



Identification of the Influence of African Dust on PM₁₀ Concentrations at the Athens Air Quality Monitoring Network during the Period 2001–2010

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ABSTRACT

The objective of this work was to identify Sahara dust intrusions and quantify their contribution to annual and daily PM₁₀ concentrations over the Athens Metropolitan Area (AMA) during the period 2001–2010. Additionally, extreme PM₁₀ values attributed to large forest fires and volcanic emissions or the synergy of the above with African dust intrusions have been identified. It was found that the concentrations of PM₁₀ during the period studied exceeded the European Union's Air Quality Standard (EU AQS) value at most of the traffic urban (TU) stations. Most of the extreme values were attributed to dust events or large forest fires. The contribution from African dust to PM₁₀ concentrations on days with events and exceedances of the EU AQS was approximately 50% at TU stations and 72% at the background suburban stations (BS). Most of the African dust events contributed about 5 µg/m³ to 34 µg/m³ to the daily PM₁₀ concentrations, whereas the average contribution to annual PM₁₀ concentrations was approximately 8.6 ± 1.8 µg/m³ i.e., 22.2 ± 7.5%. During the decade, no clear interannual relationship was observed between the African dust contribution and the frequency and intensity of events. The more frequent and severe episodes occurred during spring, while less frequent and less severe episodes occurred during summer. After the extraction of the African dust load the annual and daily AQS was exceeded in 2010 at only one TU station. The average annual contribution of African dust to the exceedances was approximately 36% at TU stations and 58% at BS stations.

Keywords: Desert dust event; Forest fire; Urban air quality; PM₁₀.

INTRODUCTION

Sahara is the more intense Aeolian dust source in the world and is responsible for half of the mineral dust emissions globally (Zender *et al.*, 2004). Guerzoni *et al.* (1999) and Kallos *et al.* (2005) found that approximately 10⁸ metric tons of dust are deposited annually over the Mediterranean Sea, and a similar amount is transferred towards Europe. In particular, the deposition rate of Saharan dust in the Mediterranean has been estimated to 3–14 g/m² per year with the largest value found for the Eastern Mediterranean (Prospero, 1996). The Eastern Mediterranean receives air masses loaded with dust particles from the Northern Africa predominately during spring (associated with the eastward passage of low pressure frontal systems) and from the Middle East during the autumn (Samoli *et al.*, 2011).

Sahara dust is transported over the Mediterranean Sea mainly because of the cyclone activity inside and around the area (Prospero, 1996). Papayannis *et al.* (2005, 2008) found

that dust particles penetrate the geographical area located between 20–40°E and 40–47°N while the source region of desert dust for southern and south-eastern Europe is at the central and eastern Sahara regions. The long range transport of pollutants is induced by the different heating between the Mediterranean and Northern Africa, the land water contrast and the landscape variability (Kallos *et al.*, 2007). Dust transport over Greece mainly occurs during the warmer months of the year when the prevailing meteorological conditions are characterized by the occurrence of western-south-western synoptic flows which favor the transport of dust from North Africa (Katragkou *et al.*, 2009). During the cold period intense outbreaks occur but dust concentrations are often attenuated due to wet deposition (Nastos, 2012). The Sahara dust intrusions in the area are favored by the formation of southerly or south-westerly flow in the lower troposphere which leads to poor dispersion over urban areas (Astitha *et al.*, 2006). Moreover, the time scales for long range transport range from 1 to 4 days resulting in air quality degradation in cities/urban areas in SE Mediterranean (Kallos *et al.*, 1998).

Sahara dust has an important impact on the environment by affecting climatic processes, nutrient cycles, soil formation and sediment cycles (Goudie and Middleton, 2001). Dust particles modify the earth's radiation budget,

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act as Cloud Condensation Nuclei (CCN), so they alter cloud microphysical processes, and neutralize acid rain (Solomos *et al.*, 2011). Their effects in the Mediterranean include serious implications for the water budget and the regional climate (Kallos *et al.*, 1998; Santese *et al.*, 2010). Additionally, Sahara dust contributes to the increase of PM₁₀ concentrations over urban and other populated areas with significant effects on human health. Dust clouds have a large impact on human health as they transport infectious microorganisms to distances up to several thousands of kilometers (Prospero, 1999). In particular, Perez *et al.* (2008) showed that coarse particles during Sahara-Sahel desert dust events increased the daily mortality in Barcelona, whereas Samoli *et al.* (2011) examined the effects from short term exposure to PM₁₀ to total (all-cause) and cause specific (cardiovascular, respiratory) mortality in Athens, Greece during the period 2001–2006. It was found that a 10 µg/m³ increase in PM₁₀ concentrations can be associated with a 0.71% increase in total mortality with the effects being more intense for the elders (> 75 y) and females (Samoli *et al.*, 2011). A recent review indicates that further research is needed as regards the health effects from Sahara dust outbreaks especially in South Europe (Karanasiou *et al.*, 2012).

In an earlier study it was shown that the natural background levels cause difficulties for AMA to meet the EU AQS (Aleksandropoulou *et al.*, 2012a). The limit values for PM₁₀ set by the European Union in order to protect human health are 40 µg/m³ for the annual averaged concentrations and 50 µg/m³ for the 24 hour averaged concentrations, a value which must not be exceeded more than 35 times in a calendar year (EU, 1999, 2008). Additionally, previous studies of PM₁₀ concentrations in Athens have shown that some of the high PM concentration events occurring can be attributed to long range transported aerosols such as desert dust events (Grivas *et al.*, 2004; Borge *et al.*, 2007). In this study the concentrations of PM₁₀ during the period 2001–2010 at the AMA monitoring network are analyzed with the objective to identify high PM₁₀ events, exceeding the PM₁₀ daily limit values, caused by natural particulate inputs produced by Sahara air mass intrusions. Exceedances of the AQS caused by other extreme natural events i.e., large forest fires and volcanic eruption are also estimated in the current approach. In addition, the concentrations are examined with regards to compliance with the annual EU AQS. Finally, the contribution from African dust intrusions to annual and daily PM₁₀ concentrations over the Athens area is quantified.

MATERIALS AND METHODS

PM₁₀ Concentration Data

The PM₁₀ concentration data analyzed in this study have been recorded at the monitoring stations of the Athens Metropolitan area (AMA) network during the period 2001–2010. Specifically, PM₁₀ concentrations were measured at the traffic urban stations Aristotelous (ARI; 37.99°N, 23.73°E; 95 a.m.s.l.), Marousi (MAR; 38.03°N, 23.79°E; 170 a.m.s.l.), Goudi (GOU; 37.98°N, 23.77°E; 155 a.m.s.l.; 2001–2004, 2006–2007) and Pireaus-1 (PIR-1; 37.94°N, 23.65°E; 20

a.m.s.l.) and at the background suburban stations Lykovrisi (LYK; 38.07°N, 23.78°E; 210 a.m.s.l.), Agia Paraskevi (AGP; 37.99°N, 23.82°E; 290 a.m.s.l.), Thrakomakedones (THR; 38.14°N, 23.76°E; 550 a.m.s.l.), Zografou (ZOG; 37.97°N, 23.79°E; 245 a.m.s.l.; 2001–2007) and Koropi (KOR; 37.90°N, 23.88°E; 140 a.m.s.l.; 2008–2010). All the stations are part of the national air pollution monitoring network operated by the Hellenic Ministry for the Environment, Energy and Climate Change (H.M.E.E.C.C., 2011). Additionally are exploited data collected during 2010 at the rural background station in Aliartos (ALI; 38.37°N, 23.11°E; 110 a.m.s.l.; 29/7/2010–31/12/2010) which is a member of the EMEP network (European Monitoring and Evaluation Programme). The assessment of PM₁₀ concentrations is performed by beta ray attenuation (FH 62 I-R, ESM Andersen GmbH) at all stations. Equivalence to the reference methods given in Directive 2008/50/EC (EU, 2008) has been previously shown in Grivas *et al.* (2004) whereas more details on the sampling procedure and calibration are given in Grivas *et al.* (2008).

Identification of Desert Dust Intrusion Events Coinciding with AQS Exceedances

The African dust outbreaks coinciding with exceedances of the daily AQS for PM₁₀ concentrations at the air quality monitoring network of AMA were identified using the methodology presented in the draft guidance to member states on PM₁₀ monitoring (EC, 2002). This methodology, which was further developed and used by e.g. Escudero *et al.* (2005) and Querol *et al.* (2009), has been recently included in the guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe (EC, 2011). In particular, the PM₁₀ time series of concentrations at the AMA network stations were examined, high PM₁₀ levels (peaks) at each station were highlighted and a list of coincident peaks was created. According to the methodology, the time series of simultaneous peaks should be compared with information on PM₁₀ concentrations from a rural/remote/EMEP area (used as a reference series) or a regional background station close to each monitoring site. In this study, the PM₁₀ concentrations at the THR background suburban station were considered as the reference series (the reasons will be discussed in the section following). Grivas *et al.* (2004) have shown that THR station can be considered as regional background location to the city of Athens due to the absence of primary anthropogenic PM sources. Thus, periods with possible desert dust influence are highlighted for each station.

The occurrence of desert dust events during the highlighted periods is confirmed by combining the above data with information on atmospheric circulation, meteorological conditions and satellite imagery derived from several tools. More specifically, Aerosol Index (AI) maps from OMI (Ozone Monitoring Instrument) measurements were collected and examined for the period October 2004–September 2010. The AI maps provide a measure of absorption of UV radiation by smoke, desert dust, and volcanic ash. In order to detect African dust events produced at surface level (not

detected by OMI) also daily images for the Mediterranean basin were obtained from the Sea-viewing Wide Field-of-view Sensor (National Aeronautics and Space Administration SeaWiFS Project). Moreover, maps on the daily dust load, the surface dust concentration and the vertical profile over Europe and the Mediterranean were collected as images from the BSC-DREAM8b (Dust REgional Atmospheric Model) model, operated by the Barcelona Supercomputing Center. The model predicts the atmospheric life cycle of the eroded desert dust. The efficiency of the model to predict significant dust outbreaks ($> 12 \mu\text{g}/\text{m}^3$) near the surface in the Eastern Mediterranean has been recently assessed (Papanastasiou *et al.*, 2010). Specifically during the period 2001–2007 the model predicted approximately 75% of significant dust outbreaks at THR station.

Additionally, air mass trajectories were obtained using the HYSPLIT4 Model (HYbrid Single-Particle Lagrangian Integrated Trajectory), developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (NOAA) (Draxler and Rolph, 2012). The 1 month meteorological data files were retrieved from NOAA (National Centers for Atmospheric Prediction National Center for Atmospheric Research; NCEP/NCAR data) and have a time resolution of 6 hours and spatial resolution of 2.5 degree latitude-longitude. The 3-dimensional trajectories (isentropic) were computed for the coordinates of each station of the AMA monitoring network (at 500 m, 1000 m and 4000 m above sea level), at 00, 06, 12 and 18 UTC (Coordinated Universal Time). The trajectories were 120 h (5 days backwards transport period) long. The three altitudes correspond to transport at surface, boundary layer height and free troposphere and are appropriate for identifying Saharan dust transport events over the Eastern Mediterranean, Greece and the greater area of Athens (Hamonou *et al.*, 1999; Alpert *et al.*, 2004; Papayannis *et al.*, 2005; Papayannis *et al.*, 2009). Also 3D trajectory data available from the Chemical Co-ordinating Centre of EMEP (EMEP/CCC) calculated using Flextra trajectory model every 6 h were retrieved. Each plot shows three trajectories at different arrival heights (500 m, 1000 m and 1500 m above sea level) calculated using the Flextra model with meteorological data provided from ECMWF (European Centre for Medium Range Weather Forecast). The conclusions drawn from the analysis of the back-trajectories were validated by inspecting synoptic meteorological charts at sea level pressure and at 850 mb geopotential height available every 6 h from the NCEP/NCAR Reanalysis Data Composites.

It must be mentioned that in certain cases of African dust incursions, i.e., when the regional air masses are not renewed due to the atmospheric conditions or intrusions are developed vertically, the African dust has an increased residence time and PM_{10} levels can be still affected 1 or 2 days following the last day of the episode (EC, 2011).

The above information was combined to identify the potential influence of African dust intrusions on the PM_{10} levels in AMA.

Quantification of the Natural Dust Load Contribution to Exceedances

The daily African PM load during dust outbreaks was calculated with the methodology presented by Escudero *et al.* (2007). According to this methodology the daily background levels can be obtained by applying a monthly moving 30th or other percentile to the PM_{10} time series of the regional background station representative of an area after a prior extraction of the days with African dust influence. Then, the daily net dust load is obtained by subtracting the calculated background value from the measured PM_{10} concentration in the regional background station for every day affected by the African outbreak. So, when the PM_{10} daily limit value is exceeded at an urban or suburban station during an African dust outbreak, the calculated African dust load in the relevant regional background station can be used to evaluate the impact of the natural contribution to that exceedance (Escudero *et al.*, 2007). In particular, the net dust load at the regional background station for that day is subtracted from the measured PM_{10} concentration at the urban or suburban station and if the result is lower than the limit value then the exceedance is attributed to the African dust contribution. In some cases the net dust load can be larger than the PM_{10} concentration recorded at the urban and suburban stations on days affected by African dust incursions (thus the result of the subtraction is a negative value). According to Escudero *et al.* (2007) this is most likely attributed to the difference in altitudes between the reference regional background station and the monitoring network stations and can be resolved with the use, as reference, of another background station in the vicinity of the monitoring network at approximately the same altitude. However, when the result is still a negative value, it is replaced by the monthly moving 40th percentile calculated from the PM_{10} series at the urban or suburban site after extraction of the days with African dust events (daily value without dust influence).

In the case of the AMA monitoring network, data on the regional background levels of PM_{10} were available at the rural background station Aliartos, at which however PM_{10} monitoring began only since 29/7/2010. Therefore, the comparison of simultaneous series of PM_{10} registered concentrations and the calculation of background levels for the AMA monitoring network could be performed only for the period 29/7/2010–31/12/2010. Hence, only data from the THR background suburban station were used as reference values for the AMA network stations. A previously mentioned, Grivas *et al.* (2004) have shown that THR station can be considered as regional background location to the city of Athens. To verify this assumption, the identified African dust events and the calculated dust load at THR station were compared with those at Aliartos station during the period with concurrent measurements (see Results and Discussion). The net dust load was calculated using the 40th percentile of PM_{10} concentrations after extraction of the days influenced by African dust and other natural pollution events (fire and volcanic ash in this case). The 40th percentile was used as a more conservative value among different percentiles ranging from the 20th to the 40th to obtain the background concentration levels. The 40th percentile is also the suggested value in the EC guidelines (EC, 2011).

It was assumed that when the net dust load exceeded the average daily value of PM₁₀ concentration registered at a specific station on a day affected by African dust, then the exceedance could be attributed to the dust event. The daily value without dust influence was set equal to the 40th percentile of the PM₁₀ concentration calculated for this specific station after extraction of the African dust and other natural pollution events (fire and volcanic ash in this case).

It must be noted that exceedances of the AQS attributed to large fires (> 50 ha) occurring in the area alone or coinciding with African dust events were treated separately while forest fires can contribute significantly to air pollution in Greece (Lazaridis *et al.*, 2008). Data on the location, duration and burnt area of fires at natural vegetation areas and landfills were obtained from the Fire service department (Aleksandropoulou *et al.*, 2012b). Additionally, some exceedances attributed to the Eyjafjallajökull eruptions during April and May 2010 or to the synergetic effect from African dust and volcanic ash intrusions were treated separately (Mamouri *et al.*, 2010).

RESULTS AND DISCUSSION

African Dust Load

The African dust load used in the analysis was calculated using THR station as reference for the regional background concentration in AMA. The results as regards African dust events and net dust load at THR station were compared with those at Aliartos station during the period with concurrent measurements (1/8/2010–31/12/2010). For the Aliartos station, the daily background levels were obtained by applying the 30th percentile to monthly moving average of PM₁₀ concentrations after a prior extraction of the days with African dust influence. The 30th percentile, adopted only in this estimation, has been previously used by Querol *et al.* (2009) for rural background stations in the Mediterranean Basin. Thus, the background levels and the corresponding African dust load during days with exceedances at Aliartos station were calculated at $8.4 \pm 1.7 \mu\text{g}/\text{m}^3$ and $29.9 \pm 23.0 \mu\text{g}/\text{m}^3$, respectively. The daily AQS after the extraction of the influence of dust events during the period 1/8/2010–31/12/2010 at the AMA monitoring network stations was exceeded in 40, 20, 28, 20, 12, 5 and 13 days at ARI, MAR, PIR-1, LYK, THR, AGP and KOR stations, respectively. On the other hand, the average African dust load at THR station (calculated using the 40th percentile as mentioned in the methodology section) during days with exceedances was calculated at $42.8 \pm 19.2 \mu\text{g}/\text{m}^3$ during the period with concurrent measurements with Aliartos station. The exceedances of the daily AQS without dust events were calculated at 30, 15, 23, 20, 9, 4 and 10 times at ARI, MAR, PIR-1, LYK, THR, AGP and KOR stations, respectively. The value of African dust is much higher at THR station compared to ALI, most likely due to the difference in PM sources and altitude of the two stations. However, THR is the only station in the area with measurements throughout the studied period which can be used as regional background (Grivas *et al.*, 2004).

The calculated average annual African dust load at THR

station varied from $18 \pm 12.5 \mu\text{g}/\text{m}^3$ in 2007 to $48.8 \pm 32.8 \mu\text{g}/\text{m}^3$ in 2010. The African dust load at THR was enhanced during the spring and lower during the autumn period. The maximum monthly average has been observed during April ($39.2 \pm 48.1 \mu\text{g}/\text{m}^3$) and the minimum during December ($23.7 \pm 19.8 \mu\text{g}/\text{m}^3$). The maximum daily value of $429.51 \mu\text{g}/\text{m}^3$ has also been registered during spring.

The contribution from African dust load to PM₁₀ concentrations on days with Saharan dust event at stations with exceedances of the daily AQS ranged from 31.4% at LYK station in 2007 to 89.2% at THR station in 2006. More specifically, the average contribution of African dust to traffic urban stations was $49.5 \pm 5.8\%$ whereas at background suburban stations it was $72.2 \pm 7.3\%$ during the Sahara dust events of the period 2001–2010. The results are in good agreement with those of previously published works which report dust contributions to PM₁₀ in the range of 60–72% at rural and urban background sites during dust events (Gerasopoulos *et al.*, 2006; Kocak *et al.*, 2007; Koulouri *et al.*, 2008; Remoundaki *et al.*, 2011). In particular, Remoundaki *et al.* (2011) which examined the elemental composition of PM₁₀ over Athens during the period 27/3/2009–3/4/2009 found that the contribution of minerals to PM₁₀ concentrations increased from 13–19% before the Sahara dust event to 79% on the 1st April when the cent peak was occurred. The average contribution of Saharan dust to PM₁₀ during the period 30/3/2009–2/4/2009 in our calculations was approximately 61%. The overall contribution from African dust on PM₁₀ concentration in AMA during the period 2001–2010 is discussed later in the manuscript.

Contribution from African Dust Load to the PM₁₀ Concentrations

The characteristics of the average annual concentration of PM₁₀ in the AMA during the period 2001–2010 at traffic urban and suburban background stations are depicted in Figs. 1(a)–1(h). The left side of the boxplots for each year shows the descriptive statistics from the whole dataset (All) whereas the right side depicts the results after prior extraction of the African dust load on days with dust intrusions (No). Each box depicts the lower, the median and the upper quartile (bottom: 25th, band: 50th and top: 75th percentile) of all the PM₁₀ concentration data for each year, whereas the point and the whiskers show the average, minimum and maximum values. It is observed that the average annual concentration of PM₁₀ during the monitoring period at each station shows fluctuations however it has decreased from the beginning to the end of the monitoring period at most of the stations besides THR. This is probably associated with the decrease in primary PM emissions and air pollutants precursor to PM in AMA during the period 2001–2010 (Progiou and Ziomas, 2011; Aleksandropoulou *et al.*, 2012c, d) as well as the absence of primary anthropogenic PM sources at THR station (Grivas *et al.*, 2004). In particular, the emissions from road traffic have decreased and their trend has been correlated with measured concentrations over AMA (Progiou and Ziomas, 2011) whereas PM_{2.5–10} and air pollutants emissions have decreased since 2001 (Aleksandropoulou *et al.*, 2012c, d).

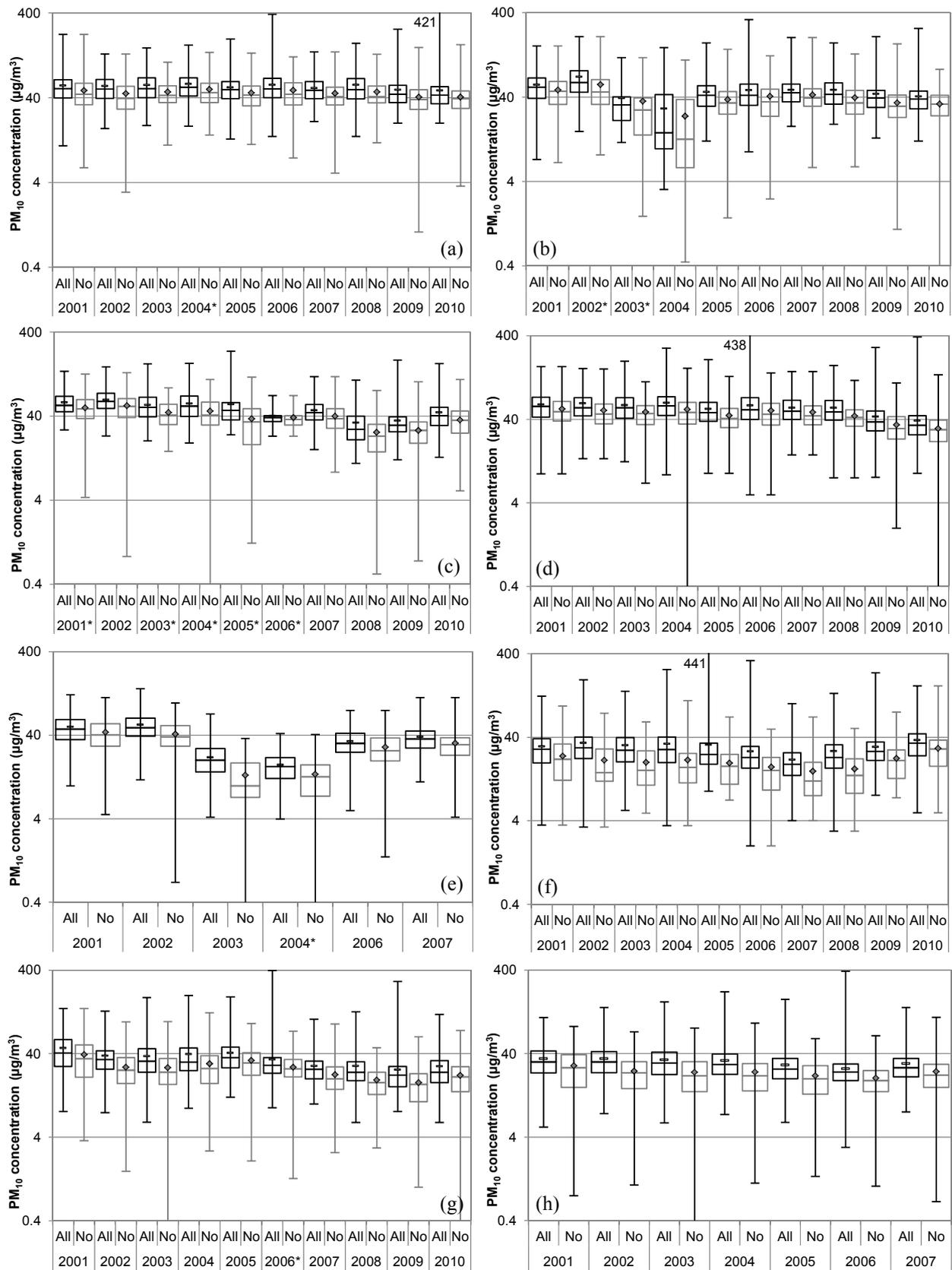


Fig. 1. Box plots of PM₁₀ concentrations at the traffic urban stations (a) ARI , (b) MAR, (c) PIR-1, (d) LYK, (e) GOU and the background suburban stations (f) THR, (g) AGP and (h) ZOG in AMA before (black) and after (grey) the extraction of the African dust load on days with dust intrusion and no large forest fire events. Data flagged with * correspond to years with data availability less than 50%.

The average annual PM₁₀ concentrations at the THR station are the lowest in the AMA network almost every year. The decadal average concentration of PM₁₀ is 30.5 ± 4.1 µg/m³ which is the lowest compared to other stations where the decadal average reached values up to 54.7 ± 7.6 µg/m³ (LYK station; standard deviation reflects variations in annual averaged values). The annual AQS was exceeded in 2010 at few stations, in particular the TU stations MAR (40.6 ± 19.5 µg/m³), PIR-1 (44.1 ± 20.2 µg/m³) and ARI (48.6 ± 29.7 µg/m³).

The interquartile range shown in the boxplots is a measure of the variability in PM₁₀ concentrations (not so clear due to the axis' lognormal scale; years with data availability less than 50% are not examined). Likewise, the variability in annual concentrations of PM₁₀ during the monitoring period at each station shows fluctuations however it has decreased from the beginning to the end of the monitoring period (e.g., ARI: from 26.4 µg/m³ in 2000 to 18.1 µg/m³ in 2010; MAR: from 28.5 µg/m³ in 2000 to 18 µg/m³ in 2010). The larger dispersion in PM₁₀ concentrations was observed in 2004 at MAR station, whereas the smaller at ZOG station in 2006. The data availability at MAR during 2004 was approximately 71% (<20% for August and no data for September) and at ZOG during 2006 was 87% (no data during April). Also, the highest fluctuations were observed at THR station most probably because of the distance of the station from anthropogenic emissions (Grivas *et al.*, 2004), whereas the lowest were found at ARI, PIR-1 and GOU stations due to their proximity to traffic and other urban sources of PM₁₀ emissions. It is also observed in Figs. 1(a)–1(h) that the median values of PM₁₀ concentration are always lower than the annual average values. This positive skewness in the distributions of PM₁₀ concentrations is an indication of the occurrence of extreme events.

The annual maximum PM₁₀ concentrations observed during the period 2001–2010 were in the range of 42 µg/m³ (GOU 2003) to 441 µg/m³ (THR 2005). Both of the above concentrations were registered during African dust events, as also applies for most of the annual maximum PM₁₀ concentrations. During periods with African dust events PM₁₀ concentrations were found higher than the average annual concentrations by 1.4 µg/m³ and up to 30 µg/m³. The maximum concentrations during African dust episodes were observed in 2006 and in 2010. In particular, on the 24/2/2006 occurred the most extreme Sahara dust event of the period 2001–2010 and all the monitoring stations recorded PM₁₀ concentration values in the range 331–438 µg/m³ while the African dust load at THR station was calculated at 321.8 µg/m³. The second most intense Saharan event occurred in 2010 during the period 17–21 February with peak on 20/2/2010 when the recorded PM₁₀ concentration values were in the range 260–421 µg/m³. The corresponding PM_{2.5} concentrations during the period 17–20 February were found as high as 100 µg/m³ (Remoundaki *et al.*, 2013). However, some of the annual peaks observed in PM₁₀ concentrations were attributed to forest fires (ARI 2007, PIR-1 2006, LYK 2001 and 2007, GOU 2007) or other sources mainly at the traffic urban areas (most probably anthropogenic pollution; ARI 2001–2002, MAR 2001–

2003 and 2007, PIR-1 2007, AGP 2001, GOU 2006).

The right side boxplots in Figs. 1(a)–1(h) reflect the descriptive statistics for the PM₁₀ concentrations after prior extraction of the African dust load on days with dust intrusion from the dataset. Therefore, the mean annual contribution of African dust to PM₁₀ concentrations in AMA is the difference between the mean values before and after the extraction of the African dust load on days with dust intrusion. It must be noted however, that the above difference does not include episodes of African dust events concurrent with large forest fires. The new decadal average concentration of PM₁₀ at THR station is 20.7 ± 3.8 µg/m³ whereas at other stations it reaches values up to 45.8 ± 7.2 µg/m³ (LYK station) and 45.8 ± 3.0 µg/m³ (ARI station). The annual AQS is still exceeded in 2010 at the TU station ARI. Likewise, the larger dispersion in PM₁₀ concentrations was observed in 2001 at ARI station, whereas the smaller at GOU station in 2003. Still there is a positive skewness in the distributions of PM₁₀ concentrations indicating the occurrence of extreme events not accounted for in the analysis. It is observed that the annually maximum PM₁₀ concentrations are lower after the extraction of African dust contributions and up to 223 µg/m³ (ARI 2001).

Additionally, the contribution to PM₁₀ concentrations from episodes of African dust concurrent to large forest fires, large forest fires and volcanic emissions, was extracted from the PM₁₀ concentrations dataset. The new decadal average concentration of PM₁₀ at THR station is 20.3 ± 3.9 µg/m³ whereas at other stations it reaches values up to 45.3 ± 3.1 µg/m³ (ARI station). The annual AQS is still exceeded in 2010 only at the TU station ARI. Moreover, a positive skewness, however smaller than that in the original dataset, was found in the distributions of PM₁₀ concentrations for most of the stations.

The average contribution of African dust to annual PM₁₀ concentrations registered at the AMA network stations was 8.6 ± 1.8 µg/m³ (i.e., 22.2 ± 7.5%; TU: 14.4 ± 5.5% and BS: 27.3 ± 6.7%), whereas the overall contribution from African dust load and forest fires occurring either alone or simultaneously with Saharan dust events (and/or volcanic emissions) was 8.8 ± 1.6 µg/m³ (i.e., 22.6 ± 7.2%). It was found that the contribution from all the above sources ranged from 5.2 µg/m³ at GOU station in 2006 to 12.8 µg/m³ at THR station in 2002. Specifically, during the period 2003–2006 the average dust contribution to PM₁₀ concentrations at THR ranged from 9.5–12.6 µg/m³. However, with regards only to days with exceedances of the daily AQS the average dust contribution during the above period ranged from 3.8–8.1 µg/m³. The results quite agree with values calculated at THR station using the SKIRON modeling system (values ranged from 2.4 to 6.4 µg/m³; Mitsakou *et al.*, 2008). In addition, Querol *et al.* (2009) found that during the period September 2004–December 2006 the dust load at Finokalia station in Southern Greece (Crete) was 10 µg/m³ which is close to the results found in this study for AMA (10.8 µg/m³).

Interannually the contribution from African dust to PM₁₀ concentrations showed significant peaks during the period 2002–2004 and in 2008 at all stations and lows in 2001, 2007 and 2010. During the periods 2002–2004 and 2007–

2008 the contribution from Sahara dust events to PM₁₀ concentrations over AMA was more frequent than during the rest of the period. In particular, the probability for an event to affect the concentrations measured at AMA network stations was 33.6% during 2001–2010 with the annual probability ranging from 18.6% in 2010 to 43% in 2002. During the years 2002–2004 and 2007–2008, the probability for an event to influence PM₁₀ concentrations in AMA was over 38% (period average 40.6%) compared to less than 30% (period average 26.6%) during the rest of the period. Moreover, the intensity of episodes varied significantly among years.

The averaged intensity of African dust events during the period 2001–2010 over the AMA network of stations is depicted in Fig. 2(a). It is observed that most of the events contribute to the daily PM₁₀ concentrations in AMA about 5 µg/m³ to 34 µg/m³. The probability for an extremely severe event (contribution > 100 µg/m³) to occur during the studied period was 1.72%. The cumulative distributions of African dust contributions to daily PM₁₀ concentrations in AMA separately for each year are depicted in Fig. 2(a). It is apparent from the Figure that more than 50% of the events contribute to PM₁₀ concentrations by not more than 24 µg/m³, with the exception of 2010 when the distribution is shifted towards higher contributions. Opposite from the cumulative distribution for 2010 is the distribution for 2007 which is shifted towards lower contributions. Additionally, the more extreme events occurred during 2009 (4.4%) and 2005 (3.7%). Hence there is no clear interannual relationship between the average contribution of African dust to PM₁₀ concentrations with the frequency and intensity of events.

Nevertheless, by examining the seasonal variability in contributions (depicted in Fig. 2(b)) it was found that the average monthly African dust contribution and the probability of occurrence for an episode have the same trend (linear relationship with Pearson *r*: 0.91). The contribution from African dust events is more enhanced during February–May and November (more than 20%, 9 µg/m³ approximately) and less during July–October and December–January (10–18%; 4.2–6.6 µg/m³ and 7.2 µg/m³ in September). Specifically, during April and May the contribution is higher than 12.5 µg/m³ thus more than 30% of the recorded PM₁₀ concentrations. In particular, during February, September, November and December the meteorological conditions favor the transport of air masses loaded with dust from north Africa over the Eastern Mediterranean (from maps of mean sea level pressure at surface over the Mediterranean), explaining the three peaks in the series of African dust contribution. Moreover, during the period September 2005–August 2006 very high levels of PM₁₀ and enhanced levels of crustal elements in PM₁₀ were recorded during February and April due to dust transport from North Africa at GOU and LYK stations (Theodosi *et al.*, 2011).

The seasonal variability of contributions is also depicted in Fig. 2(c) together with the intensity of events. It is observed that more frequent (39.5%) and more severe events (1.1% over 100 µg/m³; 4.3% over 50 µg/m³; percentages based on all the events) occur during the spring (i.e., 2.8% over 100 µg/m³ and 10.9% over 50 µg/m³). The results on the

seasonal variability of occurrence of African dust events agree with previously published research. In particular, Papayannis *et al.* (2008) found that the seasonal pattern of dust transport toward Europe is enhanced in the central and eastern Mediterranean during spring whereas Kallos *et al.* (1998) and Rodriguez *et al.* (2001) have shown that intrusions of Sahara dust in the region mainly occur during the winter and spring. Additionally, Remoundaki *et al.* (2013) examined the PM_{2.5} concentrations over Athens during 2010 and found the maximum, and with the most frequent abrupt changes, concentrations of elements which are typical constituents of dust (Si, Al, Ca, Fe, K, Mg, Ti) during spring and autumn. Extreme Sahara outbreaks occurred also during winter however their effect on PM₁₀ concentrations in AMA is for approximately 53% of the events less than 19 µg/m³. During the winter, and also the early spring period, the dust load over the eastern Mediterranean and Greece is often attenuated by precipitation (Nastos, 2012). During the summer season the dry meteorological conditions favor the occurrence of dust transport events over the area (Katragkou *et al.*, 2009). In this study the results indicate that dust events were less frequent in summer (~17.5%) and less intense (more than 37% less than 10 µg/m³, ~56% at 20–50 µg/m³; Fig. 2(c)). Overall, in summer months, as observed in Fig. 2(b), the least contribution from African dust is expected (10% in July, ~4 µg/m³, probability less than 6%; 13% in August, ~5.5 µg/m³, probability less than 6%). In autumn, Saharan dust outbreaks are more frequent (23%) however they have low impact on PM₁₀ concentration (45.8% less than 10 µg/m³; ~45.6% at 20–50 µg/m³) and only 8.6% over 50 µg/m³, most of which occur in September and November (more severe in November). The occurrence of significant dust outbreaks (contribution > 12 µg/m³) during the autumn has been also shown by Papanastasiou *et al.* (2010).

Finally, in Fig. 2(b) are also depicted the contributions from large forest fires to PM₁₀ concentrations and from African dust and large forest fires smoke from concurrent events during the studied period. The dry season dust events have been often associated with smoke plumes from forest fires over the Mediterranean (Lazaridis *et al.*, 2008). It is apparent in Fig. 2(b) the exacerbation of large forest fire events during the summer and their impact to air quality in AMA. Their contribution to average monthly PM₁₀ concentrations over AMA ranged from 2×10^{-3} µg/m³, in September, to 1.72 µg/m³, in June. The higher contributions were found in 2006 at PIR-1 station and at all stations in 2001 and 2007 when forest fire events large in intensity and duration occurred (Fire service department, 2011; Aleksandropoulou *et al.*, 2012b).

Contribution from African Dust Intrusions to the Exceedances of Daily AQS for PM₁₀ in AMA during 2001–2010

The contribution from African dust events occurring alone or concurrently with large forest fires and/or volcanic dust intrusions to the exceedances of the daily AQS during the period 2001–2010 at AMA network stations has been also examined. In Fig. 3(a) are given the exceedances of the daily AQS during the period 2001–2010 at each station

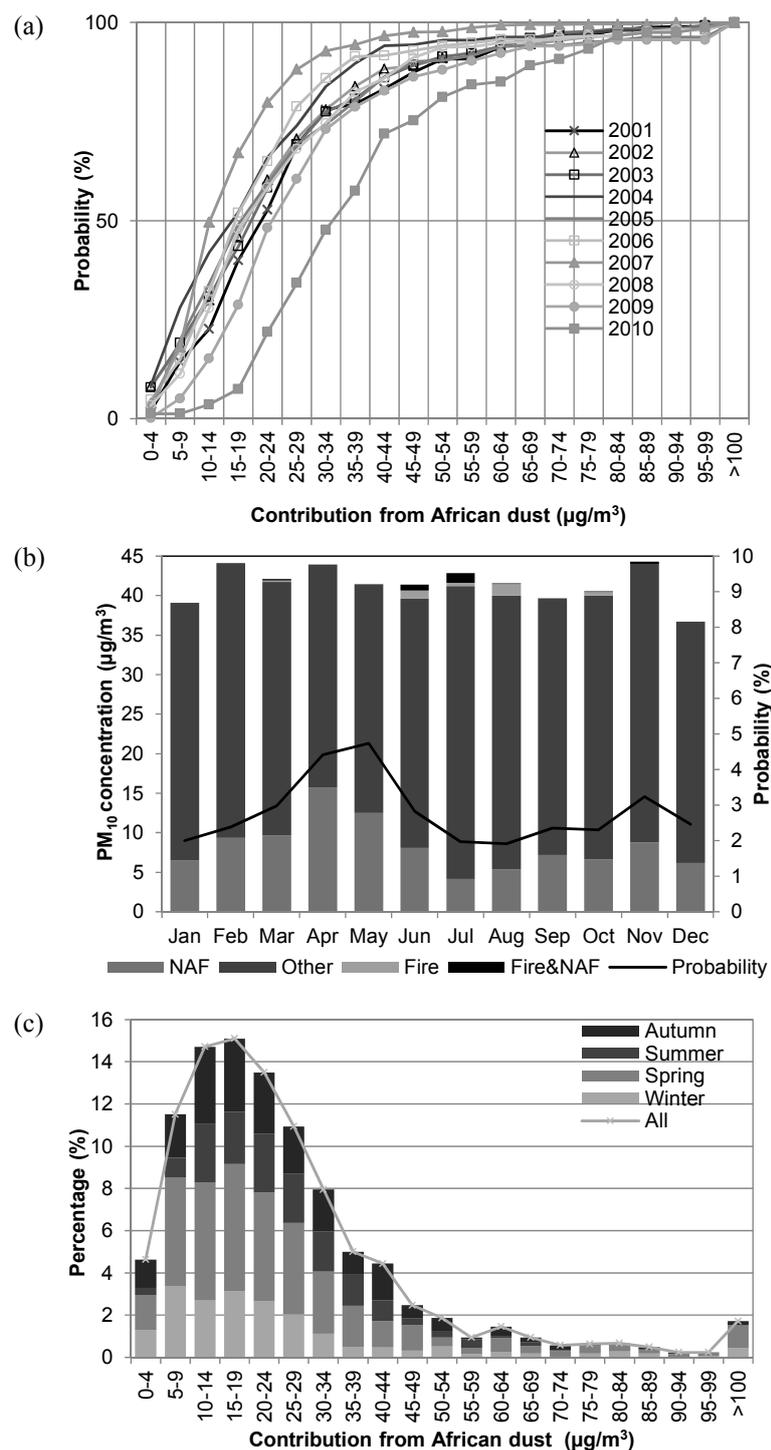


Fig. 2. (a) Annual cumulative distributions of intensity of African dust contribution to the daily PM₁₀ concentrations over AMA; (b) Average monthly PM₁₀ concentrations over AMA during the period 2001–2010 together with probability of occurrence of African dust event. Concentrations are disaggregated in contributions from Saharan dust events (NAF), large fire events, large fire events concurrent with Sahara dust intrusions and other sources; (c) Distribution of intensity of seasonal African dust contribution to the daily PM₁₀ concentrations over AMA.

classified as cases of not attainment to the daily AQS attributed to African dust (NAF), concurrent African dust and smoke from forest fire events, smoke from large fires and other sources (anthropogenic, long range and other natural). At cases attributed to large forest fires are also

included three days in 2010 with volcanic ash intrusion. The daily AQS for PM₁₀ concentrations was constantly exceeded through the period 2001–2010 at the ARI, MAR, PIR-1 (except for 2009) and LYK stations. In particular, at the BS ZOF, AGP, and THR the daily AQS was exceeded

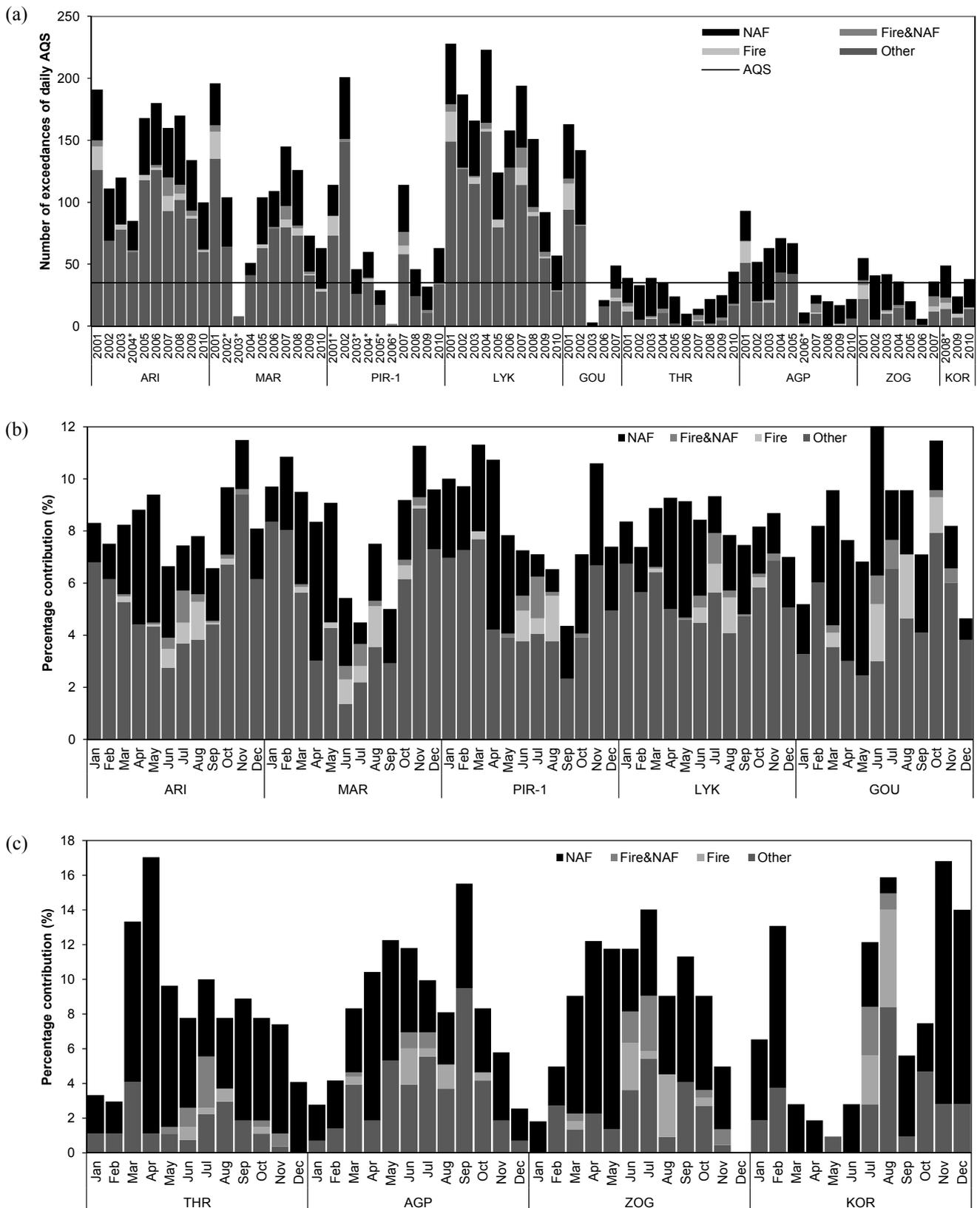


Fig. 3. Number of cases of not attainment to the AQ5 in AMA during the period 2001–2010. (a) Interannual variation at traffic urban and background suburban stations; (b) Monthly variation at traffic urban stations; (c) Monthly variation at background suburban stations. The cases are attributed to Saharan dust events (NAF), large fire events, large fire events concurrent with Sahara dust intrusions and other sources. Data flagged with * correspond to years with data availability less than 50%.

in the past however during the last years of the studied period the daily PM₁₀ concentrations are lower than the AQS. During 2010 a slight increase of average daily PM₁₀ concentrations was observed and the daily AQS was exceeded at the THR and KOR stations.

After the extraction of the cases of not attainment to the AQS attributed to African dust outbreaks it is observed in Fig. 3(a) that the number of the exceedances at BS stations is compliant with the AQS (except for AGP in 2004, 2005). On the other hand, at TU stations the extraction of cases attributed to African dust from the dataset is not enough to meet the AQS, except for 2010 at all stations besides ARI. In 2010, the daily PM₁₀ concentrations have dropped dramatically compared to the rest of the studied period at all stations. The contribution from African dust to exceedances of the daily AQS ranged from 18% at MAR in 2001 to 100% at THR in 2006. At TU stations the average annual contribution was approximately 36%, whereas at BS stations 58% (30–78%). The results agree with the results from Kallos *et al.* (2007) which indicated that 30–70% from violations of the AQS in Southern Europe occurs due to Sahara dust transport. Also, according to another analysis of PM records in Athens during the period 2001–2004, approximately at 60–70% of the violation cases the synergetic contribution of anthropogenic (urban and long range transport) and natural (Sahara dust) sources is evident (Astitha *et al.*, 2006).

The seasonal variation in the number of exceedances of the current daily AQS is depicted in Figs. 3(b)–(c). Most cases of non-attainment to AQS occur during the cold period (October–March) at TU stations and during the warm season at BS stations. Exceptions from the above trend are KOR station with enhanced number of violations during winter and GOU and LYK with almost equal contributions throughout the year. At BS stations the seasonal variation is determined by the long range transported aerosols and natural sources (except for African dust) which are exacerbated during the warm season (i.e., secondary aerosols) while African dust events and forest fires enhance this pattern. At TU stations the anthropogenic pollution (mainly space heating and traffic; Kallos *et al.*, 1998; Aleksandropoulou *et al.*, 2011) is the main source of violations of the AQS and determines the seasonal variation in exceedances. Violations of the AQS due to African dust outbreaks add to the above pattern during spring and autumn. The contribution from exceedances due to forest fire or forest fires concurrent with African dust events to the overall number of violations is low and occurs during the dry season (4–15%).

CONCLUSIONS

The objective of this study was to identify influences from natural particulate inputs produced by Sahara air mass intrusions on PM₁₀ concentrations over Athens during the decade 2001–2010. The aim was to quantify the contribution from African dust outbreaks to the annual and daily PM₁₀ concentrations over AMA in order to deduct it from the calculations for compliance of the EU Directive on air quality. The results indicated that at BS stations the African desert dust intrusions and events concurrent with large

forest fires are responsible for the exceedances of the AQS during most of the period studied. On the other hand, at TU stations is still difficult to comply with the EU AQS even after the subtraction of the African dust load from the average annual and daily PM₁₀ concentrations, indicating that emission abatement measures should be taken to improve the air quality in the Athens metropolitan area.

During 2010 due to the economic shrinking and probably the update of the fleet with new cleaner vehicles (incentives were given for the disposal and replacement of old vehicles) emissions of primary PM and air pollutants precursor to PM have dropped significantly. Consequently, the PM₁₀ concentrations remained low compared to previous years and even at TU stations (except ARI) the African dust intrusions can be considered responsible for non-compliance to the AQS.

ACKNOWLEDGMENTS

This work was supported by the European Union's LIFE Programme under grant LIFE 09 ENV/GR/000289.

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Received for review, December 21, 2012

Accepted, March 28, 2013