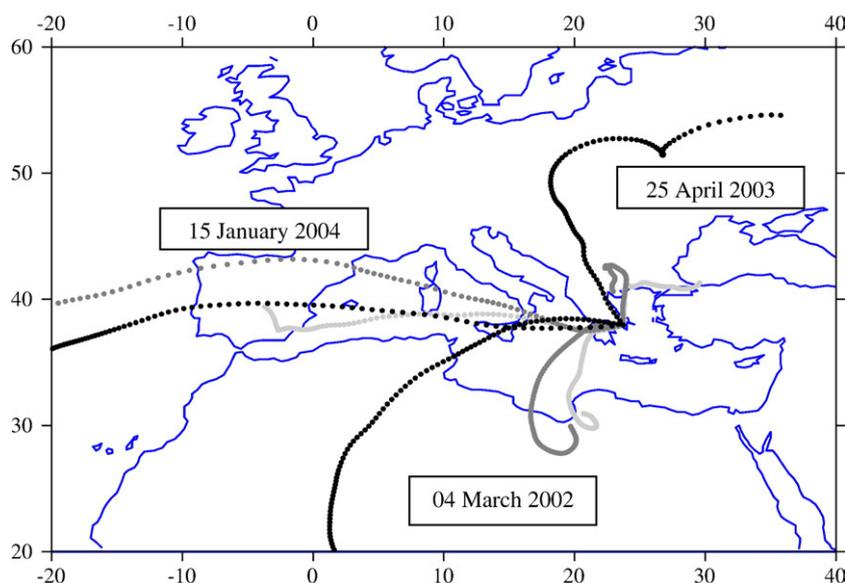


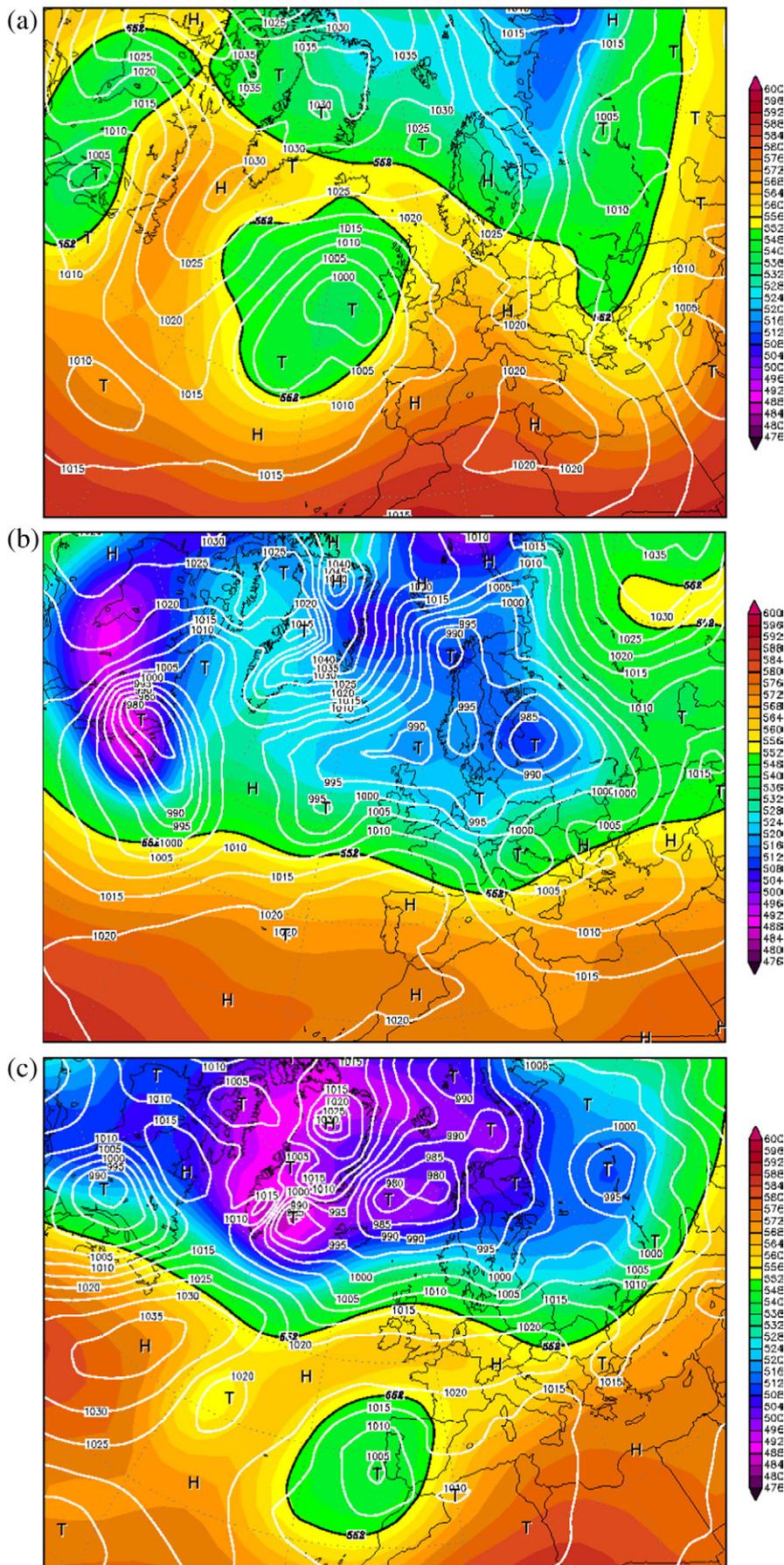
Fig. 8. Air mass back trajectories at three altitudes (500 m, 1500 m and 4000 m) from the HYSPLIT model with origins from north sector (25 April 2003), west sector (15 January 2004) and south sector (4 March 2002), favoring the formation of UI, CM and DD aerosol types, respectively, over GAA (black line: 4000 m, gray: 1500 m, light gray: 500 m).

emissions or when the atmospheric and meteorological conditions favored the dispersion of the pollutants. Therefore, the optical depth and characteristics of this aerosol type are mainly dependent on the wind field, atmospheric circulation patterns, aerosol removal processes and the origin of the arriving air masses over GAA. It was found (not presented) that the majority of them have Atlantic origin.

The coarse-mode aerosols are mainly attributed to natural sources. However, in an urban environment their optical properties can be misquoted by the mixing processes with the local emissions. Despite this, the  $AOD_{550}$  variability of the DD aerosols (Table 4 and Fig. 7), which presents a general category of coarse-mode aerosols and not only DD particles, is more pronounced than the respective UI type. This highlights the fact that the optical characteristics of the natural aerosols are mainly depended on the source regions, long-range transport mechanisms and air mass trajectories. In the period of measurements the mean  $AOD_{550}$  of the DD aerosol type is  $0.52 \pm 0.19$ , exhibiting significant seasonal and inter-annual variability. Since the DD type corresponds to natural aerosols, this variability is more significant than the respective of the UI type. Contrary to the two previous types, DD presents its maximum  $AOD_{550}$  values in spring ( $0.60 \pm 0.24$ ) and winter ( $0.57 \pm 0.31$ ) and lower in the summer ( $0.46 \pm 0.12$ ) and autumn ( $0.45 \pm 0.10$ ). Bear in mind that the frequency of occurrence of this type is higher in the summer and lower in the winter. As stated above, the DD type represents a general category of natural aerosols and not only desert dust ones. The low frequency associated with high  $AOD_{550}$  values in winter suggests that the DD type is related to Sahara dust traveling over Athens in this season. The same conclusion can be drawn for spring, too. It was found that 54% and 68% of the occurrences of the DD type in winter and spring, respectively, corresponds to Sahara dust events. Note also that springtime is the most favorable period for intense Sahara dust

outbreaks over Eastern Mediterranean (Barnaba & Gobbi, 2004). In further contrast, only a 16% of the DD occurrences in summer and autumn is associated with air masses having African origin. It was found that the majority of the air masses in late summer and early autumn are associated with the Northern sector winds (Etesians) traversing the North Aegean Sea. Fig. 8 highlights the significant variability of  $AOD_{550}$  values, which is more pronounced than the respective for the other two aerosol types. On annual basis, the higher  $AOD_{550}$  values occurred in the first years of the measuring period, as well as in the spring of 2002. From all the above, it is concluded that the  $AOD_{550}$  values are closely related to the intensity, the frequency and the duration of the Sahara dust events. Note also that all  $AOD_{550}$  values lower than 0.3 correspond to the absence of the DD aerosols in winter of 2000, thus the white gaps.

Regarding the FM values of all aerosol types, these are strongly affected by the local emissions dominated by the optical characteristics of fine- rather coarse-aerosol particles. The seasonal and inter-annual variability of the FM values is significantly lower than the respective of the  $AOD_{550}$ . Especially, the variability of the FM values for the UI type (Table 2) is nearly absent for the period of measurements, ranging from 0.82 (autumn 2001) to 0.98 (winter 2004). This is attributed to the continuous anthropogenic emissions throughout the year within the GAA and the small range of the FM values for this aerosol type (0.8–1.0). In addition, the presence of this type is favored by stagnant air masses or pollution transport from Eastern Europe. As a consequence, the aerosol field is quite well mixed over the Athens basin, strongly dominated by fine-mode anthropogenic aerosols. On the other hand, the FM variability for the natural aerosols is clearly shown. As far as the CM type is concerned (Table 3), the FM values show a decrease in the summer mainly associated with mixed and coarse-mode soil particles, whose production is favored in this dry season. However, the occurrence



of the CM type in the summer is very rare and the conclusions cannot be considered very accurate. Note also the zero values of their occurrence in the summer of 2000 and 2003. The higher occurrence of anthropogenic aerosols in spring seems to also affect the optical properties of the CM type, as suggested by the larger mean FM value in this season ( $0.58 \pm 0.10$ ). Despite the large range of FM values of the CM type (0.0–0.7), on seasonal and annual basis, the FM variability is very low. This variability is more pronounced as regards the DD type (Table 4), despite the smaller range of the FM values (0.0–0.6). The spring maximum as well as the summer minimum of the FM values for the DD type are also obvious and are attributed to the same reasons presented above, while the inter-annual variability of the FM values (Table 4) is more pronounced than for the previous aerosol types. On seasonal basis, this variability is dampened, exhibiting similar values in all seasons. The inter-annual variation is mainly attributed to the variability in dust outbreaks (Barnaba & Gobbi, 2004) and the air mass origin.

#### 4.3. Favorable meteorological conditions

In this section three cases are analyzed, each one representing the occurrence of a specific aerosol type. In an unpublished yet study it was found that the UI type is favored by stagnant air masses or the transport of fine-mode aerosols from Eastern Europe. This Northeastern sector has already been recognized as the one corresponding to fine-mode pollution aerosols over Northern Greece (Zerefos et al., 2000). Bearing in mind that the UI type is more frequent in spring, 25 April 2003 was chosen as a representative day for this aerosol type. As expected, the CM type is associated with clean air masses from the Atlantic Ocean traversing the Western Mediterranean, especially in winter. Thus, 15 January 2004 represents clean atmosphere above Athens. On the other hand, the intense Sahara dust outbreaks in Eastern Mediterranean, favored in spring by thermal cyclonic situations (Fotiadi et al., 2006; Meloni et al., 2007), are associated with high AOD<sub>550</sub> values, as clearly stated above. A significant dust transport over Athens took place on 4 March 2002 and this case is analyzed as representative of DD aerosols.

The air mass trajectories for the three days are shown in Fig. 8 at three altitudes: 500 m, 1500 m and 4000 m. The UI type is associated with local air masses within the boundary layer enriched by anthropogenic aerosols, thus presenting both high AOD<sub>550</sub> and FM values (0.54 and 0.98, respectively). The CM conditions are associated with fast-moving air masses from the Atlantic Ocean (note the longer distances to be covered compared to those for the other two aerosol types). Similar air mass trajectories and characteristics associated with clean conditions in Lampedusa have been reported by Meloni et al. (2007). The MODIS AOD<sub>550</sub> on 15 January 2002 was nearly to background conditions (0.05), also having low FM fraction (0.35), characteristic of the presence of marine aerosols. On the other hand, on 4 March 2002 the air masses at the three altitudes have a Saharan origin, carrying significant amounts of dust over

Athens. The air mass reaching Athens at 4000 m was within the Sahara boundary layer favoring the uplift of dust aerosols in the free troposphere. Nevertheless, viewing the trajectory plots a vertical distribution of dust occurs on that day, enhancing the PM<sub>10</sub> concentrations on the surface to  $113 \mu\text{g m}^{-3}$ . The columnar AOD<sub>550</sub> as derived from MODIS is extremely high, 0.98, while 51% of this is attributed to coarse-mode dust aerosols.

The synoptic meteorological conditions (500 h Pa geopotential and surface atmospheric pressure) for the aforementioned three days are depicted in Fig. 9(a–c). Concerning the day on 25 April 2003, north winds were well established all over Greece because of the combination of high surface pressure over Scandinavia and low surface pressure over Russia. Besides, in the middle atmosphere, the north current was apparent, taking into consideration the 500 h Pa geopotential pressure (Fig. 9a). These meteorological patterns carry polluted air masses from Eastern Europe over Greece, additional to the accumulation of the local emissions favoring the occurrence of the UI aerosol type. In the second examined day (15 January 2004) a clear defined west circulation was obvious in the middle atmosphere, while in the surface, a trough over north Italy navigated the air masses to Greece from the west (Fig. 9b), thus bringing clean marine air above Athens. On the 4 March 2002, an expanded trough dominated over North Africa, apparent both in the middle atmosphere and the surface. These synoptic conditions steered the air masses to Greece from the south and as a consequence Saharan dust was transported to the country (Fig. 9c).

In Fig. 10(a–f) the AOD<sub>550</sub> and FM maps derived from the Terra-MODIS data are presented covering the whole Mediterranean and the greater part of Europe. This Figure, associated with air mass trajectories (Fig. 8) and meteorological conditions (Fig. 9a–c), indicates the different aerosol load and optical characteristics on the three days considered, 25 April 2003 (Fig. 10a, d), 15 January 2004 (Fig. 10b, e) and 4 March 2002 (Fig. 10c, f). On 25 April 2003, when the UI type dominated in the Athens atmosphere, the AOD<sub>550</sub> along Eastern Europe, the Balkan countries and Greece presented larger values than over the Central and Western parts. Also, low AOD<sub>550</sub> values covered the whole Mediterranean region. On this day the air masses carried pollution aerosols from Eastern Europe towards Greece and the Aegean Sea. Quite characteristic is the map regarding the FM values (Fig. 10d), where very high values covering the European Continent, Greece, Adriatic and Aegean Seas, thus confirming the pollution exposure from Europe to East Mediterranean. On 15 January 2004, the low AOD<sub>550</sub> over whole of the Mediterranean basin (Fig. 10b) is dominated by clean maritime conditions over the region, with origin from remote oceanic areas (Smirnov et al., 2002). The FM values are higher over coastal Europe (Fig. 10e), exhibiting a large gradient between continental Europe and the Mediterranean. Nevertheless, the FM values are lower than those in the previous case. The higher FM values above coastal industrialized areas are attributed to the local anthropogenic emissions. However, air pollution exposure from continental Europe to the Mediterranean or

Fig. 9. (a–c). Spatial distribution of 500-h Pa geopotential height (gpdm) along with surface atmospheric pressure (h Pa) from NCEP reanalysis for 25 April 2003 (a), 15 January 2004 (b) and 4 March 2002 (c).

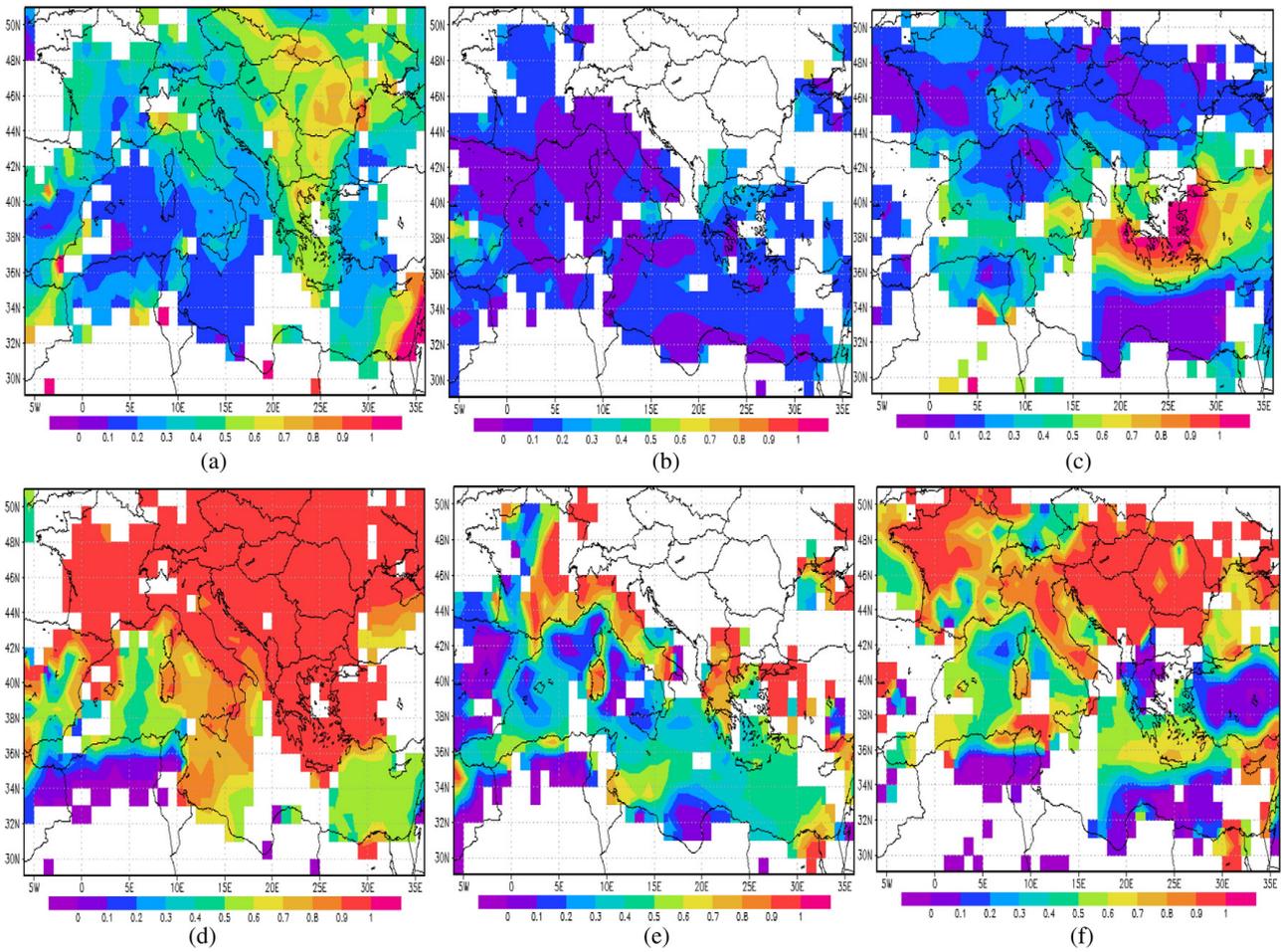


Fig. 10. (a–f). First row: AOD<sub>550</sub> values derived from MODIS on 25 April 2003 (a), 15 January 2004 (b) and 4 March 2002 (c). Second row: FM values derived from MODIS on 25 April 2003 (d), 15 January 2004 (e) and 4 March 2002(f).

Aegean Sea is not observed, as in the previous case. On the other hand, the European pollution exposure towards Eastern Mediterranean is favored in the summer months as has been highlighted in the modeling study of Duncan and Bey (2004). The Sahara dust outbreak over Greece on 4 March 2004 is highlighted by the high AOD<sub>550</sub> values (Fig. 10c) along the dusty air mass pathway (see Fig. 8). In the rest of Mediterranean and Europe, the AOD<sub>550</sub> values remain low. The presence of coarse-mode dust aerosols over Greece on this day is also confirmed by the low FM values over this area (Fig. 10f), while in the Balkans the FM values take high values directly related to anthropogenic aerosols. From this Figure, it is concluded that the MODIS data constitute a powerful tool for the discrimination and monitoring of different aerosol types in the Mediterranean basin, despite the multiple sources of aerosols. This fact has also been established by a research study of Barnaba and Gobbi (2004).

## 5. Conclusions

A six-year (2000–2005) dataset of AOD<sub>550</sub> and FM fraction from the MODIS sensor onboard the NASA-Terra satellite was analyzed in order to evaluate the seasonal variability of specific aerosol types over GAA. The long-term dataset composed of 1804 daily AOD<sub>550</sub> and FM data gives the opportunity for a climatological analysis of the aerosol types. Based on the AOD<sub>550</sub> and FM values three specific aerosol types are discriminated. Urban/industrial (UI) aerosols dominated by fine-mode particles in a turbid environment, clean maritime (CM) conditions associated with low AOD<sub>550</sub> values and coarse-mode aerosols (DD, probably desert dust) characteristic of low FM and high AOD<sub>550</sub> values. In order to quantify the mixing processes in the atmosphere and to avoid overestimation or underestimation of the specific aerosol types, an additional mixed type was assumed, which was removed from the analysis. As expected, in an urban environment, like Athens, the UI type clearly dominates (442 cases) against 337 of the DD type. In contrast, the CM conditions over Athens are relatively rare (183 cases) occurring mainly in the cold period of the year (late autumn and winter). The UI type occurred mainly in spring, directly related to the intensity of anthropogenic emissions, stagnant air masses and poor dilution of aerosols and pollutants in rain. The DD type exhibited its higher frequency in summer, when the atmospheric conditions favor the erosion of soil dust from the dry surfaces, the more stable atmospheric conditions and the absence of precipitation; all these mechanisms allow aerosols to suspend in the air. The inter-annual variability of the number of occurrences as well as the optical properties of the specific aerosol types over Athens was more pronounced for the natural aerosols (CM and DD). On the other hand, the UI type did not present great variability in its optical properties, since the anthropogenic emissions are well-defined and continuous in the GAA. On the other hand, the aerosol load and optical properties of the DD type were strongly related to the air mass pathways and natural events (e.g., Sahara dust transport over the study region), exhibiting significant variability from year-to-year. The AOD<sub>550</sub> for this aerosol type was higher in the spring due to the more intense dust events occurring in this period. Particularly high AOD<sub>550</sub> values were also observed in the winter,

despite the very low occurrence of DD type in this season. However, the occurrence of the DD type in winter was closely associated with air masses from Sahara. On the other hand, both UI and CM aerosols exhibited slightly larger AOD<sub>550</sub> values in the summer mainly attributed to the more stable atmospheric conditions favoring the residence of aerosols. The three case studies presented were quite representative of the meteorological conditions, the main pathways and the optical properties favoring the presence of each aerosol type. Therefore, MODIS can be a powerful tool for monitoring aerosols over the Mediterranean from space. In combination with additional spectral information and comparison with ground-based instruments, it is very hopeful that our knowledge on aerosols over Athens will be enriched soon.

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