Aerosol characterization in a semi-rural site in South-Italy by combined in-situ and ground-based remote sensing measurements

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High resolution radiometric together with in-situ gravimetric and aethalometer measurements have been performed in a semi-rural site in Southwest Italy (Tito Scalo, 40° 35’ N, 15° 41’ E, 750m a.s.l) to derive aerosols optical and physical properties. The measurement site is located in the Mediterranean area and this condition allows to monitor aerosol properties variation due to the influence of air masses with different origin. In particular, dust particles from Saharan desert could be present together with polluted and smoke aerosol mainly from Central and Eastern Europe. Moreover marine particles are an almost constant component both from the Mediterranean itself and from the Atlantic Ocean. Some anthropogenic local sources impact the site, such as few little plants and a main road. Columnar properties have been inferred by a high resolution (1.5 *nm*) spectroradiometer Ocean Optics (400*nm*-800*nm*) to derive Aerosol Optical Depths (AODs), Ångström turbidity parameters and size distributions, while daily mass size distributions at the ground have been obtained by a 13 stages impactor. Finally, Black Carbon (BC) concentrations have been measured during the campaign by using an aethalometer and, combining all these measurements, it has been possible to obtain a closure between aerosol optical and physical properties.

Introduction

The scientific community is in agreement recognizing aerosols as responsible of a strong impact on climate change, causing variations on both climate and environment.

As stated by IPCC 2007, direct, indirect and semi-direct effects of aerosols are a problem with a medium-low level of scientific understanding due to the unhomogeneity of their sources and to their high spatial and temporal variability. These uncertainties can be reduced by integrating different instruments: improved and intensified in situ observations along with remote sensing techniques, will allow a better estimation of the aerosol contribution to the radiative forcing.

Radiometric remote sensing technique is a powerful tool allowing to measure aerosol parameters and thus to identify aerosol sources. On this subject, Masmoudi et al. (2003) identified aerosol origin region by simply estimating and comparing AODs and Ångström exponent α variations. Moreover, from radiometric measurements number and volume size distributions on the atmospheric column can be obtained (Cachorro et al., 2008; Lyamani et al., 2005; Pavese et al., 2009; Perrone et al., 2005).

However, since in the near-ground layer anthropogenic aerosols are mostly concentrated and different atmospheric layers could be affected by different particles, in-situ measurements are desirable since they could help in assessing uncertainties on column-integrated aerosol optical and physical parameters retrievals. Previous studies on integrated radiometric and in situ measurements have led to significant results, showing in many cases a good correlation between ground-level and columnar aerosol properties as in Mukai et al. (2006), Chaudhry et al. (2007), Cheng et al. (2008), Schaap et al. (2009). The main goal of this work is to analyze optical and physical properties of aerosols by applying different observation techniques such as ground-based radiometric, in-situ gravimetric and filter-absorbing techniques. This led to compare ground-level and columnar size distributions for days of contemporary measurements, while a particular attention has been addressed to the aerosols fine fraction component by measuring Black Carbon (BC) concentration in $PM_{2.5}$ fraction. In fact, BC plays a central role in climate forcing, due to its solar radiation absorbing properties.

1. Measurements site main characteristics

The measurement site, located in South-Italy (Tito Scalo, 40.60° N, 15.72° E, 750ma.s.l.), is a very small industrial zone surrounded by a large rural area. Few local sources producing anthropogenic aerosol impact this site: some little plants and a main road far about 1 km. During the sampling period (2008-2010), mainly summer, frequent local smoke events from neighbouring woods have been registered, while aerosol loading both anthropogenic from North-Eastern Europe and mineral from Sahara are frequent. Marine particles are a quite constant component on this site, due to its closeness to Tyrrhenian, Adriatic and Ionian seas (respectively 70 km, 90 km, and 95 km).

2. Instrumentation, procedures and measurements analysis

2.1 The instruments

Measuring solar direct radiation with a high resolution radiometer (spectral range 400–800 nm and res. 1.5 nm), allows to estimate important parametrs describing columnar aerosol optical and physical properties, such as Aerosol Optical Depth, Ångström turbidity parameters α and β and number and volume size distributions. A detailed description of the procedures followed to obtain them is in Esposito et al. (2004). The radiometer, manually operated, measures solar spectra only in cloudless conditions, from the morning until the afternoon, each 15 minutes.

Aerosol mass size distributions at ground level have been measured by a Dekati Low Pressure Impactor, hereafter DLPI, with the inlet placed at about 2 m above the ground. The DLPI has a size range from 0.03 μm to 10.0 μm and a flow rate of 30 l/min with 50% stage cut off Equivalent Aerodynamic Diameters (EAD) at 0.029, 0.056, 0.095, 0.158, 0.264, 0.383, 0.610, 0.939, 1.577, 2.354, 3.928, 6.471, 9.769 μm. Aerosols have been collected on Nucleopore polycarbonate filters with 25 mm diameter for gravimetric analysis. The sampling time was 24 h. Mean daily values of air temperature, pressure and relative humidity measured at the impactor inlet were used to obtain the normalised sampled volume. Gravimetric measurements procedure has the following steps: filters conditioning in a room (50±5RH % and 20±5 °C) for 24 hours, before and after exposure. Filters weighing in the same conditioned room, before and after exposure, with a microbalance (Mettler Toledo MX5 Type – weighing accuracy of ±1 µg) for gravimetric analysis. Whenever the first two measurements on a filter were not within 5 µg, a third weighing was performed and the mean of the two closest weighing was considered as a datum useful for the analysis. Field blanks were even measured to calculate the limit of detection (LOD) for the whole procedure. Black Carbon measurements have been carried out with a seven wavelength Aethalometer (Magee Scientific, model AE31, λ 370, 470, 520, 590, 660, 880, 950 nm), whose size selective inlet (aerodynamic particle diameter <2.5 µm) was placed at about 2 m above the ground, such as for the DLPI. Aerosols have been collected on a quartz fiber tape with 5 min time resolution and with a flow rate of 3.9 lpm. The attenuation of lamps light through the filter allows to estimate BC mass concentration, considering a constant value of the mass absorption cross section equal to, 16.6 m^2 g^{-1} at 880 nm, as recommended by the manufacturer. These first BC measurements do not take in account the corrections either for both shadowing effect and scattering offset, as described in Collaud Coen et al., (2009). In fact, the results here obtained are considered useful for a first qualitative BC data analysis. As a next step, a correction procedure will be carried out for a more quantitative BC content estimation.

2.2 Aerosol Size Distributions by radiometer and DLPI

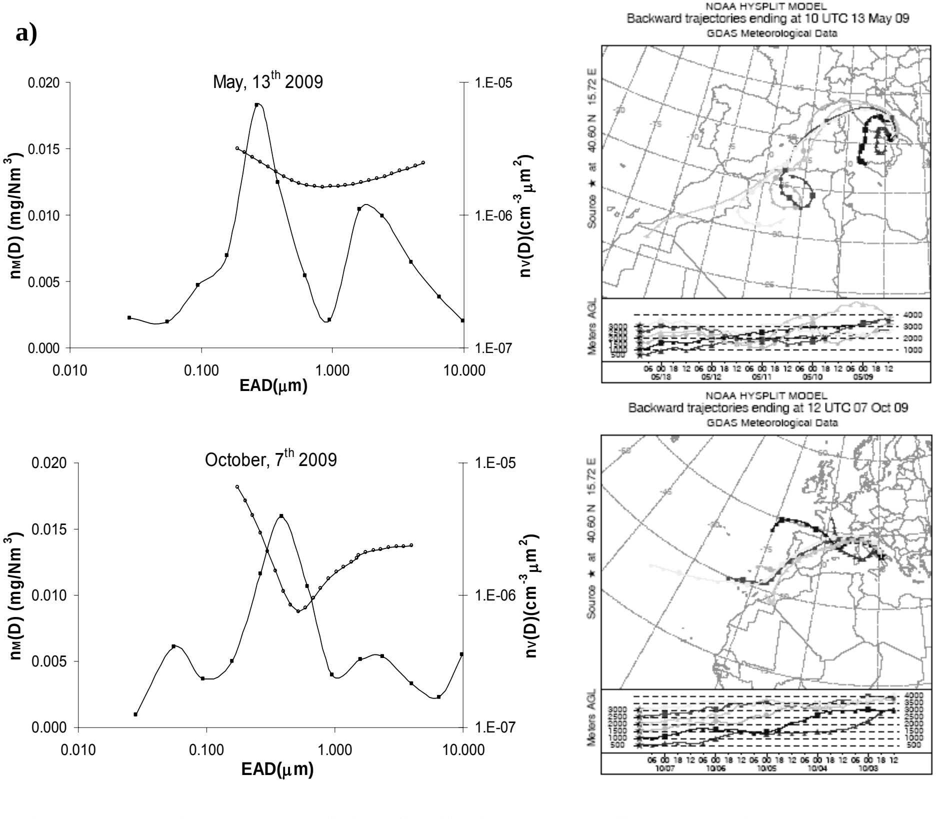

Fig.1: Comparison of aerosol size distributions from radiometer and DLPI for **a)** May, 13th 2009 and **b)** October, 7th 2009. The corresponding air masses back-trajectories are reported.

In figure 1 the size distributions, as obtained by inverting radiometric data and by gravimetric measurements for May, 13th 2009 and October, 7th 2009 are shown, along with corresponding air masses back-trajectories. It must be pointed that radiometric distributions have been averaged over the measurement day. In case a) the inverted volume size distribution shows two modes with similar amplitude, while in case b) the fine particle mode is dominating over the coarse one. The behaviour of case a) agrees quite well with DLPI size distribution which exhibits, for May, 13th 2009, a dominating fine mode, but with a well defined and wide coarse mode. In fact, the HYSPLIT backward trajectories (http://www.arl.noaa.gov/ready/hysplit4.html) shows a clear contamination of aerosols in the measurements site with dust mineral particles from Sahara desert. In case b) a three modes distribution has been obtained, where the large particles mode is still present, but less evident, if compared with the fine one. An ultra-fine mode has even been found: once more, its presence is explained by particles backward patterns, which have been moving across North-Central Italy and anthropogenic particles could join the measurements site.

2.3 Correlating radiometric, DLPI and BC measurements

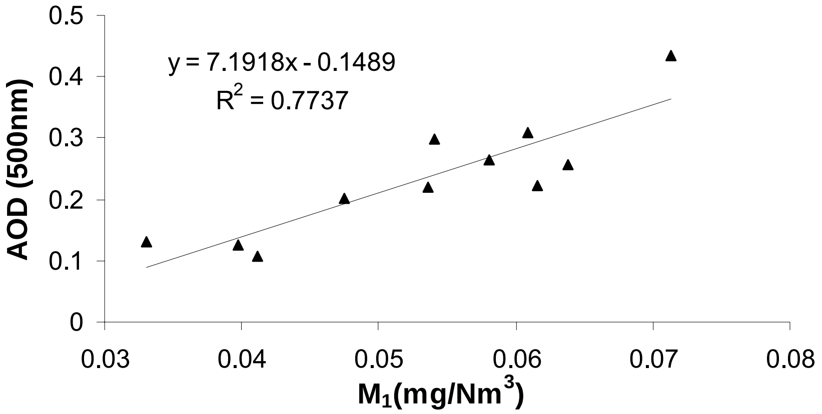

Fig.2: Correlation between *AODs* @500 nm from radiometer and fine mode concentration M_1 from DLPI.

As previously found in Calvello et al., (2010) these measurement techniques, though different, can, in some sense, be coherent and give more information on aerosol properties. Considering fine mode mass concentration M_1 (from DLPI) as the concentration of particles with EAD ≤ 1 µm, a correlation with radiometric data, namely AODs, has been searched. In figure 2, AOD @500 nm daily averaged values are reported, along with M_1 data. As can be easily verified, the scatter plot shows a good correlation (R^2 = 0.77) which tends to diminish whenever the AOD wavelength increases. This suggests that columnar optical properties in the visible range are dominated by ground aerosol fine fraction, and the fine fraction along the whole atmospheric column is characterised by similar optical properties.

Next plot shows the correlation between the fine fraction mass concentration M_1 and the BC content, as derived from particles absorption @ 880 nm. In this case the correlation coefficient is even better than the previous case (R^2= 0.85).

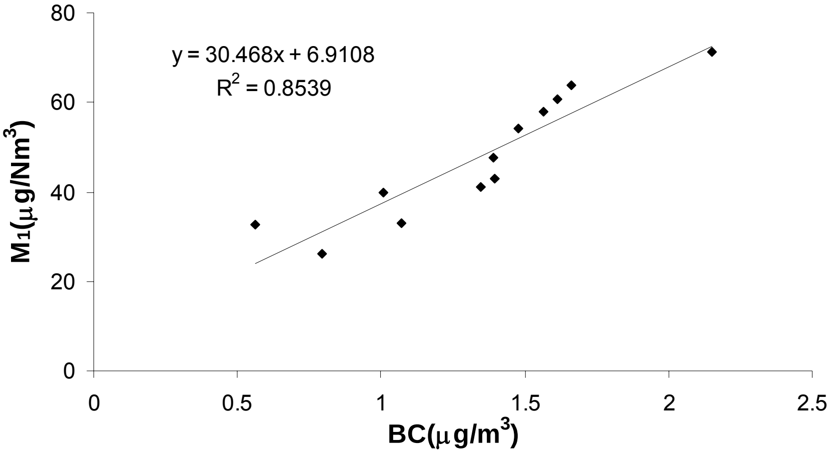

Fig.3: Correlation between the fine mode concentration M1 from DLPI and BC content

This improved correlation can be due to the fact that the air samples are more or less the same for both instruments because DLPI and aethalometer inlets are located 2 m above the ground. Moreover, since DLPI measurement lasts 24 h, the BC content has been averaged exactly over the same period of DLPI, thus allowing a total temporal coincidence of both measurements. This result suggests that anthropogenic aerosols cannot be disregarded even in a rural site and are the main component in the fine mode particles.

Conclusions

The comparison of data coming from three different instruments (radiometer, impactor and aethalometer) has been carried out. In spite of this different measurement techniques, the attempt gives encouraging results to pursuing this study. In fact, variations in the size distributions, both from radiometer and DLPI, are related to the different air masses crossing the site, while good correlations between fine fraction mass concentration with AOD and BC content, suggest, in turn, the predominance of ground small particles optical properties all along the atmosperic column and the predominance of anthropogenic particles in the ground fine fraction, although the rural site.

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