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An integrated approach to identify the origin of PM_{10} exceedances

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Abstract
Purpose This study was aimed to the development of an integrated approach for the characterization of particulate matter (PM) pollution events in the South of Italy.
Methods PM_{10} and $PM_{2.5}$ daily samples were collected from June to November 2008 at an urban background site located in Bari (Puglia Region, South of Italy). Meteorological data, particle size distributions and atmospheric dispersion conditions were also monitored in order to provide information concerning the different features of PM sources.
Results The collected data allowed suggesting four indicators to characterize different PM_{10} exceedances. $PM_{2.5}/PM_{10}$ ratio, natural radioactivity, aerosol maps and back-trajectory analysis and particle distributions were considered in order to evaluate the contribution of local anthropogenic sources and to determine the different origins of intrusive air mass coming from long-range transport, such as African dust outbreaks and aerosol particles from Central and Eastern Europe. The obtained results were confirmed by applying principal component analysis to the number particle concentration dataset and by the chemical characterization of the samples (PM_{10} and $PM_{2.5}$).
Conclusions The integrated approach for PM study suggested in this paper can be useful to support the air quality managers for the development of cost-effective control strategies and the application of more suitable risk management approaches.

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1 Introduction

The effects of particulate matter (PM) on human health have been deeply investigated in several papers (Englert 2004; Forbes et al. 2009; Pope et al. 2009; Zanobetti and Schwartz 2005); the exposure to high PM_{10} concentrations has been associated with increased risk of death for cardiovascular or respiratory causes. These effects may be largely caused by finer particles $PM_{2.5}$ that, as a consequence of their greater surface area, could be effective media to transport deeply into the lungs different kinds of pollutants (Nadadur et al. 2007; Reich et al. 2009; Sager and Castranova 2009).

In order to ensure air quality, the European regulation (Directive 2008/50/EC) established 50 μg m^{-3} which may only be exceeded 35 days/year. The yearly $PM_{2.5}$ value has been limited to 25 μg m^{-3}, and it will be reviewed by the Commission in 2013 by taking into account further information concerning the health and environmental effects of fine particles. The introduction of a new limit value for $PM_{2.5}$ would probably cause a stricter limit for daily average PM_{10} concentrations. Moreover, the directive provides member states to subtract the contribution of natural sources before comparing PM_{10} concentrations to the limit values according to the guidelines provided by the European Commission (671/11, 2011). The possibility to discount these exceedances is relevant especially for the Southern European regions that are significantly affected by Saharan dust outbreaks (Escudero et al. 2007; Meloni et al. 2008; Perrino et al. 2009; Querol et al. 2004). This area is

also characterized by pollution and smoke aerosols coming from Central and Eastern Europe that determine the increase in finer particle concentrations (Calvello et al. 2010; Glavas et al. 2008; Amodio et al. 2010b). In particular, as concerns the Puglia regional sites, PM ionic secondary specie determination has suggested the relevance of regional contribution to PM concentrations, confirmed by the East European and regional origin of back trajectories air mass analysis. Moreover, Amodio et al. (2011) pointed out the 'regional' character of sulphate concentration in PM_{10} samples as a consequence of the synergistic effect of regional and long transport contributions at Puglia Region sites.

In order to characterize the mix of aerosols coming from different sources, the project entitled 'Integrated System for the Monitoring of the Atmospheric Particulate (SIMPA)' was the first comprehensive study founded by the Puglia Regional Government (South of Italy) that aimed for the development of an 'integrated' approach in the evaluation of PM concentration levels. The project was focused on the identification of natural and anthropogenic contributions to PM_{10} and $PM_{2.5}$ samples due to local emission sources or long-range transport. The monitoring campaign included the PM mass determination, the evaluation of the dispersion properties of the lower atmospheric layers, the characterization of the optical particle properties and the chemical characterization of PM-collected samples. These data were related to information provided by aerosol models (DREAM and NAAPs) and 5-day backward trajectory analysis in order to evaluate the long-range contribution of air masses. Finally, statistical investigations allowed to identify the different optical patterns of PM events which occurred during the monitoring campaign.

The 'SIMPA' campaign was performed from June to November 2008 by collecting PM_{10} and $PM_{2.5}$ samples at an urban background site located in Bari town (Puglia Region, south of Italy). A total amount of 348 samples were collected. Meteorological data, particle number concentrations, and atmospheric dispersion conditions were also monitored at the sampling site. The data obtained by automated instruments were used to select PM_{10} and $PM_{2.5}$ samples for the chemical characterization of inorganic components and carbonaceous fraction.

The data discussed in this paper were collected from the 9th of October to the 6th of November 2008; this period was considered very interesting for PM investigations as it was characterized by local emission sources and long-range anthropogenic and natural contributions. The information provided by the 'integrated' approach was proven to be also useful for supporting the air quality managers in the development of cost-effective control strategies and the applications of more suitable risk management approaches.

2 Material and methods

2.1 PM sampling

Sampling was carried out at an urban background site close to the Chemistry Department of the University of Bari (Puglia Region). $PM_{2.5}$ and PM_{10} daily samples were collected by a dichotomous low volume sampler SWAM Dual Sampler (FAI Instruments, Rome, Italy). The samples were collected on quartz fiber filters (Whatmann, 47-mm diameter) by FAI EN 1,234.1 sampling heads operating at a flow rate of 2.3 $m^3\ h^{-1}$. The sampling was performed from the 1st of June to the 30th November 2008, and a total amount of 348 PM samples were collected. The number concentration of aerosol particles with optical diameter greater than 0.3 μm (20 size ranges) was determined by a laser scattering counter (OPC Monitor, FAI Instruments) with 5-min time resolution. For data analysis, OPC channels were clustered in order to obtain data for the most significant optical ranges (0.30–0.49 μm; 0.49–0.94 μm; 0.94–1.54 μm; 1.54–1.98 μm; >1.98 μm). The chemical characterization of PM_{10} and $PM_{2.5}$ samples (9–16 October 2008; 26 October–2 November 2008) was performed in order to determine inorganic components as ions and carbonaceous compounds.

Meteorological data (wind speed, air temperature, barometric pressure) were continuously recorded by an automated weather station. The mixing ratio of atmospheric compounds in the lower layers of planet boundary layers (PBL) was determined by PBL Mixing Monitor (FAI Instruments), a sequential automatic system able to estimate natural radioactivity levels by measuring beta activity of radon decay products with 1-h time resolution. Natural radioactivity maintains constant low values in case of convective mixing or advection and it increases when the atmospheric stability allows the radon accumulation in the low PBL layers. PBL mixing monitor data were considered in order to determine the pollutant accumulation events and to understand the temporal evolution of air pollution processes.

2.2 Inorganic analysis

The ionic PM fractions (PM_{10} and $PM_{2.5}$) were extracted by ultrasonic agitation of a quarter of sample with two aliquots of 5 mL of deionized water for 20-min periods. The obtained solutions were analyzed for chloride (Cl^-), nitrate (NO_3^-), sulphate (SO_4^{2-}), sodium (Na^+), ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}), and calcium (Ca^{2+}) ions. Analyses were performed using a Dionex DX120 (Dionex, Sunnyvale, CA, USA) ion chromatography system equipped with an electrical conductivity detector for anions and a Dionex DX600 ion chromatography system for cations. Anions analyses were carried out using a Dionex IonPac AS4A-SC column and an isocratic 1.8 mM sodium carbonate/1.7 mM sodium

bicarbonate eluent. Cation analyses were performed by a Dionex IonPac CS12A column and an isocratic 20 mM methanesulphonic acid eluent. Standard solutions were prepared by suitable dilution of their stock solutions (1,000 mg L^{-1} from Fluka, Milwaukee, WI, USA) in Milli-Q water (Millipore Corporation, Billerica, MA, USA).

2.3 Organic carbon/elemental carbon analysis

Rectangular punches (normally 1.50 cm^2) of filter deposit of PM_{10} and $PM_{2.5}$ samples were analyzed for the detection of the organic (OC) and elemental carbon by a thermal optical method (Sunset Laboratory Inc., Tigard, OR, USA). To remove the possible carbon contamination, quartz fiber filters were precleaned in a muffle furnace according to the NIOSH method 5040; in this method, the speciation of organic, carbonate, and elemental carbon is performed by temperature and atmosphere control. He/Ne laser light passed through the filter allows continuous monitoring of filter transmittance, and an optical feature corrects for pyrolytically generated OC. A flame ionization detector is used for quantification of evolved carbon and the instrument calibration is carried out by injection of a known volume of methane into the sample oven (Birch and Cary 1996).

2.4 Principal component analysis

Principal component analysis (PCA) has been widely used to characterize emission source categories observed at a receptor site (Amodio et al. 2010a; Andriani et al. 2010, 2011; Hellebust et al. 2010; Henry et al. 1984; Pant and Harrison 2011). The purpose of PCA is to identify patterns in data and to express the data in such a way as to highlight their similarities and differences. In this way, it is possible to compress the dataset in order to reduce the number of variables which explain their total variance. New orthogonal and uncorrelated variables, called principal components (PCs), are created as linear combinations of the original variables; the first PC explains the largest amount of variance of the original data. The first step of the procedure consists in calculating eigenvalues and eigenvectors of the correlation data matrix; the eigenvectors are rotated in order to obtain a clear pattern of loadings, that is, factors that are somehow clearly marked by high loadings for some variables and low loadings for others.

In this work, PCA with Varimax rotation was applied to the number particle concentrations determined by OPC during the PM_{10} exceedances occurred from the 9th of October to the 6th of November 2008. The main purpose of the procedure was to evaluate the effectiveness of PCs in capturing the cluster structure of the data and to highlight the different optical properties of aerosol particles during the considered events.

2.5 Aerosol model and air mass trajectories

Saharan dust outbreaks were identified by Dust Regional Atmospheric Model (DREAM) aerosol maps provided by Barcelona Supercomputing Center (http://www.bsc.es/projects/earthscience/DREAM/). Sulphate and smoke concentration distributions were evaluated by Navy Aerosol Analysis and Prediction System (NAAPS) aerosol maps (http://www.nrlmry.navy.mil/aerosol/). One hundred twenty-hour back trajectories were calculated with NOAA's Hysplit model. Trajectories were calculated at three different heights (Level 1, 750 m AGL; Level 2, 1,500 m AGL; Level 3, 2,500 m AGL) with arrival time 12 UTC local noon (http://ready.arl.noaa.gov/HYSPLIT.php).

3 Results and discussion

The aim of this work was to determine the main sources that caused PM_{10} exceedances at the sampling site; in order to achieve this goal, different sorts of data have been evaluated during the investigated period.

The first considered indicator was the percentage of $PM_{2.5}/PM_{10}$ ratio; it ranged from 40% to 90% depending on meteorology and emission sources. The higher values of this indicator are observed for primary directly emitted particles or secondary formed via atmospheric reaction. High concentrations of coarse particles (low $PM_{2.5}/PM_{10}$ ratio) result from natural long-range transport (Saharan dust events) that significantly influences PM_{10} concentrations than $PM_{2.5}$ fraction levels (Perez et al. 2008) and from resuspension of particulate matter emitted by motor vehicles and agricultural activities. The second indicator was the natural radioactivity; it allows to evaluate the PBL dilution potential and its effect on the temporal evolution of PM concentrations (Perrino et al. 2008). Low values of natural radioactivity are associated to advection conditions that should cause a Saharan dust event at the receptor site when strong winds coming from the south occur. The third indicator was the interpretation of daily meteorological situation with the 5-day back trajectories at three different heights provided by NOAA's Hysplit model and the evaluation of daily aerosol maps obtained by the DREAM and NAAPS models. This information allows to identify the occurrence and the duration of air mass long-range transport, such as Saharan dust events. The last indicator considered in this work is the number of particles in the ranges analyzed by the optical particle counter (>0.30 μm) that was linked to the different sources of aerosol particles. The source-like pattern was also confirmed by applying principal component analysis to the dataset.

These indicators allowed to suggest the different origins of PM exceedances which occurred during the sampling

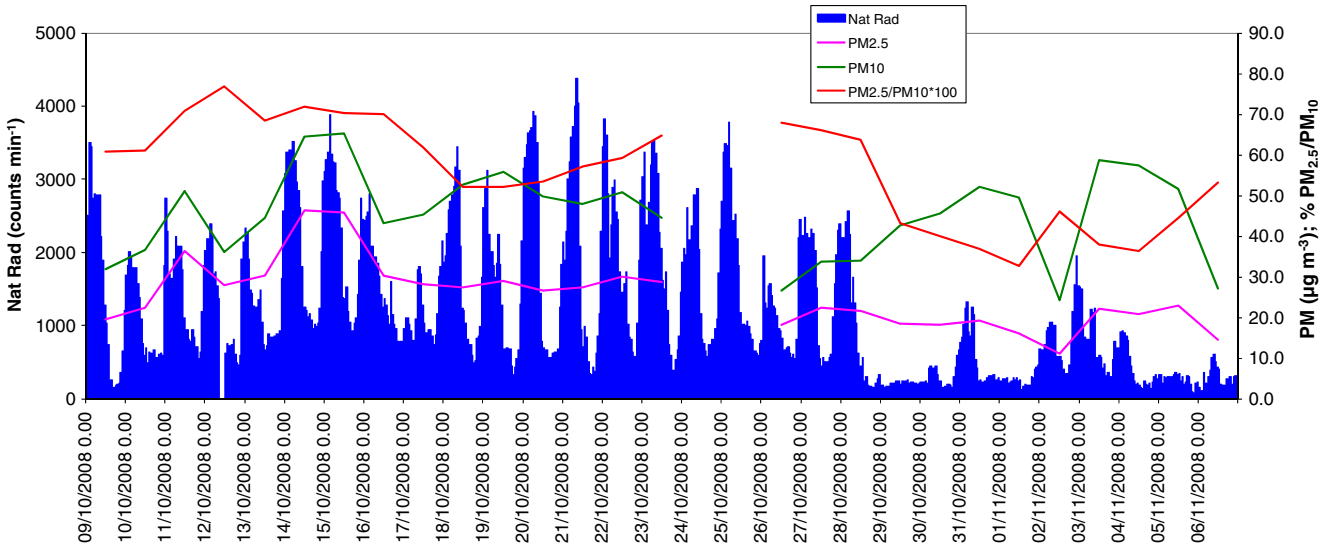


Fig. 1 Daily PM_{10} and $PM_{2.5}$ concentrations and time pattern of natural radioactivity (9 October–6 November 2008)

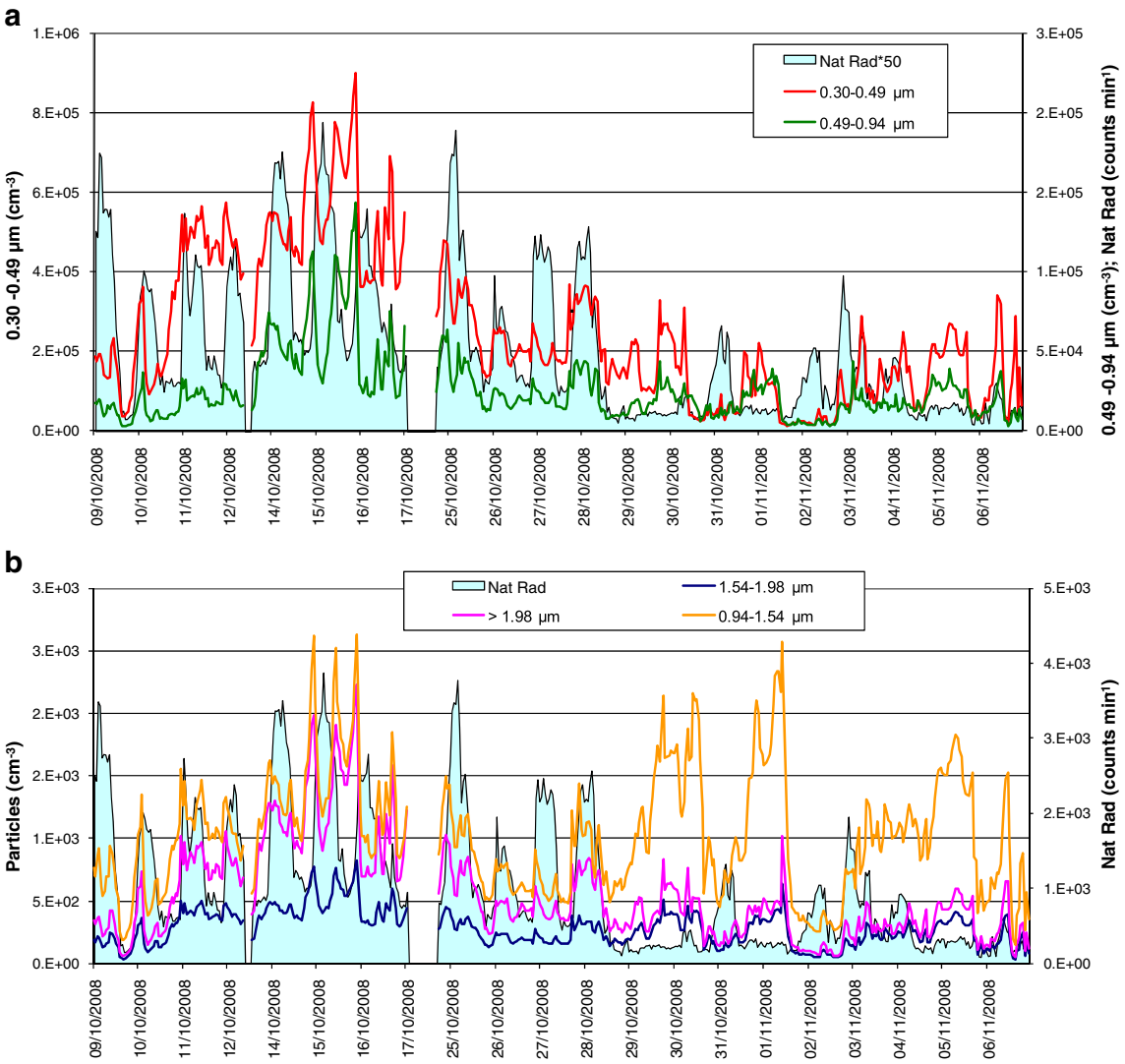


Fig. 2 Temporal pattern of natural radioactivity and particle number concentrations in the size range 0.30–0.94 µm (**a**) and 0.94–10 µm (**b**)

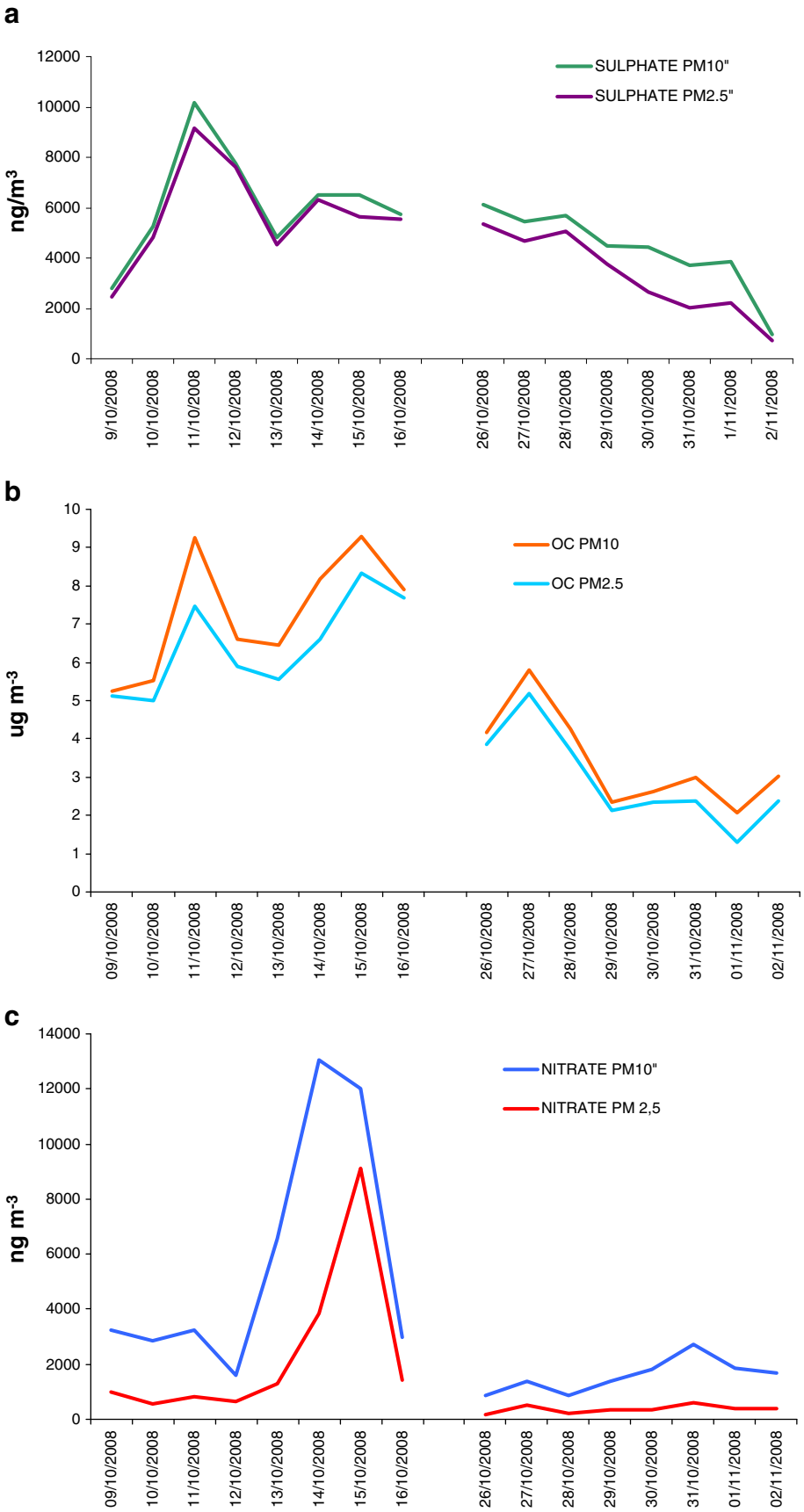


Fig. 3 Daily concentration of sulphate (SO_4^{2-}) (**a**), organic carbon (OC) (**b**) and nitrate (NO_3^-) (**c**) in $PM_{2.5}$ and PM_{10} samples

campaign. In order to confirm them, the chemical characterization of the samples collected from the 9th to the 16th of October 2008 and from the 26th October to the 2nd of November 2008 was evaluated. This approach to PM study was performed in order to assess the use of different automatic monitoring instruments and information provided by aerosol models for PM_{10} exceedance origin. The chemical speciation of PM samples was carried out in order to support the information provided by the considered indicators, and it should be performed when the indicators are not sufficient to clearly identify the different PM features.

3.1 PM event characterization

The data collected from the 9th of October to the 6th of November 2008 are discussed. As shown in Fig. 1, PM_{10} concentrations ranged from 24.3 to 65.2 μg m^{-3} with an average level of 45.3 μg m^{-3} and ten exceedances of the limit value for daily average PM_{10} concentrations. The $PM_{2.5}$ concentrations ranged from 11.2 to 45.2 μg m^{-3} with an average level of 25.2 μg m^{-3}.

The data collected in the first period (9–16 October) showed the good correlation between natural radioactivity and PM levels; the mean ratio of $PM_{2.5}/PM_{10}$ was equal to 0.6 (Fig. 1) and PM mass concentrations reached the maxima values on the 11th and the 14th to 15th of October, when the $PM_{2.5}$ was about 75% of PM_{10}. Typical daytime evolution of the atmospheric boundary layer in Bari was determined from the 9th of October to the 28th of October; convective air motions that caused intense turbulent mixing in the morning and low atmospheric dispersion conditions in the night were observed. This periodic trend depending on day–night difference in solar irradiation was not affected by advective motion of air masses and, in particular, the highest levels of radioactivity of the sampling period were found in these days. The meteorological data (not shown) allowed also to evaluate high pressure and calm wind conditions. These atmospheric properties favored air pollutants accumulation (Manigrasso et al. 2010). Aerosol maps highlighted high sulphate ground concentration, and back trajectory analysis allowed to determine air mass transport from North–East Europe, in particular on the 11th of October. Particle number concentrations also showed maxima on the 11th and the 14th to 15th of October but at different size ranges (Fig. 2a, b). While on the 11th of October the highest particle number of the annual mean value was obtained for a 0.30–0.49-μm range, on the 14th to 15th of October, the increase of particle concentrations was observed for all the considered size ranges. These results suggested local anthropogenic origin for PM exceedances determined in the first period; an additional contribution due to a synergistic effect of regional and long transport was also considered, especially on the 11th of October for the increasing of the finest particle concentrations. Moreover, this day was characterized by natural radioactivity levels lower than those determined in the other days of the first period that should have favored long-range apportion.

The chemical characterization of $PM_{2.5}$ and PM_{10} fractions confirmed these findings. High concentration of secondary particulate components, such as sulphate, organic carbon, and nitrate was found in this period. Although negative artifacts due to the nitrate volatilization were observed (Huang et al. 2004), ammonium and nitrate ionic concentrations were still considered for PM sources characterization. In particular, high sulphate content of the samples was determined on the 11th of October and high concentrations of organic carbon and nitrate were observed on the 14th to 15th October (Fig. 3a–c). These results confirmed the local anthropogenic contribution to PM. High coarse nitrate concentration was observed in these 2 days; its origin may be due to PM resuspension and pollutant accumulation occurred in lower dispersive atmospheric conditions (Pakkanen 1996; Zhuang et al. 1999). Finally, the high sulphate content

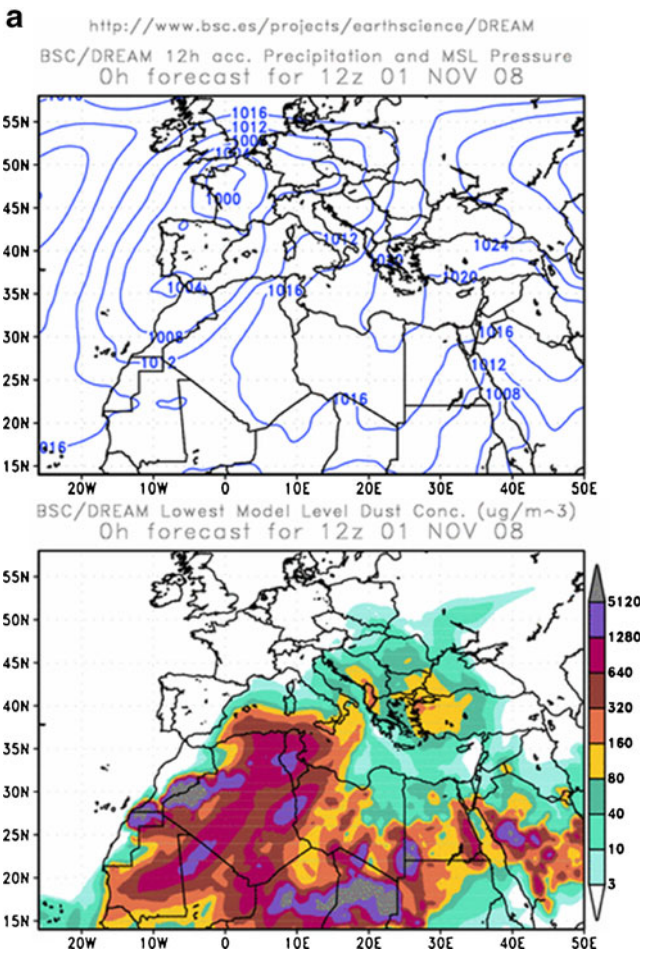
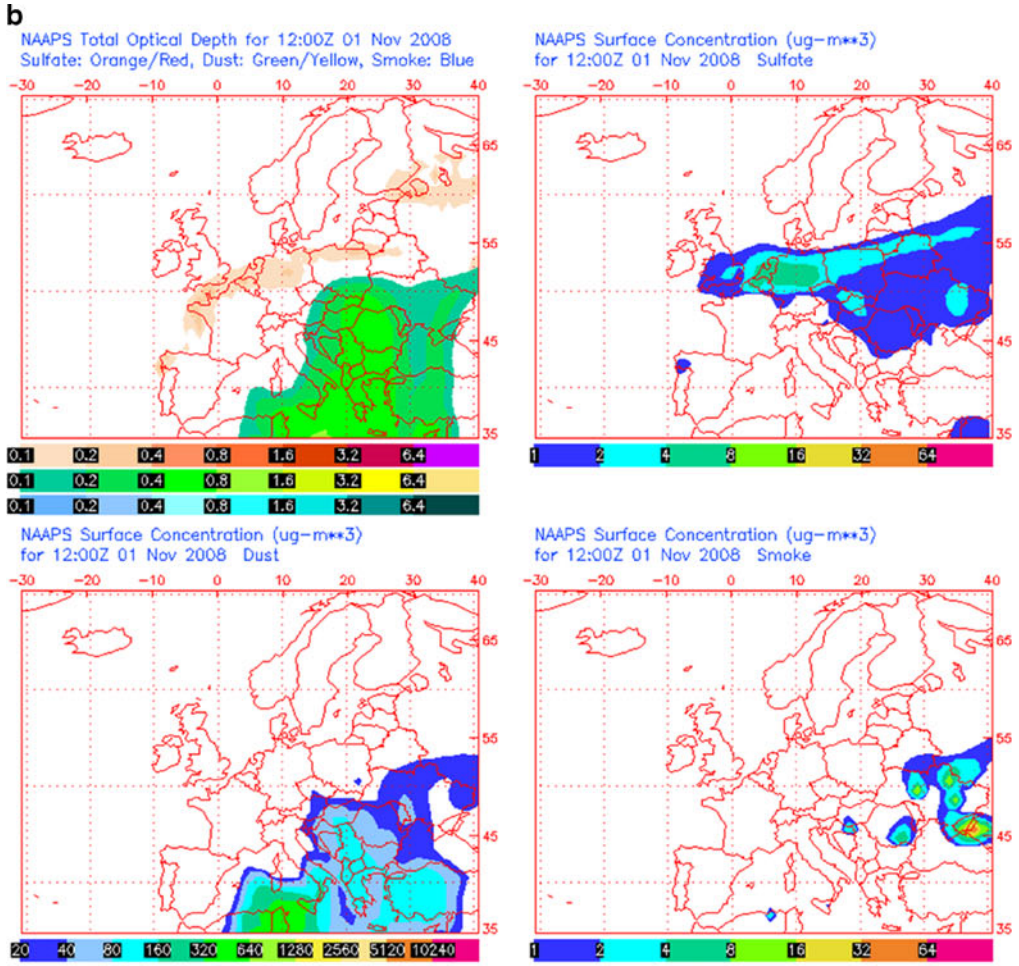


Fig. 4 Daily aerosol map by DREAM (**a**) and NAAPS (**b**) models on November 1st 2008 (**a**); backward trajectories at three different heights (750, 1,500, 2,500 m) by NOAA's Hysplit model on November 1st 2008 (**c**)



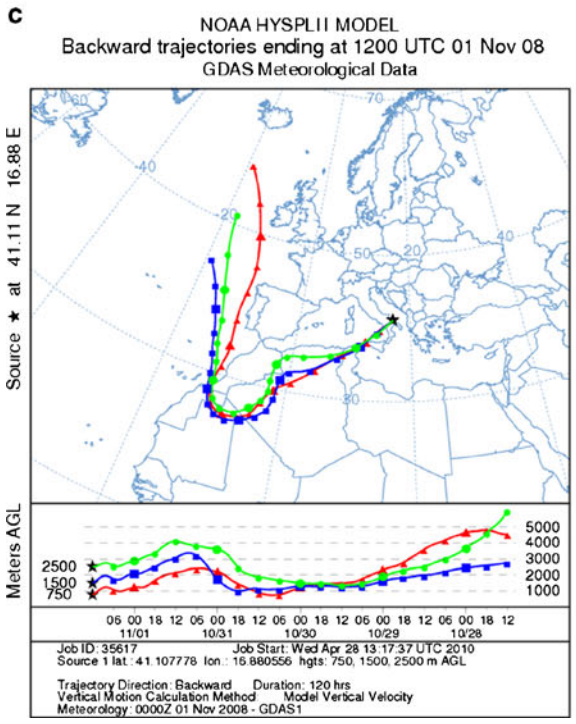


Fig. 4 (continued)

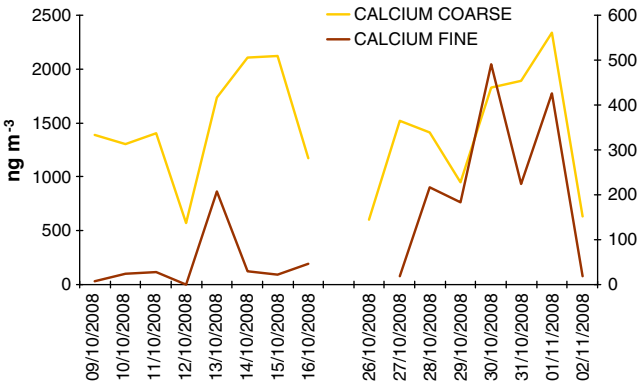


Fig. 5 Daily calcium concentration (Ca^{2+}) in fine ($PM_{2.5}$) and coarse (PM_{10}–$PM_{2.5}$) fraction

determined on the 11th of October pointed out the anthropogenic contribution to particulate matter due to regional or long-range transport (Amodio et al. 2011).

The atmospheric scenario changed from the 29th of October to the 6th of November. This period was characterized by the mean ratio of $PM_{2.5}/PM_{10}$ equal to 0.4 (Fig. 1). PM_{10} concentrations also reached high values but the same behaviour was not observed for to $PM_{2.5}$ concentrations. In fact, the mean PM_{10} concentration was 48 μg m^{-3} in the first period and 42 μg m^{-3} in the second one; the mean $PM_{2.5}$ concentration was 30 and 18 μg m^{-3}, respectively. The lowest level of natural radioactivity and more dispersive atmospheric conditions were observed. Southern winds with higher speed than the previous period were detected; the increase in wind speed was followed by higher temperature and lower pressure. These conditions favoured air pollutant concentration abatement and long-range transport processes. Information provided by aerosol models (DREAM and NAAPs), and 5-day backward trajectories confirmed the outbreak from the Saharan desert in the southern regions of Italy (Fig. 4a–c). In these days, the OPC data pointed out a relevant increase of particles which optical diameter ranging between 0.94 and 1.54 μm (Fig. 2b). This behaviour has been found typical of natural dust events (de Gennaro et al. 2011).

The chemical characterization of $PM_{2.5}$ and PM_{10} collected samples was also evaluated. In particular, from the 29th of October to the 1st of November 2008, the ionic coarse fraction exceeded the 50% of PM_{10} ionic fraction (its mean contribution is equal to 30%) and a good correlation between PM_{10} calcium and carbonate was observed (Querol et al. 2001). The sulphate content in coarse fraction tends to increase with calcium concentrations and suggested that the uptake of precursor gases on the aerosol dust particles is favoured by their large surface area (Hayami 2004). The decrease in ammonium fine concentrations was explained according to the role of dust aerosol in neutralizing the acid species such as nitric and sulphuric acids (Harrison et al. 1994; Querol et al. 2001). The Saharan dust outbreak was also pointed out by the similar trend observed for calcium in fine and coarse PM fractions and by the increasing of Ca^{2+} concentration in $PM_{2.5}$ samples (Fig. 5) (Amodio et al. 2008).

Table 1 Loadings and eigenvalues in PCA on particle number concentrations during PM_{10} exceedances (9 October–6 November 2008)

	PC1	PC2
0.30–0.49 μm	0.93	0.33
0.49–0.94 μm	0.73	0.59
0.94–1.54 μm	0.37	0.92
1.54–1.98 μm	0.78	0.61
>2 μm	0.90	0.44
Eigenvalues	4.44	0.38

3.2 PCA analysis

Principal component analysis with Varimax rotation was applied to the dataset containing the hour mean of particle number concentrations in the most significant size ranges collected (0.30–0.49, 0.49–0.94, 0.94–1.54, 1.54–1.98, >1.98 μm) during PM exceedances which occurred from the 9th of October to the 6th of November 2008. Since the variables varied by several orders of magnitude, PCA was applied to the normalized data matrix. The main purpose of the procedure was to evaluate the effectiveness of PCs in capturing the cluster structure of the data.

Loadings and eigenvalues obtained for each of the components are summarized in Table 1. Two PCs were calculated explaining up to 90% of the total variance. Factor loadings of the variables were used to distinguish the exceedances caused by natural and anthropogenic contributions to PM collected samples (Table 1). PC1 was characterized by high loadings for accumulation and coarse mode of aerosol particles, suggesting the role of stagnant

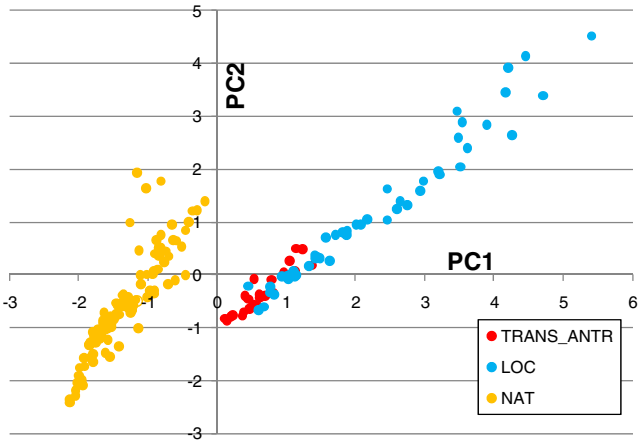


Fig. 6 Score plot of two components obtained by applying PCA on numerical particle concentrations during PM_{10} exceedances (9 October–6 November 2008)

atmospheric conditions on total particle concentration emitted by anthropogenic sources (Mazzei et al. 2007; Ruuskanen et al. 2001). PC2 showed a high loading for coarse mode particles, especially for 0.94–1.54 μm size range, and highlighted the natural contribution to PM samples. These features can be also evaluated by taking into account the score plot in Fig. 6. Two different groups of scores were obtained: high score values on the PC1 were observed for samples collected when local anthropogenic contributions to PM samples occurred in the first period ('LOC', blue dots); moreover, it was possible to identify those characterized by high sulphate content due to regional recirculation or long-range apportion ('TRANS_ANTR', red dots). High score values on the PC2 was evaluated for samples collected during Saharan dust outbreaks in the second period ('NAT', yellow dots).

The results allowed to confirm that, when African dust outbreaks occurred, it was possible to determine an increase in the number concentration of aerosol particles in the range 0.94–1.54 μm. This information should also be considered for the identification of dust outbreaks even if they do not determine PM_{10} exceedances.

4 Conclusions

The results discussed in this paper were obtained in the framework of the project entitled 'Integrated System for the Monitoring of the Atmospheric Particulate (SIMPA)' that aimed to the development of an 'integrated' approach in the evaluation of PM levels.

Differences in particulate matter concentrations were found to be significantly affected by meteorological and atmospheric dispersion conditions. In particular, high PM concentrations were observed during atmospheric stability and long-range transport event. Four indicators were proposed to characterize the different PM exceedances. $PM_{2.5}$/PM_{10} ratio, natural radioactivity, aerosol maps and back trajectory analysis and optical particle distributions allowed to identify the main features of PM contribution: local emission sources, anthropogenic and natural long-range transport. Finally, the chemical characterization of PM samples was used to confirm the different origin of high PM during the sampling campaign, and it should be performed when the four indicators are not sufficient to deeply describe PM sources. The approach suggested in this paper should be adopted by air quality managers (regional and national government) for the evaluation of PM origin and for the implementation of policies complying with European and national regulations.

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