

# AEROSOL CHARACTERIZATION OVER NORTHERN GREECE; AEROSOL LOADING DERIVED FROM SATELLITE OBSERVATIONS AND GROUND-BASED MEASUREMENTS

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## ABSTRACT

With the synergistic use of ground-based measurements and modelling predictions, the applicability of the Aerosol Index, as extracted from OMI/Aura observations, is assessed as a potential parameter to quantitatively describe the aerosol load over Thessaloniki, Greece. Even though it is rightly assumed that the spade of satellite measurements will highly complement and aid the use of ground-based observations, great care is required in the interpretation of their joint findings. Ground based measurements of aerosol optical depth with a sun photometer and lidar system and spaceborne observations of the aerosol index during 2005 were utilised in this work to study the varying aerosol load over Thessaloniki. The city is situated in a unique sea-side location, while being frequently affected by biomass burning and desert dust particles arriving at the location. Local and regional pollution further affect the air quality and the tropospheric optical depth. Two categories of discrete atmospheric aerosol loading were examined in detail with the combined use of dust loading modelling over the Mediterranean area and lidar measurements at days with significant *Saharan dust loading*. Furthermore, ATSR-2 World Fire Atlas observations over Europe and Russia were superimposed over back-trajectory calculations of air-masses arriving over Thessaloniki, hence identifying *biomass burning events*. Finally, results from a three-month observational campaign during the summer of 2005, combining OMI/Aura overpasses with detailed sun photometer, cloud cover and lidar measurements are presented.

## 1. INTRODUCTION

Aerosols found over the Mediterranean basin result from the superimposition of a marine component (sea spray from the Mediterranean Sea itself), a mineral dust component (outbreaks of Saharan dust and local dust suspension) and an anthropogenic component (local- and long-range transported pollution) [1],[2],[3],[4]. These different aerosol types may exhibit either absorbing or non-absorbing characteristics and hence leave a distinct signature in the observation of the atmospheric aerosol load. High aerosol optical depths may be measured during the presence of either absorbing or non-absorbing particles and hence various sources of information are necessary in order to assess both the detailed nature and the optical properties of such a complex aerosol load, as the one over the region. The basic scientific question behind this work is whether the OMI-derived Aerosol Index [AI] can act as a solid indicator of the aerosol load over regions that receive a number of different aerosol particles (local and regional pollution particles, Saharan dust, biomass burning particles transported from distant sources and maritime matter). It has been shown in previous studies [5],[6],[7],[8] that a high linear correlation exists between the AI and the Aerosol Optical Depth [AOD] measured by ground based instruments in sites near discrete sources of aerosol. However, in the case of the city of Thessaloniki, a region at the cross-roads of a variety of aerosol loaded air masses, this remains to be examined for the Aura/OMI products as was for the Earth Probe/TOMS measurements presented in [20]. In the following, we present the efforts undertaken to study the air-born particles over the Eastern European region using satellite observations, ground-based measurements and model calculations.

## 2. THE DATA SETS

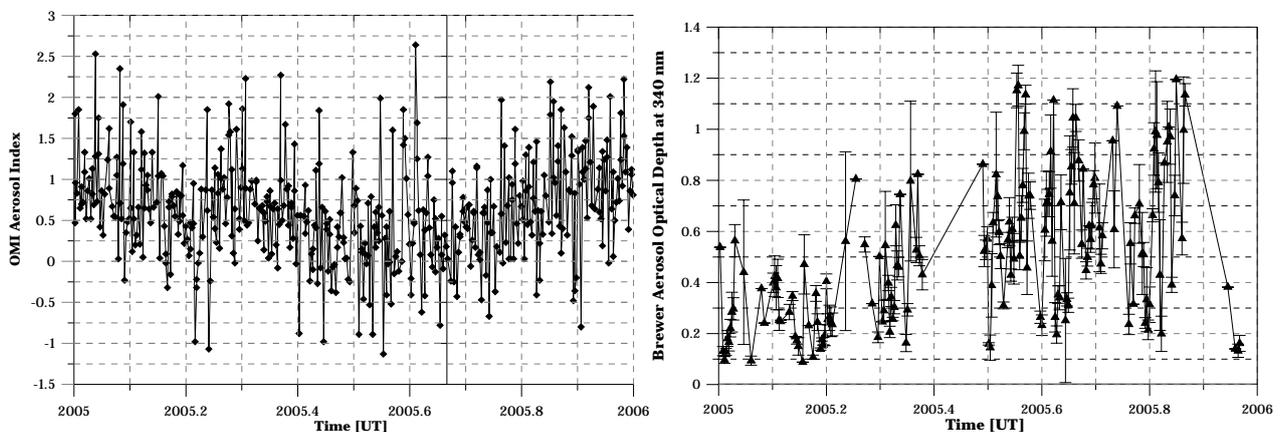
### 2.1. Aerosol Index [AI] from the Ozone monitoring Experiment on Aura/NASA

The Ozone Monitoring Instrument [OMI] on board the AURA/NASA satellite provides the Aerosol Index [AI], a measure that indicates the presence of absorbing aerosols in the troposphere [23]. It separates the spectral contrast at two ultraviolet (UV) wavelengths caused by absorbing aerosols from that of other effects, including molecular (Rayleigh) scattering, surface reflectivity, gaseous absorption, and scattering from aerosol and clouds [5]. In this work, the ratio of the upwelling radiance between observations at 331 nm and 360 nm, wavelengths where gaseous absorption of UV radiation is weak, is used. UV absorbin aerosols include smoke produced by biomass burning, black carbon from

urban and industrial activities, agricultural dust, mineral dust from arid and semi-arid regions (desert dust), volcanic aerosols and ash. Non-absorbing aerosols are primarily sulphate ( $\text{H}_2\text{SO}_4$ ) aerosols. The aerosol index is very useful for tracking global transport of smoke aerosols and dust particles, for it is not as affected by clouds as are most other aerosol products. In Fig 1 [left], the aerosol index, retrieved from the database for Aura/OMI overpasses for Thessaloniki is shown [<http://avdc.gsfc.nasa.gov/Data/Aura/index.html>]. The spread of values are from -1.13 to 2.64 with a mean of  $0.588 \pm 0.613$ . As can be noted, the vast majority of AI values are positive, showing larger values during winter, but distinct peaks are observed also during spring and summer.

## 2.2. Aerosol Optical Depth [AOD] from the Brewer Spectrophotometer

The Aerosol Optical Depth [AOD] is spectrally retrieved from a Brewer spectroradiometer situated on the roof of the Department of Physics of the Aristotle University of Thessaloniki at  $40.63^\circ\text{N}$  and  $22.96^\circ\text{E}$  and 60 m above sea-level [9]. Direct irradiance spectral measurements in the range 290-365 nm and in steps of 0.5 nm are used to retrieve the total AOD column. Brewer measurements are taken under the most stringent cloud free conditions that permit the calculation of the AOD. In Fig 1 [right], daily mean values for the AOD at 340 nm measured during 2005 over Thessaloniki are shown. The highest observed AOD under clear sky conditions, reaching values of almost 1.2, were seen during summer and the lowest value of 0.088 during winter, whereas the annual mean is  $0.510 \pm 0.28$ . The gaps in the data set represent days when no measurements were possible.



**Fig. 1. Left:** Aerosol Index over Thessaloniki derived by the Ozone Monitoring Instrument [OMI] for the year 2005. **Right:** Aerosol Optical Depth at 340 nm over Thessaloniki derived by the Brewer Spectrophotometer for the year 2005.

## 2.3. Aerosol Optical Depth [AOD] from the Raman Lidar

The Aerosol Optical Depth [AOD] at Thessaloniki is also measured at 355 nm with a UV Raman lidar and is used to investigate separately the aerosol properties in the Planetary Boundary Layer [PBL] and the Free Troposphere [FT] [10]. The basic idea is to estimate the mean aerosol properties in the layers above and below 2000 m and consider them as FT and PBL contributions, respectively. Apart from the nominal night-time measurements taken with the LIDAR, coincident day-time measurements were taken through the summer of 2005 at the time of the Aura overpass. From the study performed by [10] where data from 2001 to 2004 were analysed and a pronounced seasonal pattern was found in the retrieved AOD, with higher values close to 1.0 occurring mainly in early spring and late summer. This pattern was attributed to the enhanced contribution of the free tropospheric aerosol component to the total AOD reaching almost 50% in cases of Saharan dust outbreaks.

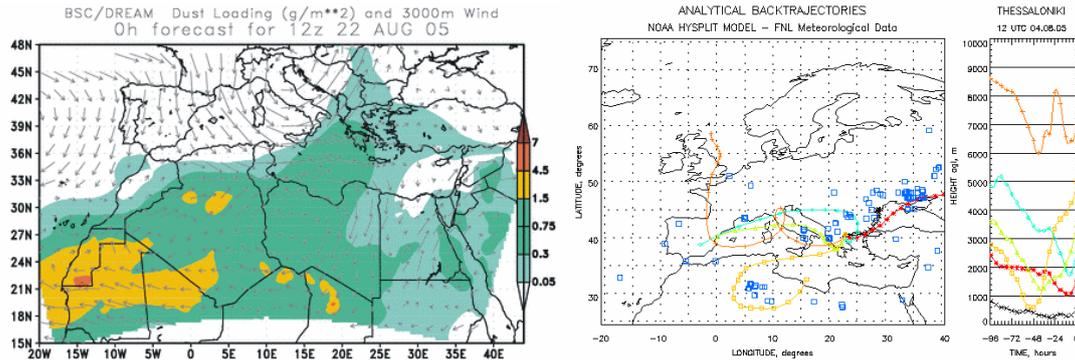
## 3. THE AEROSOL SOURCES OVER THESSALONIKI

The atmosphere above Thessaloniki, located in the northern Aegean Sea region, is affected by aerosols originating from both local and regional pollution sources, Saharan dust outbursts, long-range transport of biomass burning particles and maritime particles [11],[12],[13],[14]. Among these particles, sea-spray and sulphate aerosols are the most common non-absorbing aerosols [15], whereas carbonaceous (smoke) particles can have both absorbing and non-absorbing

characteristics [16], [15] and mineral or dust aerosols, transported from nearby dry regions, represent the clearest possible absorbing signature [17]. Two main sources of transported aerosols are Saharan dust and biomass burning particles and these have been studied in more detail.

### 3.1. Saharan Dust

In order to identify events of elevated dust loading over the region of Thessaloniki, the Dust REgional Atmospheric Modelling (DREAM) model was employed [18]. The predictions, available by the Barcelona Supercomputing Centre, <http://www.bsc.es/projects/earthscience/DREAM/>, for the entire year of 2005, depict the geographical region of Northern Africa and Southern Europe, as seen in Fig.2 [left]. The heaviest cases of Saharan dust loading occurred during the months of January to March 2005 and only moderate dust loading was observed between July and September of the same year.



**Fig. 2. Left.** The dust loading predictions by the DREAM model for August the 22<sup>nd</sup>, 2005 are shown as the colour contours. The onset of a Saharan dust storm which lasted several days and transported dust particles over the Eastern Mediterranean can be noted. However, the range of dust loading over Thessaloniki never exceeded  $0.75 \text{ g m}^{-2}$ . **Right.** Four-day back-trajectory calculations using the HYSPLIT model appear as the coloured lines of the left panel. The blue squares depict the possible fire hot spots identified by the AATSR-2 observations. In the right panel the altitude of precedence of the air masses at six different heights for which HYSPLIT was run are shown.

### 3.2. Biomass Burning

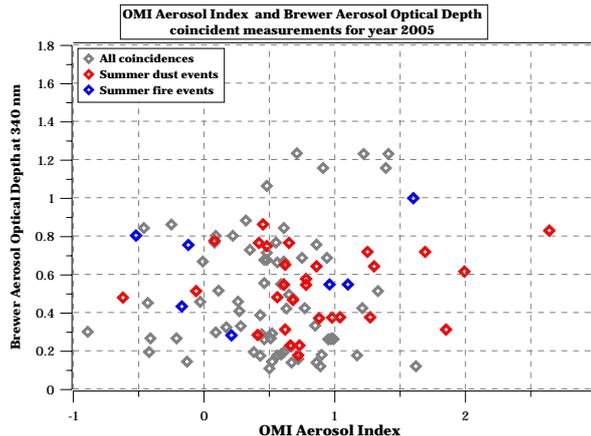
Four-day back trajectories were computed for the days of interest, using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [19], which employs the meteorological data produced by the National Weather Service ETA Model to compute advection and dispersion of air parcels. We used HYSPLIT to generate 4-day back trajectories for air-parcels arriving over the Thessaloniki site at altitudes from 500m to 6000m for the year 2005. The atmospheric trajectories were calculated for an arrival time of 12:00 UTC, which is the time closest to the average of the overpass times of the OMI/Aura to the routine Brewer observations. Biomass burning events that transport smoke particles of various origins over the city of Thessaloniki have been identified using the Along Track Scanning Radiometer (AATSR-2) World Fire Atlas online database [<http://dup.esrin.esa.int/ionia/wfa/index.asp>]. The identified events were super-imposed to the back-trajectory analysis performed using the HYSPLIT model and days when air parcels containing smoke particles arrived over Thessaloniki at 12 UTC were hence isolated. In Fig. 2 [right], the air masses at 500m and 1500m on August 4<sup>th</sup>, 2005, are seen to have traversed regions surrounding the Black Sea of heavy fire activity during the four days before arrival over the city.

## 4. THE SUMMER CAMPAIGN

During the summer of 2005, an observational campaign was hosted in the Laboratory of Atmospheric Physics of the Aristotle University of Thessaloniki including coincident ground-based measurements to the OMI overpass over the region. The routine Brewer spectrophotometer measurements were performed every fifteen minutes in the hour preceding and following the predicted OMI overpass time and Raman lidar measurements were scheduled to be taken within fifteen minutes of the named overpass time. As noted briefly in the introduction, theoretical modelling studies have been performed by reference [5] which predict the expected correlation between the AI and AOD values

depending on the aerosol type and the height of the aerosol layer. They showed that for an aerosol layer centred at either 1.5 or 3 kilometres altitude the relationship between the aerosol optical depth and the aerosol index is linear up to optical depths of 4 for weakly absorbing smoke, 2 for moderately absorbing smoke and 1 for large dust particles. For an optical depth of 0.5, which is the annual mean value observed over Thessaloniki, weakly absorbing aerosols are expected to have a near zero AI value, moderately absorbing aerosols an AI of 1 and large dust particles an AI of almost 2 and the relation between the two physical quantities is expected to be linear.

#### 4.1. OMI AI vs Brewer AOD

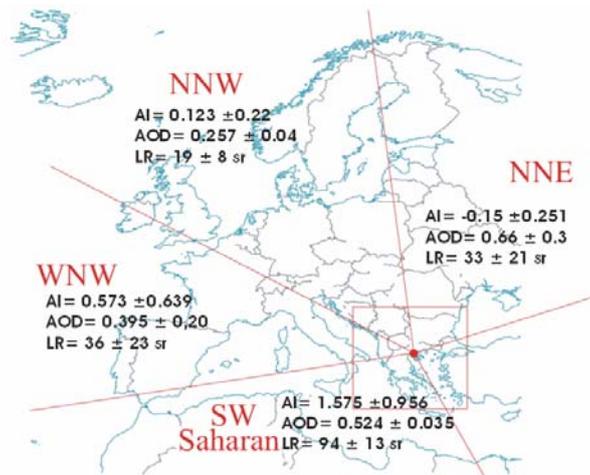


**Fig. 3.** Comparison of OMI Aerosol Index and Brewer Aerosol Optical Depth at 340nm coincident measurements from July to September 2005. The grey diamonds represent all available data, the red diamonds the measurements on days identified as carrying Saharan dust particles and the blue diamonds the measurements on days with biomass burning particles over Thessaloniki.

Based on the aforementioned theoretical assumptions the relationship between the AI and the Brewer AOD is depicted for the summer months of 2005 in Fig. 3. The entire dataset available, namely 52 coincidences, is shown as the grey diamonds, whereas the days when the air bore Saharan dust particles predicted by the DREAM model (17 coincidences) are shown in red and the days when the air bore biomass burning particles as stipulated by the four-day back-trajectories and AATSR-2 observations are shown in blue (13 coincidences). The reflectivity, derived from the OMI measurements, was constrained to the 20% limit. The spread in absolute values of both physical variables appears promising in identifying different types of aerosols, however the limited amount of data does not allow to establish a significant linear correlation between AOD and AI. A similar study performed using the Earth Probe/TOMS Version 8.0 Aerosol Index for the city of Thessaloniki but for a range of coincident Brewer measurements during 1997 to 2001 [20] has yielded more promising leads, since a tentative correlation between the AOD and AI values for 223 biomass burning events was found to be of the order of 0.67 whereas a correlation of 0.57 was deduced for the 37 cases of Saharan dust loading. It is possible that the higher number of coincident cases and the stringiest selection criteria have led to a better data set for the proposed comparisons.

#### 4.2. OMI AI vs Lidar AOD

Twenty-two coincident OMI overpass and lidar measurements during the same summertime campaign were used for this comparison. Four-day back trajectory calculations on these coincidences were performed using HYSPLIT as described in Section 3.2. Furthermore, the data were grouped using a cluster analysis algorithm [21] which segregated the computed atmospheric back-trajectories into different groups, the so-called clusters. From this analysis four distinct clusters were identified, as seen in Fig. 4.: the NNE cluster which corresponds to air masses from Eastern Europe, the SW cluster, which corresponds to air masses originating from the Sahara desert, the NNW cluster, which corresponds to air masses mostly advected from central Europe, and finally the WNW cluster which corresponds to air masses from Italy, Spain and possibly the Atlantic Ocean. As seen from Fig. 4, the lowest mean AOD values ( $0.257 \pm 0.04$ ) correspond to the NNW cluster which represents central European air and possibly fast transport from the Atlantic Ocean. For this cluster the AI also shows low values ( $0.123 \pm 0.22$ ) indicating less absorbing aerosols.



**Fig. 4.** Average properties of the aerosols during summer 2005 over Thessaloniki relative to the origin of the air-masses.

High AOD and AI values are associated with the WNW cluster, namely  $0.573 \pm 0.639$  and  $0.395 \pm 0.20$ . For these coincidences an increased free-tropospheric contribution to the total AOD is also observed, which further points that higher aerosol layers are better detectable by OMI [23]. The lowest AI ( $-0.15 \pm 0.251$ ) values are associated with aerosols originating from the NNE cluster and with large variability of the optical depth ( $0.66 \pm 0.30$ ). NNE flow transports pollution from the Balkans and Eastern Europe and during summertime this is associated with regions with large emissions from biomass burning episodes and heavy industrial activity. Free tropospheric contribution was found to be significant for this cluster and the small AI values observed are consistent with single scattering albedo (SSA) estimates of  $0.95 \pm 0.03$  that indicate less absorbing aerosols for this cluster. Finally, the SW cluster which is mostly associated with Saharan dust transport is related to higher free tropospheric contribution to the total AOD and the absorbing features of dust are better represented by the AI ( $1.57 \pm 0.035$ ), which shows the lowest variability.

Complementary studies might enlighten the deductions for two of the abovementioned clusters. Reference [22] have calculated the SSA using global spectral irradiances taken by the Brewer spectrophotometer and have shown that the SSA for the NNW cluster suggests the presence of more absorbing aerosols which however correspond to low AOD ( $0.257 \pm 0.04$ ) and AI ( $0.123 \pm 0.22$ ) values. Since the main aerosol load for this cluster is thought to be dominated by the absorbing aerosols found in the boundary layer, the SSA should have been also affected. This may point to the fact that the AI fails to represent the more absorbing characteristics of Thessaloniki's boundary layer aerosol load, since any aerosols below about 1000m is unlikely to be detected. On the contrary, the NNE cluster (associated with large summertime emissions from biomass burning episodes) is well represented in the AI with negative values ( $-0.15 \pm 0.251$ ) and high variability of the optical depth ( $0.66 \pm 0.30$ ), also seen in the same study of [22], where the SSA for this airflow was found to be  $0.95 \pm 0.03$  identifying mainly scattering particles in the air.

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