Reply to reviewers

acp-2015-902

Detection of Saharan dust and biomass burning events using near real-time intensive aerosol optical properties in the northwestern Mediterranean.

The authors would like to thank the reviewer for their comments and suggestions, which helped improving the quality of this work. A new version of the manuscript has been prepared following the suggestions. We provide below detailed replies to each of the comments.

Anonymous referee #1

Reviewer#1. General comment 1). This paper by Ealo et al. presents a very interesting idea for the real-time detection of dust and biomass burning events. However, one major concern I see with this technique is the difficulty to differentiate between the dust and biomass burning events, both dust and biomass being strong absorber in UV. This issue might be bigger in summer when the co-occurrence of SDE and Wildfire events may be highly probable. Due to re-circulation, these events may not be differentiated over prolonged time scales. Please discuss.

Reply to Reviewer#1. General comment 1). The authors agree with the referee that the concomitance of biomass burning and wildfires episodes during Saharan dust events (SDE) may be an issue. As stated in the manuscript, one of the objectives of this paper is to study the limitations of the proposed technique, firstly developed by Collaud Cohen et al. (2004), and to highlight the necessity of a better estimation of the different episodes through a multidisciplinary approach.
For example, as we show in the manuscript, wildfires events can be detected by means of different tools such as back-trajectories, forecast models and remote sensing data (satellite images, ceilometer/LIDAR and sun photometers measurements), and considered as isolated events (Fig. 8 in the manuscript). Concerning biomass burning events, the scattering Ångström exponent (SAE) parameter is certainly a useful parameter to establish differences between mineral dust (coarse material) and biomass burning (finer aerosol). Moreover, it is also possible to consider online PMx mass data in order to assess the SAE parameter regarding the changes in the size of aerosols. In addition, the black carbon concentration, which considerably increases during biomass burning events (See Fig. 8 describing a wildfire episode in the manuscript), can be also investigated. Furthermore, offline chemical filter analysis is a valuable input for the identification of iron oxides contained within the mineral dust to characterize the atmospheric situation.
It should be also considered that biomass burning is an active source in winter in the area under study and that during the study period (2012-2014) only three SDE were identified in winter. Furthermore, biomass burning concentration is relatively lower compared to northern areas in Europe; annual average contribution was estimated in 36%. During these winter SDE we did not see a marked daily cycle of AAE, therefore the dominance of mineral dust appears to be larger with respect to biomass burning regarding the effects on intensive optical properties.

In order to take into account the Referee's comment the following sentence has been added to section 5.3.3 in the revised manuscript:

"The concomitance of biomass burning and wildfire episodes during SDE may be an issue, being both dust and biomass burning strong absorbers in the UV. The SAE is a useful parameter that should be considered in order to establish differences in near real-time between mineral dust (coarse material) and biomass burning (finer aerosol). However, since relatively low biomass burning concentration was found in the area under study, the dominance of mineral dust appears to be larger with respect to biomass burning regarding the effects on intensive optical properties. Furthermore the co-occurrence of SDE and biomass burning winter emissions is not usual. Whereas for differentiating wildfires and SDE, both frequently occurring during summer, wildfires events can be considered as isolated events and detected by means of different tools such as back-trajectories, forecast models and remote sensing data".

Reviewer#1. General comment 2). This technique makes use of intrinsic properties of the aerosol species like Absorption, Scattering and Single Scattering Albedo Angstrom Exponents. However, these properties are influenced by environmental factors like temperature, RH, aerosol aging time, which is not discussed in this study. A part of difference in aerosol optical properties between MSY and MSA may be due to the fact that aerosol processing at these locations may be different and aerosol may have different properties. These concerns are highlighted especially during the re-circulation events. Please discuss.

Reply to Reviewer#1. General comment 2). The most important ambient parameter affecting scattering measurements is RH. High values of RH produce hygroscopic growth on particles, enhancing the scattering coefficient. It should be note that nephelometer measurements were made at constant RH, lower than 40%, following standards recommended by ACTRIS in order to avoid possible scattering enhancement due to hygroscopicity. Moreover, also the temperature (changing with RH) is controlled in the nephelometer cell and kept at almost constant values throughout the year.

Certainly, the aging of the aerosols affects the aerosol intensive optical properties (IOP). However, the study of aerosol aging is far beyond the scope of this work. In this manuscript we are mainly interested in studying how IOP change during SDE and biomass burning events, which dominate the IOP during their occurrence. Moreover, as we show in the manuscript, the proximity to anthropogenic sources of fine PM is the main affecting the SSAAE.

Despite this fact, the effect of aerosol aging time on the intensive optical properties is partially considered and discussed in the text within the air mass origin analysis, where Atlantic advections, Saharan dust events and re-circulation episodes were considered (Fig. 2a and 2d in the manuscript). Atlantic advections, occurring more frequently during winter, are characterized by dominance of fresh aerosols. MSA is usually within free troposphere during the colder period and influenced by clean air masses from Atlantic advections. However re-circulation episodes usually take place during warmer months, when both stations are frequently within the planetary boundary layer. These episodes are identified by stagnated air masses, high insolation and lack of precipitation lasting for few days, resulting in a dominance of aged aerosol. Probably the aerosol aging time is more pronounced at MSA, since it is more isolated from fresh anthropogenic pollutant sources transported mainly from Barcelona urban and industrial areas. Then pollutants at MSA present larger atmospheric residence time and thus also probably stronger aging process than at MSY.

Nevertheless, for a deeper evaluation of how these variables independently affect the intensive optical properties it would be necessary to perform specific studies, such as evaluating the aerosol aging time and coating processes.

Reviewer#1. Specific comment 1). The nephelometer instrument was calibrated only 3 times a year and zero adjust was carried out once a day may possibly insufficient for unbiased measurements. Some plots or data showing the stability of the instrument can be helpful in supporting the frequency of calibration and zero adjustments.

Reply to Reviewer#1. Specific comment 1). We calibrated both nephelometers following the standards proposed by ACTRIS network in order to obtain high quality data, and comparability among all ACTRIS stations. Both instruments have participated in workshop intercomparisons organized by ACTRIS and showed satisfactory comparability with other EU nephelometer and "reference" instruments.
As an example, Figure 1 shows the stability of nephelometer measurements at MSA station. Figure 1 shows the average scattering coefficients at the three wavelengths for different percentiles (0.05, 0.25, 0.50, 0.75, 0.95) during zero check calibration for the year 2013. Average ± standard deviation are reported for each wavelength during zero check. Since acceptable limits for zero check are set in ±2 Mm^{-1}, our data are within the standards recommended by the manufacturer.
It should be noted that some problems concerning the stability of measurements were found after long-distance transportation. However, most of these problems were solved through a full calibration of the nephelometer.

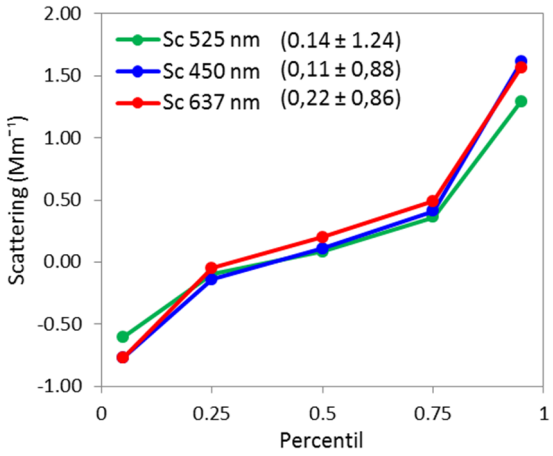

Figure 1. Average scattering coefficients for the calculated percentiles (0.05, 0.25, 0.50, 0.75, 0.95) during zero check calibration measurements at 450, 525 and 635 nm for the year 2013 at MSA station. Average and standard deviation of scattering for the three wavelengths during the whole period are also reported.

Reviewer#1. Specific comment 2). In order to help the reader, please provide average and standard deviation values in parentheses while comparing the optical properties in different events or between the two stations.

Reply to Reviewer#1. Specific comment 2). We agree with the Referee that this information is missing in the manuscript. It should be considered, however, that data within the Ångström matrix are displayed on hourly base whereas air mass origin was identified once per day, therefore average values during a whole day could include some hourly data points which are not representing these situations. Nevertheless, ranges of SAE, AAE and $\%PM_{1\text{-}10}$ for different air mass origin provided in the text are meaningful in order to consider limit values purely representing these atmospheric situations.
In order to consider the Referee's comment, the frequency distribution plots of SAE and AAE for SDE, regional episodes (REG) and Atlantic advections (ATL) will be added in the supporting material (Fig. 2a and 2b for MSY and MSA). These plots will help the reader to interpret the variability of the intensive properties and establish differences among atmospheric scenarios at both stations.

Accordingly to the referee's suggestion, the following sentence was added to section 5.2 of the Manuscript: "Further information providing the frequency distribution and average value of SAE and AAE for each atmospheric scenario is reported in Figs. S1a and S1b for MSY and MSA.

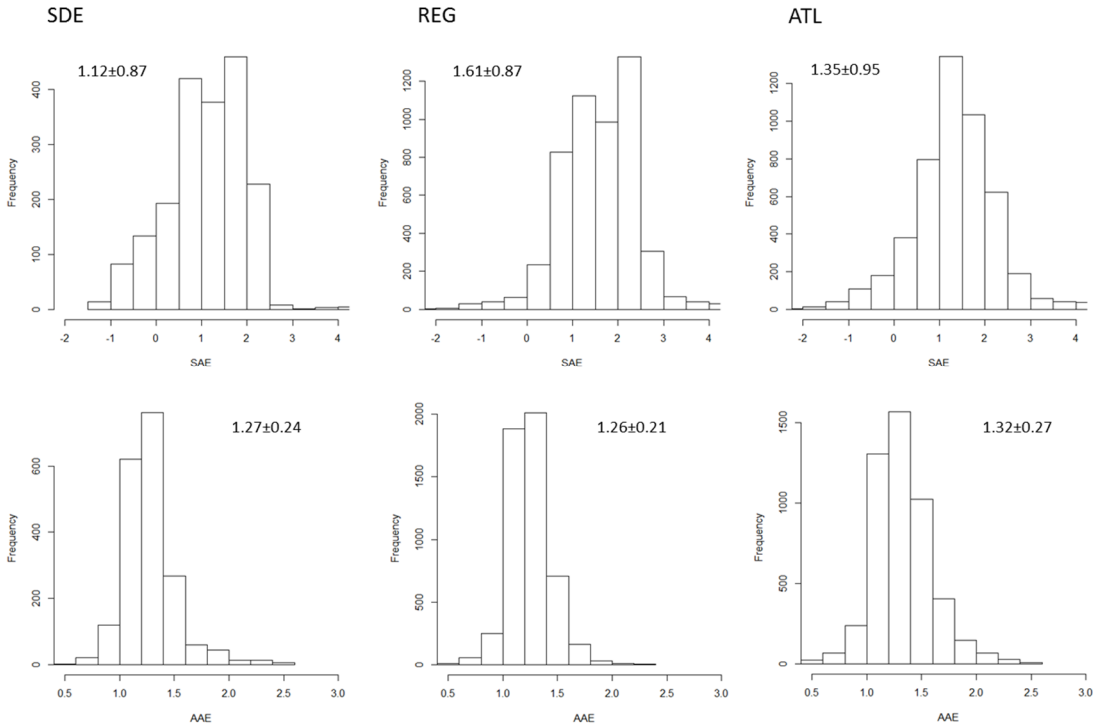

Figure 2a. Frequency distribution of SAE and AAE parameters for the three atmospheric scenarios (SDE REG, ATL) displayed in the Ångström matrix at MSY. Average and standard deviation of these parameters are also reported for the whole period (2012-2014).

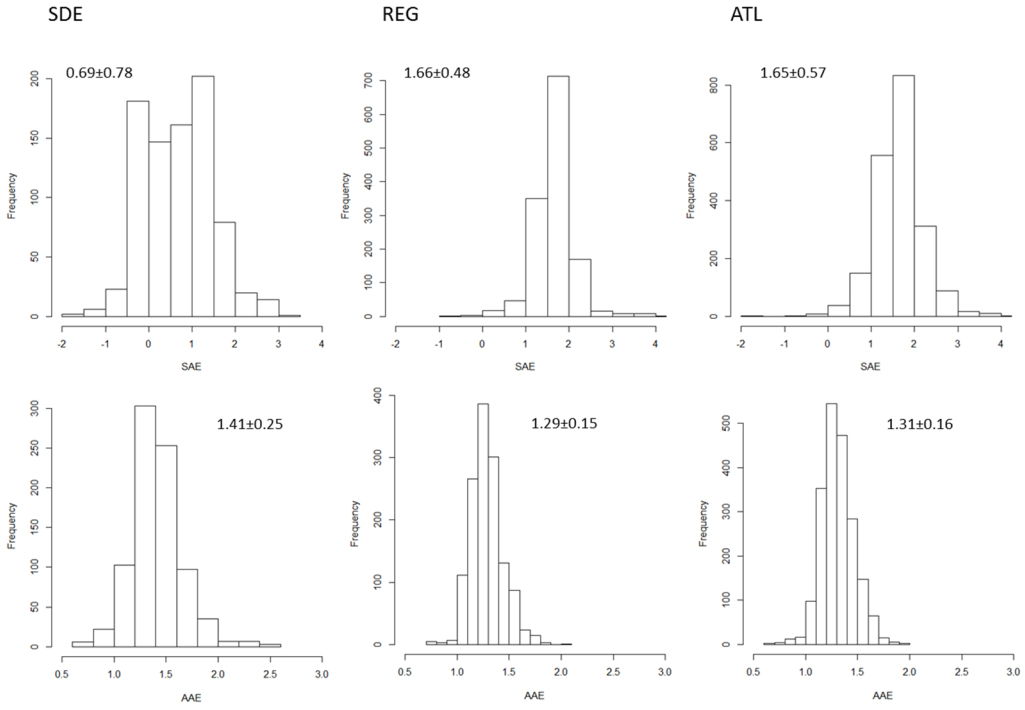

Figure 2b. Frequency distribution of SAE and AAE parameters for the three atmospheric scenarios (SDE, REG, ATL) displayed in the Ångström matrix at MSA. Average and standard deviation of these parameters are also reported for the whole period (2013-2014).

Reviewer#1. Specific comment 3). Line 511: "bellow" correction: below.

Reply to Reviewer#1. Specific comment 3). It has been corrected.

Reviewer#1. Specific comment 4). Lines 592 and 593: please provide the abbreviated station names in the heading.

Reply to Reviewer#1. Specific comment 4). It has been corrected.

Reviewer#1. Specific comment 5). Lines 553- Lines 560: Thee fraction of BBOA and HOA in previous studies may be dependent upon the time of the year those measurements were made. So how fair it is to make those assumptions based on the observations in previous study?

Reply to Reviewer#1. Specific comment 5). Effectively, it is not far enough to consider the POA to SOA ratios from previous studies carried out during not identical periods of the year. Unfortunately, as stated in the manuscript, we did not performed any experiment, simultaneously to the study period, for differentiating primary from secondary organic sources (such as using levoglucosan as biomass burning tracer), and then we had to resort to results obtained in previous studies since it is the unique information we have to establish POA to SOA relations. In order to differentiate primary from secondary OM_{bb}, we considered the proportion of SOA originated from BBOA reported by Cubison et al. (2011). This ratio was primary applied to the results of the source apportionment to ACSM performed at MSY station (Minguillón et al., 2015), which were also used in this study. For OM_{ff}/HOA ratio, we considered results from previous studies performed also at MSY using ^{14}C technique (Minguillón et al., 2009).
Thus, we do not have the necessary information to evaluate how much the use of non-simultaneous ancillary data is affecting our results. However, even though these assumptions concerning POA to SOA ratios are not from measurements carried out in the same study, the considered results for AAE_{ff} and AAE_{bb} (1 and 2 respectively) satisfactory agreed with those coefficients used in previous studies deploying the Aethalometer model.

Following the referee 's suggestion section 5.3.2 in the revised manuscript has been modified as follows:

"Since simultaneous experimental data to the study period were not deployed for differentiating POA to SOA ratios, we have considered results previously reported for MSY measurements. SOA formation from biomass burning emissions can be up to 25% of the BBOA emitted, as shown by Cubison et al. (2011) using Aerosol Mass Spectrometer (HR-ToF-AMS) data. This ratio was primarily applied by Minguillón et al. (2015) to the results obtained from the source apportionment to ACSM performed at MSY station, which were also used in this study."

Reviewer#1. Specific comment 6). Lines 651 and 678: "leaded" correction: lead

Reply to Reviewer#1. Specific comment 6). It has been corrected as "led" instead of "lead".

Reviewer#1. Specific comment 7). Supplementary tables S1 should be numbered S1 (a) and S1 (b) as they are discussed in the text.

Reply to Reviewer#1. Specific comment 7). It has been corrected.