Temporal patterns and trends of particulate matter over Portugal: a long-term analysis of background concentrations

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Abstract

Air quality management regarding PM concentrations in the atmosphere is a complex problem to tackle. In this paper, we aim to characterize the temporal patterns and trends of aerosol background levels over Portugal. Hourly data from the national air quality monitoring network, gathered from 2007 to 2016, is analysed using statistical methods. Data from 20 monitoring stations was processed to prepare datasets with different time scales, and results were grouped by their type of surrounding area (urban, suburban or rural). Urban and suburban background sites are characterized by strong seasonal patterns, with higher monthly mean concentrations in winter than in summer. In contrast, rural background PM10 concentrations are highest during August and September. This study suggests that urban background concentrations are significantly influenced by anthropogenic non-combustion sources, which contribute to the coarser aerosol fraction (PMc). PMc is about 3 $\mu g \, m^{-3}$ higher during week-days than during Sundays, at urban sites. However, there is no clear relationship between the value of the PM2.5/PMc ratio and the type of monitoring station. During the 10-year period of study, a decrease of 1.83%/year, 3.58%/year and 4.89%/year was registered in PM10 concentrations at Portuguese rural, urban and suburban areas, respectively. Despite the higher decrease at suburban monitoring stations, those sites present the highest 10-year mean PM10 concentrations. This work provides an import insight on temporal variations of PM10, PM2.5 and PMc concentrations over Portugal and summarizes trends through the last decade, contributing to the discussion on sources and processes influencing those concentrations.

Keywords: particulate matter, Portugal, seasonal patterns, air quality, PM2.5/PMc ratio

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1. Introduction

Atmospheric aerosols affect air quality and climate, their impacts have been evidenced in many studies across urban, regional, and global scales (Calvo et al. 2013; Fuzzi et al. 2015). Furthermore, exposure to atmospheric aerosols may affect human health (WHO-OCDE 2015). In the past decades, many studies highlighted the role of ambient airborne particulate matter (PM) as an important environmental pollutant for cardiopulmonary diseases and lung cancer (e.g. López-Villarrubia et al. 2016; Valavanidis et al. 2008).

In addition to primary emission sources, PM concentrations can be significantly influenced by secondary aerosol formation in the atmosphere, which is not only dependent on the precursor emissions but also on meteorological conditions and geographical location. Furthermore, in addition to local pollution sources, air quality is affected by long-range transport of air masses that contribute to regional background pollution levels and air pollution episodes. In this context, North African dust outbreaks may influence air quality in Europe, especially on the Mediterranean Basin (Pey et al. 2013). Thus, air quality management regarding PM concentrations in the atmosphere is a complex problem to deal with.

Due to the geographic location of Portugal and the dominant wind regime (Valverde et al. 2015), influenced by the presence of the semi-permanent Azores high-pressure and the Icelandic low-pressure systems over the North Atlantic Ocean, it is expected that, most of the year, transport of maritime air promotes a decrease of anthropogenic and mineral PM concentrations (Almeida et al. 2013). Nevertheless, Portugal has been facing air quality problems, being PM10, together with O_3, the main critical pollutants. Furthermore, results from Monteiro et al. (2017b) evidence that human health protection will be even more critical in the future, particularly for PM; in 2050, due to the warmer and dryer conditions and the expected increase of background concentrations, degradation of air quality is expected to occur.

Several studies intend to characterize PM long-term trends in European regions (e.g., Barmpadimos et al. 2012; Cusack et al. 2012; Guerreiro et al. 2014). However, to our knowledge, no study was published focusing on long-term detailed analysis for Portugal. Specifically for Portugal, PM levels were previously characterized by Cruz et al. (2016), throughout a 3-year period only. Other studies focused on the characterization of PM levels and composition over

specific cities or regions, based on intensive experimental campaigns. PM composition and sources have been documented for Lisbon (Almeida et al. 2006, 2009) and Porto (Custódio et al. 2016; Diapouli et al. 2017).

The aim of this study is to provide a characterization of the aerosol background levels in Portugal, based on the long-term observations of PM10 and PM2.5 concentrations. Background levels or concentrations are pollution levels which are not influenced significantly by any single source, but rather by the integrated contribution from all upwind source areas. This is the first attempt to characterize aerosol over Portugal with such a long record (10 years) of observations and a large number of monitoring stations. The results of this study bring new insights on the temporal patterns and trends of PM levels over Portugal, and also on the distribution between fine and coarse particulate matter.

2. Methodology

Surface data from the Portuguese air quality monitoring network (see http://qualar.apambiente.pt/) is used in this paper to characterize PM levels in Portugal. We focus on PM10 and PM2.5 concentrations collected over the last 10 years, more precisely, observations recorded from January 2007 to December 2016. The aerosol coarse fraction, PMc (the size fraction between 2.5 and 10 μm), was estimated through the subtraction of PM2.5 from collocated PM10.

Air quality data is analysed in terms of temporal patterns, long-term trends and legal limit compliances. Temporal patterns (which include daily, weekly and monthly cycles) and long-term trends of concentrations were performed using the OpenAir package for R (Carslaw and Ropkins 2012; Ropkins and Carslaw 2012).

2.1. Data

In Portugal, the air quality stations from the national air quality monitoring network are classified by their type of influence (traffic, industrial or background) and the area or environment type: (urban, suburban or rural). Within the scope and aim of the present study, we consider the background influenced sites, e.g., sites which are not influenced significantly by any single source, but rather by the integrated contribution from all upwind source areas. Background sampling points shall, as a general rule, be representative for several square kilometres. Sites from Azores

and Madeira islands are not included in this characterization.

Data quality objectives for air quality assessment are defined in the EU Directive 2008/50/EC (EU 2008) as a percentage of hourly data availability along the year. The minimum percentage of data capture for PM10 and PM2.5 is 90%. Despite the minimum 90% of data quality objectives, in some cases, measurements from stations with a minimum annual data capture of 75% are used to achieve data continuation over time. Between 2007 and 2016, we could gather data for 8 rural, 4 suburban and 8 urban sites, distributed along mainland Portugal (see Figure 1, which depicts the 10-year mean concentrations observed in each of the monitoring stations). Details on the location and classification of the stations used can be found in Table 1, together with the information on data completeness relative to the period 2007-2016.

[Table 1 about here.]

[Figure 1 about here.]

Among the selected stations, 13 only measure PM10 and 7 (4 rural and 3 urban) measure both PM10 and PM2.5 concentrations. All the selected monitoring stations are automatic and use the beta attenuation method to measure PM10 and PM2.5 concentrations. This method stands on beta-ray absorption in a sample captured on filtering material. The difference between the beta-ray absorption of the exposed and non-exposed filtering material, which is proportional to the mass of the captured suspended particle matter, gives the information on its concentration. However, the data cannot always be considered equivalent to the manual gravimetric reference method, which is required in Europe for compliance measurements. Correction procedures are employed to obtain reference equivalent PM10 data series from automatic beta-attenuation monitors. Most of the monitoring stations use the Environnement S.A MP101M dust monitor. FRO and VCO use the Verewa F-701-20 Ambient Air Dust Concentration Monitor, and MVE and TER use the Thermo ESM Andersen ESM FH 62 IR.

2.2. Methods

By simply plotting data in different ways, a visual analysis can often provide valuable insights. As such, a comparison of PM concentrations between different locations or monitoring station

types can reveal for example, information concerning the likely sources.

Hourly measurements from the selected monitoring stations were processed to prepare datasets with different time scales. Moreover, in this analysis, data were grouped according to station type. In the study of PM temporal cycles or patterns, four different plots are produced: (i) day of the week variation, (ii) mean hour of day variation, (iii) a combined hour of day – day of the week plot and (iv) a monthly plot. In these plots, relative to PM10, PM2.5 and PMc concentrations, the mean and the 95% confidence interval in the mean are depicted, relative to the 10 years data, aggregated by the time scale and the station classifications. The daily cycle of PM concentrations (mean hour of the day variation) is also plotted split by season.

PM10, PM2.5 and PMc long-term temporal trends have been estimated with the Openair's TheilSen function, which quantifies monotonic trends in unit/year, and calculates the associated p value through bootstrap simulations. Trend is estimated for mean monthly values, and the 95% confidence interval of the slope is presented. In this analysis, data has been deseasonalized using the seasonal-trend decomposition procedure based on locally weighted scatterplot smoothing, LOESS (Cleveland et al. 1990).

3. Results and Discussion

3.1. Temporal patterns in PM concentrations

Urban and suburban sites are characterized by higher PM levels in comparison with rural ones (see Figure 1) and exhibit common typical temporal cycles. Figures 2, 3 and 4 show the monthly variability through an average year, the daily variability through an average week and the diurnal variability per average week and per average day of PM10, PM2.5 and PMc concentrations, respectively, observed between 2007 and 2016 and grouped by station type.

[Figure 2 about here.]

[Figure 3 about here.]

[Figure 4 about here.]

In general, during weekdays, urban and suburban sites show a dip in PM10 concentrations during the night (between 00h and 07h) which then rise between 07h and 10h, dropping again

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between 10h and 14h. During the afternoon, concentrations increase and the highest peak of the day is achieved between about 20h and 23h. This profile is similar to those found by Harrison et al. (2012) relative to specific UK urban background stations. On Sundays, on the same group of stations, the peak in the early morning is not significant (probably due to the absence of the morning rush-hour traffic) and concentrations reach their minimum values in the beginning of the afternoon (around 14h). They then rise until around 21h, achieving the highest peak of the day, which is, as expected, lower than during week-days.

The combined hour of day – day of the week plot for PM2.5 shows a similar cycle to the PM10 one. Apart from the magnitude of concentrations, the main differences rely on the morning rise of concentrations, which is weaker for PM2.5, and on the differences between week-days and Sundays. The larger difference, between week-days and Sundays, in the magnitude of PM10 concentrations during late evening peaks at urban sites, reflects the importance of coarse particles of anthropogenic origin in Portuguese urban areas. Figure 4 shows that PM coarse fraction is about to 3 μg m^{-3} higher during week-days than during Sundays, at urban sites.

The late evening peak shown in the mean hour of day plots is related with both the daily evolution of the atmospheric boundary layer, which gets thinner during the night, and evening contribution of domestic sources such as heating (Borrego et al. 2010; Gonçalves et al. 2012; Vicente et al. 2015) and cooking (Ots et al. 2016). In addition, according to Harrison et al. (2012), there might exist a contribution of semivolatile material condensing on ambient particles with the lower night-time temperatures. All these causes are more important during winter (due to thinner and more stable boundary layers, more emissions from heating, colder night-time temperatures), which may explain why the intensity of this late evening peak is much stronger during winter (see in Figure 5 the PM10 concentrations daily variability split by season).

[Figure 5 about here.]

It is at suburban sites that we find the larger differences between seasonally averaged daily cycles. On those stations, during winter, the PM10 late evening peak concentration is 20 μg m^{-3} higher than during summer. This fact suggests a greater use of residential wood combustion in urban suburbs than in city centres, as it happens in other European cities such as London (Fuller et al. 2014) and Berlin (Wagener et al. 2012). It is important to notice that in this analysis we are

including monitoring stations with different geographical locations (see Figure 1): the 4 suburban monitoring stations are located in the North of Portugal (in Aveiro, Porto and Braga districts), while the other 8 urban monitoring stations, 6 are located in Great Lisbon region (Lisbon and Setúbal districts), one in Coimbra and another one in the Porto district. The different geographical locations might as well contribute to the differences found between PM concentrations measured in urban and suburban groups of stations.

As shown in Figures 2 and 5, rural sites present higher PM10 levels during summer. Most of the rural sites (FPO, FRN, FUN and TER) exhibit pronounced seasonal patterns, with the highest PM10 concentrations observed during August or August/September, when the monthly mean PM10 concentrations are approximately 6 to 8 μg m^{-3} higher than the rest of the year. Cruz et al. (2016), who analysed PM levels in Portugal throughout a 3-year period, attributed this behaviour to the influence of high-altitude mountain stations. In that study, only two monitoring sites (FUN and Douro Norte, a station which is not considered in our characterization due to non-compliance with the data coverage threshold) present the highest PM10 concentrations during summer. The authors relate this with enhancement of secondary particulate formation, arising from photochemical reactions between biogenic VOC compounds and anthropogenic precursors transported from populated coastal areas in the north and the centre of the country.

In our results, we also observe this seasonal trend in low altitude stations (FPO and TER). We agree that the summer maximum might partially result from photochemically driven secondary formation of aerosols (Alves et al. 2001), which is, according to our results, not exclusive to high-altitude stations. Figure 3 also shows an increment in rural PM2.5 concentrations during summer. However, our data analysis suggests, with PMc higher summer levels at rural sites (see Figure 4), that other factors are influencing particulate mass increase in the rural summer atmosphere. Possible reasons which may also contribute for the PM10 summer maximum are local dust emissions from dry soils, the impact of forest fires and long-range transport of African desert dust. In addition, differences in the precipitation patterns between winter and summer may play a huge role in PM concentrations, due to the effect of precipitation scavenging on PM removal from the atmosphere.

Other sites, such as ERM, ERV, FRO, ILH and VCO are characterized by higher average PM10 concentrations during winter. In this group of stations all types of areas are included: urban (ERM),

suburban (FRO, ILH and VCO) and rural (ERV).

3.2. Relationship between PM2.5 and PM10 and between PM2.5 and PMc

PM2.5 and PM10 mass concentrations are correlated and slope values for individual sites range from 0.44 to 0.54 (not shown). These values are lower than the ones calculated by Dingenen et al. (2004) for 11 European sites, using "binned" PM2.5 and PM10 24-h values, which range from 0.57 to 0.89. Putaud et al. (2010) observed "binned" PM2.5/PM10 ratios between 0.44 and 0.90, in 34 European sites, having not found any clear relationship between PM2.5/PM10 ratio and the type of site or its location in Europe. In our study, we also found no clear relationship between the value of the PM2.5/PM10 ratio and the type of monitoring station. Ratios computed for individual stations range from 0.44 to 0.52 for urban and from 0.44 to 0.54 for rural stations.

Lower PM2.5/PM10 ratios are associated with spatial and temporal conditions where the observed PM levels are dominated by coarse particles. Higher ratios are observed at sites where secondary aerosol sources, which produce fine particles, are predominant (Dingenen et al. 2004), or where fuel combustion is amongst the main sources of particulate matter emissions. The values we calculated for the Portuguese background monitoring stations indicate that we have a significant contribution of the coarser fraction to the total PM10.

In order to analyse with more detail the fine and coarse contribution to the total aerosol mass, Figure 6 shows the daily variability split by season of the PM2.5 and PMc concentrations, at rural and urban background stations. In general, background stations exhibit similar values of PM2.5 and PMc concentrations.

[Figure 6 about here.]

PM2.5 concentrations are higher during winter. The relative contribution of the PM2.5 fraction to the total aerosol mass is on average higher in rural than in urban background stations (mainly during winter). In other words, Portuguese urban background stations show a contribution of anthropogenic emissions to the PMc concentrations. These findings are in accordance with Almeida et al. (2006), which found that during autumn/winter, a predominant fraction of coarse soil dust observed in an urban area (in the outskirts of Lisbon) originates from anthropogenic activities. Most cities with pollution from transport and other combustion sources show a non-combustion source which is mainly attributed to dust re-suspension (from vehicle non-exhaust

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sources, construction activities, soil dust, etc.), which forms part of the coarser fraction PMc.

Figure 7 shows PM2.5 vs. PMc daily mass concentrations for the 7 sites with data from 2007 and 2016. There is no clear relationship between the value of the PM2.5/PMc ratio and the type of monitoring station. At urban sites, the correlation coefficient between fine and coarse particulate matter concentrations indicates a robust relationship between the two variables. This relationship may denote similar PM sources in the atmosphere.

[Figure 7 about here.]

The rural sites ERV and FUN show lower values of correlation between PM2.5 and PMc. The ERV monitoring station is located 5 km from the seashore, which means that it might be influenced by sea-spray. However, sea-spray influences mainly large particles (Cesari et al. 2018; Slezakova et al. 2007). The scatter plot of PM2.5 and PMc concentrations shows a large number of points with larger PM2.5 than PMc concentrations, which indicates that sea-spray is not the main contributor in these cases.

3.3. Long term temporal trends

In previous sections PM temporal patterns were presented. Here, the trend in PM concentrations through the last decade are discussed. Figure 8 shows the trend in PM2.5, PM10 and PMc concentrations observed at background stations between 2007 and 2016, according to type of station. Statistically significant decreasing trends (p<0.001) are estimated for the evolution of average PM10 concentrations of each area type (rural background, suburban background and urban background) for the last ten years. The decrease of concentrations is stronger in urban (-0.96 μg m^{-3}/year) and suburban (-1.46 μg m^{-3}/year) stations than in rural ones (- 0.35 μg m^{-3}/year); while in 2007 there was a large difference between PM10 levels observed at suburban, urban and rural stations, ten years later, in 2016, the three types of stations measure a similar range of mean concentration values. PM10 concentrations decreased by 1.83%/year, 3.58%/year and 4.89%/year at Portuguese rural, urban and suburban areas, respectively, between 2007 and 2016.

[Figure 8 about here.]

Trend estimates for PM2.5 and PMc concentrations are statistically significant (p<0.001) at rural background stations and urban background stations, respectively. Over the last decade and

according to our results, mean PM2.5 concentrations at rural background stations registered a reduction of approximately -0.25 µg m^{-3}/year. Urban stations registered a huge decrease (-0.79 µg m^{-3}/year) in the aerosol coarse concentrations, PMc.

The improvement in the air quality might be partly related to the Portuguese economic crisis. Monteiro et al. (2017a) found a relationship between the reductions in energy consumption (registered at transport, industry and residential sectors) and PM10 concentrations in Lisbon and Porto municipalities. Moreover, the economic crises severely affected construction activity within cities, which may positively affect PM10 concentrations due to a reduction in dust production and re-suspension.

4. Conclusions

The aerosol background levels in Portugal are analysed based on data gathered from 2007 to 2016. Statistically significant decreasing trends (p<0.001) are observed in the average PM10 concentrations of each area or environment type. The decrease of concentrations is higher in urban (-0.96 µg m^{-3}/year) and suburban (-1.46 µg m^{-3}/year) stations than in rural ones (-0.35 µg m^{-3}/year). Our analysis shows that the main factor contributing to the PM10 decrease in urban areas is the decrease in the PMc concentrations.

PM10 and PM2.5 concentrations are characterized by typical temporal patterns. Throughout the day, concentrations follow the expected patterns with rural sites exhibiting less variability and urban and suburban sites characterized by two main peaks during the day: one during the morning, and the largest one late in the evening.

The results of this study point out the importance of coarse particles of anthropogenic origin in Portugal. This conclusion is supported by the larger differences between weekdays and Sundays in PM10 rather than PM2.5. PMc is approximately 3 µg m^{-3} higher during week-days than during Sundays, at urban sites.

It is on suburban sites that the larger differences between the PM10 characteristic daily cycle of each season are found. In this group of stations, the highest daily concentrations (observed at late evening / during the night) during winter are on average 20 µg m^{-3} higher than during summer. PM2.5 concentrations could not be assessed at suburban stations.

The methodology implemented in this work allows to obtain deeper characterization of PM

levels in Portugal and provides a different view on pollution patterns that was not identified in previous studies. Namely, our results suggest that rural background sites in Portugal reveal higher average PM10 concentrations during summer periods. This finding is relevant in the context of identification of main pollution sources and definition of future pollution abatement strategies. This work provides important insights on temporal variations of PM10, PM2.5 and PMc concentrations over Portugal and summarizes trends during the last decade. Our study contributes to the characterization of background levels of particulate matter and to the ongoing discussion on sources and processes influencing those concentrations.

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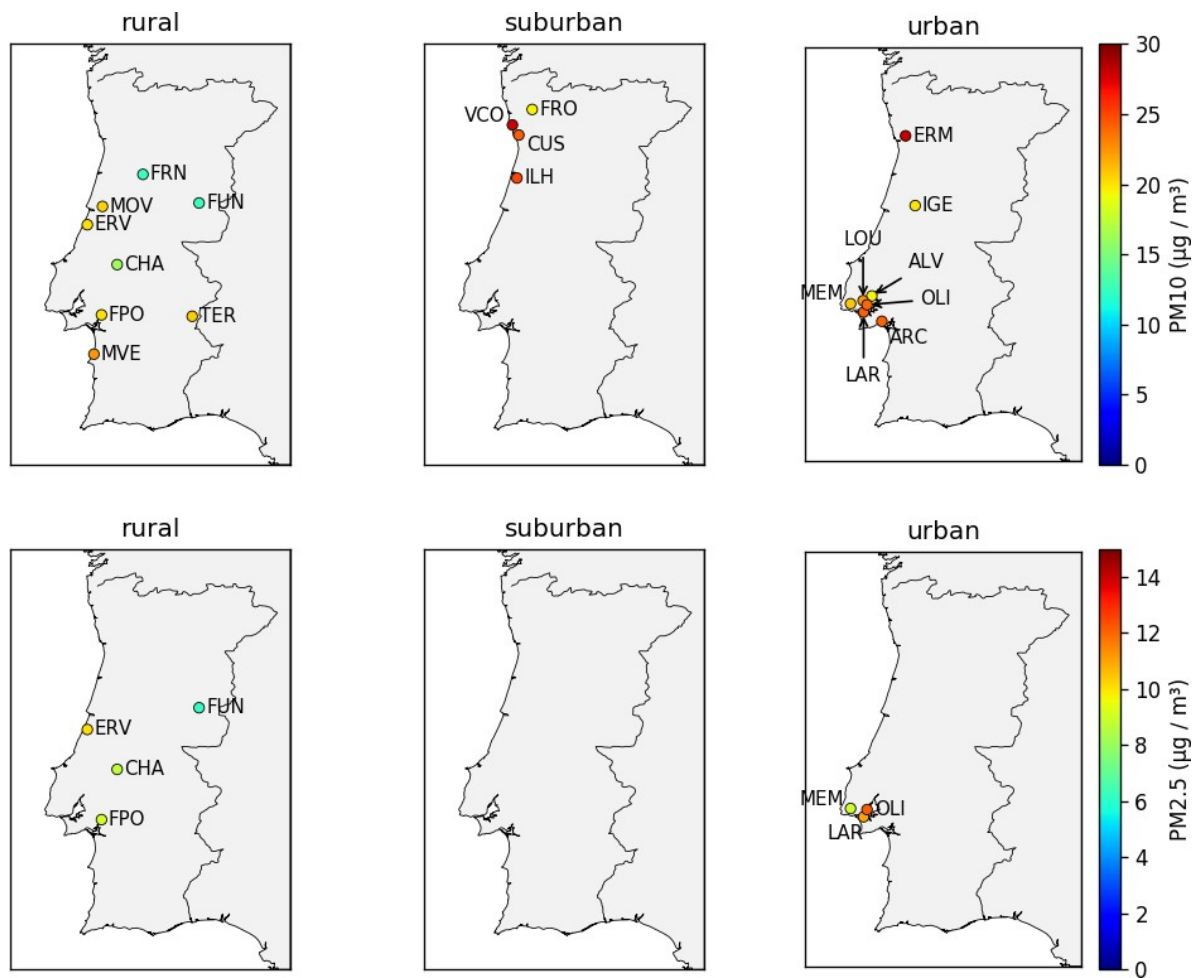
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Figure 1: Spatial variation of the PM10 and PM2.5 mean concentrations observed at background stations between 2007 and 2016, according to the monitoring station environmental classification.
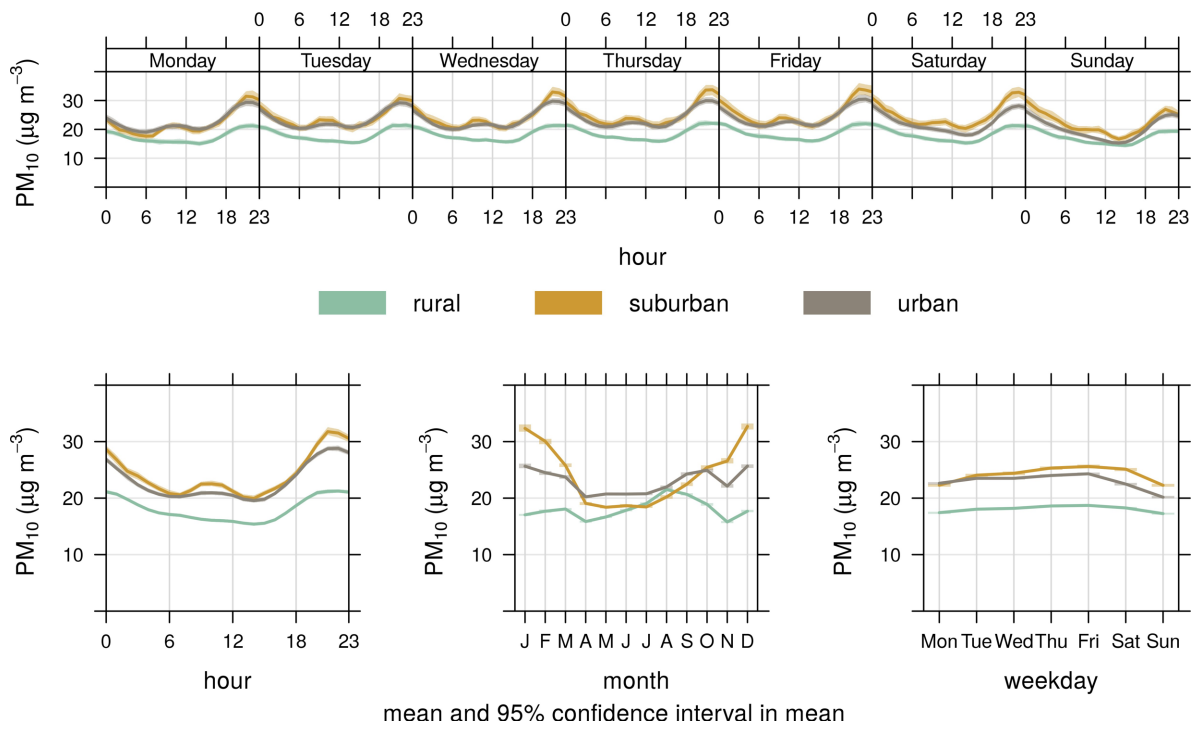
Figure 2: Hourly, daily and monthly variability of the PM10 concentrations observed at background stations between 2007 and 2016.
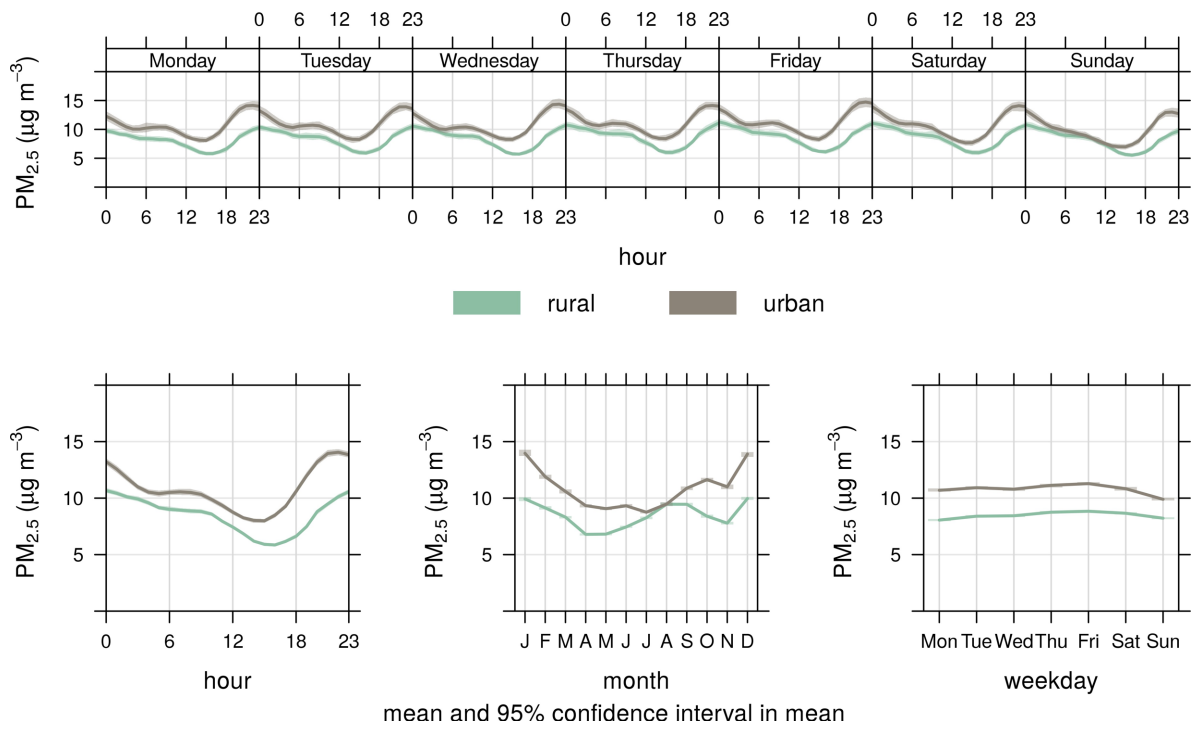
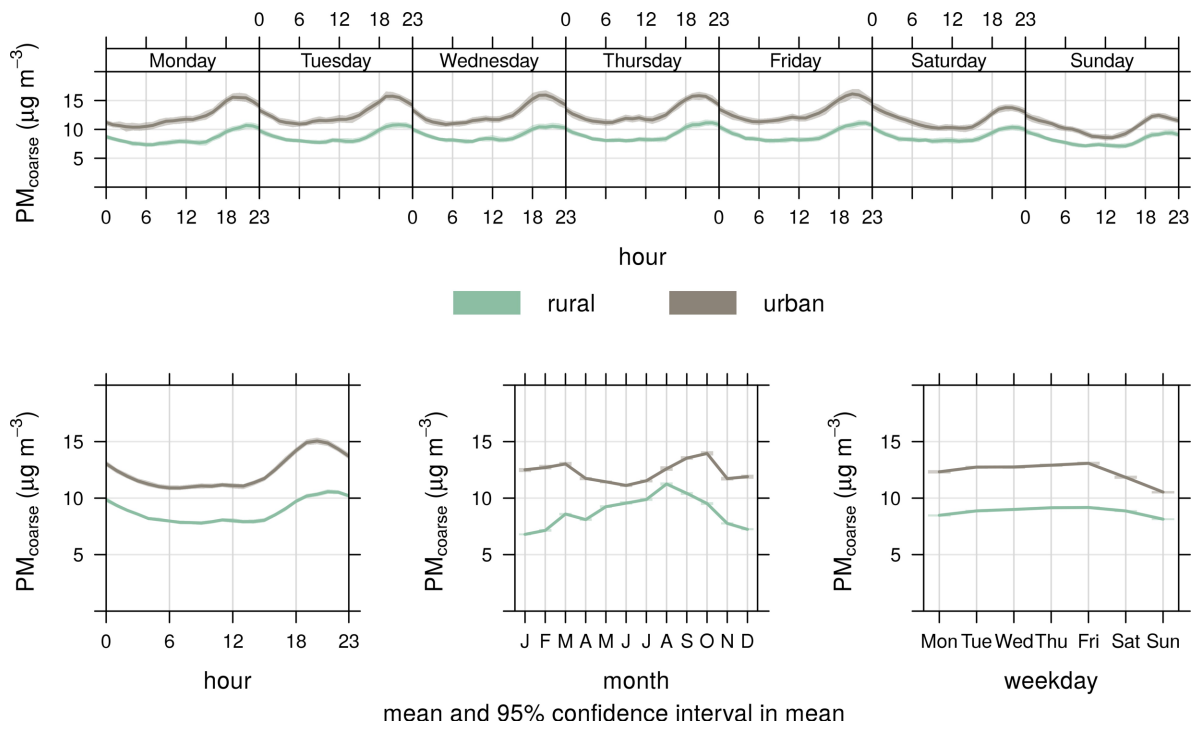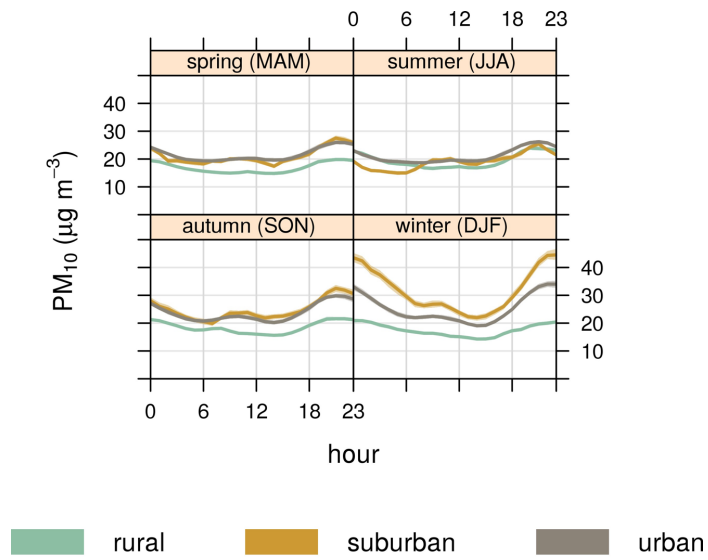Figure 3: Hourly, daily and monthly variability of the PM2.5 concentrations observed at background stations between 2007 and 2016.

Figure 4: Hourly, daily and monthly variability of the PMc concentrations estimated at background stations between 2007 and 2016.

Figure 5: Daily variability split by season of the PM10 concentrations observed at background stations between 2007 and 2016.
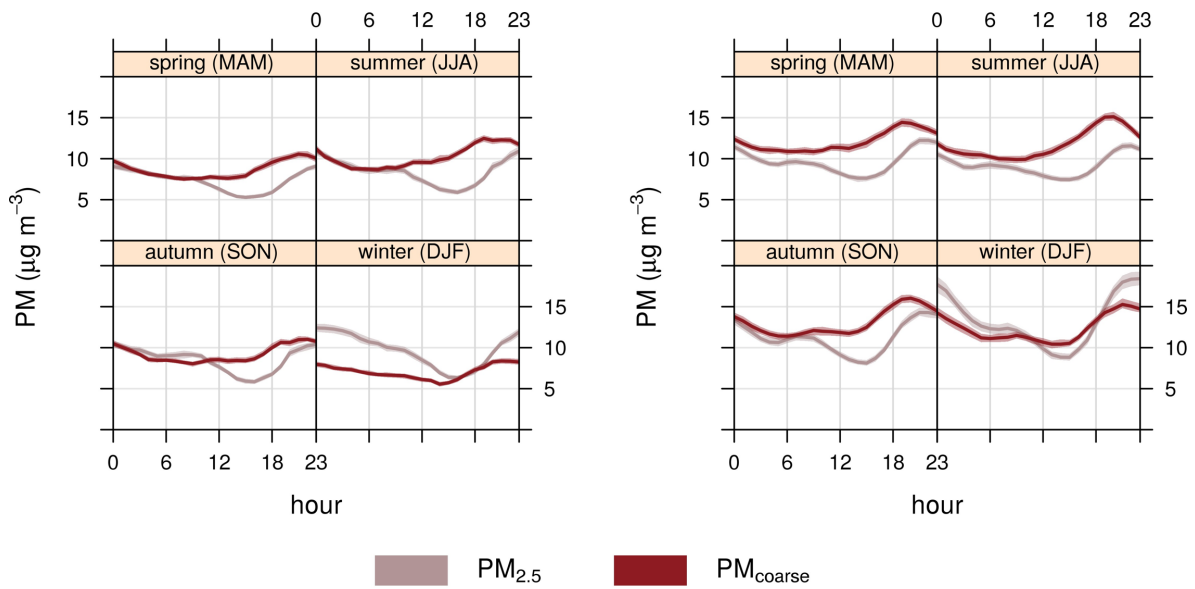
Figure 6: Daily variability split by season of the PM2.5 and PMc concentrations observed at rural (left plot) and urban (right plot) background stations between 2007 and 2016.
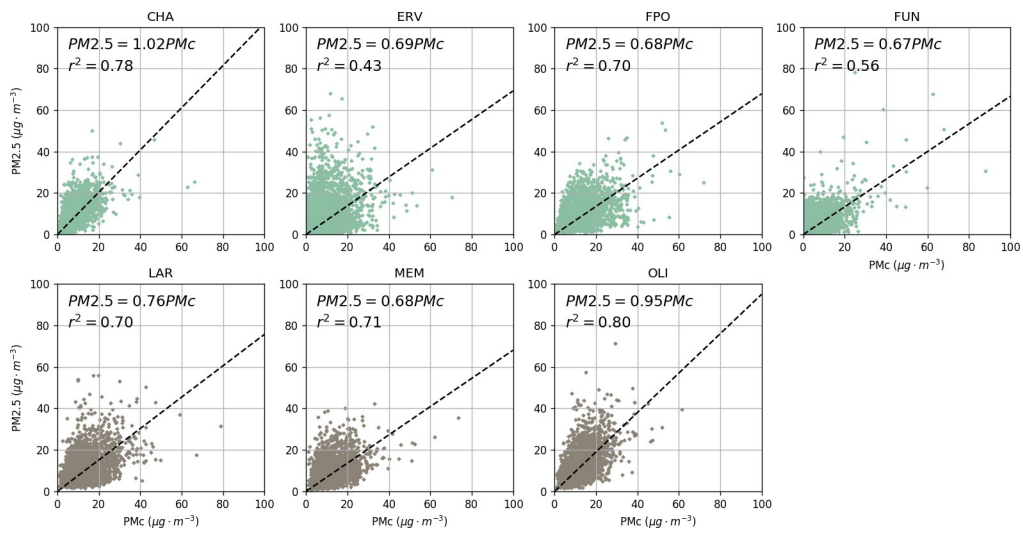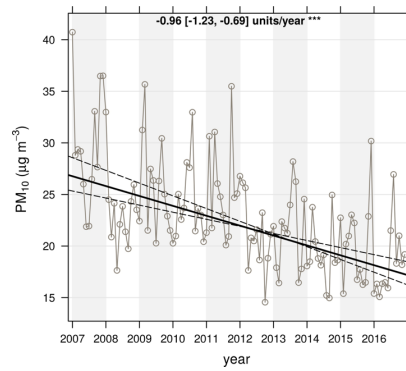
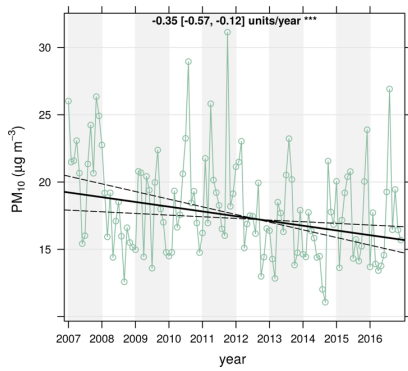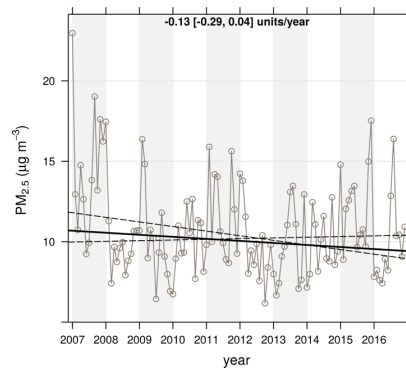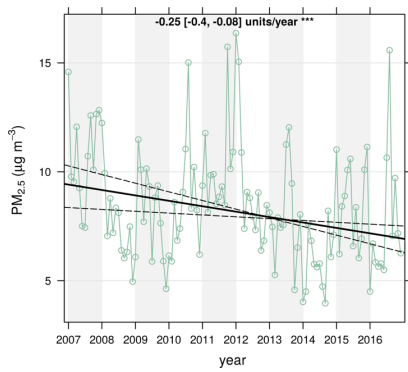Figure 7: Ratio between PM2.5 and PMc concentrations observed at individual background monitoring stations between 2007 and 2016. Rural stations data is represented with green dots in the top panel while data from urban stations is represented by grey dots in the bottom panel.
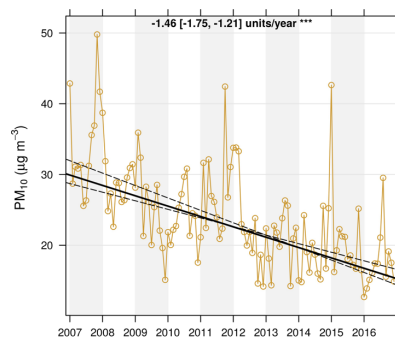
(a) PM10 at rural background stations. (b) PM10 at urban background stations.

(c) PM2.5 at rural background stations. (d) PM2.5 at urban background stations.
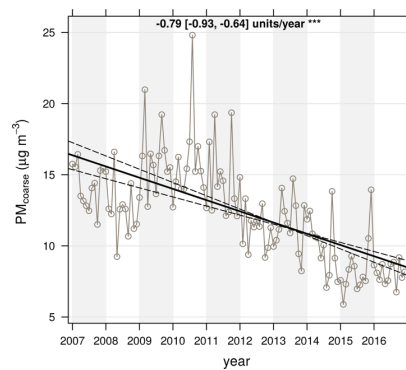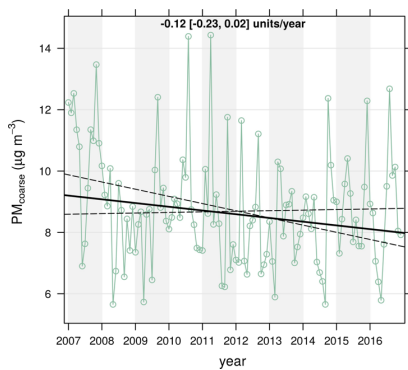
(e) PMc at rural background stations.   (f) PMc at urban background stations.

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(g) PM10 at suburban background stations.

Figure 8: Trends in PM10, PM2.5 and PMc at background stations. The solid black line shows the trend estimate and the dashed lines show the 95% confidence intervals for the trend based on resampling methods. The overall trend and the 95% confidence intervals in the slope are shown at the top of each plot. The *** are shown when the trend is significant to the 0.001 level.

List of Tables

Table 1: List of QUALAR background stations with data completeness > = 75% during the period 2007-2016. Coordinates and surrounding environment type classification are included.

Code	Name	LON	LAT	height (m)	Classification	PM10 data	PM2.5 data
ALV	Alverca	-9.040	38.896	22	urban	78.0 %	-
ARC	Arcos	-8.894	38.529	2	urban	79.1 %	-
CHA	Chamusca	-8.468	39.353	143	rural	95.9 %	92.2 %
CUS	Custóias Matosinhos	-8.645	41.201	100	suburban	82.6 %	-
ERM	Ermesinde Valongo	-8.551	41.217	140	urban	83.6 %	-
ERV	Ervedeira	-8.893	39.922	60	rural	93.4 %	92.9 %
FPO	Fernando Pó	-8.691	38.636	57	rural	87.3 %	84.3 %
FRN	Fornelo do Monte	-8.100	40.640	741	rural	95.9 %	-
FRO	Frossos - Braga	-8.454	41.566	51	suburban	87.7 %	-
FUN	Fundão	-7.300	40.232	473	rural	97.2 %	94.0 %
IGE	Inst. Geof. Coimbra	-8.412	40.206	145	urban	93.8 %	-
ILH	Ílhavo	-8.672	40.588	32	suburban	92.7 %	-
LAR	Laranjeiro	-9.159	38.663	63	urban	94.2 %	78.1 %
LOU	Loures-Centro	-9.166	38.828	10	urban	86.1 %	-
MEM	Mem Martins	-9.348	38.784	173	urban	87.3 %	84.5 %
MOV	Montemor-o-Velho	-8.677	40.183	96	rural	87.9 %	-
MVE	Monte Velho	-8.799	38.076	53	rural	94.4 %	-
OLI	Olivais	-9.109	38.768	32	urban	90.6 %	87.7 %
TER	Terena	-7.398	38.616	187	rural	82.8 %	-
VCO	Mindelo V. Conde	-8.736	41.345	25	suburban	82.9 %	-