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Indicators reflecting local and transboundary sources of $PM_{2.5}$ and PM_{COARSE} in Rome – Impacts in air quality

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HIGHLIGHTS

- PM_{10} transportation in Rome along with slow moving air masses is suggested.
- Intrusions of $PM_{2.5}$ were originated from Balkan Peninsula due to combustion.
- Exogenous sources of PM_{COARSE} are scattered across Mediterranean and North Africa.
- PCA strongly associated local $PM_{2.5}$ emissions with vehicular combustion.
- Secondary local sources of PM_{COARSE} (natural, dust resuspension) were indicated.

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ABSTRACT

The keystone of this paper was to calculate and interpret indicators reflecting sources and air quality impacts of $PM_{2.5}$ and PM_{COARSE} (PM_{10}–$PM_{2.5}$) in Rome (Italy), focusing on potential exogenous influences. A backward atmospheric trajectory cluster analysis was implemented. The likelihood of daily PM_{10} exceedances was studied in conjunction with atmospheric patterns, whereas a Potential Source Contribution Function (PSCF) based on air mass residence time was deployed on a grid of a $0.5^\circ \times 0.5^\circ$ resolution. Higher $PM_{2.5}$ concentrations were associated with short/medium range airflows originated from Balkan Peninsula, whereas potential PM_{COARSE} sources were localized across the Mediterranean and coastal North Africa, due to dust and sea spray transportation. According to the outcome of a daily Pollution Index (PI), a slightly increased degradation of air quality is induced due to the additional quantity of exogenous PM but nevertheless, average levels of PI in all trajectory clusters belong in the low pollution category. Gaseous and particulate pollutants were also elaborated by a Principal Component Analysis (PCA), which produced 4 components: [Traffic], [photochemical], [residential] and [Secondary Coarse Aerosol], reflecting local sources of air pollution. $PM_{2.5}$ levels were strongly associated with traffic, whereas PM_{COARSE} were produced autonomously by secondary sources.

1. Introduction

The inhalable fraction of airborne Particulate Matter (PM) includes aerosols with diameter less than 10 um (PM_{10}). Increased mortality and morbidity levels were associated in the past with elevated PM_{10} concentrations (Tao et al., 2014; Curtis et al., 2006). PM_{10} and primarily $PM_{2.5}$ (diameter less than 2.5 um), are able to penetrate in the human respiratory system (Cachon et al., 2014; Delgado-Buenrostro et al., 2013) inducing respiratory, cardiovascular and pulmonary diseases (Zhou et al., 2014; Guo et al., 2010). If the chemical composition of PM_{10} includes metalloid elements, these chemical properties are considered as responsible for contingent carcinogenic impacts (Garcia-Aleix et al., 2014, Fang et al., 2013). In addition, the demonstrated solubility of PM metal traces in lung fluids indicates an enhanced pulmonary toxic potential upon their inhalation (Wiseman and Zereini, 2014). In order to protect the European population, the European Union (EU) established a 50 ug/m^3 average daily concentration limit for PM_{10} that should not be exceeded more than 35 times per year, whereas the annual limit is set at 40 ug/m^3. For $PM_{2.5}$, only a 25 ug/m^3 annual average concentration limit is recommended by EU regulations.

Principal Component Analysis (PCA) is a statistical method widely applied, in order to organize the air pollution mixture to main components and reduce the dimension of the input data (Minguillon et al., 2013; Singh and Sharma, 2012). High contributions of gaseous pollutants in specific PCA components are

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indicators of distinct emission sources affecting air quality (Yoo et al., 2011; Slezakova et al., 2013). Generally CO, NO, NO_2 and Benzene (C_6H_6), are pollutants which are indicative of gasoline combustion (Traffic), SO_2 is attributed to oil/natural gas combustion (households, industries etc), whereas O_3 is an indicator of photochemical reactions (Statheropoulos et al., 1998; Vardoulakis and Kassomenos, 2008). Increased correlations between gaseous and particulate air pollutants are commonly used as markers for the identification of PM sources (Juda-Rezler et al., 2011; Dimitriou and Kassomenos, 2013). Hence, high loadings for PM in PCA components, along with enriched gaseous influences, suggest the origin of the emissions (Yoo et al., 2011; Kassomenos et al., 2014).

Long range transportation of PM in urban areas is usually studied by air mass trajectories (Makra et al., 2011; Kassomenos et al., 2012). In the Mediterranean basin, Saharan dust outbreaks and Mediterranean sea spray are the principal exogenous sources of dust and maritime aerosols in urban areas (Valenzuela et al., 2012; Remoundaki et al., 2011; Kocak et al., 2007; Almeida et al., 2005), whereas other external sources as power plant combustion and biomass burning are also indicated (Argyropoulos et al., 2012; Gerasopoulos et al., 2011). Slow moving air masses can more effectively absorb and transfer particulates in urban areas, due to their longer residence time over regions where natural and anthropogenic PM emissions occur (Karaca et al., 2009; Fleming et al., 2012; Dimitriou and Kassomenos, 2014). Thus, the amount of time air spends over a region is linearly related to that region's contribution to pollutants measured at the receptor site (Xu et al., 2006; Kavouras et al., 2013; Chalbot et al., 2013). Consequently, short range trajectories describing the course of slow moving air parcels are associated with increased PM concentrations in urban areas (Borge et al., 2007; Karaca and Camci, 2010; Dimitriou and Kassomenos, 2013).

The keystone of this paper was to calculate and interpret indicators reflecting sources and air quality impacts of $PM_{2.5}$ and PM_{COARSE} (PM_{10}–$PM_{2.5}$) in Rome (Italy), focusing on potential exogenous influences. Initially, PM_{10} concentrations corresponding to backward atmospheric trajectory clusters produced at 500 m Above Ground Level (AGL) were analyzed by two statistical indices (Borge et al., 2007; Murena, 2004), reflecting the frequency of occurrence of daily PM_{10} exceedances and air quality degradation respectively. Subsequently, the residing time of airflows suspicious for PM transportation was analyzed on a $0.5^\circ \times 0.5^\circ$ resolution grid, in order to produce a Potential Source Contribution Function (PSCF) indicating potential source areas (Kong et al., 2013; Polissar et al., 2001; Karaca et al., 2009; Kocak et al., 2011). PSCF was independently implemented for $PM_{2.5}$ and PM_{COARSE}, aiming to reveal possible different transboundary sources of fine and coarse particles affecting air quality in Rome. Additionally, daily concentrations of gaseous (NO_2, SO_2, CO, O_3 and C_6H_6) and particulate ($PM_{2.5}$ and PM_{COARSE}) air pollutants were also elaborated by a PCA. Gaseous air pollutants were considered as indicators of local $PM_{2.5}$ and PM_{COARSE} emissions.

2. Data and methodology

2.1. Data and sampling sites

For this study, PM_{10} and $PM_{2.5}$ levels measured at the historical urban/background sampling site "Villa Ada" (IT0953A) [lon: 12.506.945, lat: 41.932.777] in Rome (Italy) were analyzed. Due to the absence of hourly data for $PM_{2.5}$, daily concentrations of PM were used. In addition, daily concentrations of gaseous pollutants (CO, NO_2, SO_2, O_3 and C_6H_6) were also studied. Statistics and deficiencies (%) of PM_{10}, $PM_{2.5}$, CO, NO_2, SO_2, O_3 and C_6H_6 daily mean concentration data series during cold periods (CP: 1 October–31 March) and warm periods (WP: 1 April–30 September), are included in Table 1. A seven years dataset was studied, extending through the time interval 2006–2012. All data were downloaded from the website of EU Air Quality Database (Airbase) in ugr/m^3, except of CO (mg/m^3). IT0953A station is equipped with Beta Ray Attenuation analyzers for the monitoring of PM concentrations, whereas CO, NO_2, SO_2, O_3 and C_6H_6 levels are measured with infrared absorption, chemiluminescence, UV fluorescence, UV absorption and chromatographic analyzers respectively. All appliances are expected to operate within 15% of uncertainty bounds, according to EU guidelines.

The selection of IT0953A station for this study prevailed, due to the long available series of PM_{10} and $PM_{2.5}$ concentrations in its archives, and also due to the geographical position of the station which is suitable for the identification of long range transport impacts. The sampling site is situated inside Rome's major green park (Villa Ada), and it is not directly influenced by local emission sources but it's characterized by a homogeneous pollution that can be considered a background for Rome (Avino and Brocco, 2004). The station is located approximately 500 m away from main traffic arteries (Gobbi et al., 2007), neighboring to the northern ("Via del Foro Italico") and eastern ("Via Salaria") boundaries of the park. The area that surrounds the park is mainly residential.

2.2. Methodology

2.2.1. PCA Analysis

The principal purpose of this paper, is to define atmospheric pathways possibly contributing to increased $PM_{COARSE} = PM_{10}$–$PM_{2.5}$ and $PM_{2.5}$ levels in Rome. Nevertheless, local PM sources also had to be studied, in order to complement and support the findings of long range transport analysis. Daily concentrations of particulate (PM_{COARSE} and $PM_{2.5}$) and gaseous (CO, NO_2, SO_2, O_3 and C_6H_6) air pollutants were elaborated by a PCA, in order to define local factors related to PM emissions (Yoo et al., 2011). CO and C_6H_6 were considered as indicators of traffic (Avino and Manigrasso, 2008; Deacon et al., 1997), whereas SO_2 and O_3 were markers of domestic/industrial and photochemical air pollution respectively (Vardoulakis and Kassomenos, 2008). NO_2 is produced by various combustion sources. PCA was implemented separately in CP and WP, aiming to reveal possible seasonal trends of local PM emissions (Statheropoulos et al., 1998).

2.2.2. Air mass trajectory classification

Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT) of the NOAA Air Resources Laboratory was used, in order to produce 4-day backward air mass trajectories approaching Rome during the 7-years period 2006–2012 (Obrist et al., 2008). The trajectories reached the city at 500 m Above Ground Level (AGL), an altitude suitable for the identification of long range transport impacts, ensuring that the trajectory starts in the near ground atmospheric boundary layer (Karaca et al., 2009; Makra et al., 2011). The time of every air parcel's arrival in the two cities was set at 12:00 UTC. Only days with available data of daily mean PM_{10} and $PM_{2.5}$ concentrations at the IT0953A station's archives were used as

Table 1
Statistics and deficiencies (%) of PM_{10}, $PM_{2.5}$, CO, NO_2, SO_2, O_3 and C_6H_6 daily mean concentration data series of IT0953A station, during CP and WP of the time interval 2006–2012.

		$PM_{2.5}$	PM_{10}	SO_2	O_3	CO[a]	NO_2	C_6H_6
CP	Average (ug/m^3)	23.0	30.5	1.4	24.2	0.6	47.6	2.1
	Stan-dev (ug/m^3)	12.9	15.3	1.2	15.8	0.2	15.3	1.2
	Deficiencies (%)	6.7	3.8	13.9	0.9	0.5	1.3	8.1
WP	Average (ug/m^3)	15.5	24.6	1.2	55.5	0.4	31.8	0.9
	Stan-dev (ug/m^3)	5.8	9.1	0.9	16.2	0.1	11.3	0.5
	Deficiencies (%)	5.0	5.6	14.0	2.0	1.0	0.7	11.7

[a] CO concentrations are presented in mg/m^3.