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Spatial and temporal (short and long-term) variability of submicron, fine and sub-10 μm particulate matter (PM_1, $PM_{2.5}$, PM_{10}) in Cyprus

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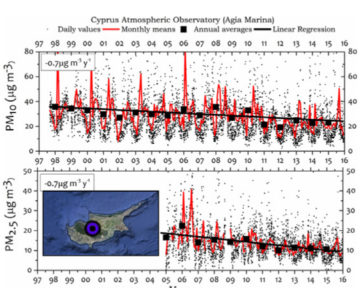
GRAPHICAL ABSTRACT



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ABSTRACT

Long-term particulate matter (PM) mass concentration measurements have been performed in Cyprus at three major cities, one industrial area and two remote stations covering the entire southern part of the island in an effort to assess; i) the spatial and temporal variability of sub-10 μm (PM_{10}), fine ($PM_{2.5}$) and submicron (PM_1) particulate matter in the eastern Mediterranean, ii) the main source areas contributing to their levels and iii) the relative contribution of regional and local anthropogenic and natural sources to PM levels. It was found that dust is responsible for the 33.6 ± 5.2% or about 10 $\mu g\,m^{-3}$ of the annual PM_{10} levels reported in background stations; the latter underlines the significant contribution of natural sources on the ambient PM_{10} amounts in the eastern Mediterranean region. A significant ($p < 0.001$) decreasing trend of 0.7 ± 0.1 $\mu g\,m^{-3}\,y^{-1}$ was observed when both PM_{10} and $PM_{2.5}$ annual values are considered, indicating contribution from both natural and anthropogenic sources to this tendency. By considering the PM_x (with x = 1, 2.5 and 10) mass concentrations obtained at the background station of Agia Marina as representative of the regional influence, the local influence of the urban and industrial sites on the measured PM_x levels can be estimated. On average, 36–44% of the observed PM_{10} levels at the urban and industrial locations is estimated to originate from local anthropogenic and/or natural emissions including vehicle, biomass-burning, shipping emissions (in Limassol), airport related emissions (in Larnaca), resuspension of dust and sea-salt (in coastal locations). These local emissions are almost

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equally distributed in the fine and coarse fractions as 40–50% of the local PM_{10} amounts are due to fine particles emissions. The above results highlight significant emissions from both fine mode (e.g. residential heating and traffic) and coarse mode urban emissions (e.g. dust resuspension, wear and tear in brakes and tires, respectively) in urban and industrial locations in Cyprus.

1. Introduction

The amounts and properties of ambient particulate matter (PM) suspended in air over various environments has been at the forefront of ongoing environmental research studies. Aerosols are minute particles, whose diameters range from a few nm to hundreds of μm. They play a key climatic role by altering Earth's radiative budget (Baker and Peter, 2008; Leibensperger et al., 2012; Myhre et al., 2013) decrease visibility, act as a source of nutrients to continental and marine environments (Duce et al., 1991; Galloway, 2003) and finally they are associated with air pollution which affect, through respiration, human health (Lelieveld et al., 2015). The latter resulted in legislation concerning the PM's concentration levels. Initially, legislation focused on coarser particles with diameter smaller than 10 μm (PM_{10}) and later on, on particles with diameters smaller than 2.5 μm ($PM_{2.5}$; also referred as fine particles), because $PM_{2.5}$ can penetrate the lungs deeper than PM_{10} due to their smaller size.

Particle size largely dictates PM lifetime that spans from a few minutes to several days allowing long-range transport that often exceed national physical boundaries. Several studies have focused on apportioning between locally produced and regionally transported PM (Karagulian et al., 2015; Pikridas et al., 2013; Viana et al., 2008). In that respect, the Eastern Mediterranean basin is of specific interest because it lies on a crossroad of diverse air masses (Kanakidou et al., 2011; Ladstätter-Weißenmayer et al., 2007; Lelieveld et al., 2002) from anthropogenic and natural sources of Europe, Asia and N. Africa. An important, circulation pattern observed in the Eastern Mediterranean, is southerly winds caused by the temperature contrast between cold marine waters and the warm continental surfaces of N. Africa (Moulin et al., 1998). This pattern is typically observed during spring and early summer and is responsible for dust transport from arid areas in N. Africa (e.g. Saharan desert). Dust is also transported to the eastern Mediterranean from the Arabian desert located eastwardly. All the above dust intrusions are frequently associated with elevated PM_{10} concentrations above legislated standards (Escudero et al., 2006; Gerasopoulos et al., 2006; Mitsakou et al., 2008). The west side of the Mediterranean Sea is also impacted by dust transport but due to a different circulation pattern (Escudero et al., 2006). The net result, over the entire basin, is a decreasing gradient from east to west and from south to north with respect to PM_{10} (Querol et al., 2009b).

The importance of dust transport in the Mediterranean, which is often episodic (the so called dust events), has led the World Meteorological Organization (WMO) to establish the Sand and Dust Storm Warning Advisory and Assessment System (SDS-WAS) portal (http://www.wmo.int/pages/prog/arep/wwrp/new/Sand_and_Dust_Storm.html), where several transport models are used to forecast dust concentrations over the Mediterranean basin.

This work focuses on Cyprus, an island located on the east Mediterranean. The majority of the PM studies in the eastern Mediterranean reported PM_{10} measurements (e.g. Andreae, 2002; Gerasopoulos et al., 2006; Kallos et al., 2014; Karanasiou et al., 2009; Koçak et al., 2004; Kleanthous et al., 2009) and to a lesser extent $PM_{2.5}$ (e.g. Sarnat et al., 2010; Schneidemesser et al., 2010; Bardouki et al., 2003; Koulouri et al., 2008; Sciare et al., 2005; Remoundaki et al., 2013). Several studies in the Asian part of the east Mediterranean focus on both size fractions (Abdallah et al., 2018; Krasnov et al., 2014; Massoud et al., 2011; Saliba et al., 2010; Shahsavani et al., 2012). Measurements in areas of limited anthropogenic activity (remote/rural) in the region, reported elevated PM_{10} (Gerasopoulos et al., 2007, 2006; Koçak et al., 2007; Safar and Labib, 2010) and $PM_{2.5}$ levels (Koçak et al., 2007; Pikridas et al., 2012; Schneidemesser et al., 2010) compared to corresponding sites in central Europe (see Putaud et al., 2010). Studies involving particles with diameters smaller than 1 μm (PM_1) are sparse (Koçak et al., 2011). Regarding Cyprus, PM_{10} levels variability has been reported in only three works focusing on the capital of the island Nicosia (Achilleos et al., 2014; Kleanthous et al., 2009; Mouzourides et al., 2015). Mouzourides et al. (2015) reported kerbside and urban PM_{10} levels for two consequent 3-year periods in an effort to explain how ambient pollutants, including PM_{10}, are affected by local meteorology and anthropogenic activities. Dust events were also followed using the WMO portal discussed above. Achilleos et al. (2014) reported the contribution of dust events to the temporal PM_{10} cycle and based on PM_{10} measurements performed at a background site estimated the contribution of anthropogenic activities to the capital.

In this work, spatial and temporal variability of sub-10 μm, fine and submicron PM (PM_{10}, $PM_{2.5}$ and PM_1, respectively), are reported for Cyprus for three major cities, an industrial area and two remote stations covering the entire southern part of the island. To the best knowledge of the authors this is the first time that $PM_{2.5}$ and PM_1 levels are reported for Cyprus. The phenomenology of each size fraction is reported including annual trends, when multi-year long observations were available, along with diurnal and seasonal cycles. The effect of air mass origin on PM_{10} is examined and the contribution of anthropogenic emissions, with respect to $PM_{2.5}$ and PM_{10}, in urban areas are additionally discussed.

2. Monitoring sites

This work focuses on the Republic of Cyprus where four major cities are located; the capital Nicosia (400,000 inhabitants), Limassol (235,000 inhabitants), Larnaka (140,000 inhabitants) and Paphos (66,000 inhabitants, Fig. 1).

Apart from Nicosia that is placed inland, in between two mountain complexes, Troodos (1850m) to the southwest and Pentadaktylos (1020m) to the northeast, all other major cities are coastal. Limassol hosts the largest shipping port of the island and Larnaca the busiest airport. The only industrial area is located at the southeastern at Zygi

Fig. 1. The distribution of sites in Cyprus. Red, blue, yellow and white circles depict urban, background, industrial and free tropospheric stations, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(see Fig. 1), where a cement factory and a power plant are operating. Note that the west to east and north to south distances in the Republic of Cyprus are 250 km and 80 km respectively.

Background PM concentrations were primarily monitored at the Cyprus Atmospheric Observatory (CAO) at Agia Marina Xyliatou. CAO is a regional station from the global atmospheric watch (GAW) operating under ACTRIS, the European research infrastructure and the network of the "co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe" (EMEP). CAO is located at an altitude of 532 m a. s. l., 1 km south of an evergreen (pine) forest and inside sclerophylous vegetation, 1 km southerly from the village of Agia Marina (≈630 inhabitants) and 40 km from Nicosia (Fig. 1). Background-free PM concentrations have been also monitored at the Troodos Mountain site. The Troodos site is located near the top of the Troodos mountain, the highest mountain complex in the island, at 1819 m a.s.l., and is representative of the free troposphere of Cyprus.

PM observations were also conducted at the four major urban centers of the island, Nicosia, Larnaca, Limassol and Paphos. In all cities, PM were measured at kerbside stations located at 4 m from the ground and at least 25 m from a major route. In addition to the urban areas, the industrial area of Zygi was also monitored (Fig. 1).

The data reported in this work originate from the air quality network of Cyprus operated by the Department of Labour Inspection (http://www.airquality.dli.mlsi.gov.cy/). Because the observations of all sites did not start simultaneously, each site corresponds to a different time period and different size fraction, a summary of which can be found in Table 1. This study examines observations no later than January 1st, 2016. In our paper, each station will be referred with an acronym, consisting of six letters, the first three are based on the station's location (e.g. CAO station at Agia Marina Xyliatou is AGM) followed by a subscript of three more indicating the station's type (Table 1). In total, four station types have been used namely, kerbside (KRB), industrial (IND), remote/rural background (BKG) and free troposphere (FTP).

3. Methods

3.1. Air masses origin and classification

Source region analysis was conducted using the CAO station as receptor site (AGM_{BKG}) for the entire measurement period (1997–2015). The source region classification scheme is similar to that presented by Kleanthous et al. (2014) and Debevec et al. (2017) for the same receptor site and period and involves 6 sectors namely SW Asia, Middle East, Europe, Marine, N. Africa and Local shown in Fig. 2a. In this work, classification was performed using the particle dispersion model FLEXPART (Stohl et al., 2005) in backward mode. Retro-plumes, a replacement of the simple back trajectory, were calculated every 6 h and followed back in time for 5 days (Stohl et al., 2002). Air masses were categorized based on the potential emission sensitivity (PES) for the lowest 1 km. A PES value (ns kg^{-1}), over an area, indicates the mixing ratio at the receptor site if a source of 1 $kg\,m^{-3}\,s^{-1}$ was active at that area. Additional information about PES calculations can be found in Seibert and Frank (2004).

Initially, the average PES was calculated for the entire study period (Fig. 2b) which allowed visualization of the extent and strength of the major retro-plume paths based on which the above 6 source regions were determined (Fig. 2a). Following the classification method of Pikridas et al. (2010, 2012), a retro-plume was attributed to a region if that region exhibited PES values above a given threshold. For periods in which more than one source region had PES values above this threshold, the air mass was attributed to that having the larger area with PES value above that threshold. In this work the threshold value was equal to the 90^{th} percentile based on the distribution of all PES values calculated for the study period. In Fig. 2b, PES values are normalized by the geometric average PES (PES values follow a lognormal distribution), and therefore the relative contribution of each area to the receptor site is shown.

As an example, a particulate source of constant output throughout the year and located in Turkey is expected to increase the aerosol concentration at the receptor site by 1–2 orders of magnitude compared

Table 1
Information summary of the 7 monitoring sites in Cyprus presented in this study. The largest sampling period of PM_x for each station is highlighted bold.

Station - Type	Acronym	Coordinates	Height (m a.s.l.)	Method used	Period
Agia Marina-- *Regional Background*	AGM_{BKG}	35 02′ 17″ N – 33 03′ 28″ E	532	TEOM, Gravimetric	**PM_{10} TEOM: 1998–2015** $PM_{2.5}$ TEOM: 2010–2015 PM_{10} Grav: 2005–2015 $PM_{2.5}$ Grav: 2005–2010, 2015 $PM_{1.0}$ Grav: 2009–2010, 2013–2015
Nicosia *Kerbside*	NIC_{KRB}	(1998–2008) 35 18′ 10″ N – 33 21′ 17″ E (2009-today) 35 09′ 07″ N – 33 20′ 52″ E	176	TEOM, Gravimetric	**PM_{10} TEOM: 1998–2015** $PM_{2.5}$ TEOM: 2011–2015 PM_{10} Grav: 2005–2010, 2013–2015 $PM_{2.5}$ Grav: 2005–2008, 2013–2015 $PM_{1.0}$ Grav: 2013–2015
Larnaca *Kerbside*	LAR_{KRB}	35 54′ 60″ N – 33 37′ 39″ E	15	TEOM, Gravimetric	**PM_{10} TEOM: 2003–2015** $PM_{2.5}$ TEOM: 2010–2015 PM_{10} Grav: 2005–2015 $PM_{2.5}$ Grav: 2005–2008 $PM_{1.0}$ Grav: 2011
Limassol *Kerbside*	LIM_{KRB}	34 41′ 10″ N – 33 02′ 08″ E	19	TEOM, Gravimetric	PM_{10} TEOM: 2006–2015 **PM_{10} Grav: 2005–2015** $PM_{2.5}$ Grav: 2005–2008 $PM_{1.0}$ Grav: 2009–2011, 2013–2015
Paphos *Kerbside*	PAP_{KRB}	34 46′ 31″ N – 32 25′ 19″ E	75	TEOM, Gravimetric	PM_{10} TEOM: 2006–2015 **PM_{10} Grav: 2005–2015** $PM_{2.5}$ Grav: 2005–2009 $PM_{1.0}$ Grav: 2011, 2013–2015
Zygi *Industrial*	ZYG_{IND}	34 44′ 14″ N – 33 17′ 24″ E	9	TEOM, Gravimetric	**PM_{10} TEOM: 2003–2015** $PM_{2.5}$ TEOM: 2010–2015 PM_{10} Grav: 2011, 2013–2015 $PM_{2.5}$ Grav: 2011
Troodos *Free Troposphere*	TRO_{FTP}	34 56′ 36″ N – 32 51′ 50″ E	1819	Gravimetric	**PM_{10} Grav: 2012–2015** **$PM_{1.0}$ Grav: 2012–2015**

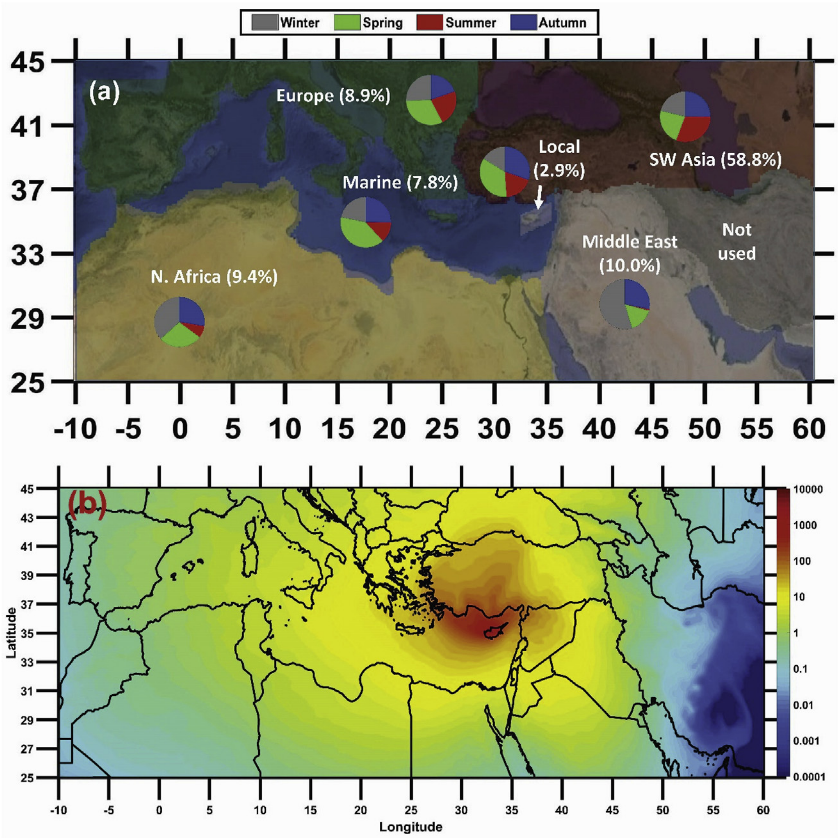

Fig. 2. Identified source regions (a) and normalized potential emission sensitivity (b) derived by FLEXPART retroplumes. The seasonal (pie charts) and annual distribution of air mass origin from each source region are shown in the top panel. In total 6 source regions were identified; N. Africa, Marine, Europe, SW Asia, Middle East and Local. An area of the map is not used because the orography prevents air masses to reach the receptor site (AGM_{BKG}). The main travel paths of the air masses on their way to the receptor site are also shown (b).

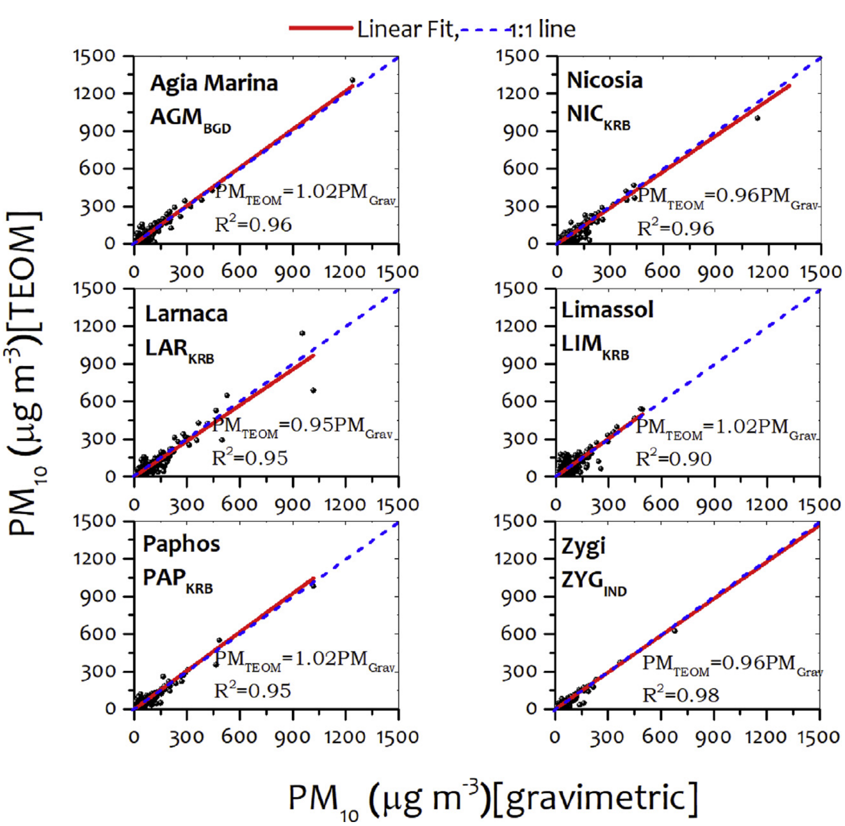

Fig. 3. Comparison of PM_{10} measurements obtained by a Tandem Elementary Oscilating Microbalance (TEOM) against gravimetric methods. A good agreement between the two methods is observed. The dashed blue line represents the 1:1 line and the red line the linear regression forced to 0. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

to the same source located in the north-east coast of Africa or Greece (Fig. 2b).

3.2. Instrumentation and data analysis

Tapered Element Oscillating Microballance measurements (TEOM): The majority of the measurements presented in this study have been conducted online using a TEOM with a 15 min resolution. The samples obtained by TEOM prior to 2006, were heated at 50 °C prior to entering the sampling chamber (R&P model 1400A). Afterwards, TEOM was replaced by a Filter Dynamics Measurement System (FDMS; Thermo model 1405DF), which incorporates additionally a Nafion dryer as part of the inlet system, at LAR_{KRB}, AGM_{BKG}, ZYG_{IND}, and NIC_{KRB}. During 2010, the remaining instruments using a heated inlet were replaced with those employing nafion dryer.

Gravimetric Analysis of Particulate Matter: Aerosol samples were also collected on pre-weighed filters (Whatman Cellulose 7194-004 till 2008; Whatman Quartz 1851-150 from 2008 till 2015; Pall Tissuquartz 2500 QAT-UP since 2015) using an autonomous filter sampler (Leckel SEQ 47/50 and Digitel DHA-80) for mass determination with daily resolution (from midnight to midnight according to local standard time). Particle mass on the filter substrates was determined gravimetrically before and after the sampling, under constant conditions dictated by protocol EN12341 with the use of a 6 digits precision analytical balance (Mettler Toledo, Model XP26C). According to that protocol filters were subjected to 50 ± 5% and 45–50% relative humidity at 20 ± 1 °C for 48 h, prior and after to 17 October 2014, respectively. Table 1 summarizes the method used at each site and the size fractions examined.

Intercomparion between TEOM and gravimetric PM_{10} concentrations: In five sites, AGM_{BGR}, NIC_{KRB}, LAR_{KRB}, PAP_{KRB} and ZYG_{IND}, PM_{10} was monitored using both methods, allowing an intercomparison, for a period of at least three years. Excellent correlation between daily averaged PM_{10} measured using the two independent methods (TEOM vs gravimetric) was observed at all sites with the calculated slopes being approximately equal to unity (0.95–1.02, Fig. 3).

This finding allowed us the consistent use of both datasets in the following analysis. Limited overlapping of gravimetric and TEOM fine mode data did not allow us to perform the same analysis for $PM_{2.5}$ except for the case of AGM and NIC where slopes of 0.83 and 0.82 were computed, respectively.

Trends and dust contribution estimation: Trends were estimated using the non-parametric Mann-Kendall method (Gilbert, 1987). Dust contribution was calculated following the method of Querol et al. (2009b) using back trajectory simulations. This method has been applied for the AGM_{BKG} site before and has been verified by the good correlation of the estimated dust load with the measured crustal concentration (Querol et al., 2009b).

4. Results and discussion

The following text presents collective information, for the first time to our knowledge, on the ambient amounts of sub-10 μm (PM_{10}), fine ($PM_{2.5}$) and submicron (PM_1) particles in Cyprus and on their temporal variability including multiannual, seasonal and diel behavior, wherever possible. Reported PM observations are based on the background and urban stations described before.

4.1. PM_1, $PM_{2.5}$ and PM_{10} concentrations in Cyprus and trend analysis

4.1.1. Background PM in Cyprus

Background-rural PM_{10}: The temporal variation of PM_{10} at AGM_{BKG} (regional background site) in the course of the 18 years (1998–2015) is examined in this work. Fig. 4 depicts all daily values (black dots), monthly means (red line), annual means (squares, Fig. 4 and blue bars, Fig. 5) together with the deseasonalized monthly means computed using the Brockwell and Davis. (2002) method.

A significant ($p < 0.001$) decreasing trend (black line) was observed on PM_{10} annual values. PM_{10} decreased by almost 0.7 ± 0.1 μg m^{-3} y^{-1} between 1998 and 2015 (Table 2), which is in agreement with the decrease reported in 5 rural sites in Europe during 1998–2010 that ranged between 0.1 and 1.3 μg m^{-3} y^{-1} (Barmpadimos et al., 2012).

Overall, the period under investigation (1998–2015) can be divided, into three sub-segments. Starting from 1998, PM_{10} decreased linearly ($R^2 = 0.96$) by 2.2 μg m^{-3} y^{-1} till 2002; then remained on average stable at 30 μg m^{-3} between 2003 and 2010. Similar trend that includes a strong decline in PM_{10} during 1990s followed by a plateau during the 2000s has been reported for the UK (Harrison et al., 2008). This plateau, with respect to PM_{10}, during the 2000s has been also recently reported in the majority of European sites (Guerreiro et al., 2014). During the last five years examined in this study (2011–2015), the annual PM_{10} averages dropped by 25% compared to the previous years, at 22.8 ± 2.1 μg m^{-3}. This drop was largely attributed to lower levels of regionally transported dust particles reaching Cyprus. Indeed calculated regional dust levels at AGM_{BKG} for the period 1998–2015 were equal to 9.7 ± 2.9 μg m^{-3}. On average, dust is responsible for the 33.6 ± 5.2% of the annual PM_{10} levels reported in AGM_{BGR}; the latter underlines the significant contribution of natural sources on the ambient PM_{10} amounts. If the complete 1998–2015 period is considered no significant trend in dust amounts is observed. However, this is misleading as two opposing sub-trends in dust are identified; an annual increase of 0.4 μg m^{-3} starting from 1998, the beginning of PM measurements in Cyprus, till 2010 followed by a subsequent sharp decrease by approximately 6.8 μg m^{-3} during 2011. In fact, during 2011 and 2012, regionally transported dust particles contributed less than 6 μg m^{-3} to the PM_{10}, the lowest amount since 1999. Compared to the 2011–2012 decrease, dust particle concentration has slightly increased reaching 8.5 μg m^{-3} during 2014 and 2015. The observed abrupt decline in PM_{10} levels after 2010 (Fig. 5), coincides with the sharp decline of more than 50% in the computed dust concentration based on the Querol et al. (2009b) method.

It should be noted that the reported annual increase of transported dust reported until 2010 (0.4 μg m^{-3}) may seem small but refers to an amount accumulated each year until 2010, i.e 12 years. Similar increasing trends in dust amounts during the first period have been

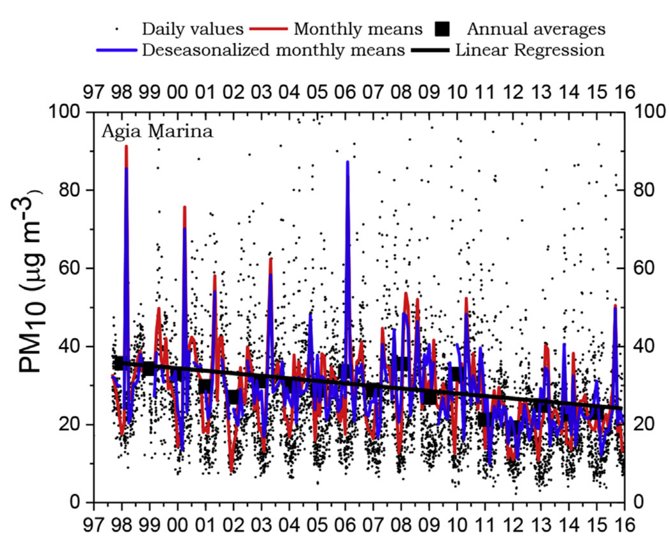

Fig. 4. Daily (black dots), monthly (red line) and annual (blue circles) means of PM_{10} concentrations observed at the background Agia Marina station (AGM_{BKG}) during the temporal period 1998–2015. Blue line and black line depicts the deseasonalized data and the linear regression, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

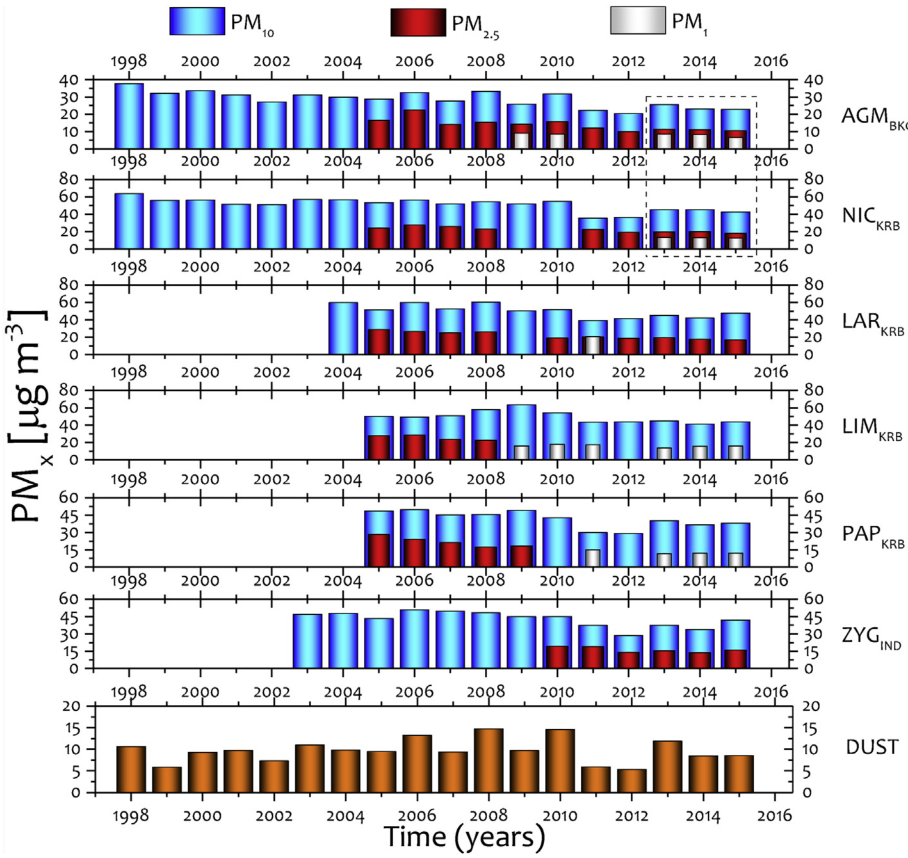

Fig. 5. Annualy averaged PM_x in Cyprus observed urban, industrial and background stations. PM_{10} are shown with blue bars, $PM_{2.5}$ with red and PM_1 with grey-white bars. Dust concentrations computed using the Querol et al. (2009b) method are shown in orange. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Mann Kendall analysis result. With the exception of Limassol (LIM_{KRB}), a decreasing trend is observed at the monitored sites with at least 95% confidence ($p < 0.05$).

Monitoring station	Period [years]	Slope (Q) [in μg m^{-3}]	Level of significance
AGM_{BGR} – PM_{10}	1998–2015	−0.67	***
AGM_{BGR} – $PM_{2.5}$	2005–2015	−0.69	**
NIC_{KRB}– PM_{10}	1998–2015	−0.93	**
NIC_{KRB}– $PM_{2.5}$	2005–2015	−0.85	**
LAR_{KRB}– PM_{10}	2003–2015	−1.42	*
LAR_{KRB}– $PM_{2.5}$	2005–2015	−1.20	**
LIM_{KRB}– PM_{10}	2006–2015	−1.11	NS
PAP_{KRB}– PM_{10}	2006–2015	−1.22	*
ZYG_{IND}– PM_{10}	2002–2015	−1.34	*

***$p < 0.001$, **$p < 0.01$, *$p < 0.05$, NS $p \geq 0.1$.

reported by Ganor et al. (2010) and Klingmüller et al. (2016) concerning dust transport from the Saharan and Arabian deserts, respectively.

If the entire period under investigation is considered (1998–2015) the average annual PM_{10} of 28.7±5.0 μg m^{-3} is higher than the majority of rural sites in central and northern Europe (Barmpadimos et al., 2012; Putaud et al., 2010; Van Dingenen et al., 2004) and the west Mediterranean basin (Querol et al., 2009a; Ripoll et al., 2015). On a regional level, the remote site Finokalia in Crete, Greece located west of Cyprus exhibited approximately the same PM_{10} levels (28–31 μg m^{-3} for 2001–2006; Gerasopoulos et al., 2006, 2007; Querol et al., 2009a), and the rural site Erdemli in Turkey, located north of Cyprus, higher (36 μg m^{-3} for 2001–2002; Koçak et al., 2007). Higher levels have also been reported for rural sites in western Saudi Arabia (95 ± 78 μg m^{-3}; Lihavainen et al., 2016) and background sites outside of Cairo for the period 1998–2007 (ranging between 120 and 150 μg m^{-3}; Safar and Labib, 2010). These results are consistent with Querol et al. (2009a) that reported a positive gradient in the Mediterranean from west to east.

PM_{10} was additionally monitored, for more than three years period (since June 2012), at the Troodos site (TRO_{FTP}), at 1819 m altitude representative of the free troposphere. At this remote site, PM_{10} daily variation was in good agreement with AGM_{BKG} ($R^2 = 0.73$) mainly because both sites experienced periods of high concentrations, such as dust events, simultaneously. During dust events PM_{10} at TRO_{FTP} was equal or higher than that monitored at AGM_{BGR}. In addition, in the Cyprus area, pollution transport from Turkey that would not be detected at altitudes lower than 1 km a.s.l. has been reported (Nisantzi et al., 2014). During these events, which are seldom (45 recorded in the course of 4 years) but last for several days, mineral dust and, in half of the reported cases, smoke from wildfires was transported, elevating PM_{10} at the free tropospheric site only. As a result, average PM_{10} values for the period June 2012–Dec 2015 at the TRO_{FTP} site (19.5 ± 8.4 μg m^{-3}) were close to the ones recorded for the AGM_{BGR} site (22.9 ± 8.2 μg m^{-3}). These results suggest that PM_{10} measurements at AGM_{BKG} represent a lower regional transport bound in the Cyprus area for that size fraction. The PM_{10} levels reported in this work at TRO_{FTP}, are higher than that reported for free tropospheric sites in the western Mediterranean (Galindo et al., 2017; Ripoll et al., 2015). Ripoll et al. (2015) reported PM_{10} levels of two sites at 1570 and 720 m a.s.l. and with concentrations equal to 11.5 ± 9.3 μg m^{-3} and 15.5 ± 7.9 μg m^{-3}, respectively. The concentration difference (≈4 μg m^{-3}) between these sites is similar to the one reported in this work (3.4 μg m^{-3}). However, such amounts constituted a larger fraction of the observed PM_{10}, as the levels in the west Meditteranean were lower than in the eastern one.

Background-rural $PM_{2.5}$: The annual average $PM_{2.5}$ concentrations of the 2005–2015 period at AGM_{BKG} (red bars in Fig. 5) were equal to 14.0 ± 3.6 μg m^{-3}.

This number lies on the high end of the observed annual means not only in the central and northern Europe but also in the Mediterranean Region where annual $PM_{2.5}$ values are below 13 $\mu g\, m^{-3}$ (Querol et al., 2009a; Koçak et al., 2007). A notable exception was Finokalia, where annual $PM_{2.5}$ during 2004–2006 was slightly higher at 18.2 $\mu g\, m^{-3}$ (Gerasopoulos et al., 2007) and Rachma, Jordan (25 ± 7 $\mu g\, m^{-3}$ for 2007). Similar to PM_{10}, significant trend ($p < 0.01$) was observed at AGM_{BKG} for $PM_{2.5}$ during the 11-year period of data (2005–2015) with an annual decrease of 0.69 ± 0.21 $\mu g\, m^{-3} y^{-1}$. For the same time period (2005–2015), the decrease of PM_{10} was 1.1 ± 0.42 $\mu g\, m^{-3} y^{-1}$. Similar to PM_{10}, $PM_{2.5}$ levels remained relatively stable till 2010, followed by a sharp decrease that persisted till 2015. For the period 2005–2010 and 2011–2015, $PM_{2.5}$ levels were 16.4 ± 3.1 $\mu g\, m^{-3}$ and 12.0 ± 2.0 $\mu g\, m^{-3}$, respectively. This decrease which is similar to that observed for the PM_{10} fraction indicates contribution from natural sources to this tendency.

Background-rural PM_1: No trend assessment with respect to PM_1 concentrations (white bars in Fig. 5) at the AGM_{BGR} station was performed due to limited years of observations (2009–2010 and 2013–2015). During the above period, annual average PM_1 was 8.2 ± 1.9 $\mu g\, m^{-3}$ similar to other Mediterranean sites (Gerasopoulos et al., 2007; Querol et al., 2009a; Ripoll et al., 2015). Notably, a moderate correlation ($R^2 = 0.42$) was observed with the concurrent PM_1 measurements at the TRO_{FTP} station for a three years period of observations (2013–2015). It was found that the free-tropospheric site exhibited only 27% lower PM_1 amounts (5.8 ± 2.4 $\mu g\, m^{-3}$) in comparison to the boundary layer-based AGM_{BGR} ones (7.8 ± 1.9 $\mu g\, m^{-3}$). The above observation is indicative of limited contribution of submicron particles in Cyprus from local sources and highlights the importance of regional sources associated with long-range transport phenomena.

4.1.2. Urban PM in Cyprus

Urban PM_{10}: Notably, similar PM_{10} levels have been observed for the three major urban centers of Cyprus (Nicosia, Larnaca and Limassol) even though the population of Nicosia is almost twice that of Limassol and a three-fold higher than of Larnaca. More specifically, the annual average PM_{10} concentration for the common period of measurements (2006–2015) at the kerbside sites of Nicosia, Larnaca and Limassol were equal to 47.3 ± 7.6 $\mu g\, m^{-3}$, 49.0 ± 7.4 $\mu g\, m^{-3}$ and 49.2 ± 7.2 $\mu g\, m^{-3}$, respectively. The respective annual averaged PM_{10} at PAP_{KRB} and ZYG_{IND} were 20% lower compared to the above kerbside stations and equal to 40.5 ± 7.2 $\mu g\, m^{-3}$ and 41.7 ± 7.3 $\mu g\, m^{-3}$, respectively.

Table 3
Annual or multi annual average concentration of PM_1, $PM_{2.5}$ and PM_{10} from various urban areas in Europe and the Middle East. All values are in $\mu g\, m^{-3}$.

City Name	Country	Period	PM_{10}	$PM_{2.5}$	PM_1	Reference
South Europe						
Nicosia	Cyprus	See Section 4.1.2	47.3 ± 7.6	21.9 ± 3.2	13.4 ± 5.2	This study
Larnaka	Cyprus	See Section 4.1.2	49.0 ± 7.4	21.1 ± 3.8	21.1 ± 5.3	This study
Limassol	Cyprus	See Section 4.1.2	49.2 ± 7.2	25.5 ± 3.0	15.9 ± 4.6	This study
Paphos	Cyprus	See Section 4.1.2	40.5 ± 7.2	21.7 ± 3.1	12.8 ± 3.1	This study
Nicosia	Cyprus	12/2002 -8/2003	45	N/A	N/A	Kleanthous et al., 2009
Nicosia	Cyprus	2008–2013	32.7	N/A	N/A	Mouzourides et al., 2015
Athens	Greece	06/2005–09/2006	N/A	26.4	N/A	Eleftheriadis et al., 2014
Athens	Greece	2001–2004	55.7 ± 23.0	N/A	N/A	Grivas et al., 2008
Piraeus	Greece	2001–2004	58.8 ± 20.8	N/A	N/A	Grivas et al., 2008
Patras	Greece	2004	42.3 ± 14.7	N/A	N/A	Pikridas et al., 2013
Patras	Greece	12/2008–5/2012	N/A	22.2 ± 11.1	N/A	Pikridas et al., 2013
Heraklion	Greece	11/2001–9/2005	51 ± 33	N/A	N/A	Gerasopoulos et al., 2006
Istanbul	Turkey	10/2007-6/2009	39.1	N/A	N/A	Theodosi et al., 2010
Barcelona	Spain	2003–2008	40	26	20	Pey et al., 2010
North Europe						
Belfast	UK	2000–2005	25.9	15.0	N/A	Abdalmogith et al., 2006
Glasgow	UK	2000–2005	23.8	14.1	N/A	Harrison and Jones, 2005
Manchester	UK	2000–2005	31.0	17.1	N/A	Harrison and Jones, 2005
Birmingham	UK	2001–2005	26.1	17.1	N/A	Harrison and Jones, 2005
London	UK	2001–2004	44.6	26.5	N/A	Abdalmogith et al., 2006
London	UK	2012	N/A	N/A	9.8 ± 10.0	Young et al., 2015
Helsinki	Finland	1996–1997	16.6	9.2	N/A	Vallius, 2005
Helsinki	Finland	2009	N/A	N/A	6.6–6.9	Carbone, 2014; Timonen et al., 2011
Central Europe						
Bern	Switzerland	2005–2008	32.5	19.5	N/A	Hueglin et al., 2005
Bern	Switzerland		39			Barmpadimos et al., 2011
Basel	Switzerland	2005–2008	20.8	15.6	N/A	Hueglin et al., 2005
Zuerich	Switzerland	2008	19.6	14.7	N/A	Hueglin et al., 2005
Augsburg	Switzerland	2005–2007	21.1	19.8	N/A	Cyrys et al., 2008; Pitz et al., 2008
Leipzig	Germany	1999–2001	N/A	22.4	N/A	Wehner et al., 2002
Dresden	Germany	2003–2007	37.0	N/A	N/A	Löschau, 2006
Middle East						
Beirut	Lebanon	2006–2007	86.81	27.63	N/A	Saliba et al., 2010
		2004–2005	103.81	38.86	N/A	Saliba et al., 2010
		2003	71.34	40.95	N/A	Saliba et al., 2010
		2014	37.9	28.8	N/A	Abdallah et al., 2018
Ahvaz	Iran	4/2010-9/2010	319.6 ± 407.07	69.5 ± 83.2	37.02 ± 34.9	Shahsavani et al., 2012
Tel Aviv	Israel	2007	N/A	30.7 ± 11.2	N/A	Sarnat et al., 2010
Haifa	Israel	2007	N/A	19.8 ± 7.7	N/A	Sarnat et al., 2010
Eliat	Israel	2007	N/A	21 ± 4	N/A	Schneidemesser et al., 2010
Amman	Jordan	2007	N/A	34.9 ± 15.1	N/A	Sarnat et al., 2010
Zarka	Jordan	2007	N/A	37 ± 7	N/A	von Schneidemesser et al., 2010
Jeddah	Saudi Arabia	2012	104 ± 162	34 ± 45	13 ± 11	Hussein et al., 2014

A significant fraction of these averages emanate from regional sources. By considering the results obtained at CAO station at Agia Marina as representative of the regional influence, the local influence of the urban and industrial sites on the PM_{10} levels can be estimated by subtracting the regional PM_{10} amounts from the measured PM_{10} ones at each location. On average, 44.0 ± 9.7%, 44.3 ± 9.7%, 45.9 ± 13.1%, 33.9 ± 12.6%, 35.5 ± 11.6% of the observed PM_{10} levels in Nicosia, Larnaca, Limassol, Paphos and Zygi, respectively is expected to originate from local anthropogenic and/or natural emissions including traffic, biomass-burning, shipping emissions (in Limassol), airport related emissions (in Larnaca), resuspension of dust and sea-salt (in coastal locations).

Because Larnaka and Limassol are coastal sites they are susceptible to sea spray emissions which is not the case at the inland Nicosia. In addition Nicosia, even though it engulfs a larger population, lacks commercial traffic such as that encountered at Limassol (harbor related emissions) and Larnaka (airport related emissions). These differences can explain why PM_{10} levels are similar at these 3 cities despite the population difference.

The above annual averages of the PM_{10} levels in the urban areas of Cyprus are amongst the highest when compared to a) the central and northeastern Europe (ranging between 20 and 40 $\mu g\,m^{-3}$; Putaud et al., 2010) and b) the majority of the cities in the west Mediterranean (ranging between 40 and 50 $\mu g\,m^{-3}$; Putaud et al., 2010; Pey et al., 2010). However, reported PM_{10} levels of urban areas lying further east of Cyprus such as Beirut, and Jeddah are higher (see Table 3).

Notably, the urban PM_{10} annual averages in Cyprus exceeded the PM_{10} levels reported for two of the most populated cities (> 5×10^6 inhabitants) in southeastern Europe, namely Athens and Istanbul (ranging between 35 and 45 $\mu g\,m^{-3}$; Grivas et al., 2008; Pateraki et al., 2010; Theodosi et al., 2010; Vardoulakis and Kassomenos, 2008) but are lower than those reported for Cairo that often exceed 150 $\mu g\,m^{-3}$ (Safar and Labib, 2010). In Table 3, annual average PM_x levels from various urban areas in northern, central, and southern Europe, including results from this study, are shown.

Based on the statistical Mann-Kendall trend-analysis of available data (Table 2), most urban sites exhibited a significant annual decrease in PM_{10} levels. In Nicosia, PM_{10} decreased by1.0 ± 0.2 $\mu g\,m^{-3}\,y^{-1}$ ($p < 0.01$), in Larnaca by 1.5 ± 0.4 $\mu g\,m^{-3}\,y^{-1}$ ($p < 0.05$), in Paphos by 1.6 ± 0.5 $\mu g\,m^{-3}\,y^{-1}$ ($p < 0.01$) and in Zygi by 1.2 ± 0.4 $\mu g\,m^{-3}\,y^{-1}$ ($p < 0.05$). Kerbside PM_{10} measurements conducted at Limassol exhibited a non-significant ($p \geq 0.1$) declining trend equal to 1.5 ± 0.6 $\mu g\,m^{-3}\,y^{-1}$. These decreasing trends are higher than that observed in the background site and indicate not only decrease in regional but also to some of the local sources mentioned above. More work is needed to understand the reasons of the decrease in the local sources.

Urban $PM_{2.5}$: $PM_{2.5}$ monitoring started at all urban stations in 2005 (see Fig. 5; red bars) and at the industrial site (Zygi) in 2010. NIC_{KRB} and LAR_{KRB} reported the longest record of data of ten and nine years, respectively. At the remaining kerbside sites (Limassol and Paphos) $PM_{2.5}$ measurements were interrupted and replaced by PM_1 during or before 2009. The annual average $PM_{2.5}$ concentration in Nicosia was equal to 21.9 ± 3.2 $\mu g\,m^{-3}$ (2005–2015), in Larnaca equal to 21.1 ± 3.8 $\mu g\,m^{-3}$ (2005–2015), in Limassol equal to 25.5 ± 3.0 $\mu g\,m^{-3}$ (2005–2008), in Paphos 21.7 ± 3.1 $\mu g\,m^{-3}$ (2005–2009) and in Zygi 16.2 ± 2.4 $\mu g\,m^{-3}$ (2010–2015). The observed urban $PM_{2.5}$ values in Cyprus are elevated compared to the majority of urban areas in the northwestern Europe (see also Table 3) which typically exhibit an annual $PM_{2.5}$ average lower than 20 $\mu g\,m^{-3}$ (Putaud et al., 2010; Barmpadimos et al., 2012) but in the same range with many central European cities where annual average $PM_{2.5}$ ranges between 18 and 26 $\mu g\,m^{-3}$ (Putaud et al., 2010). However, urban $PM_{2.5}$ in Cyprus is lower than that reported for most urban areas in the Middle East (see Table 3) with the notable exception of Haifa, Israel (Sarnat et al., 2010).

No trend analysis was conducted for the stations with limited annual data (LIM, PAP, ZYG) whereas for the NIC_{KRB} and LAR_{KRB} sites and for the period 2006–2015, significant declining trends ($p < 0.01$) were found equal to 0.9 ± 0.1 and 1.2 ± 0.1 $\mu g\,m^{-3}\,y^{-1}$, respectively.

Urban PM_1: In addition to the above presented sub-10 µm and fine PM amounts, the submicron PM concentrations in Nicosia (2013–2015), Larnaca (2011), Limassol (2009–2015) and Paphos (2011–2015) were measured and found to be equal to 13.4 ± 5.2 $\mu g\,m^{-3}$, 21.1 ± 5.3 $\mu g\,m^{-3}$, 15.9 ± 4.6 $\mu g\,m^{-3}$ and 12.8 ± 3.1 $\mu g\,m^{-3}$, respectively. The annual levels at all cities are higher than those reported for urban background stations in northern Europe (Table 3) such as London (9.8 ± 10.0 $\mu g\,m^{-3}$; Young et al.,

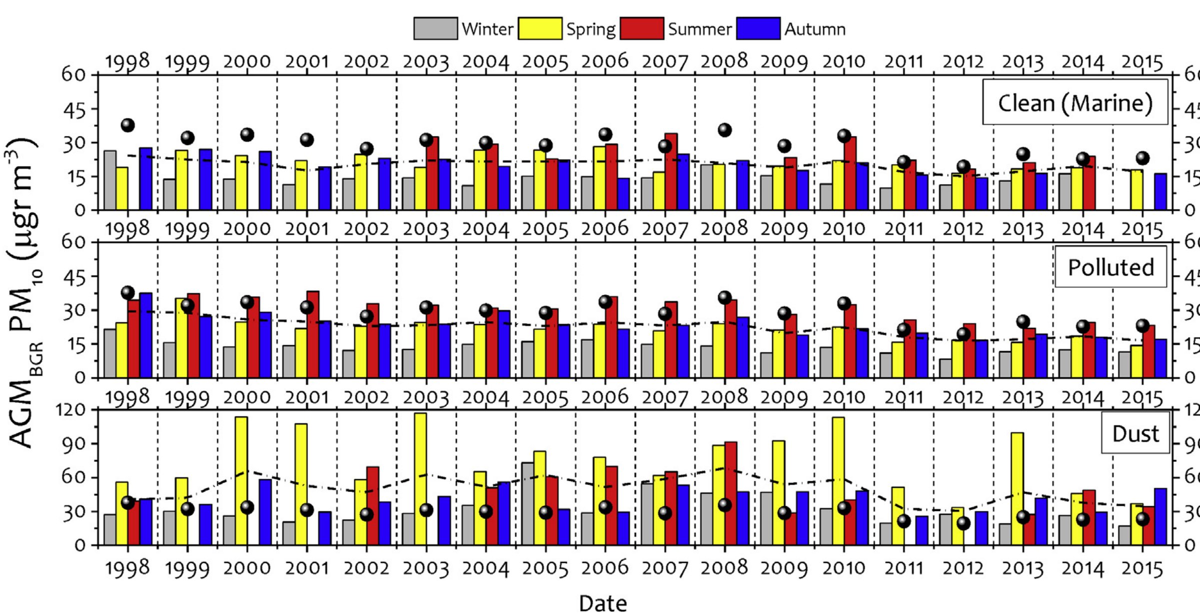

Fig. 6. Seasonally averaged PM_{10} at the Agia Marina regional background site (AMX_{BKG}) with respect to air masses reaching from westerly (clean) air masses (top panel), the northerly polluted regions (middle panel) and from dust-related easterly and southerly air masses (bottom panel). Dashed lines represent the annual average if only air masses for the corresponding source regions were reaching the site. Black circles show the measured annual average.

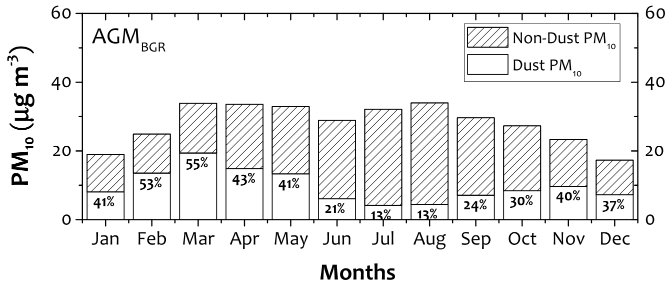

Fig. 7. Seasonal variability of PM_{10} 1998–2015 mass concentrations at the Agia Marina monitoring station (AGM_{BGR}). The dust content has been computed using the Querol et al. (2009b) method (see text). The patterned bars depict the remaining PM_{10} fraction (measured PM_{10} minus dust).

2015) and Helsinki (ranging between 6.6 and 6.9 μg m^{-3}; Carbone, 2014; Timonen et al., 2011) or in Middle East such as Jeddah in Saudi Arabia (13 ± 11 μg m^{-3}; Hussein et al., 2014). Similar PM_1 levels to those monitored in Limassol, have been reported for Barcelona (20 μg m^{-3}; Pey et al., 2010), a major city in south Europe. The urban area of Ahvaz, Iran has been associated with higher, approximately a factor of 2, super-micron levels, which do not correspond to a complete calendar year (Shahsavani et al., 2012). In Europe, long term submicron sampling is mainly performed by the ACTRIS network, which involves background and rural stations along with a few urban sites (see eg Crippa et al., 2013; Lanz et al., 2010). Notable exceptions can be found in the west Mediterranean basin (Galindo et al., 2018; Nicolas et al., 2015; Yubero et al., 2015).As a result, reports of PM_1 annual averages concerning European urban sites are scarce. Data coverage resulting from this work is not enough to calculate any annual trend.

4.2. Influence of air mass origin on ambient PM_{10} levels

The most common wind pattern in the East Mediterranean is northerly "Etesian" winds; in Cyprus they account on average for 68% of the air masses crossing the island. The remaining 31% corresponds to air masses from N. Africa (9%), Middle East (10%), westerlies (8%) that even though they originate from Europe or less frequently from Africa spend several days above the sea before reaching Cyprus, and to stagnant (low wind speed) conditions (3%). An additional 2% could not be attributed to a specific source region. The Etesians transport pollutants either from Europe (notably from Eastern Europe) or Turkey (Kleanthous et al., 2014). The west side of Turkey is more industrialized than the remaining country and frequently (9% on annual average) air masses cross over that region prior to reaching Cyprus. However, the most frequent pattern is air mass transport from the mainland of Turkey and the areas north of it (marked with red on Fig. 2a and are referred as source region "SW Asia") accounting for more than half (58.8%) of the air masses reaching the island on a yearly basis.

Seasonal PM_{10} averages at AGM_{BKG} for each year from 1998 to 2015 have been calculated individually for each of the source regions presented on Fig. 2. To simplify the analysis, air masses from N. Africa and Middle East have been lumped because they are often related to dust events and will be referred as dust-dominated source regions (Fig. 6). Similarly, source regions Europe and South West Asia have also been

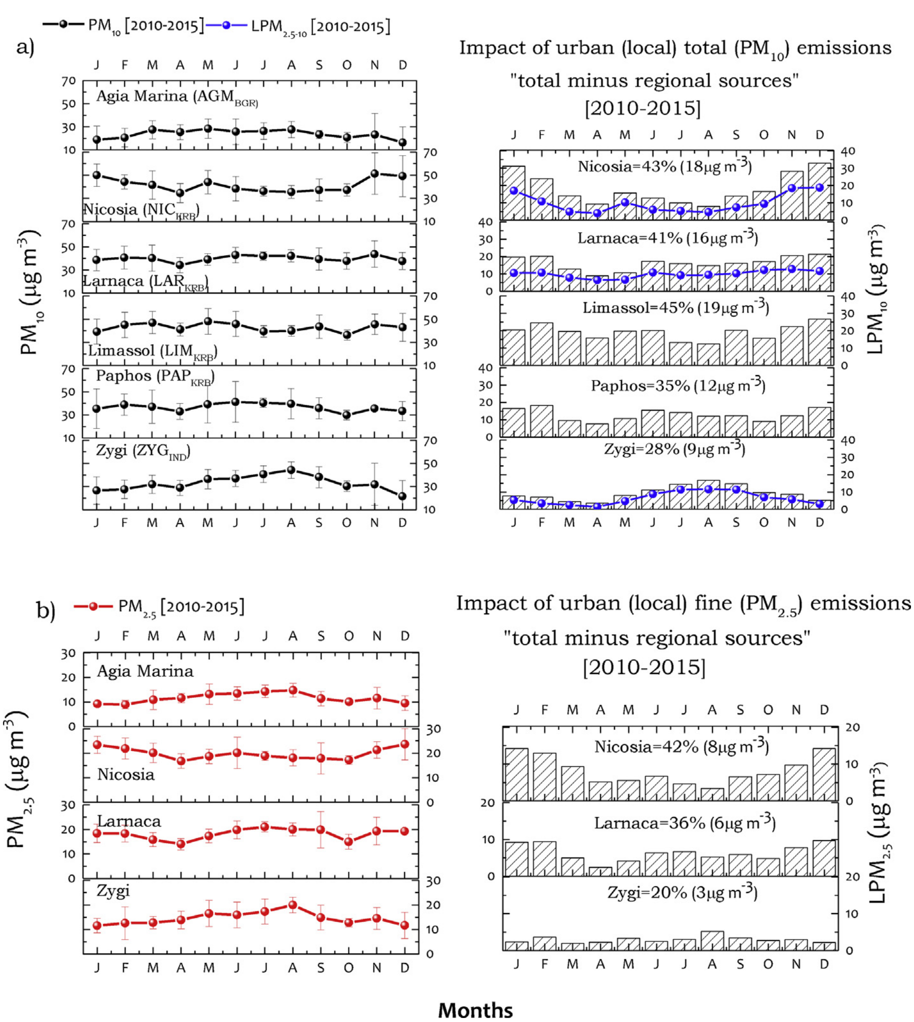

Fig. 8. The upper top panel presents the seasonally averaged PM_{10} mass concentrations (2010–2015 period; black line) for the AGM_{BGR}, NIC_{KRB}, LAR_{KRB}, LIM_{KRB}, LAR_{KRB} and the ZYG_{IND} stations. The bottom left panel shows the seasonally averaged $PM_{2.5}$ concentrations (2010–2015 period; red line) and for the AGM_{BGR}, NIC_{KRB}, LAR_{KRB} and ZYG_{IND} stations. The top and bottom right panels present the calculated local LPM_x emissions (x = 10, top right panel; x = 2.5, bottom right panel) by assuming a common regional background represented by the AGM_{BGR}. The blue line of the right top panel depicts the local coarse fraction of PM ($LPM_{2.5\text{-}10}$) with $LPM_{2.5\text{-}10}$ = LPM_{10}-$LPM_{2.5}$. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

lumped representing anthropogenic sources and are referred as polluted source regions. Finally, air masses originating from the marine (west) sector (shown with light blue in Fig. 2a) have been considered as cleaner, because they spend at least one day above the sea before reaching Cyprus. The majority of these air masses have also spent several days crossing Europe and were inevitably influenced by regional emissions in addition to maritime emissions that includes ship's exhaust. If a season for a certain year was represented by less than 10% of the corresponding days (≈ 10 days) it was omitted from analysis. Based on the seasonal averages for each of the three lumped source regions, the estimated annual PM_{10} average of polluted, marine, and dust-influenced air masses at AGM_{BKG} is calculated and presented as dashed lines in Fig. 6. For comparison, black dots depict the (measured) AGM_{BGR} PM_{10} annual average.

Polluted sources: Air masses from polluted source regions, located northerly of AGM_{BKG}, exhibited on average 8.0% higher PM_{10} compared to the measured annual average only during the summer months (June–August). During the remaining year, PM_{10} associated with these source regions was on average 31.7% lower. As a result, if air masses reached AGM_{BKG} originated exclusively from the north it would result on an annual average PM_{10} of $22.4 \pm 4.0\,\mu g\,m^{-3}$, 21.8% smaller compared to the measured PM_{10} of $28.7 \pm 5.0\,\mu g\,m^{-3}$, (see Section 4.1).

Marine air masses: The annual averaged PM_{10} would reduce at $20.0 \pm 2.6\,\mu g\,m^{-3}$, if only air masses of westerly origin (the source region is shown with light blue on Fig. 2a), reached AGM_{BKG}. Although significantly reduced, compared to the measured annual average because air masses have spent at least one day over the sea, it remains however a non-negligible PM_{10} level. As explained before this does not imply the complete lack of regional influence. Following a stricter definition of the marine source region, that excludes any regional influence, the annual average PM_{10} should reduce further. However, such clean air masses are rare ($\approx 1\%$) and the remaining sample is not enough for any accurate statistical analysis.

Dust emissions: During each of the 15 years investigated in this study, air masses associated with dust-dominated source regions (shown with yellow and brown in Fig. 2a) always exhibited elevated monthly-averaged PM_{10} compared to polluted or marine source regions, regardless of season. Overall if air masses exclusively originating from the Middle East and N. Africa region was reaching AGM_{BKG}, the estimated annually averaged PM_{10} would be $49.9 \pm 11.7\,\mu g\,m^{-3}$ or 75% above the actual measured annual PM_{10} levels.

On a seasonal basis (Fig. 7) dust follows a bi-modal distribution peaking in spring (March–May) and autumn (September–November), similar to what is observed at Crete island in Greece (Kalivitis et al., 2007). Up to more than half of the PM_{10} during February, March and April, months of the maximum dust influence, was due to regional dust transport. During autumn and winter, PM_{10} emanating from dust is still high ranging from 24 to 41% (Fig. 7). As depicted in Fig. 6, during summer air masses influenced by dust dominated source regions, also exhibit elevated PM_{10} ($52.2 \pm 19.6\,\mu g\,m^{-3}$ on average), yet because they are less frequent the impact is smaller, accounting only for 15.6% of the observed PM_{10} at AGM_{BKG} (Fig. 7).

4.3. Temporal variability of $PM_{2.5}$ and PM_{10} in Cyprus

4.3.1. Seasonal PM variation

Seasonal PM_{10} and $PM_{2.5}$ patterns (Fig. 8a and b, respectively) were obtained using monthly means for the period 2010–2015, when concurrent $PM_{2.5}$ measurements were reported for the NIC_{KRB}, LAR_{KRB} and ZYG_{KRB} stations in addition to AGM_{BKG}.

Background Agia Marina site: The regional background site (AGM_{BKG}) exhibited a distinct seasonal variability with higher PM_{10} concentrations during the warm period of the year and minimum during winter (Fig. 8). The observed maximum plateau of around $27\,\mu g\,m^{-3}$ is caused by the combination of dust-influenced air masses primarily occurring during spring and autumn as shown before, and the enhanced transport of polluted air masses from the north sector during the Etesians. Part of the observed variability could also be explained by local dust re-suspension caused by wind erosion from local cropland. More than half (58% according to Zoumides et al., 2013) of Cyprus' croplands are cultivated with rain-fed cereals and fodder crops, which are usually planted in November and harvested in April–May (Zoumides et al., 2013). This leaves the land bare or with little cover during the remainder of the year. During drought years, the crop area shrinks (for example in the 2008 drought year the harvested cropland was 23% below the 1995–2009 average (Zoumides et al., 2013)) and rain-fed crops are often harvested early (March), leaving soils vulnerable to wind erosion in spring. This is especially the case for the croplands around the AGM_{BKG} station, which are dominated by fine textured soil (clay $< 2\,\mu m$; Camera et al., 2017). Contrary, during winter, when precipitation maximizes, a minimum was observed. Seasonal $PM_{2.5}$ variation at AGM_{BKG} (Fig. 8b) follows the one of PM_{10}. Monthly averaged $PM_{2.5}$, based on 5-year measurements (2010–2015) depict a winter-to-summer increase of 50%. This behavior can be explained a) by the enhanced photochemical conditions during summertime leading to secondary aerosol formation, b) the absence of precipitation, which increases particles lifetime and c) the prevalent Etesian winds transporting air masses rich in fine mode anthropogenic pollutants from Turkey or to a lesser extent from Europe.

The Agia Marina site is located inland 40, 52.5, 38.3 and 63 km away from the urban centers of Nicosia, Larnaka, Limassol and Paphos, respectively. Based on the prevailing wind patterns (see Fig. 2) the site is expected to be impacted by emissions from Paphos, the smallest city with respect to area and number of inhabitants and furthest away from AGM_{BKG} compared to the remaining urban areas under investigation, at least 75.5% of the time. Southerly and easterly winds are prevalent only when air masses originate from Middle East and N. Africa (19.4% on annual basis).

As mentioned above, AGM_{BKG} station is surrounded by fine clay soils that typically erode during spring and summer. Besides Paphos, that is upwind, all other urban areas are expected to be affected by the transport of local eroded soil. However, the distribution is not expected to be homogeneous throughout the island. Pikridas et al. (2013) has shown that differences in the amounts of sulfate should also be expected between the urban and rural sites. These discrepancies result in underestimation of urban emissions and thus the calculations presented below should be regarded as a lower estimate.

Urban sites: The seasonal variability of the ambient PM_{10} and $PM_{2.5}$ amounts at the anthropogenic influenced monitoring sites could be assessed by using the locally estimated PM_{10} (LPM_{10}) and $PM_{2.5}$ ($LPM_{2.5}$) levels; with regional background concentrations being subtracted from the urban ones, (Fig. 8a-right panel). These concentrations represent local (anthropogenic) emissions but have a physical meaning only if the subtracted means are statistically different. To ensure this hypothesis, a two-tailed *t*-test for samples of unequal variances at the 95% confidence level was applied for each calendar month using measurements from AGM_{BGR} as a reference both for $PM_{2.5}$ and PM_{10}. All urban areas were statistically different from the background for every calendar month. Only at the industrial site (ZYG_{IND}) the difference was not significant during March and April with respect to PM_{10} and during March, with respect to $PM_{2.5}$. During March and April, regional dust transport maximizes and the effect of anthropogenic activities at ZYG_{IND} becomes negligible.

In NIC_{KRB}, the observed pattern of LPM_{10} and $LPM_{2.5}$ emissions follow a distinct cycle, with their highest values in winter (Fig. 8a and b). This variation, indicative of anthropogenic emissions, was not solely attributed to fine mode ($LPM_{2.5}$) increase during winter, but also to the concurrent increase of local coarse mode mass concentrations ($LPM_{2.5\text{-}10} = LPM_{10}$ - $LPM_{2.5}$: blue line in Fig. 8a). Notably, 54% of the LPM_{10} in Nicosia are found exclusively in the coarse $LPM_{2.5\text{-}10}$ mode. This shows that local fine mode urban emissions (e.g. residential heating and traffic) and coarse mode urban emissions (e.g. dust resuspension, wear and tear in brakes and tires, respectively) are of almost equal importance in Nicosia. Overall, LPM_{10} and $LPM_{2.5}$ in Nicosia account, for the period 2010–2015, for the 43% (18 μg m^{-3}) and 42% (8 μg m^{-3}) of the PM_{10} and $PM_{2.5}$, respectively, monitored at NIC_{KRB} as pointed out with the bars in Fig. 8a and b.

Similar but less pronounced seasonality in the PM_{10} amounts is observed in Larnaca, Limassol and Paphos cities. The calculated $LPM_{2.5}$ and LPM_{10} mass concentrations in Larnaca denote that the winter peak is retained suggesting enhanced emissions from traffic and residential heating. A second, less pronounced, peak is observed during summer possibly related to ongoing touristic activities. The reader should note that the population of all coastal sites (LAR, LIM, PAP) increases significantly during summertime due to domestic and international tourism, enhancing thus anthropogenic emissions. LPM_{10} in Larnaca consist, on average, of 61% $LPM_{2.5\text{-}10}$ (Fig. 8a) and 39% of $LPM_{2.5}$. This shows that Larnaca is primarily affected by coarse mode local emissions such us dust resuspension and sea salt emissions. Contrary to mainland Nicosia, all coastal cities are exposed to sea salt emissions under favorable wind directions. Local emissions account for the 41% and for the 36% of the PM_{10} and $PM_{2.5}$ observed in $LAR_{KRB.}$

The seasonal variability of PM_{10} at the industrial area of Zygi (ZYG_{IND}) presents, similar to AGM_{BGR}, i.e a winter-minimum to summer-maximum gradient. However, PM_{10} mass concentrations measured at ZYG_{IND} were on average 29% higher than the AGM_{BKG} ones. Excluding the regional background contribution quantified using $PM_{2.5}$ and PM_{10} AGM_{BGD} concentrations, the largest fraction (65%) of this difference is attributed to local sources caused mainly (68%) by coarse mode particles ($LPM_{2.5}$-$_{10}$) compared to the fine ($LPM_{2.5}$) ones (32%). The dominance of the local $LPM_{2.5\text{-}10}$ coarse mode underlines once again the importance of dust resuspension, this time caused by the unpaved roads in the Zygi site and possibly the transport of sea salt

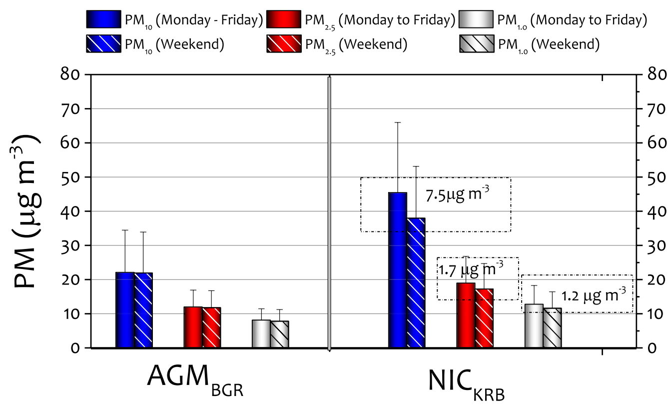

Fig. 10. Daily mean PM_x (x = 1, 2.5 and 10) values at the AGM_{BGR} and the urban NIC_{KRB} stations calculated for the period 2013–2015 separating weekdays (Monday-Friday, non-pattern bars) weekends (Saturday-Sunday, pattern bars).

emissions. Our findings suggest that the ZYG_{IND} area is, on average, mostly affected by regional pollution as only 28% (9 μg m^{-3}) of the recorded PM_{10}, and 20% (3 μg m^{-3}) of the $PM_{2.5}$ is caused by local emissions.

4.3.2. Diurnal variations

The calculation of the diurnal profiles, shown in Fig. 9, was restricted by the availability of TEOM measurements because gravimetric measurements were conducted on a daily and not hourly basis. Note that such higher resolution measurements were not initiated simultaneously at all sites. Therefore, the diurnal profiles presented on Fig. 9,

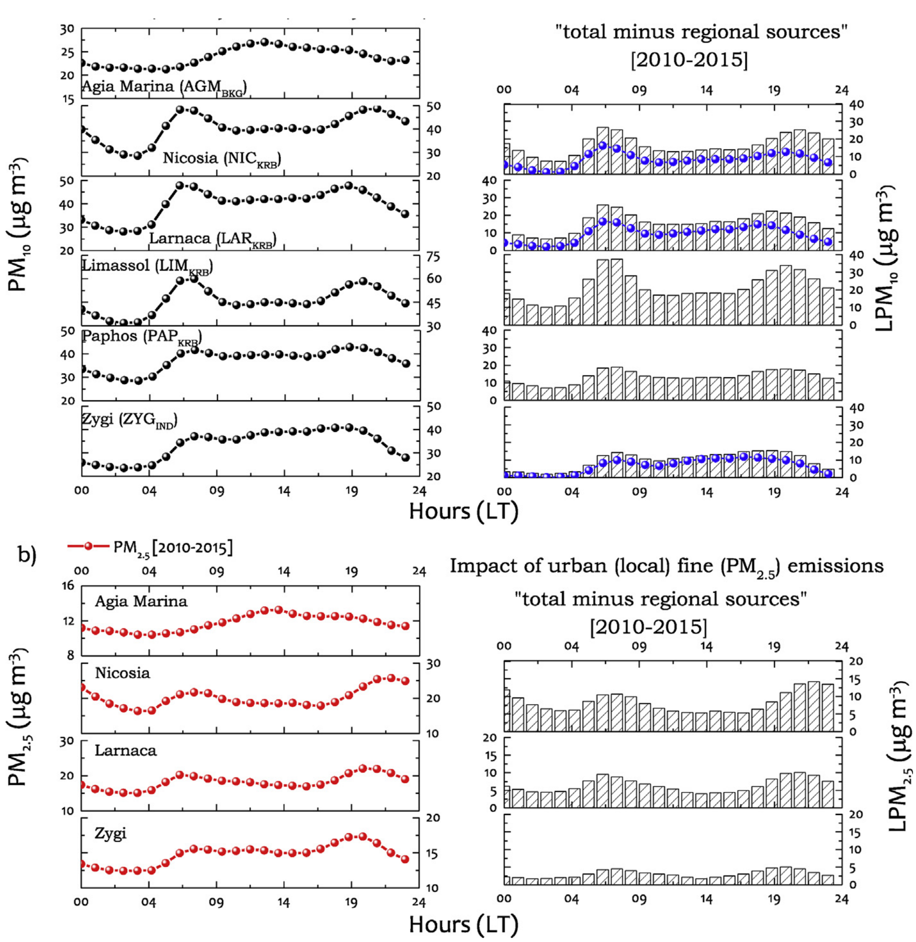

Fig. 9. Diurnal cycle of PM_{10} (black dots, top left panel) and $PM_{2.5}$ (red dots, bottom left panel) during the 2010–2015 period. Local emissions for both size fractions (top and bottom left figures, respectively) were calculated by assuming a common regional background represented by the AMX_{BKG} station, that was subtracted. Similarly to Fig. 8, the blue line (right panel of Fig. 9a) presents the coarse fraction of local PM, or else the $LPM_{2.5\text{-}10}$. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

correspond to a 6-year period (2010–2015) when simultaneous measurements were conducted at most sites and at both PM_{10} and $PM_{2.5}$ size fractions. No $PM_{2.5}$ measurements were performed in Limassol and Paphos.

The regional background site (AGM_{BKG}) exhibits a very small diurnal variability for both PM_{10} and $PM_{2.5}$ fractions which does not exceed 5.8 $\mu g\,m^{-3}$ and 2.8 $\mu g\,m^{-3}$, respectively with a slight maximum occurring at 13:00 (LT). Due to lack of local sources at the background site, the origin of the observed maximum around noon can be attributed to regional transported polluted air masses and the in situ production of secondary aerosol due to the intense photochemical activity in the Mediterranean (Hildebrandt et al., 2010; Nicolas et al., 2015).

PM_{10} diurnal profiles at all urban stations followed a uniform pattern that included; a minimum at 03-04:00 LT, similar to the $PM_{2.5}$ profiles, a maximum at 06:00–08:00 LT and a second maximum between 18:00–20:00 LST. Additionally, at all urban stations the morning peak was the highest, increasing the PM_{10} levels by 70%, 70%, 89% and 50% at NIC_{KRB}, LAR_{KRB}, LIM_{KRB}, PAP_{KRB}, respectively. To assess the above variability, the local variability of $LPM_{2.5}$, $LPM_{2.5\text{-}10}$ and LPM_{10} mass concentrations at each city is calculated by eliminating the regional influence quantified at the Agia Marina station (Fig. 9, right panels).

All fractions follow the presented bimodal pattern with distinct morning and evening peaks. This behavior can be explained based on the combined effect of the evolution of the mixing layer and the emission of pollutants due to anthropogenic activities (see e.g. Pikridas et al., 2015). During morning, the mixing layer height starts to increase as the case of anthropogenic activities (e.g. rush hour traffic, commuting). The net effect is an increase of the measured concentration. However, maximum atmospheric mixing is reached in the afternoon and those primary pollutants are diluted resulting in a concentration decrease. The mixing height remains constant until nighttime, when it decreases, along with after-work traffic/commuting and an increase in household heating, resulting in a second increase in the levels of those primary pollutants. The relative contribution of local fine-to-local sub-10 μm particulate matter ratio (R_{LF}: $LPM_{2.5}/LPM_{10}$) was estimated, using this time only TEOM data at both fractions (2.5 and 10). In Nicosia, 50% of the local PM_{10} amounts emanate from fine particles and the remaining 50% from coarse particles while in Larnaca, R_{LF} was found to be equal to 41%. In both cities, the fine fraction is linked to traffic and heating emissions, especially in winter and the high percentage of the coarse mode denotes the existence of sources leading to emission of large particles i.e. from brakes/tires wearing and dust resuspension. The difference of up to 10% in the computed R_{LF} enclose, most probably, the contribution of sea salt emissions in the observed PM_{10} at Larnaca.

A weak bimodal diel cycle of PM_{10} mass concentrations is observed in Paphos and an even weaker in Zygi with an almost constant, peak-to-peak variability. For the Zygi area, concurrent PM_{10} and $PM_{2.5}$ observations revealed that roughly 30% of the PM_{10} of the region emanates from local sources and from that, only around 30% is linked to fine-mode aerosols. Contrary, $LPM_{2.5\text{-}10}$ plays a more important role in the region and controls almost exclusively the diurnal LPM_{10} variability (Fig. 9) The variability of $LPM_{2.5\text{-}10}$ mass concentrations at Zygi follows a diel cycle with very low values during night, a morning increase coinciding with the rush hour peak, a small decrease due to the increase of the boundary layer and then constant values until the evening, when it gradually drops again to zero. Apart from the discussed apparent daytime anthropogenic activities contributing to the observed coarse mode aerosols, the above cycle with day-to-night contrast and negligible nighttime emissions points to sea breeze circulation with local winds bringing during daytime sea salt to the station.

4.3.3. Weekly cycles

Weekly cycles were calculated for the NIC_{KRB} and the regional background station AGM_{BKG} using all available PM_{10}, $PM_{2.5}$ and PM_1 data during 2013–2015, when measurements from all size fractions were available (Fig. 5). Data are clustered to weekdays (Monday-Friday) and weekends (Saturday and Sunday). At AGM_{BGR} no difference is observed between weekdays and weekends (Fig. 10) in all fractions coherent with the background character of the station. Contrary, at the NIC_{KRB} station, the most populated city in Cyprus, weekends exhibit lower PM levels. It was found that the decrease is higher for the PM_{10} (16%, or 7.5 $\mu g\,m^{-3}$), and smaller for the $PM_{2.5}$ (9% or 1.5 $\mu g\,m^{-3}$) and PM_1 (9% or 1.2 $\mu g\,m^{-3}$) fractions. The above shows that most of the observed weekend reduction in Nicosia is due to the decline in coarse-mode mass concentrations most probably from traffic re-suspension. Similar trends have been reported in an urban area of the west Mediterranean (Galindo et al., 2018).

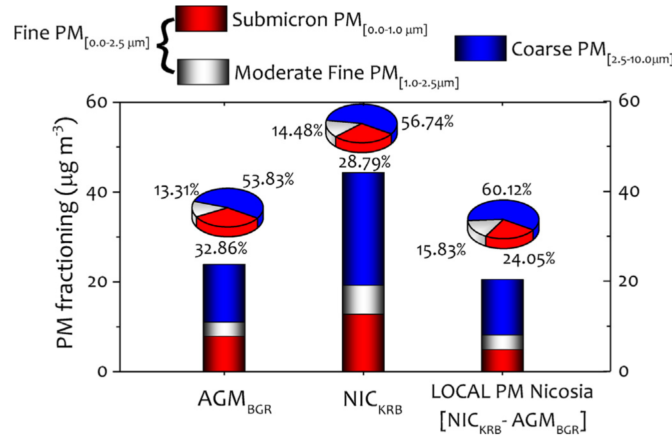

Fig. 11. Submicron (0.0–1.0 μm, red bars), fine (1.0–2.5 μm, white bars) and coarse (2.5–10.0 μm, blue bars) PM fractions at the AGM_{BGR} and the NIC_{KRB} monitoring stations. For the analysis, daily PM_1, $PM_{2.5}$ and PM_{10} concentrations of the common 2013–2015 period were used. The left and middle pie charts depict the calculated percentages represented by the submicron, moderate fine and coarse PM fractions at the AGM_{BGR} and NIC_{KRB} stations, respectively. Similarly, the right bar and pie chart show the computed local PM fractions in Nicosia. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

4.4. Fractioning of PM in urban and background sites in Cyprus

The above three-year dataset (2013–2015) of submicron (PM_1), fine ($PM_{2.5}$) and sub-10 μm (PM_{10}) were used to quantify the fractioning of particulate matter in coarse particles ($PM_{2.5\text{-}10} = PM_{10}\text{-}PM_{2.5}$), moderate fine ($PM_{1\text{-}2.5} = PM_{2.5}\text{-}PM_{1.0}$) and submicron ($PM_1$). The analysis was conducted for the background station Agia Marina and the urban Nicosia station Fig. 11. At AGM_{BGR}, coarse and fine modes explain the 46% and 54%, respectively of the PM_{10} mass; with the PM_1 mode accounting for the 70% of the $PM_{2.5}$ fraction.

In Nicosia the coarse particles dominate (57%) followed by the PM_1 ones (29%) and the $PM_{1\text{-}2.5}$ (14%). Similarly to Agia Marina, the submicron fraction is the dominant fraction of the fine mode. To account for the fractions of local PM sources in NIC_{KRB}, the regional contributions of all PM fractions at the background AGM_{BGR} station were subtracted from the respective ones at the NIC_{KRB} site. The results depict that a) 45% of the PM in Nicosia is of local origin (as also shown in section 4.3.1), b) most of the LPM_{10} (60%) is found in the coarse mode, c) 60% of the local fine mass emanates from submicron particles. These findings, suggesting that particles produced locally at Nicosia are primarily in the coarse ($PM_{2.5\text{-}10}$) mode from dust re-suspension and to a lesser extent in submicron mode, are in agreement with mass size distributions measured at an urban site in Sweden (Van Dingenen et al., 2004).

5. Conclusions

The amounts and properties of ambient particulate matter (PM) suspended in air over various environments has been in the forefront of ongoing environmental research studies and strict legislation regarding their levels has been imposed by international bodies such as WHO, EU etc. Information regarding aerosol sources (natural vs anthropogenic), origin (local vs regional), spatial and temporal variability are clearly needed by policy makers to propose mitigation strategies. Consequently, studies such as the one presented here, providing detailed information on the spatial and temporal variability of sub-10 μm, fine and submicron PM_x fractions (PM_{10}, $PM_{2.5}$ and PM_1, respectively) at various environments, are of great importance; especially if they are performed at areas, such as the Eastern Mediterranean region, where PM_{10} levels are among the highest reported ones for Europe. To the best knowledge of the authors this is the first time that $PM_{2.5}$ and PM_1 levels are reported for Cyprus; such studies are clearly missing in the area. In addition, the phenomenology of each size fraction is reported including annual trends, when multi-year long observations were available, along with diurnal and seasonal cycles. The effect of air mass origin on PM_{10} is also examined and the contribution of local anthropogenic emissions, with respect to $PM_{2.5}$ and PM_{10}, in urban and industrial areas are additionally discussed.

Several conclusions have been reached from this study:

- If the entire period under investigation is considered (1998–2015), the average annual PM_{10} of 28.7±5.0 $\mu g\,m^{-3}$ observed at regional background sites is higher than the majority of rural sites in central and northern Europe and the west Mediterranean basin. Similar PM_{10} levels have been observed for the three major urban centers of Cyprus (Nicosia, Larnaca and Limassol) even though the population of Nicosia is almost twice that of Limassol and a three-fold higher than of Larnaca indicating significant contribution from regional sources. More specifically, the annual average PM_{10} concentration for the common period of measurements (2006–2015) at the kerbside sites of Nicosia, Larnaca and Limassol were equal to 47.3 ± 7.6 $\mu g\,m^{-3}$, 49.0 ± 7.4 $\mu g\,m^{-3}$ and 49.2 ± 7.2 $\mu g\,m^{-3}$, respectively. The respective annual averaged PM_{10} at Paphos city and Zygi industrial site were 20% lower compared to the above kerbside stations and equal to 40.5 ± 7.2 $\mu g\,m^{-3}$ and 41.7 ± 7.3 $\mu g\,m^{-3}$, respectively.
- Dust from regional transport from eastern Asia and Northern Africa is by far the most important natural source in the area contributing up to 33.6 ± 5.2% or about 10 $\mu g\,m^{-3}$ to the annual PM_{10} levels reported in background stations; the latter underlines the significant contribution of natural sources on the ambient PM_{10} amounts in the eastern Mediterranean region.
- To assess the importance of regional sources, seasonal PM_{10} averages from 1998 to 2015 at the background site of Agia Marina have been calculated individually for each of the three source regions representing anthropogenic sources, clean air masses and dust emissions from both E. Asia and N. Africa. Air masses from polluted source regions exhibited on average 8.0% higher PM_{10} amounts compared to the measured annual averages only during the summer months (June–August). During the remaining year, PM_{10} associated with these source regions was on average 31.7% lower. As a result, if air masses would reach Cyprus exclusively from the polluted sector it would result on an annual average PM_{10} value of 22.4 ± 4.0 $\mu g\,m^{-3}$ or ~20% smaller compared to the measured PM_{10} (28.7 ± 5.0 $\mu g\,m^{-3}$) The annual averaged PM_{10} would further reduced at 20.0 ± 2.6 $\mu g\,m^{-3}$, if only air masses from the clean sector would reach Cyprus. Finally, if air masses would exclusively originate from the Middle East and N. Africa regions, annually averaged PM_{10} values of 49.9 ± 11.7 $\mu g\,m^{-3}$ (75% above the measured PM_{10} levels) would have been observed.
- A significant ($p < 0.001$) decreasing trend of 0.7 ± 0.1 $\mu g\,m^{-3}\,y^{-1}$ was observed when both PM_{10} (1998–2015) and $PM_{2.5}$ (2005–2015) annual values are considered, indicating contribution from both natural and anthropogenic sources to this tendency. By considering that the PM results obtained at Agia Marina are representative of the regional influence, the local influence of the urban and industrial sites on the PM_{10} levels can be estimated. On average, 44.0 ± 9.7%, 44.3 ± 9.7%, 45.9 ± 13.1%, 33.9 ± 12.6%, 35.5 ± 11.6% of the observed PM_{10} levels in Nicosia, Larnaca, Limassol, Paphos and Zygi, respectively are expected to originate from local anthropogenic and/or natural emissions including vehicle, biomass-burning, shipping emissions (in Limassol), airport related emissions (in Larnaca), resuspension of dust and sea-salt (in coastal locations).
- These local emissions are almost equally distributed between the fine and coarse fractions as 50–60% of the local PM_{10} amounts are due to coarse particles emissions (with the higher amounts observed at coastal sites). The above results highlight significant emissions from both fine mode (e.g. residential heating in winter and traffic all year round) and coarse mode urban emissions (e.g. dust resuspension, wear and tear in brakes and tires, respectively) in urban and industrial locations in Cyprus.

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