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MEASURING OF PARTICULATE MATTER LEVELS IN THE GREATER AREA OF ATHENS AT TWO DIFFERENT MONITORING SITES

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ABSTRACT

Simultaneous PM_{10} and $PM_{2.5}$ sampling was conducted for a six months period, at two sites of different characteristics. The first site was located in an urban background location in the North-Eastern part of GAA, affected by primary emission sources and particle transport from other parts of the GAA basin. The second site was located in central Athens, at a busy roadway, affected by heavy traffic-related and commercial activities. Additionally, continuous field measurements of PM fractions were also performed using direct-reading monitor in parallel with gravimetric samplers. The mean PM_{10} and $PM_{2.5}$ concentrations for the sampling period were 26.2 and 13.7μg m^{-3} and 40.1 and 22.8 μg m^{-3} for ZOG and ARI, respectively. The $PM_{2.5}/PM_{10}$ ratio found 0.52 and 0.57 for ZOG and ARI, respectively. The coefficient of variation calculated equal to 0.40 for both fractions. Additionally, the weekday/weekend discernment of the particulate concentration levels for each site display different characteristics of the emission sources and composition, while the diurnal distribution of particulate levels demonstrated the dependence of the PM levels on anthropogenic activities and habits.

Both the local meteorological conditions and the air mass history indicating long-range atmospheric transport of particles are significant parameters that influence the levels of PM.

Keywords

PM_{10}; $PM_{2.5}$; spatial variation, diurnal distribution; long range transport

1. Introduction

Over the last two decades a large number of epidemiological studies conducted in many countries around the world observed associations between ambient particle concentrations and human health risks (Nelin *et al*., (2012), Pope and Dockery (2006)). These results highlighted the importance of continuous monitoring of ambient particles, the inhalable (PM_{10} fraction) particles and especially the fine ($PM_{2.5}$ fraction) particles, which penetrate deeper in the respiratory track and might have a direct harmful impact on human health.

To address these critical issues for the Greater Area of Athens (GAA) systematic efforts have been conducted by Chaloulakou *et al*., (2003, 2005), Grivas *et al*., (2004, 2008), Diapouli *et al*., (2008) over the last years. The present work is focusing on parallel measurements in two sites of different characteristics, for both PM fractions (PM_{10} and $PM_{2.5}$). The spatial, the temporal and the diurnal variation of PM_{10} and $PM_{2.5}$ concentrations as well the intra-fraction correlation were explored together with the association between PM fractions and meteorological parameters, allowed a further insight on the factors affecting the measured ambient particulate levels.

2. Experimental

2.1 Study area characteristics

Sampling sites were located within the Athens Basin (450 km^2), the largest and most populated plain of the Attica peninsula. The basin is surrounded by high altitude terrain in the north, which becomes lower towards the Saronic Gulf in the S–SW. Parnitha Mt. (1413 m asl) covers the northern sector of the basin, Penteli Mt. (1109 m asl) lies in the northeast and Hymettus Mt. (1026 m asl) in the east extends to the sea. The mountainous arc surrounding the basin closes with Aegaleo Mt. (468 m asl) in the N–NW. Mountains are connected with moderately high saddles, at the main exits of the basin to the Mesogeia plain in the East and the Thriassion plain in the West. The large industrial complexes are located in the Thriassion plain, several kilometres to the west of the GAA. They are separated from the Athens basin by mount Aigaleo that acts as a physical barrier preventing most of the exchange of air pollutants between the industrialized area and the city (Melas *et al*., 1998). It is apparent that the complex topography of the basin entails significant difficulty in the dispersion of air masses over the city, especially during low wind periods, favoring high particulate matter concentrations, often reported during the past years (Chaloulakou *et al*., 2003, Grivas *et al*., 2008).

2.2 Sampling sites and method

PM_{10} and $PM_{2.5}$ sampling was conducted for a six months period (February to September 2011), using Harvard impactors at a flow rate of 10lpm, at two sites of different characteristics (fig. 1). The first site was located in Zografos (ZOG), an urban background type location in the North-Eastern part of GAA. The site is located at ground level, inside the National Technical University of Athens (NTUA) campus, the surrounding roads experience little traffic which is minimized during the afternoon hours and weekends and is 0.337 and 0.438 km away from two of the major avenues of Athens, mainly affected by primary emission sources and particle transport from other parts of the GAA basin. The second site was located in central Athens, at Aristotelous Str. (ARI), a busy roadway close to a three-roadway junction, affected by heavy traffic-related and commercial activities. The samplers were installed 7m above the ground, on the first floor balcony of the Greek Ministry of Health.

Figure 1. Overview of the Greater Athens Area. Sampling locations are noted on figure

PM was collected on pre-weighted 37 mm Teflon-coated glass-fiber filters (2μm pore size). The sampling program resulted in 98 filters being collected in ZOG and 82 in ARI. A series of field blank filters were collected throughout the campaign, replicating sampling conditions. Both blank and field filter samples were conditioned in a clean room at constant temperature (20^{o}C) and relative humidity (54.5%) for at least 48 h prior to being weighted.

Additionally, continuous field measurements of PM fractions (PM_{10} and $PM_{2.5}$) were also performed at ARI site, using direct-reading automated monitor (TSI DusTrak DRX 8533) in parallel with gravimetric samplers. The instrument was calibrated by the manufacturer using standard ISO 12103-1, A-1 test dust (formerly Arizona Test Dust). For that reason a calibration method indicated by the manufacturer was used to calibrate the instrument to actual measured aerosol.

Meteorological data from the nearest, to each site, monitoring station were also obtained, in order to detect any interactions of the PM concentrations with the weather conditions.

In order to examine possible relationships between ground level PM concentrations and long-range transport of polluted air masses over the GAA, days with daily $PM_{10}/PM_{2.5}$ ratio higher than two were identified and plotted the three-day long kinematic back trajectories (with arrival time 12.00 UTC). The kinematic back trajectories were modeled with the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT, version 4) developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (further information on the model can be found at: http://www.arl.noaa.gov/ready/hysplit4.html). The meteorological data used for the computation of the trajectories were obtained from the NOAA reanalysis database (http://www.arl.noaa.gov/archives.php), and the trajectory arrival height was 750 m above ground level in all cases. It must be noted that the accuracy of an individual trajectory is limited by uncertainties as meteorological inputs and analysis errors.

3. Results and discussion

3.1. PM levels

Descriptive statistics for PM_{10} and $PM_{2.5}$ concentrations, as measured at the two sites are summarized in table 1.

Table 1. Descriptive statistics for daily average PM_{10} and $PM_{2.5}$ concentrations at the two sampling sites. (All values in μg m^{-3})

Site	**ZOG**		**ARI**	
PM Fraction	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$
Mean	26.3	13.8	40.1	22.9
SD (±)	9.8	5.1	14.8	8.2
Min	8.4	4.2	10.4	5.3
10-percentile	14.9	7.1	20.9	11.7
Median	25.1	13.3	39.1	22.6
90-percentile	39.0	20.0	59.5	33.7
Max	49.8	27.1	72.1	39.2

The observed PM_{10} concentration levels at ARI site were lower (25%) than those reported in a study by Aleksandropoulou *et al*., (2012), including data retrieved from European Air quality database for the period 2001–2008 and from the Hellenic Ministry for the Environment, Energy and Climate Change (H.M.E.E.C.C.) for 2009 and 2010. In comparison to the PM levels reported by Grivas *et al*., (2008), in a measuring campaign (January 2001-December 2004), the decrease on the observed, in this study, PM_{10} concentration levels found to be 28%. However, the reported levels by Grivas *et al*., (2008) were lower (23-32%) than those reported by Chaloulakou *et al*. (2003) and Grivas *et al*. (2004), on previous studies (the two studies cover the periods January 2000–December 2000 and June 2001–May 2002, respectively). Compared to these studies, the present study recorded lower PM_{10} concentration levels by 46% and 51%, respectively. Additionally, Aleksandropoulou *et al*., (2012) and Grivas *et al*., (2008) have recorded higher PM_{10} concentrations (11 and 21%, respectively) in a similar site in Zografos. A yearly decrease on PM_{10} levels is appeared, reported also by Aleksandropoulou *et al*., (2012), as it is also indicated in the above. The use of natural gas for domestic heating purposes, which has increased lately and new technology vehicles equipped with particle traps producing noticeable decreased PM emissions, can be designated as potential factors, among others, responsible for this significant decrease in atmospheric particle concentration during the last decade in Athens. Concerning the 24-h limit value (50 μg m^{-3}), no exceedance in 49 cases was recorded in ZOG and 10 exceedances in 41 cases in ARI (24.4%), overcoming, proportionally, the EU air quality standard (AQS) annual maximum margin of tolerance (10%). $PM_{2.5}$ levels ranged between 4.2 and 27.1 μg m^{-3} (mean 13.8 μg m^{-3}) and 5.3 and 39.2 μg m^{-3} (mean 22.9 μg m^{-3}) at ZOG and ARI site, respectively. The presence of a higher mean concentration in ARI is not unexpected, given that the site is located in a busy roadway in the center of Athens. However, the recorded levels are lower (42%) compared to those reported by Chaloulakou *et al*., (2003), in the same site. The average concentration levels of both sites were found lower than the EU air quality standard value of 25 μg m^{-3} (Directive 2008/50/EC), however these levels overcome the annual USEPA NAAQS standards (15 μg m^{-3}). At that point should be emphasized that the EU and USEPA standards for each of the PM fraction are referring in annual and in 3-years basis, respectively.

3.2. Spatial variation of PM_{10} and $PM_{2.5}$ concentrations.

PM_{10} and $PM_{2.5}$ concentrations, for the measurement period, were strong correlated in both sites with Pearson correlation coefficients' of 0.76 and 0.89, for ZOG and ARI, respectively. The reported coefficients and their slight differentiation ($r_{ARI} > r_{ZOG}$) match to those reported by Querol *et al*., (2004), Putaud *et al*., (2010) and Harrison *et al*., (2012), for similar type of sites and r_{ARI} was similar to those reported in the same site by Chaloulakou, *et al*., (2003). Since $PM_{2.5}$ accounts for a major proportion of PM_{10} (Harrison *et al*., 2012), the observed strong consistency in the correlation coefficients' at the two different sites, indicates that meteorological conditions and emissions from sources, which are effective over all the area (e.g. traffic), rather than specific local sources and events, dominate the relative variations of the concentrations for the measurement period. Furthermore, the Pearson correlation coefficients between sites were high ($r_{PM10} = 0.78$ and $r_{pm2.5} = 0.80$), suggesting regionally homogeneous sources (Wilson *et al*., 2005), possibly related to the relatively uniform distributions of $PM_{2.5}$ concentrations over large areas (USEPA, 1998).The coefficient of variation (CV) between sites, based on daily averaged concentrations found equal to 0.40 for both PM fractions. This is a value relatively high, as compared to studies examining spatial variations in large metropolitan areas (Martuzevicius *et al*., 2004). It is, however, validated by previous findings regarding the degree of PM_{10} spatial variation in the area of Athens (Grivas *et al*., 2004; Grivas *et al*., 2008).

The $PM_{2.5}/PM_{10}$ ratio was found to be 0.52 and 0.59 at ZOG and ARI, respectively. The slight increase on the ratio observed in ARI compared to ZOG site is attributed on the higher PM_{10} levels recorded on that site and are in agreement with ratios reported by Van Dingenen *et al*., (2004), Putaud *et al*., (2010) and Harrison *et al*., (2012) for similar type of sites. The above observation affects the ratio since fine particles have a major contribution to PM_{10} mass concentrations (Harrison *et al*., 2012).

3.3 Monthly and weekly variation

Monthly fluctuations in PM_{10} and $PM_{2.5}$ concentrations are shown in figure 2.

Higher PM_{10} concentrations in ZOG were recorded in February, March and April, months with low ambient temperature. The negative correlation between PM_{10} and mean daily temperature (r = -0.44), for these months, was reasonably, suggesting that lower temperature resulted in excessive use of domestic heating. The highest PM_{10} concentrations in ARI were recorded in September. High levels of PM_{10} concentrations in Athens in September have also been recorded by Kalabokas *et al*., (2010), including data retrieved from the Greek National Air Pollution Monitoring Network (NAPMN) for the 4-year period 2001-2004. Nevertheless, September 2011 had a peculiarity since it was a month with intense political turmoil in the center of Athens, since days with demonstrations were followed by days when the public transportations were on strike, causing extended *use* of cars and intense traffic emissions. In particular, a day describing the above-mentioned situation was the 30th of September, where the recorded levels were 57.7 and 29.5 μg m^{-3} for PM_{10} and $PM_{2.5}$, respectively, while a usual day, with similar meteorological conditions, was the 28th of September, where the recorded levels were 37.6

and 20.5 μg m^{-3} for PM_{10} and $PM_{2.5}$, respectively. Regarding the $PM_{2.5}$ fraction in both sites, a slight monthly variability was observed. A decline, on both fractions of PM in both sites, was observed in late July, a period dominated by the well-established etesian winds, a generally strong (25% greater than the average) northerly wind flow phenomenon resulting a dilution effect (Kassomenos, *et al*., 2003).

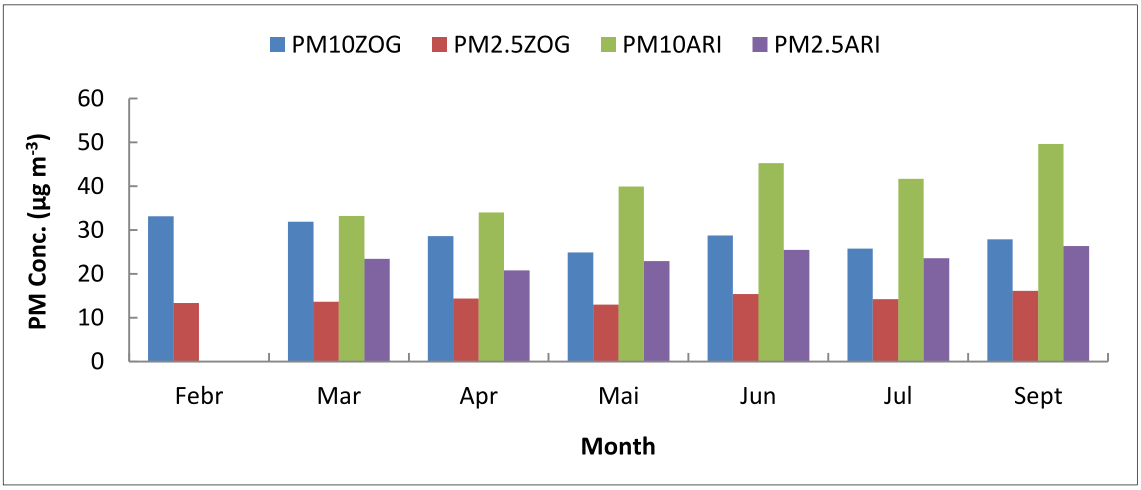


Figure 2. Mean monthly variation of PM_{10} and $PM_{2.5}$ concentrations for both sites

The "day of week" pattern (figure 3) reveals that higher PM_{10} and $PM_{2.5}$ concentrations were observed on weekdays (Monday to Friday) compared to weekends (Saturday and Sunday) for both sites. This is reasonable since traffic- and other human- related particle generating activities are reduced during Saturdays and to a greater extent during Sundays. Additionally, in the case of $PM_{2.5}$, uniformity is observed for concentrations from Monday to Saturday for both sites, with only concentrations on Sunday being significantly lower (up to 25%) in ARI, indicating the contribution of traffic emissions to $PM_{2.5}$ levels. Additionally, the concentration levels in ARI was 35% higher in weekends compared to concentration levels in ZOG for both PM fractions while the corresponding deference between the two stations for weekdays was 35% and 40% for PM_{10} and $PM_{2.5}$ concentration levels, respectively. The recorded "day of week" PM_{10} and $PM_{2.5}$ concentration levels in ARI found significant lower than those reported by Chaloulakou *et al*. (2005) in the same site. The percentage reduction was 47% and 43% on weekdays, 46% and 43% on Saturdays and 39% and 49% on Sundays for PM_{10} and $PM_{2.5}$, respectively. The clear reduction in levels in an urban traffic and commercial site characterized by the Ministry of Environment, Energy and Climate Change, analyzed in two parameters derived from the same source, the economic crisis. The first is the clear effect of the economic crisis in the use of cars and heating systems, while the second parameter refers to the decline on commercial use.

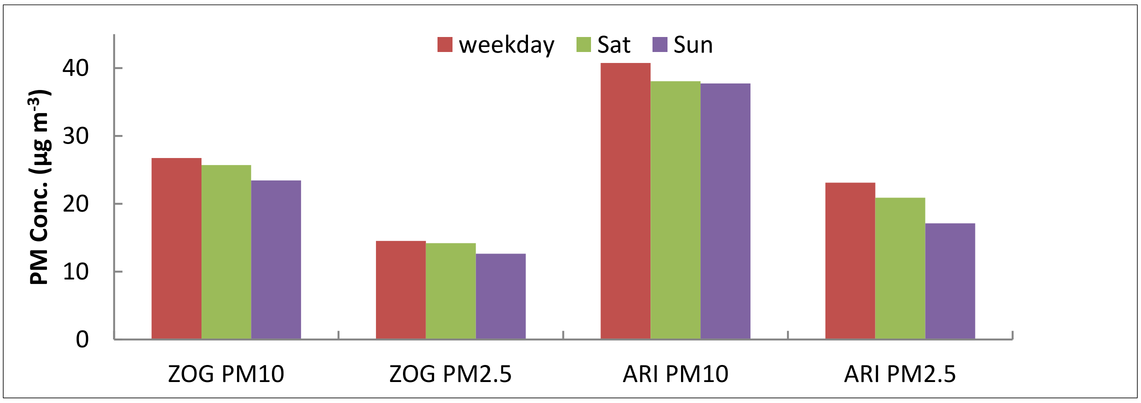


Figure 3: PM_{10} and $PM_{2.5}$ concentrations on weekdays and weekends for both sites

3.3 Diurnal variation

Daily cycles of PM_{10} and $PM_{2.5}$ performed with DustTrak DRX are presented on figure 4. The DustTrak measurements were recalculated using a site specific calibration factor for each measurement (Park *et al.*, 2009). The gravimetric concentration and integrated DustTrak DRX concentrations were highly correlated (r_{PM10} = 0.76 and $r_{PM2.5}$ = 0.77).

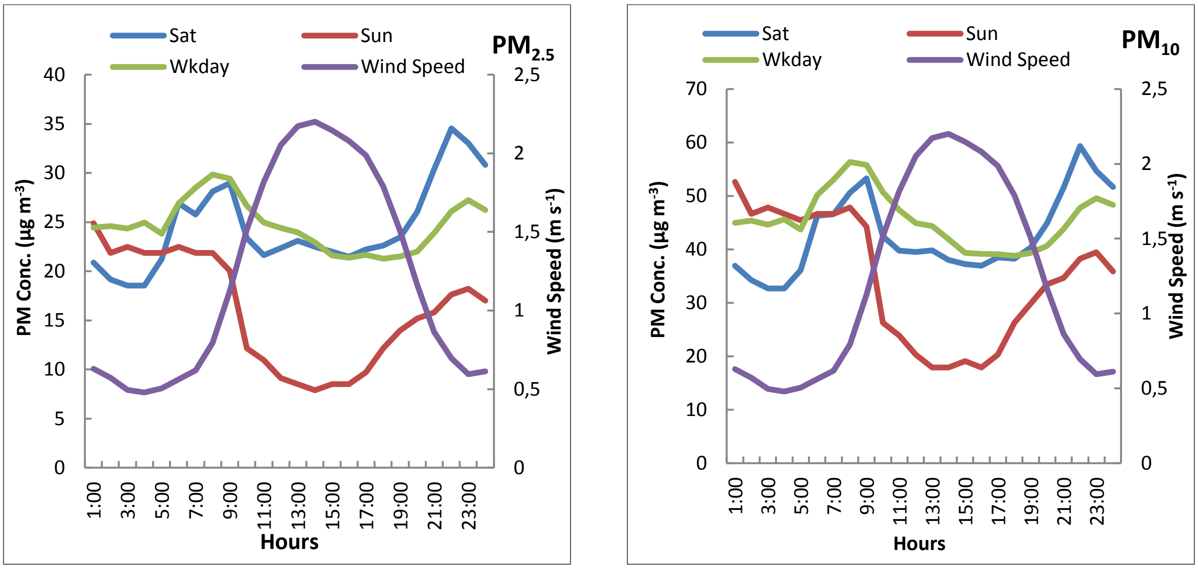


Figure 4. Diurnal cycles of PM_{10} and $PM_{2.5}$ concentrations on weekdays and weekends and wind speed in ARI

The morning build up of emissions from local anthropogenic activities and especially go-to-work traffic was responsible for an early morning peak (07.00–09.00 h). The morning concentration peak was followed by a gradual decrease due to meteorological conditions. The increase in wind speed till late afternoon enhances the dilution of pollutants, while rising temperatures during the day, lead to atmospheric mixing and the elevation of the mixed layer height, resulting in the better dispersion of pollutants. Thus, pollutant concentrations decreased in the late morning and remained low throughout the afternoon and early evening. From that point, PM concentration increased up to nocturnal values where, another significant peak in PM concentrations was observed during night-time which was related to the drop in the mixed layer height. Transport of aerosols from other parts of the region, is also possible under favoring meteorology. Saturday's diurnal cycle follows the weekdays' pattern since the site is characterized by commercial activity while a second distinct peak due to night social activities is observed. Sunday's diurnal cycle demonstrated a completely different pattern compared to the weekdays' due to the absence of an intense source of pollution such as traffic.

3.4 Relationships of PM and meteorological parameters

PM_{10} and $PM_{2.5}$ daily average concentrations recorded important negative correlations with wind speed (r values exceeding -0.44, for both fractions, in both sites). Strong inverse correlations of PM with wind speed have been reported in many cases, for urban sites in Athens (Chaloulakou *et al.*, 2005) and are indicative of the wind dilution effect.

Additionally, concentration data were binned according to daily average wind speed (figure 5). While $PM_{2.5}$ daily concentrations decline markedly with increasing wind speed, PM_{10} concentrations reduce initially until a wind speed threshold but then increase due to re-suspension of coarse particles (Kassomenos *et al.*, 2012).

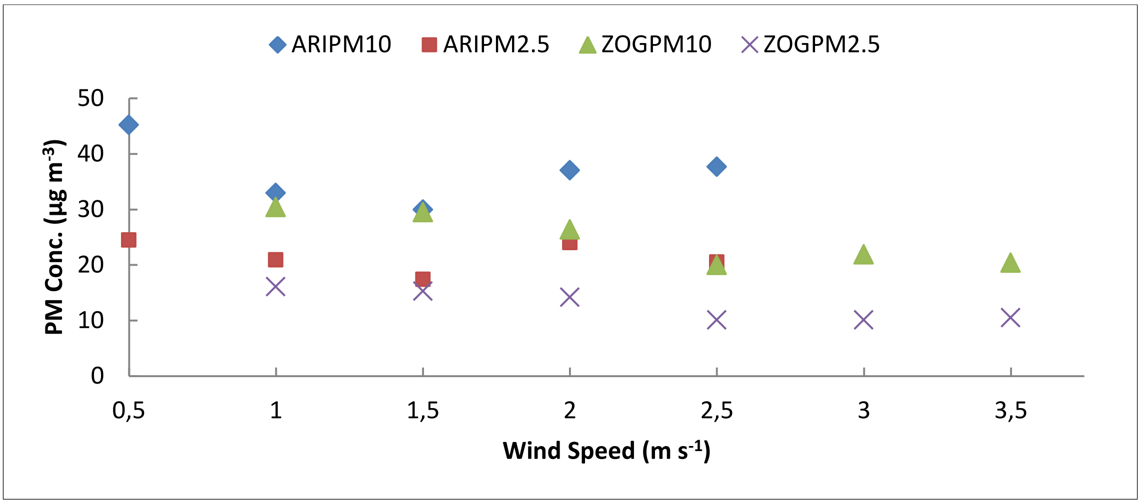


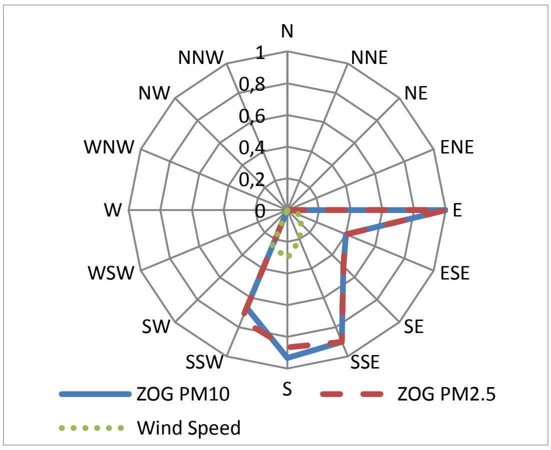
Figure 5. Wind speed dependence of PM concentrations for both sites

The influence of wind direction is presented at figure 6, by histogram plots of the conditional probability function (CPF) for 16 directions. The CPF has been used for finding the directions of pollution sources (Kim *et al.*, 2004) and estimates the probability that concentrations observed under a specific wind direction range, exceed a predetermined threshold value. It is described by the equation:

$$CPF\Delta\Theta = m\Delta\Theta / n\Delta\Theta \qquad (1)$$

where mΔΘ is the number of number of exceedances of the threshold during winds from the sector Δθ sector and nΔΘ the total number of concentration data from the same wind sector. Calm conditions (wind velocity < 1 m s^{-1}) were not appeared. The 25th percentile of concentrations was set as the threshold (fig. 6).

Regarding the frequency of winds from specific directions in ZOG, winds from the southern sector (SSW-SSE) dominated, having a cumulative frequency of 89 and 92% for PM_{10} and $PM_{2.5}$, respectively. The occurrence of high PM levels during the prevalence of southwestern flows was related to the limited ventilation of the basin on those days (Kassomenos *et al.*, 1998a) and the transfer of particulate matter from the densely populated and trafficked center of the city. On the other hand, the increased PM levels during wind flows from the eastern sector were attributed in the highway located to this orientation.



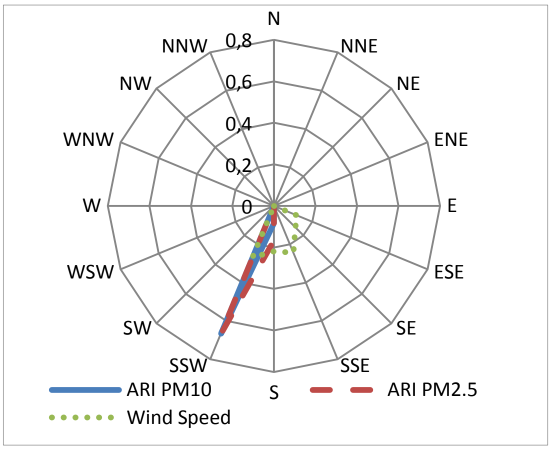


Figure 6. Conditional probability function (CPF) of PM_{10}, $PM_{2.5}$ and wind speed, for 16 wind directions, for both sites

In ARI, high levels of both PM_{10} and $PM_{2.5}$ were observed during winds of southern sector (SSW-S), under prevalence of weak southerly flow conditions. The appearance of weak southerly flow is associated with limited ventilation conditions. (Kassomenos *et al*.,1998a). Consequently, in the area of the site, accumulation of PM_{10} and $PM_{2.5}$ produced in the S-SW part of the city (which includes the industrial and harbor zones) was possible.

In order to examine relationships between the recorded ground concentration levels and possible long-range transport of polluted air masses, days with daily $PM_{10}/PM_{2.5}$ ratio higher than two were identified and the three-day long kinematic back trajectories were plotted. The analysis of the back trajectories arriving in the urban background site (ZOG) in Athens revealed two patterns, the first consisting of 50% of the high $PM_{10}/PM_{2.5}$ days is associated with trajectories with origin over the valleys of south Russia and Ukraine. This area is considered a reservoir of particulates, both anthropogenic and naturally oriented, coming under certain atmospheric circulations from the surrounding industrial areas of Russia and Ukraine. Grivas *et al*. (2008), have also reported significant long range transportation of pollution from this area to GAA in the past. The second (37.5%) consists of air masses coming from the maritime area between Crete and the North African Coast.

4. Conclusion

PM_{10} and $PM_{2.5}$ sampling was conducted during the period of February through September. Sampling was conducted, in parallel, at two sites of different characteristics in order to obtain a thorough estimate of population exposure.

The percentage of days when PM_{10} concentrations exceeded the EU AQS 24-h limit value of 50 μg m^{-3} was 24% in the urban site, overcoming proportionally the annual maximum margin of tolerance (10%), while no excedance recorded in the urban background site. Concerning the $PM_{2.5}$ levels, the average concentration levels of both sites were found lower than the EU AQS value of 25 μg m^{-3}, however these levels overcome the annual USEPA NAAQS standards (15 μg m^{-3}). Nevertheless, the EU and USEPA standards for each of the PM fraction are referring in annual and in 3-years basis, respectively.

The performed statistical analysis led to similar findings, concerning the spatial differentiation, to those reported by other researchers for similar type of sites, indicating the potential sources and factors underlying the observed levels. PM_{10} and $PM_{2.5}$ concentrations showed higher correlation in the city center, indicating higher participation of traffic in the recorded levels. Additionally, both sites were affected by the transfer of PM from more polluted areas, when southwestern wind directions prevailed, due to their relative position in the GAA. However, the urban background site found to be influenced by more prevailing wind directions, due to its orientation towards other significant local sources.

Higher PM_{10} concentrations in the urban background site was recorded in months with low ambient temperature as a result of extensive impact of domestic heating in the recorded levels of that site, while higher PM_{10} concentrations in the urban traffic site was recorded in September, a month with political turmoil in the center of Athens. Regarding the $PM_{2.5}$ fraction in both sites, a slight variability was observed.

The weekly variation of PM levels demonstrated the expected behavior, displaying higher levels on weekdays compared to Saturdays and Sundays, for both sites, while the diurnal variation showed significant correlation between PM levels and anthropogenic activities and habits. Higher levels of PM concentrations are directly related to the prevailing meteorological conditions, particularly to wind direction, indicating the important role of both local sources and characteristics as well as the relevance of regional sources. Additionally, sources of the PM were also investigated using atmospheric back trajectory modeling. The modeling results indicate that there are significant long-range transport sources of particles in GAA, in the measurement period, in addition to local sources. High PM_{10} levels were mainly associated with air masses originating from continental Europe (north central and northeast) and from north Africa.

Findings that might be attributed to the effects of economic crisis could be extracted by the significant reduce on PM levels compared with previous studies. The reduce in the use of cars and heating systems, the possibility of using cheaper but lower quality fuels and the change in consumption habits from the one and the impending use of diesel vehicles in the center of Athens from the other, shaping a new momentum in the GAA in which the continuous monitoring of PM levels would be a useful tool towards an estimation of population exposure.

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