Trends in the aerosol load properties over south eastern Italy

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Abstract. The long-term (2003-2013) variations in columnar aerosol properties at Lecce, a site representative of the central Mediterranean, have been analysed for trend assessment. The study focuses on aerosol optical thickness (AOT) at 340, 440, 500 and 1020 nm and Ångström exponent (AE) for the pair 440-870 nm, retrieved from a sun photometer operating within the Aerosol Robotic Network (AERONET). A non-parametric trend analysis of the monthly mean, median and upper and lower tails (90th and 10th percentiles) suggests that the aerosol load has decreased during the study period, while the mean particle size remained unchanged. The characteristic advections reaching the study site were found by clustering analysis of back trajectories at 500, 1500 and 3000 m. Despite the strong influence they have on aerosol load and particle size, neither of the trends in advection routes could explain the tendencies found in the columnar aerosol properties. However, trends in aerosol data by advection type allow understanding the overall trends. Aerosol properties under flows with high residence time over continental Europe present differences according to the specific residing area. More specifically, no trend is found when flows arrive from Ukraine and the Balkans, while under advections from north-western/central Europe there are downward trends in the background levels and a reduction of the fine fraction. Negative trends are also found under flows with high residence time over the Mediterranean and northern Africa, again with differences according to the residing area.

1. Introduction

Atmospheric aerosols are an important component of the atmosphere due to their role in the Earth’s radiation budget and in cloud and precipitation processes, and due also to their impact on air quality. Aerosol effects largely depend on the aerosol optical and microphysical properties. The long term analysis of such properties is of particular importance to investigate the changes of natural and anthropogenic aerosol contributions and to better evaluate the aerosol impact on the surface radiation balance.

Numerous studies on the temporal variation of aerosols and the assessment of trends have been conducted at different spatial and time coverage. Works using measurements at the global scale with satellite sensors and those dealing with data from a large number of monitoring stations may give (within intrinsic limitations and differences) a broad picture of the existence of trends in aerosol load and properties. To this end, studies of AOT measured by satellite sensors such as the Advanced Very High Resolution Radiometer (AVHRR) \cite{1}, the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) \cite{2}, the Moderate Resolution Imaging Spectroradiometer (MODIS) \cite{3, 4, 5} and the Multispectral Imaging Spectroradiometer (MISR) \cite{4, 5} were performed, in addition to AOT studies using ground-based measurements by sun photometers at a large number of sites belonging to AERONET \cite{6, 7}, and
aerosol optical parameters at the ground level retrieved with nephelometers and aethalometers within the GAW and the Interagency Monitoring of Protected Visual Environments (IMPROVE) networks [8]. Studies of long-term observations of PM10 concentrations at the EMER (European Monitoring and Evaluation Programme) monitoring sites [9], and of aerosol number concentrations at GAW (Global Atmosphere Watch) and ACTRIS (Aerosols, Clouds, and Trace gases Research Infrastructure Network) sites [10] were also performed. These studies found, in general, downward trends over Europe and North America (mostly in their western and eastern parts, respectively), and mostly upward trends over South and East Asia. These tendencies are consistent with the emission-inventory assessments in these regions. As for the trends observed in dust emissions and export from source areas, it is found a positive trend in the Arabian Peninsula while in northern Africa the trend is negative.

We present an analysis of AOT and AE data for the period 2003-2013 at Lecce, south eastern Italy, for long-term trend assessment. The monitoring site can be considered as representative of coastal sites in the Central Mediterranean away from large sources of local pollution. The dependence of the columnar aerosol optical properties on the synoptic-scale advection patterns is analyzed. The observed strong dependence motivates the subsequent exploration for the existence of trends both in the frequencies of the major advection types and in the aerosol load and its optical properties by advection type.

2. Data and methods

A ten-year (March 2003-February 2013) dataset of AOT at wavelengths 340, 440, 500 and 1020 nm and AE for the 440-870 nm pair is considered. Level 2 data, cloud screened and quality assured, were retrieved from direct solar measurements with a CIMEL CE 318 sun/sky photometer operating within AERONET at the University of Salento (40.33 N, 18.10 E, 27 m asl; Lecce, southeastern Italy) and are available at http://aeronet.gsfc.nasa.gov. The methods used to measure solar radiation and the instrument description are given, e.g. in [11]. The algorithms for cloud-screening and the retrieval of aerosol properties are described in [12] and [13]. The measurement error of the aerosol optical thickness is estimated to be in the range of 0.01–0.02 for the visible channels and 0.02 for UV [11, 14].

The assessment of monotonic trends was performed by non-parametric procedures based on the Mann-Kendall test in combination with the Theil-Sen (TS) slope estimate, over the time series of monthly mean, median, 10th and 90th percentile values. The Mann-Kendall test allows working with non-normal data and in situations with many missing values, and it is resistant to extreme cases. It, however, requires independence between observations; therefore, short-term persistence and seasonality alter the significance levels and may lead to wrong conclusions about the presence of a trend. The seasonal Kendall (seasK) test [15], as well as the trend-free pre-whitening (TFPW) procedure [16] applied over the seasonally adjusted monthly series (deseas+TFPW), were used. The first test is robust against seasonality but not against serial dependence, while the second one removes the lag-1 autocorrelation from the deseasonalized time series before applying the Mann-Kendall test. Two-tailed tests at 95 and 90% significance were conducted. The deseasonalized time series used in the TFPW procedure were obtained from a seasonal-trend decomposition of the series based on LOESS (locally weighted low-degree polynomial regression) applied recursively to the seasonal and trend components [17].

Kinematic 96-hour back-trajectories were calculated with the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model v 4.8 at 500, 1500 and 3000 m asl, every 6 hours (00, 06, 12, 18 UTC) during the study period. ERA-Interim data of 1.5° resolution was used as meteorological input. The trajectories were subsequently classified into homogeneous groups by a robust clustering procedure based on the k-means algorithm. Hourly longitudes and latitudes were the input variables and the similarity metrics was based on great-circle distances. A detailed description of the procedure can be found in [18] or [19]. Each trajectory was associated to the closest AERONET data in time to study aerosol properties by advection type.
Figure 1. Single data (grey circles), monthly means and 10th and 90th percentile (pct10 and pct90, respectively) time series of AOT and AE in the study period. The linear trends were estimated with the Theil-Sen slope.
3. Results

3.1. AOT and AE trends
According to both tests there exist significant downward trends in all the AOT time series, at least at the 90% confidence level, while no trend is found in the AE calculated at the 440 and 870 nm wavelength pair (see Figure 1 and Table 1). These results suggest that the aerosol load has decreased over the study period while the mean particle size likely remained unchanged from 2003 to 2013.

The trends reported by Li et al. [7] for the AOT(440) and AE monthly medians are similar to the ones reported here. It is also observed (Table 1) that the (negative) AOT trends are stronger at lower wavelengths. In addition, at each wavelength the strongest trends are found for the 90th percentile time series while the weakest ones are found for the 10th percentile values. This means that the number of intense episodes have decreased over the study period, while the background aerosol load shows a much lesser lowering if any. The trends in the 90th percentile of the AOT at 1020 nm (that is more sensitive to coarse mode particles) and of the AOT at 340 nm (more sensitive to fine mode particles) would indicate that the concentration of both coarse and fine mode particles are decreasing over time.

Table 1. TS trend estimates for the monthly AERONET time series that present significant trends at 95% (in bold) or 90% confidence level according to the SeasK and deseas+TFPW tests. Trends were calculated on the original and the seasonally-adjusted series, respectively.

| Aerosol parameter | seasK Trend (decade⁻¹) | deseas+TFPW Trend (decade⁻¹) |
|------------------|------------------------|-------------------------------|
| Mean AOT(1020)   | -0.026                 | -0.023                        |
| Median AOT(1020) | -0.017                 | -0.013                        |
| pct10 AOT(1020)  | -0.008                 | -0.007                        |
| pct90 AOT(1020)  | -0.061                 | -0.065                        |
| Mean AOT(500)    | -0.057                 | -0.051                        |
| Median AOT(500)  | -0.052                 | -0.041                        |
| pct10 AOT(500)   |                        | -0.022                        |
| pct90 AOT(500)   | -0.115                 | -0.010                        |
| Mean AOT(440)    | -0.062                 | -0.053                        |
| Median AOT(440)  | -0.058                 | -0.045                        |
| pct10 AOT(440)   |                        | -0.025                        |
| pct90 AOT(440)   | -0.115                 | -0.103                        |
| Mean AOT(340)    | -0.084                 | -0.073                        |
| Median AOT(340)  | -0.084                 | -0.067                        |
| pct10 AOT(340)   | -0.050                 | -0.040                        |
| pct90 AOT(340)   | -0.133                 | -0.123                        |

3.2. Major advection patterns and trends in their monthly occurrence
Advection at the synoptic scale has a strong impact on the aerosol optical properties and mass concentration, as it has been shown at the study site for different, shorter, time periods [19, 20, 21]. The major advection routes were identified by a cluster analysis of back-trajectories, which resulted in 7, 6, and 7 major routes at 500, 1500 and 3000 m asl, respectively (see Figure 2). They are named according to their geographical pathway.
The airflows with a significant residence time over the Mediterranean are W500/WSW1500,3000, which cross the western Mediterranean after passing over the Iberian Peninsula or the northern-most part of Africa; and S which are slow-moving flows arriving from the southern side of the central Mediterranean basin. While WSW flows are common throughout the year with slightly higher frequency in spring and autumn, S are less frequent in the central months.

The advection routes with continental Europe characteristics are NE flows, which pass over Ukraine and the Balkans before reaching the study site and are more frequent in spring and autumn, while in March they are almost absent; and sNW 500/WNW1500/sWNW3000 slowly-moving flows that pass over the Italian Peninsula, are the most common type of advection and present strong seasonality due to their strong predominance in summer at the study site; and NW 500 which cross over northwestern/central Europe after arriving from the North Sea.

There are two types of fast advectons influencing SE Italy: NNW flows, which are of Arctic/continental polar nature, and WNW 500/fWNW1500,3000 flows that correspond to polar maritime air masses traced back to north America. Both are the less frequent advection routes and show a strong seasonal behavior as are mostly found from November to March. NNW are more frequent in March, while the polar maritime air masses show a maximum in December/January.

Further details on the synoptic and the local meteorology associated to each advection route can be found in [20], with minor dissimilarities due to differences in the study time periods. Figure 3 shows the basic statistics of the AERONET aerosol properties by advection route. Three contrasting behaviors can be appreciated: (1) S and W500/WSW1500,3000 flows have the lowest AE and the highest AOT values, indicating loaded situations with coarse particles. (2) NE and sNW 500/WNW1500/sWNW3000 flows present the highest AE values and high AOT levels (these latter are higher the lower the wavelength and AOT340 are comparable to those of S and W500/WSW1500,3000) and correspond to fine particles of anthropogenic origin. (3) NNW, as well as WNW 500/fWNW1500,3000 advectons, show the lowest AOT and intermediate AE levels that are lower for the polar maritime air masses, and therefore are clean air masses.

The trend analysis of the monthly frequencies reveals monotonic trends in only a few advection types: NNW flows reaching the study site at 3000 m show downward trend (at the 95% confidence level) and also at 1500m (at the 90% confidence) with both analyses (Table 2). The relative reduction of NNW3000 is strong: 7 cases per decade for a mean of 11 cases per year. In turn, sWNW and fWNW at 3000 m present upward trends but at lower significance, as it is found for S flows at 500 m. Neither of these trends in advection routes explains the trends found in the columnar aerosol parameters.

### Table 2. TS trend estimates for the monthly frequencies of occurrence of the advection patterns that present significant trends at 95% (in bold) or 90% confidence level according to the SeasK and deseas+TFPW tests. Trends were calculated on the original and the seasonally-adjusted series, respectively.

| Parameter          | seasK Trend (# decade⁻¹) | deseas+TFPW Trend (# decade⁻¹) |
|--------------------|---------------------------|--------------------------------|
| Freq S500          | -4.2                      | -3.8                           |
| Freq NNW1500       | -3.2                      | -3.8                           |
| Freq NNW3000       | -7.1                      | -7.7                           |
| Freq fWNW3000      | 2.0                       | -7.7                           |
| Freq sWNW3000      | 4.6                       | 5.8                            |

The airflows with a significant residence time over the Mediterranean are W500/WSW1500,3000, which cross the western Mediterranean after passing over the Iberian Peninsula or the northern-most part of Africa; and S which are slow-moving flows arriving from the southern side of the central Mediterranean basin. While WSW flows are common throughout the year with slightly higher frequency in spring and autumn, S are less frequent in the central months.
3.3. Trends in AOT and AE by advection route

We focus on the results for the advections reaching the study site at 500 and 3000 m. It should be noted that in some cases the SeasK test could not be applied to the data segregated by advection type due to the lack of data in some seasons; the analysis in these cases is made with the deseas+TFPW procedure and it is checked that the confidence intervals of the TS slope do not include the zero slope.

The analysis of the aerosol parameters by advection at 500 m reveals (Table 3) mostly negative trends both under advections associated to fine aerosols and under advections associated to the coarse fraction. AOT values under sNW and NW flows present downward trends; however, no trend is found under NE flows which are also associated to anthropogenic aerosols. Under sNW, trends in mean and median AOT are similar to those of the overall AOT at 1020 and 500 nm, although they become lower and less significant at lower wavelengths. The trends in the AOT upper tails (90th percentile) may indicate a reduction of episodic events during sNW advections. Under NW flows, the 10th percentile AE is found to have a significant negative trend (95% confidence) by the two tests; similarly, the 10th percentile of all the AOT values show a decreasing trend at the 95% confidence after the
deseas+TFPW test, with trend magnitudes that decrease with decreasing wavelength. This may indicate a reduction in the background concentrations and also a reduction of the fine mode fraction. In turn, the lower tails in NNW show positive trends of small magnitude after the deseas+TFPW test.

![Box plots of AOT and AE levels by advection type.](image)

**Figure 3.** AOT and AE levels by advection type. Mean (dot), median (horizontal bold line), 25th and 75th percentiles (lower and upper box boundaries) and 5th and 95th percentiles (whiskers) are shown.

Regarding the aerosol properties under advections associated to coarse particulates like African dust and sea-salt particles, the mean, median and 10th percentile AOT at 500, 440 and 340 nm under S flows present negative trends of much higher magnitude than the overall AOT trends, while the trends for the upper tails are of the same magnitude. Under W flows, trends behave mostly like S advections but with higher significance for the AOT(1020) values.

Trends in the aerosol parameters by advections reaching Lecce at 3000m show some differences (Table 4): no trend is found under S advections, while under WSW there are negative trends in all the AOT parameters and an upward trend in the 90th percentile AE that would imply a reduction in dust load associated to these flows and also a decrease in the coarse-dominated episodes. The AE parameters present downward trends also under NNW (mean and 90th percentile) and fWNW advections (all the parameters). While NNW show no trends in AOT, fWNW present trends in AOT(340) and AOT(440) and mWNW flows present downward trends in all the AOT parameters.
Finally, like in the case of S flows, the other two types of slower advections, i.e. NE and sWNW, show no trend.

### Table 3. TS trend estimates for the monthly time series by advection type reaching the study site at 500 m, with significant trends at 95% (in bold) or 90% confidence.

| Aerosol parameter | seasK Trend (decade⁻¹) | desea+YP Trend (decade⁻¹) | Aerosol parameter | seasK Trend (decade⁻¹) | desea+YP Trend (decade⁻¹) |
|-------------------|-------------------------|---------------------------|-------------------|-------------------------|---------------------------|
| **W₅₀₀**          |                         |                           | **NNW₅₀₀**        |                         |                           |
| mean AOT(1020)    | -0.052                  | -0.077                    | pct10 AOT(1020)   | 0.009                   |
| median AOT(1020)  | -0.049                  | -0.076                    | pct10 AOT(500)    | 0.018                   |
| pct10 AOT(1020)   | -0.040                  | -0.052                    | pct10 AOT(440)    | 0.026                   |
| pct90 AOT(1020)   | -0.060                  | -0.112                    | pct10 AOT(340)    | 0.032                   |
| mean AOT(500)     | -0.105                  | -0.140                    |                  |                         |
| median AOT(500)   | -0.093                  | -0.128                    |                  |                         |
| pct10 AOT(500)    | -0.072                  | -0.088                    | pct10 AOT(1020)   | -0.010                  |
| pct90 AOT(500)    | -0.115                  | -0.146                    | pct10 AOT(500)    | -0.037                  |
| mean AOT(440)     | -0.113                  | -0.146                    | pct90 AOT(500)    | -0.050                  |
| median AOT(440)   | -0.104                  | -0.141                    | pct10 AOT(440)    | -0.042                  |
| pct10 AOT(440)    | -0.082                  | -0.101                    | pct90 AOT(440)    | -0.052                  |
| pct90 AOT(440)    | -0.127                  |                         |                  | -0.063                  |
| mean AOT(340)     | -0.138                  | -0.177                    | Mean AE(440,870)  | -0.184                  |
| median AOT(340)   | -0.129                  | -0.135                    | pct10 AE(440,870) | -0.418                  |
| pct10 AOT(340)    | -0.118                  | -0.135                    |                  | -0.355                  |
| pct90 AOT(340)    | -0.150                  |                         |                  |                         |
| **S₅₀₀**          |                         |                           |                  |                         |
| pct10 AOT(1020)   | -0.040                  | -0.088                    | pct90 AOT(1020)   | -0.046                  |
| mean AOT(500)     | -0.108                  | -0.146                    | mean AOT(500)     | -0.060                  |
| median AOT(500)   | -0.115                  | -0.146                    | median AOT(500)   | -0.055                  |
| pct10 AOT(500)    | -0.102                  | -0.146                    | pct90 AOT(500)    | -0.022                  |
| mean AOT(440)     | -0.106                  | -0.146                    | pct90 AOT(440)    | -0.087                  |
| median AOT(440)   | -0.124                  | -0.146                    | mean AOT(440)     | -0.061                  |
| pct10 AOT(440)    | -0.107                  | -0.146                    | pct90 AOT(440)    | -0.094                  |
| pct90 AOT(440)    | -0.112                  | -0.146                    | mean AOT(340)     | -0.074                  |
| mean AOT(340)     | -0.140                  | -0.146                    | median AOT(340)   | -0.073                  |
| median AOT(340)   | -0.157                  | -0.146                    | pct10 AOT(340)    | -0.042                  |
| pct10 AOT(340)    | -0.135                  | -0.146                    | pct90 AOT(340)    | -0.112                  |
| pct90 AOT(340)    | -0.133                  |                         |                  | -0.102                  |
| **sWNW₅₀₀**       |                         |                           |                  |                         |
| pct90 AOT(340)    |                         | -0.133                    | pct90 AOT(340)    | -0.067                  |

### 4. Summary and conclusions

The long-term variations in aerosol properties at a site representative of the central Mediterranean have been explored in this work. A nonparametric trend analysis of monthly statistics of AOT and AE gives statistical evidence that the aerosol load has decreased over 2003-2013 in SE Italy, while the mean particle size remained unchanged.

The synoptic-scale advections have a strong influence on the aerosol load and particle size. However, their trends in frequency of occurrence cannot explain the observed trends in the aerosol properties. It should be noted that meteorology shows a strong interannual variability that is not well described in terms of linear tendencies. The trends in AERONET aerosol properties by advection type allow to understand the origin of the trends in the overall time series. Trends under advections with high residence time over continental Europe present clear differences depending on their specific residing area: no trend is found with flows from Ukraine and the Balkans (NE), while aerosols under
advections that arrive from anywhere in the Italian Peninsula (sNW) present trends similar to the overall AOT series, and advections from north-western/central Europe (NW) show downward trends in the background levels and a reduction of the fine fraction that would be a signal of the reduction of the emissions in EU. The flows with high residence time over the Mediterranean, which may also carry African dust to the study site, present also some differences in the corresponding aerosol trends: the aerosol load have decreased under W_{500}/WSW_{3000} advections while such a reduction is found only under S at the lowest level. Fast advections, of polar continental/arctic or polar maritime origin show a reduction in the fine fraction when they arrive at 3000 m; and while the WNW_{500}/fWNW_{3000} show a decrease in AOT(340), there is an increase in the background aerosol load under NNW reaching the site at the lowest height.

The present exploratory study highlights that the analysis of the aerosol properties at a site by large-scale advection routes may show separately the long term changes in emissions at different source areas. Future work on PM10 mass concentration may provide insight on the presence of aerosols inside/above the ABL and would help to further understand the causes of long-term variation. However, differences in sampling scheme with respect to the columnar measurements, as well as the influence of cloud disturbance and onset and end times of pollution episodes on the latter need to be accounted for to properly compare measurements.

Table 4. TS trend estimates for the monthly time series by advection type reaching the study site at 3000 m, with significant trends at 95% (in bold) or 90% confidence.

| Aerosol parameter | deseas+TFPW Trend (decade^{-1}) | Aerosol parameter | seasK Trend (decade^{-1}) | deseas+TFPW Trend (decade^{-1}) |
|-------------------|----------------------------------|-------------------|--------------------------|----------------------------------|
| **WSW\textsubscript{3000}** |                                  | **mWNW\textsubscript{3000}** |                          |                                  |
| mean AOT(1020)    | -0.063                           | mean AOT(1020)    | -0.030                   | -0.027                           |
| median AOT(1020)  | -0.075                           | median AOT(1020)  | -0.024                   | -0.021                           |
| pct10 AOT(1020)   | -0.054                           | pct10 AOT(1020)   | -0.024                   | -0.026                           |
| pct90 AOT(1020)   | -0.063                           | pct90 AOT(1020)   | -0.042                   | -0.033                           |
| mean AOT(500)     | -0.111                           | mean AOT(500)     | -0.083                   | -0.072                           |
| median AOT(500)   | -0.116                           | median AOT(500)   | -0.068                   | -0.056                           |
| pct10 AOT(500)    | -0.086                           | pct10 AOT(500)    | -0.056                   | -0.057                           |
| pct90 AOT(500)    | -0.114                           | pct90 AOT(500)    | -0.116                   | -0.096                           |
| mean AOT(440)     | -0.115                           | mean AOT(440)     | -0.096                   | -0.081                           |
| median AOT(440)   | -0.123                           | median AOT(440)   | -0.079                   | -0.066                           |
| pct10 AOT(440)    | -0.093                           | pct10 AOT(440)    | -0.065                   | -0.067                           |
| pct90 AOT(440)    | -0.120                           | pct90 AOT(440)    | -0.132                   | -0.109                           |
| mean AOT(340)     | -0.140                           | mean AOT(340)     | -0.139                   | -0.119                           |
| median AOT(340)   | -0.150                           | median AOT(340)   | -0.122                   | -0.106                           |
| pct10 AOT(340)    | -0.118                           | pct10 AOT(340)    | -0.106                   | -0.109                           |
| pct90 AOT(340)    | -0.162                           | pct90 AOT(340)    | -0.183                   | -0.161                           |
| pct90 AE(440,870) | 0.181                            | pct90 AE(440,870) |                         |                                  |
| **NNW\textsubscript{3000}** |                                  | **fWNW\textsubscript{3000}** |                          |                                  |
| mean AE(440,870)  | -0.194                           | median AOT(440)   | -0.056                   |                                  |
| median AE(440,870)| -0.213                           | median AOT(340)   | -0.071                   |                                  |
| pct90 AE(440,870) | -0.194                           | pct10 AOT(340)    | -0.086                   |                                  |
|                  |                                  | mean AE(440,870)  | -0.054                   |                                  |
|                  |                                  | median AE(440,870)| -0.287                   |                                  |
|                  |                                  | pct10 AE(440,870) | -0.270                   |                                  |
|                  |                                  | pct90 AE(440,870) | -0.276                   |                                  |
|                  |                                  |                    | -0.268                   |                                  |
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