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NO_2, PM10 and O_3 urban concentrations and its association with circulation weather types in Portugal

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HIGHLIGHTS

- Objective classification scheme of the atmospheric circulation affecting Portugal.
- Synoptic characteristics and frequency of 10 circulation weather types (CWTs).
- Circulation-to-environmental approach.
- Identification of links between CWTs and air pollution episodes.
- Higher-than-average concentrations are predominantly associated to eastern CWTs.

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ABSTRACT

High levels of atmospheric pollutants are frequently measured in Portugal, a country which has been affected by several pollution episodes, exceeding PM10, O_3 and NO_2 legal limits repeatedly during the last decade. The occurrence of these episodes is often related to either local-scale conditions or regional-scale transport. In order to better understand the atmospheric factors responsible for poor air quality, the relationships between air pollution and meteorological variables or atmospheric synoptic patterns represent an important research area.

Here an objective classification scheme of the atmospheric circulation affecting Portugal, between 2002 and 2010, is presented, where daily circulation is characterized through the use of a set of indices associated with the direction and vorticity of the geostrophic flow in the lower atmosphere. The synoptic characteristics and the frequency of ten basic circulation weather types (CWTs) are discussed and a framework that permits the identification of the main characteristics associated to the occurrence of pollution episodes is mapped based on the identified patterns. The relationship between CWTs and poor air quality allowed distinguishing between which types are most frequently associated to pollution episodes. It is shown that the anticyclonic and north types, although being the most frequent classes during the majority of the year, do not prevail during pollution episodes that are dominated by easterly types. In general, higher concentration of all three pollutants and the two extreme events analysed occur associated predominantly with synoptic circulation characterized by an eastern component and advection of dry air masses. Moreover, results on the link between CWTs and air quality for Lisbon and Porto urban areas suggest that air quality regimes are generally similar for the northern and southern regions considered with the exception of spring and autumn PM10. Results obtained highlight the existence of strong links between the interannual variability of daily air quality and interannual variability of CWTs.

1. Introduction and background

Urban air pollution (UAP) has emerged as a major health, economic and social problem, since cities expand at unprecedented rates across the world. The complex mixture of toxic components in the urban atmosphere may induce acute and chronic responses

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from sensitive groups, such as children and people with previous heart and respiratory insufficiencies (Krzyzanowski et al., 2002; Díaz et al., 2004; Pope and Dockery, 2006; Martins et al., 2012a). According to a recent report by the Organisation for Economic Co-operation and Development (OECD), by the year 2050, outdoor air pollution is projected to be the world's top environmental cause of mortality, ahead of dirty water and lack of sanitation (OECD, 2012). Facing this direct impact of UAP on human mortality and morbidity, exposure to pollutants is currently a key environment-related health concern (EEA, 2013). UAP is determined by the combination between different factors, namely, emissions, physical constrains and meteorological conditions (e.g. Fenger, 1999; Demuzere et al., 2009; Pearce et al., 2011). Meteorological conditions importance in constraining the atmospheric processes of dilution, transformation, transport and removal of pollutants has been referred previously by several authors (e.g. Boubel et al., 1993; Baumbach, 1996; Dayan and Levy, 2002; Pearce et al., 2011). In order to better understand the atmospheric factors which are responsible for poor air quality (AQ), the relationships between meteorological variables or synoptic patterns and surface air pollution have become an important research area.

Over the last few decades, the association between weather conditions and air pollution has been investigated extensively. Early work funded by European Union projects (MECAPIP, RECAPMA, SECAP) has been discussed by Millán et al. (1996, 1997), focussing on photooxidant dynamics in the Iberian Peninsula and in the Mediterranean basin. Millán et al. (2000) presented an overview of previous studies focused on the atmospheric physical and chemical processes that govern the dynamics of air pollutants in the Mediterranean basin and in the Iberia Peninsula. A number of authors have investigated the relationships between weather conditions and air pollution in Portugal (Barros et al., 2003; Carvalho et al., 2006; Evtyugina et al., 2006; Borrego et al., 2013). The tools used in the studies for Portugal and for other geographical regions include atmospheric and air pollution models (e.g. Carvalho et al., 2006; Pearce et al., 2011; Borrego et al., 2013) or synoptic patterns and back-trajectories approaches (e.g. Dayan and Levy, 2002, 2004; Carvalho et al., 2006; Demuzere et al., 2009; Saavedra et al., 2012), and were applied to historical data (e.g. Pearce et al., 2011; Saavedra et al., 2012), or to climate scenarios (e.g. Dias et al., 2012).

Preceding studies revealed that a number of meteorological variables can be relevant to characterise air pollution, namely: temperature, wind speed and direction, relative humidity, cloud cover, dew point temperature, sea level pressure, precipitation and mixing layer height (Hooyberghs et al., 2005; Demuzere et al., 2009; Pearce et al., 2011). However, the majority of the research focused on individual meteorological variables and non-automated procedures of variables' selection. The last two decades have witnessed a growing interest by meteorological and climatological oriented groups to develop automated classifications of regional atmospheric circulation patterns, usually entitled circulation weather types (CWT). These CWTs are generally specific to a given region as they result from the examination of synoptic weather data (e.g. sea level pressure (SLP) or geopotential height at 500 hPa), usually on regular gridded fields (Ramos et al., 2011). Most classification procedures are based on the application of statistical selection rules (e.g. cluster analysis and regression trees) but can also be based on the determination of physical parameters related with the prevailing atmospheric circulation pattern (e.g. Huth et al., 2008; Philipp et al., 2010; Beck and Philipp, 2010). Moreover their applications to environmental analysis have been adopted increasingly in recent years. In particular several authors have applied different methodologies to western Iberia (e.g. Zhang et al., 1997; Trigo and DaCamara, 2000; Santos et al., 2005). The purpose of these studies varies considerably but the focus tends to be threesome: i) climatic variability, including trends and extreme years, ii) environmental and iii) weather driven natural hazards. Thus CWTs objective classification has successfully been applied to Portugal mainland by Trigo and DaCamara (2000) and Santos et al. (2005), both studies linking CWTs to precipitation. Pereira et al. (2005) and Ramos et al. (2011) analyse the impacts of atmospheric circulation, respectively, on fire activity and on lightning activity over Portugal. Several applications to the Iberian Peninsula were mostly focused on climatic trends (Paredes et al., 2006; Lorenzo et al., 2008) or associated to outstanding drought events (García-Herrera et al., 2007) or very wet years (Vicente-Serrano et al., 2011), or even to assess the changes in present and future CWTs frequency (Lorenzo et al., 2011).

In recent years, several studies have been published establishing objective links between CWT and air quality (Dayan and Levy, 2002; Demuzere et al., 2009). In these applications it is expectable that each circulation pattern dictates the long-range transport, linking a particular air mass to dispersion conditions and also to the mesoscale meteorological behaviour that controls the regional transport of air pollution (Dayan and Levy, 2004). The concentrations of pollutants in the atmosphere have been shown to be linked to the occurrence of certain synoptic weather conditions (Demuzere et al., 2009; Carvalho et al., 2010; Saavedra et al., 2012) and to the regional wind flow pattern induced by mesoscale meteorological processes such as land-sea breezes (Dayan and Levy, 2004). A specific application to AQ in the Iberian Peninsula was developed by Saavedra et al. (2012), which presented a very detailed description of the relationship between synoptic pressure patterns and high-ozone incidents in northwest Galicia. Carvalho et al. (2010) adopts a different approach, not assuming an *a priori* objective CWT classification, but relating synoptic patterns to air quality. This study refers to the characterization of the atmospheric conditions, namely synoptic patterns anomalies and back trajectories clusters, which lead to ozone-rich episodes at one specific location in northern Portugal (Lamas de Olo). According to Carvalho et al. (2010), the analysis of the ozone concentrations indicates that northeast circulation pattern, together with the southern flow, are responsible for the occurrence of the highest ozone peak episodes. Moreover, this study also suggests that long-range transport of atmospheric pollutants is the main contributor to the ozone levels registered at Lamas de Olo. However, to the best of our knowledge there are no studies in the literature focussing on the application over the Iberian Peninsula of objective automatic classification procedures of weather types to the air quality. For detailed information on different CWTs classification and applications over different regions of Europe please see the revision works by Huth et al. (2008), Philipp et al. (2010) and Beck and Philipp (2010) and references therein.

The main objective of this paper is to present an automated classification of pre-defined and widely used CWTs affecting Portugal and provide a framework that is useful to characterise the occurrence of pollution episodes, namely its inter-annual and intra-annual variability, as well as during the occurrence of extremes. Here a detailed analysis is conducted for two densely populated areas (Porto and Lisbon) and two rural areas in Portugal.

The paper is organized as follows. Section 2 discusses the methodology defined for the automatic classification of atmospheric circulation and its application to atmospheric air quality, and describes the different data sets used in the analysis. In Sections 3.1 and 3.2 the air quality in the study area and the CWTs are characterized, while in Section 3.3 the relation between CWT and AQ is analysed. Section 4 is devoted to the analysis of two specific extreme episodes. Finally, Section 5 presents the main conclusions.

2. Data and methodology

2.1. Air quality data

The European Union (EU) has a solid legislation, developed over 30 years, that establishes a common demand level for environmental norms and practices in all Member States. The need to deliver cleaner air has been recognized for several decades with actions taken at national and EU level and also through active participation in international conventions. As a result, European authorities have produced legislation in order to reduce ambient air pollution, namely the Directive 2008/50/EC on ambient AQ and cleaner air for Europe. Directive 2008/50/EC corresponds to present day Europe's main air quality legislation for 12 regulated pollutants including nitrogen dioxide (NO_2), ground-level ozone (O_3), and particulate matter (PM10), regulating the existence of AQ monitoring stations and also imposing the need for mitigation plans whenever the legal values are exceeded.

For assessment and management purposes and complying with EU regulations, continental Portugal is covered by several air quality monitoring stations, with major incidence on areas where health protection and spatial coverage are fundamental, i.e. large urbanized regions. In Portugal, the two major urban areas are Lisbon and Porto, with a population of around 2 million and 1.2 million people, respectively. Both regions are located close to the Atlantic Ocean from where most of the moisture affecting western Iberia arrives (Gimeno et al., 2010) with the prevalent winds and low pressure systems, particularly in winter months (Trigo et al., 2002). Despite this impact of the ocean that diminishes the effects of aerosols and pollution, both regions have been affected by several pollution episodes in the last two decades, exceeding PM10, O_3 and NO_2 legal limits repeatedly (APA, 2008).

Lisbon's urban area is covered by a total of 8 urban background monitoring stations. The Porto region located in Northern Portugal includes a total of 6 background monitoring stations, 3 of which classified as urban stations, and 3 as suburban stations (Fig. 1). Besides these urban and suburban air quality monitoring stations, two rural background stations were also selected for analysis: Chamusca belonging to the Lisbon's Metropolitan Area located in Central Portugal, NE of Lisbon; and Lamas de'Olo located in the Northern Region, E of Porto (Fig. 1). The analysis of data from these two rural stations will complement the analysis conducted for the urban areas.

In the next section, data from the above mentioned background monitoring stations from 2002 to 2010 are analysed (Fig. 1). The dataset comprises PM10, O_3 and NO_2 air concentrations recorded hourly by the monitoring stations, from which daily mean concentrations were calculated for PM10 and NO_2 and the daily 8-hourly maximum mean was calculated for O_3. The monitoring stations were chosen with the expectation that, by selecting only background stations, non-local correlations would be more clearly revealed and that the confounding effect of local urban vehicular NO_x emissions would be limited (Demuzere et al., 2009).

Similarly to the situation that can be find in many other countries, the air quality monitoring database has changed over the years in the number of monitored pollutants and number of monitoring stations. The number of missing values registered also diminished during the last decade. The minimum data capture considered valid for air quality assessment on annual basis is 90% (Annex I, Directive 2008/50/EC). Additionally, 5% extra can be considered if the calibration and maintenance of instruments, adding up to a total minimum of 85% of valid measured data annually. Thus, in this study, the analysis was restricted to the period spanning between 2002 and 2010, considering only the stations which present less than 15% of missing values.

2.2. Large scale meteorological fields

Daily mean sea level pressure (MSLP), relative humidity and temperature at the 1000 hPa level, and geopotential height at the 1000 hPa level values were extracted from ERA Interim Reanalyses dataset (Dee et al., 2011) for the 1981–2010 period on a grid of 1° latitude by 1° longitude for Portugal (40°W–30°E, 20°–70°N). The period between 1981 and 2010 was used to perform a 30 year climatology that included the air quality period under analysis (2002–2010).

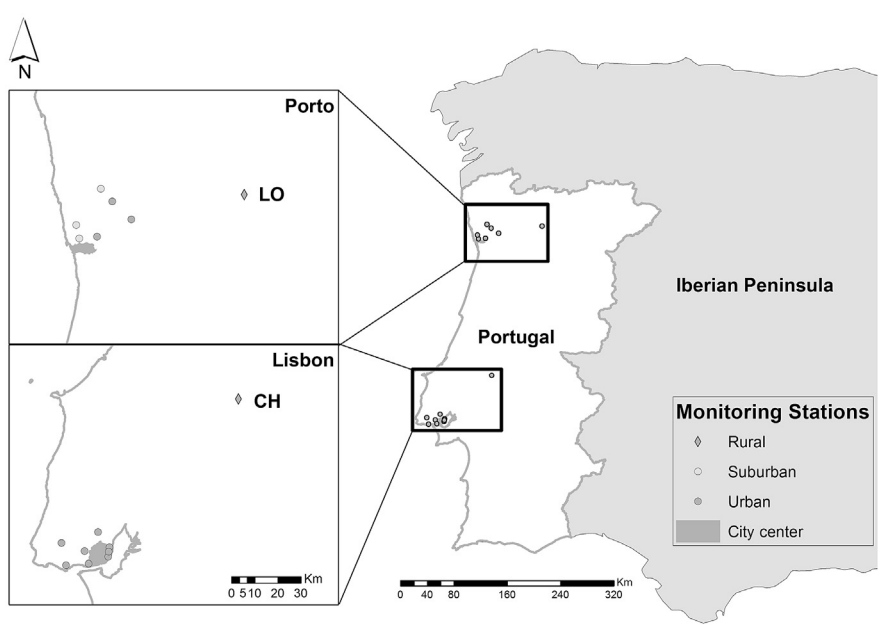

Fig. 1. Case study area.

2.3. Daily circulation weather type classification

Classification procedures based on the determination of circulation weather patterns and their applications for environmental analysis have been used frequently in recent decades (e.g. Trigo and DaCamara (2000), Demuzere et al. (2009), Ramos et al. (2011)). During a long period of time, classification procedures have been divided into manual and automated classifications (Yarnal, 1993). More recently, subjective versus objective procedures or even hybrid methods have been considered as the main dividing framework (Philipp et al., 2010). The use of so-called objective methods to classify CWTs, such as those based on indices derived from atmospheric pressure fields, represent an advantage over more subjective studies (e.g. classification of Lamb 1972). Thus, different ways and different sorts of circulation-based classifications may be used for describing atmospheric circulation at the synoptic scale. For a more detailed overview of the available methods often used in Europe see Philipp et al. (2010).

Based on the large-scale fields, prevailing CWTs at regional scale where determined using the simple geostrophic approximation according to the methodology proposed by Trigo and DaCamara (2000). Following the referred approach, 26 circulation weather types (CWT) are initially identified. However, in order to obtain a more structured and statistical representatively analysis, the 26 CWTs are reassembled in just 10 classes, excluding any undefined classification. Thus, only ten distinct CWTs are therefore considered, eight of which are driven by the direction of the flow (NE, E, SE, S, SW, W, NW, and N) and two by the shear vorticity (cyclonic C or anticyclonic A). A short description for the observed SLP features of each type is presented in Table 1 and a thorough description of this methodology including the simplification from 26 to 10 classes may be found in Trigo and DaCamara (2000).

The daily CWT for the 2002–2010 periods was computed based on Trigo and DaCamara (2000) approach by means of the daily SLP retrieved from ERA Interim.

2.4. HYSPLIT model application

In addition to the CWT classification method described previously, there are several other methods that can classify circulation weather patterns according to clusters of provenience classes, such as the backward air trajectories method. The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) (http://ready.arl.noaa.gov/HYSPLIT.php) is a complete simulation system able to determine air parcel trajectories, calculate single scattering processes and perform complex simulations of deposition. HYSPLIT uses historical meteorological data and dispersion of a pollutant is calculated by assuming either puff or particle dispersion (Draxler and Rolph, 2013; Rolph, 2013). The model has been applied recently to Saharan desert advection to the Iberian Peninsula (Negral et al., 2012) where air-mass backtrajectories for low AQ episode days or conductive to African dust outbreaks have been calculated to describe the pollutant sources and long-range transport. In the present work, the air-mass trajectories were estimated using the HYSPLIT model in order to complement the CWT classification, employing NCEP/NCAR reanalysis as meteorological input files. The HYSPLIT model was applied in order to compute backtrajectories for the analysis of specific air pollution episodes (Section 4). Ensemble backtrajectories were calculated for each episode, ending at 1000 m of height. The HYSPLIT trajectory ensemble option starts multiple trajectories from the selected starting/terminus location. Each member of the trajectory ensemble is calculated by offsetting the meteorological data by a fixed grid factor (one grid meteorological grid point in the horizontal and 0.01 sigma units in the vertical) and is represented in the figure by a track line. A tracking time of 24 h was applied and, for each episode the terminus of the trajectories is considered to be located at the sampling site (Lisbon, 38.77°N–9.13°W), and the arrival time corresponds to the previous midnight of the date of the maximum pollutant value measured. The model was run using the meteorological model's vertical velocity fields.

3. Results and discussion

3.1. Air quality characterization

The air quality data collected by the background monitoring stations in the regions of Lisbon and Porto (Fig. 1) and the evolution of the monthly values of NO_2, O_3 and PM10 were analysed.

In order to compare the seasonal cycle of the 3 pollutants at different monitoring stations in both study areas their intra-annual variability was analysed in a single figure. The box-plots of Fig. 2 show the monthly distributions of pollutants concentrations throughout the year for the entire studied period (2002–2010), with stations located in the Lisbon urban area in Fig. 2a and those from the Porto urban area depicted in Fig. 2b. As expected, the rural stations present very different monthly distributions of pollutants concentrations throughout the year when compared with the urban and suburban stations and therefore will be analysed separately in Section 3.3.2.

The boxes present the median, the first and third quartiles, while the whiskers represent the minimum and maximum values. The pollutant NO_2 reaches the highest peak and median values during winter months (December, January and February, DJF), while lowest daily medians are found in summer months (June, July and August, JJA). On the other hand, O_3 presents the highest median and maximum values during April, May and June. This is in accordance to the presence of a spring and summertime midlatitudes maximum (Demuzere et al., 2009). Fig. 2 also shows that O_3 lowest daily medians of ozone concentrations are found in winter months,

Table 1
Short description for the observed SLP features of CWT type.

CWTs	Synoptic situation characterized by:
A (anticyclonic)	An extended high pressure centre between the Iberian Peninsula and the Azores Islands
C (cyclonic)	A low pressure centre over the western Portuguese coast, sometimes accompanied by a blocking anticyclone located between Iceland and the British Isles
NE (northeasterly)	An extended Azores high pressure to the northeast and by low pressure over the Mediterranean region
E (easterly)	An anticyclone between the British Isles and the Iberian Peninsula
SE (southeasterly)	A low pressure region extending from Madeira to the east of the Azores Islands and high pressure over Northern Europe
S (southerly)	Low pressure east of Azores, and by high pressure over central Europe
SW (southwesterly)	A weakening of the Azores anticyclone and a strong low pressure located between Iceland and the Azores
W (westerly)	Setting of the Azores high pressure around 30°N and by low pressure centres west of the British Isles
NW (northwesterly)	The location of the Azores high pressure between the Azores and Madeira and by low pressure centres over northern France
N (northerly)	The presence of the Azores high pressure near the Azores Islands and low pressure over southern Europe and the Mediterranean

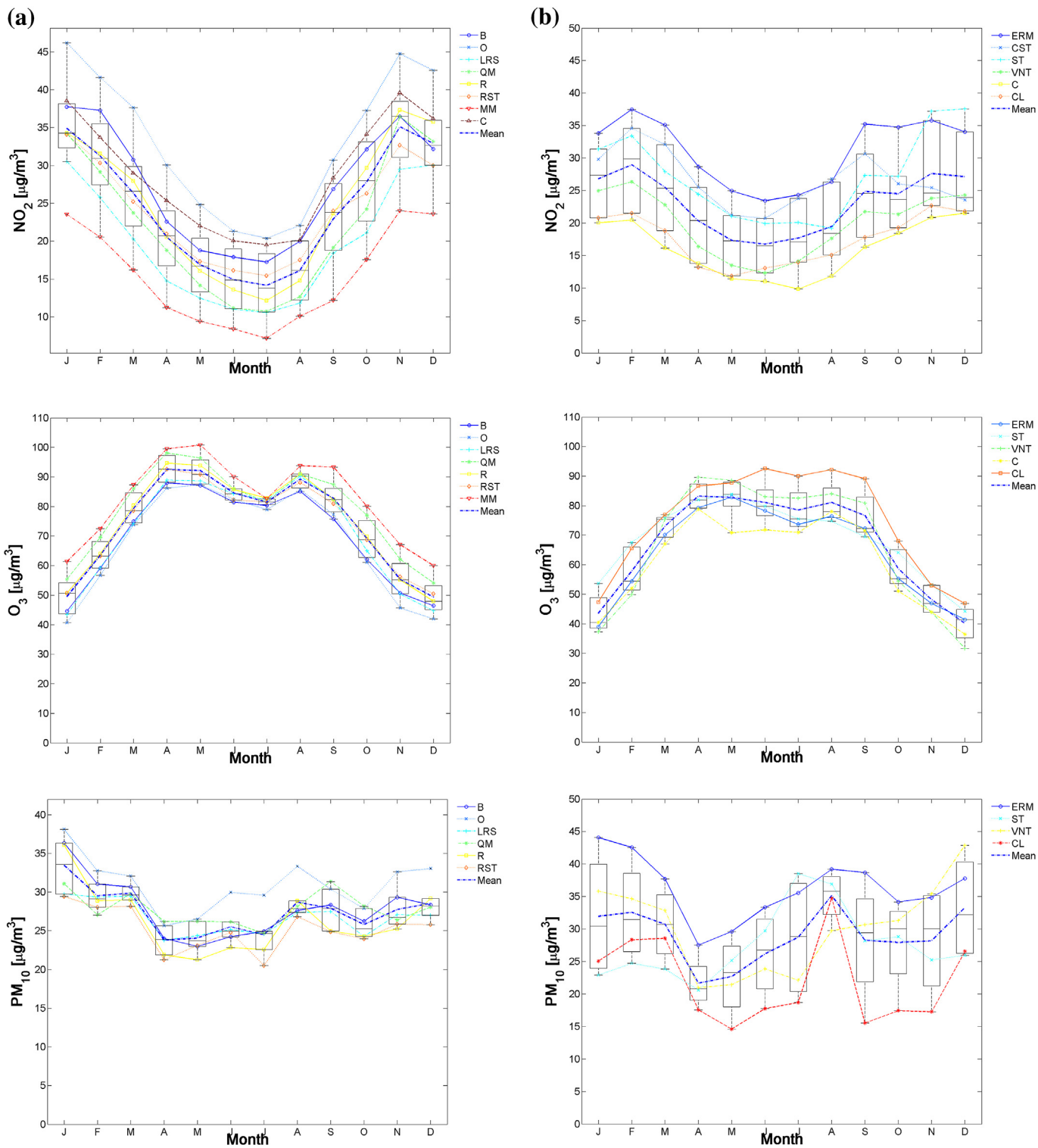

Fig. 2. **(a)** Monthly mean distributions of NO_2, O_3 and PM10 concentrations for the period 2002–2010 in Lisbon; (b) Monthly distributions of NO_2, O_3 and PM10 concentrations for the period 2002–2010 in Porto.

while PM10 time series do not present a clear annual cycle (except the existence of a moderate minimum in spring months).

Considering that the annual distribution of NO_2, O_3 and PM10 is similar among the selected monitoring stations (Fig. 2), the circulation type-specific NO_2, O_3 and PM10 concentrations are analysed for a new time series that corresponds to the spatial average of all the background stations (Fig. 3). Fig. 3 shows the cycles that can be observed for the spatial average of the three pollutants in the city of (a) Lisbon and (b) Porto. Seasonal and annual cycles can be observed through the analysis of the time series, namely for O_3 and NO_2. Additionally, there seems to be a small but visible negative trend of PM10 values throughout the decade analysed.

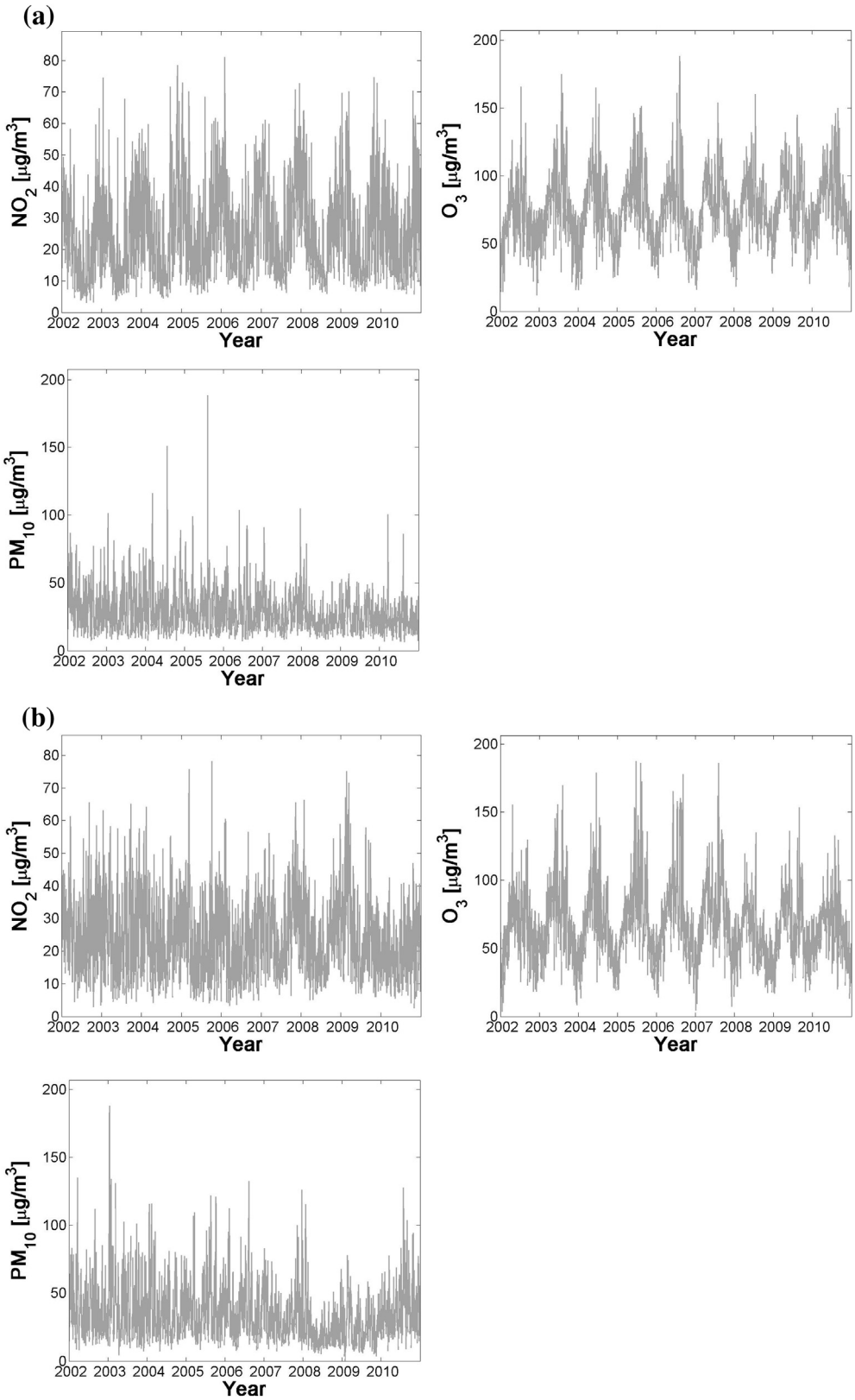

Fig. 3. Time series of daily mean values for surface NO_2, O_3, and PM10 between the period 2002–2010 (a) in Lisbon; (b) in Porto.

3.2. CWT characterization

Based on the large-scale standard pressure fields, the prevailing circulation patterns where determined. The relative frequency of each CWT over the period 2002–2010 is shown in Table 2. The anticyclonic (A) type is the most frequent circulation pattern throughout the year, except for the summer month of August, which is also highly influenced by the northeast (NE) and north (N)

circulation types. The anticyclonic (A) type predominance is mainly due to the migration of the Azores anticyclone towards the peninsula (Tomás et al., 2004).

The meridional circulation types north (N) and south (S) present very distinct behaviours: while S is rather insignificant, especially from April to August; N is one of the dominant CWT, particularly during spring and summer months. The directional CWTs with a significant southern component (i.e. S, SE, SW) are the least frequent of all the weather types throughout the year, although the SW regimes increase during winter and early spring. NW, W, and E circulation types present relatively stable frequencies during most of the year. Throughout the year, the relative frequency of cyclonic (C) situations does not change significantly reaching two relative peaks, one in March and another in October. In previous works it has been shown that the precipitation regime in Portugal is controlled by the monthly frequency of a few CWTs, namely C, W, and SW (Trigo and DaCamara, 2000) and that very dry (wet) years are characterized by lower (higher) than average occurrence of these CWTs (García-Herrera et al., 2007; Vicente-Serrano et al., 2011). In addition, C, W, and SW are also the CWT's contributing to the highest fraction of spring (MAM) and autumn (SON) precipitation over Iberia (García-Herrera et al., 2007). Although, the anticyclonic type is the most frequent CWT affecting Portugal, it presents a low contribution to winter precipitation (Paredes et al., 2006).

Fig. 4 presents a selection of CWTs for winter and summer months, namely the cyclonic (C), westerly flow (W), and anticyclonic (A) CWTs for winter (DJF) and north flow (N), northeasterly flow (NE), and anticyclonic (A) CWTs for summer (JJA). The referred CWTs were chosen among the 10 available CWTs for each season following the criteria of maximizing the CWTs' relative importance and frequency of occurrence. The anticyclonic (A) CWT is the most frequent circulation pattern for both seasons and the north (N) and northeasterly (NE) flows are the most frequent CWTs during summer (JJA) (Table 2). The cyclonic (C) and the westerly flow (W) were also chosen because these two CWTs contribute significantly to the precipitation regime in Portugal.

3.3. Relationship between the circulation weather types and air quality – the circulation-to-environmental approach

3.3.1. Lisbon and Porto urban areas

Having obtained all CWTs patterns and time series for the selected period, possible associations to observed AQ patterns were evaluated. This circulation-to-environmental approach (Yarnal, 1993) should allow identifying clear links between circulation patterns and AQ variables, which could be used as a downscaling tool for air quality assessment (Demuzere et al., 2009). Additionally, it may also permit a direct comparison between air quality forecasts and observations in order to evaluate the strength of the circulation-to-environmental approach.

Figs. 5–7 depict the box plot of these three pollutants according to their respective weather type (or class), at the seasonal scale. Since some CWT clusters are not sufficiently represented, a few percentile bars/outliers are absent.

In general, the highest median and maximum concentrations of the 3 pollutants can be observed when an eastern component is present (E, NE and SE). The exceptions occur for the maximum concentrations of (1) O_3 during DJF, which are associated to SW (Lisbon) or NW (Porto) configurations; (2) NO_2 during DJF in Lisbon and PM10 during DJF in Porto, which are associated to the A configuration.

When high median values of NO_2 and O_3 are observed associated to the same CWT (e.g. summer) that implies that the measured O_3 values result from long distance transport rather from local photochemistry. The increased transport of O_3 precursors during summer months from the Iberian Peninsula (IP) continental area can lead, in combination with positive temperature anomalies, to higher O_3 formation and concentrations. Similar results were also reported for Holland (Davies et al., 1992; Demuzere et al., 2009).

For PM10, SE, S, E, NE and A appear to be associated to the highest median and maximum concentrations. Similar results were also reported for winter in Holland (Demuzere et al., 2009). In MAM, the highest PM10 concentrations are associated with the CWTs ranging from northeast to south directions, which could be associated to the transport of PM10 from IP continental area. In autumn, the highest levels of PM10 can again be associated with E and SE flow patterns.

Thus, a major preliminary conclusion is that although anticyclonic and north component CWTs are the most frequent during the year, the highest concentrations of the three pollutants tend to occur associated with less frequent CWT, namely those characterized by an easterly component (E, NE, SE). These CWTs are complementary to those presented by Trigo and DaCamara (2000), Paredes et al. (2006) and García-Herrera et al. (2007) relatively to the CWTs representing the highest fraction of winter (DJF), spring (MAM) and autumn (SON) precipitation over Iberia (C, SW, W, NW). As expected, the water content in the atmosphere is an important factor to consider, with the highest concentrations of the three pollutants being inversely (directly) associated with wetter (dryer) CWT's.

In order to obtain a more in-depth understanding of the physical mechanisms behind the relationship between AQ and CWTs, seasonal composite maps of atmospheric pressure were calculated for each CWT class best associated with the median values of each pollutant (Fig. 8). For each season, the circulation type with the highest median concentration of each pollutant is identified, and the corresponding mean circulation pattern is depicted for the period 2002–2010 together with the MSLP, temperature and relative humidity anomalies (computed in reference to the normal period from 1971 to 2000). Only the pressure related figures are shown.

For NO_2, all the seasons present an eastern component. The winter, summer and autumn surface SLP composite maps show in both cases the presence of an anticyclone located between the British Isles and the Iberian Peninsula. This setting results in predominant eastern winds in IP, and negative anomalies of temperature and humidity, bringing colder and dry air masses from Spain and Central Europe. During spring, the resulting configuration corresponds roughly to the SE type, which is characterized by the

Table 2
Circulation weather type (CWT) frequency during the 2002–2010 period for Portugal on a seasonal basis.

	A	C	N	S	W	E	NW	NE	SW	SE	Total
DJF	**40.7%**	8.6%	5.2%	4.2%	8.3%	7.0%	5.4%	7.0%	8.7%	4.8%	100%
MAM	**33.2%**	11.8%	**11.9%**	1.2%	7.6%	7.0%	8.5%	**12.0%**	4.5%	2.3%	100%
JJA	**31.4%**	6.4%	**20.5%**	0.7%	4.4%	3.5%	7.8%	**23.6%**	1.2%	0.7%	100%
SON	**33.4%**	10.8%	5.9%	2.8%	7.4%	8.0%	8.2%	**13.3%**	7.8%	2.4%	100%

Values highlighted in bold represent the most significant CWTs.

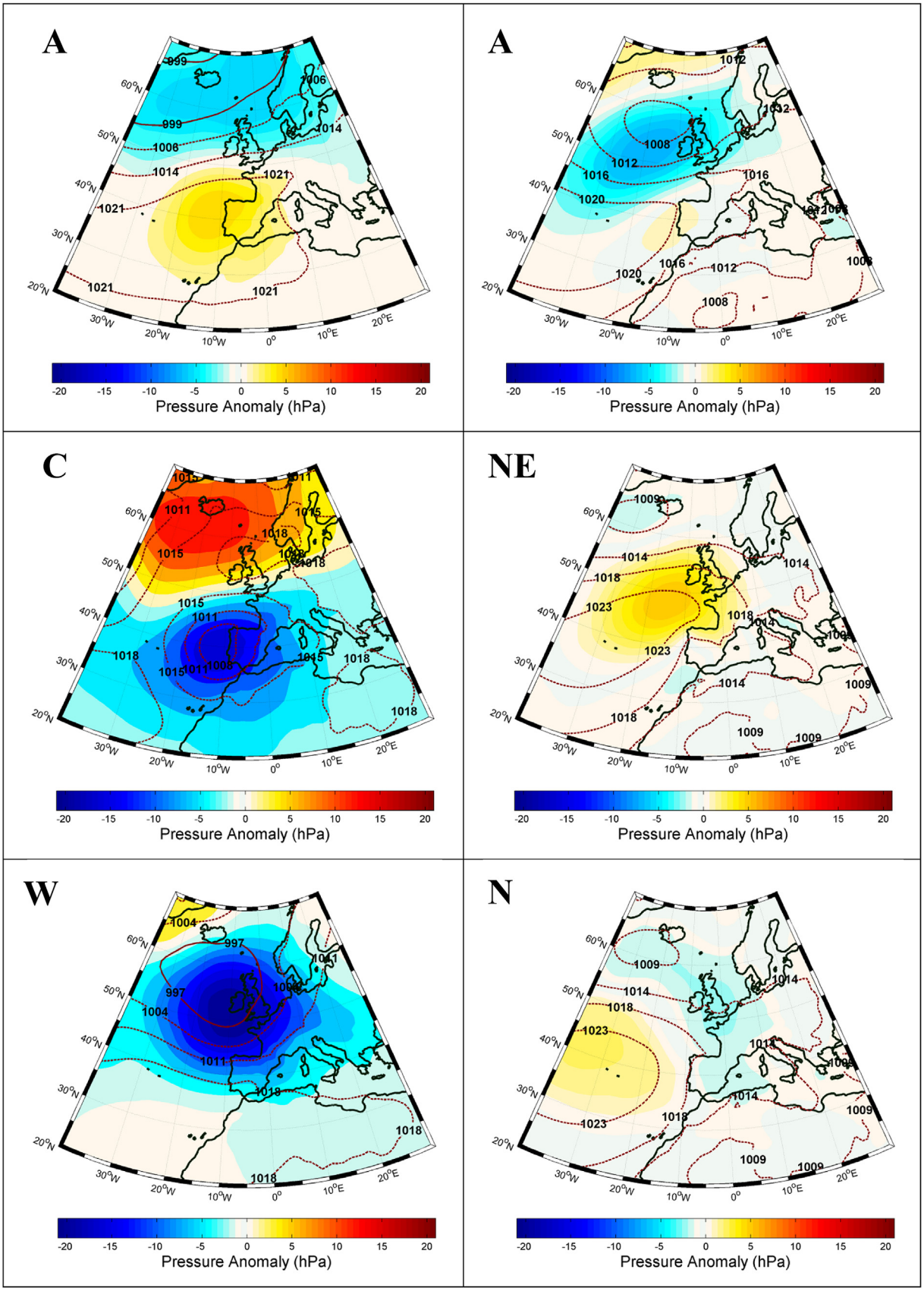

Fig. 4. Composite maps of MSLP and anomaly SLP fields for winter (DJF): (left panel) and summer (JJA): (right panel) months during the period 2002–2010. Dashed lines display MSLP levels (hPa).

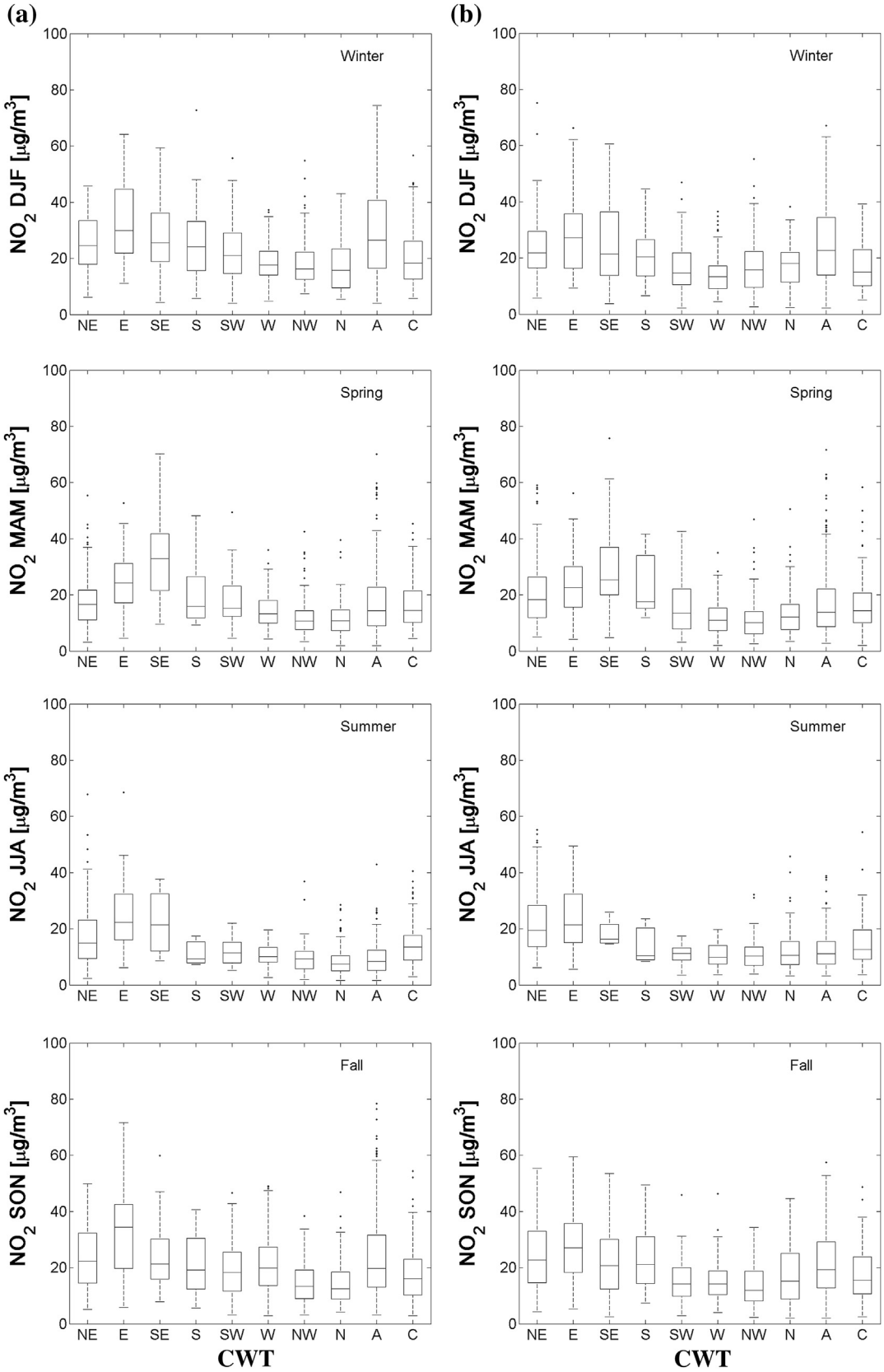

Fig. 5. Box–Whiskers plots with the concentrations of NO_2 coherent with the results obtained through the CWT methodology for (a) Lisbon and (b) Porto, according to the CWT type classes per season and year, averaged over the period 2002–2010. The box and whiskers present the median, the first and third quartiles, the minimum and maximum value and possible outliers.

presence of a high-pressure over the British Isles. Negative humidity anomalies are associated with the transport of dry air masses from the southeast of IP (not shown). The positive (negative) temperature (humidity) anomaly is associated with the transport of warmer and dry air masses from the southern Spain and northern Africa (not shown).

High values of ozone are clearly related with the predominance of CWTs with a strong easterly component. MSLP maps show again the presence of an anticyclone located between the British Isles and the Iberian Peninsula. The prevailing easterly winds promote the transport of hot and dry air masses from Spain and Central Europe (Saavedra et al., 2012). These results are in agreement with the findings of Beck et al. (1998) and Saavedra et al. (2012), which state that during most spring and summer high-ozone episodes in the Iberian Peninsula there is a high surface pressures located to the north of the Iberian Peninsula while a high-altitude ridge of high pressure crosses the Peninsula from North Africa.

According to Saavedra et al. (2012), in most of the O_3 episodes, the eastern component veered slightly to the south, causing S–SE winds due to the establishment of a thermal low over the Iberian Central Plateau, especially in the middle of the day and/or to shallow Atlantic depressions producing SE winds as they slowly approached the Portuguese coast. In some cases, the resulting S–SE circulation drives the hot dry North African air masses, causing one or more days of extremely high temperatures in this region. Pressure gradients and corresponding anomalies during summer are generally weak, which is also consistent with the negative relationship between high O_3 levels and wind speed.

For PM10, high levels occur during those occasions where air masses are advected from the southeast or east. In DJF and JJA, the most favourable CWT corresponds to the E pattern associated with the presence of an anticyclone located between the British Isles and the Iberian Peninsula. Such pattern results in an eastern flow and advection of dryer continental air. The Saharan dust transport over the western Mediterranean is prominent in summer (Pérez et al., 2006) and has a major impact on the concentration and transport of the particulate matter concentration (Querol et al., 2009) which may impact on human health. The mobilization of Saharan dust and its northward advection are related to specific synoptic conditions favourable to the extraction from the surface, the lifting above the boundary layer and the transport (Gaetani and Pasqui, 2012). During spring months, the prevailing CWTs are the eastern (Porto) and south-eastern (Lisbon). Thus, the favourable inducing conditions are represented by the presence of an anticyclone located between the British Isles and the Iberian Peninsula that affects the Porto region and by a low pressure region extending from Madeira Island to the east of the Azores Islands and high pressure over the northern Europe which affects the region of Lisbon. The prevailing winds are steered from east/southeast promoting the transport of dry air masses from Spain and northern Africa. These patterns are often associated with the generation of storm situations, especially in mountainous areas due to orographic forcing (Tomás et al., 2004). Particularly, in spring transport is associated with the development of Saharan thermal lows south of the Atlas, which travel eastward along the northern Africa coastline, while in summer the transport is favoured by the coupling between the Balearic low and the Saharan high (Gaetani and Pasqui, 2012). During the autumn, the dominant patterns affecting the IP also have an eastern component, which are respectively eastern (Lisbon) and south-eastern (Porto).

These results are consistent with the results obtained by Demuzere et al. (2009) who stated that PM10 concentrations are higher than normal during summer under conditions of high temperatures and dry weather. During spring months, the pressure gradient is more intense, resulting in stronger winds than in the other seasons. Winds with low intensity hamper the dispersion of pollutants and may give rise to pollution episodes. In summary the occurrence of PM10 extreme values is predominantly associated with situations characterized by an eastern component and advection of dryer air masses.

3.3.2. Chamusca and Lamas de Olo

As a complement to the analysis performed for the urban/suburban background stations, the circulation type-specific NO_2, O_3 and PM10 concentrations are analysed for the rural background stations of Chamusca (Lisbon Metropolitan Area) and Lamas de Olo (Northern Portugal). These two stations were chosen due to their proximity to the urban areas analysed; also they both present long data series with low number of missing values and both measure the three pollutants considered. Fig. 9 depict the box plot of these three pollutants according to their respective CWT, considering the whole year. As expected and reflecting the Chamusca (CH) and Lamas de Olo (LO) locations, NO_2 values are considerable low; on the contrary, O_3 and PM10 present several exceeding episodes.

For Chamusca, the results obtained for the three pollutants enhance the importance of the CWTs with an eastern component. In fact, for the three pollutants maximum and median concentrations are observed when an eastern component is present (E, NE and SE), which is in agreement with the previous results (see Figs. 5–7). Again, here the measured O_3 values result especially from long distance transport and not from local photochemistry.

For Lamas de Olo, the results present some similarities to those for Lisbon, especially for the O_3 and PM10 values, which again enhance the importance of the CWTs with an eastern component. Nevertheless, some notable differences occur, especially associated to the NO_2 concentrations. The results for the highest NO_2 concentrations for winter at the seasonal resolution are dominated by S and N types. During spring, summer and autumn, maximum concentrations are observed when an eastern component is present (NE and SE) or when the N types dominates. The median results at the seasonal resolution enhance the importance of the CWTs with an eastern component, except for the autumn months which are dominated by the N type. The median results obtained for O_3 enhance the importance of the CWTs with an eastern component. The highest O_3 values, which occur during summer months, are mainly associated to C and NE types (not shown). The dominant CWTs may be responsible for the transport of the secondary pollutants and its precursors from the North of Spain and from the central IP to Northern Portugal. The increased transport of O_3 precursors during summer months can lead, in combination with positive temperature anomalies, to higher O_3 formation and concentrations. Similar results were also reported for Lamas de Olo by Carvalho et al. (2010). In contrast to what happens in CH, higher median values of NO_2 and O_3 are associated to different eastern component CWTs, which means that the measured O_3 values result primarily from local photochemistry, as a result of the presence of locally produced biogenic VOC's and NO_2 resulting from long distance transport.

4. Extreme episodes

Episodes of air pollution in urban areas can pose a serious danger to human health (Pohjola et al., 2004). Under episodic conditions, concentrations of air pollutants in the atmosphere may considerably exceed established limit values.

Boundary layer inversions, which promote air stagnation situations, are particularly important in the occurrence of extreme pollution episodes, which are in many cases responsible for high levels of pollution (Piringer and Kukkonen, 2002). Regional and long-distance transport of pollutants can also lead to surpassing

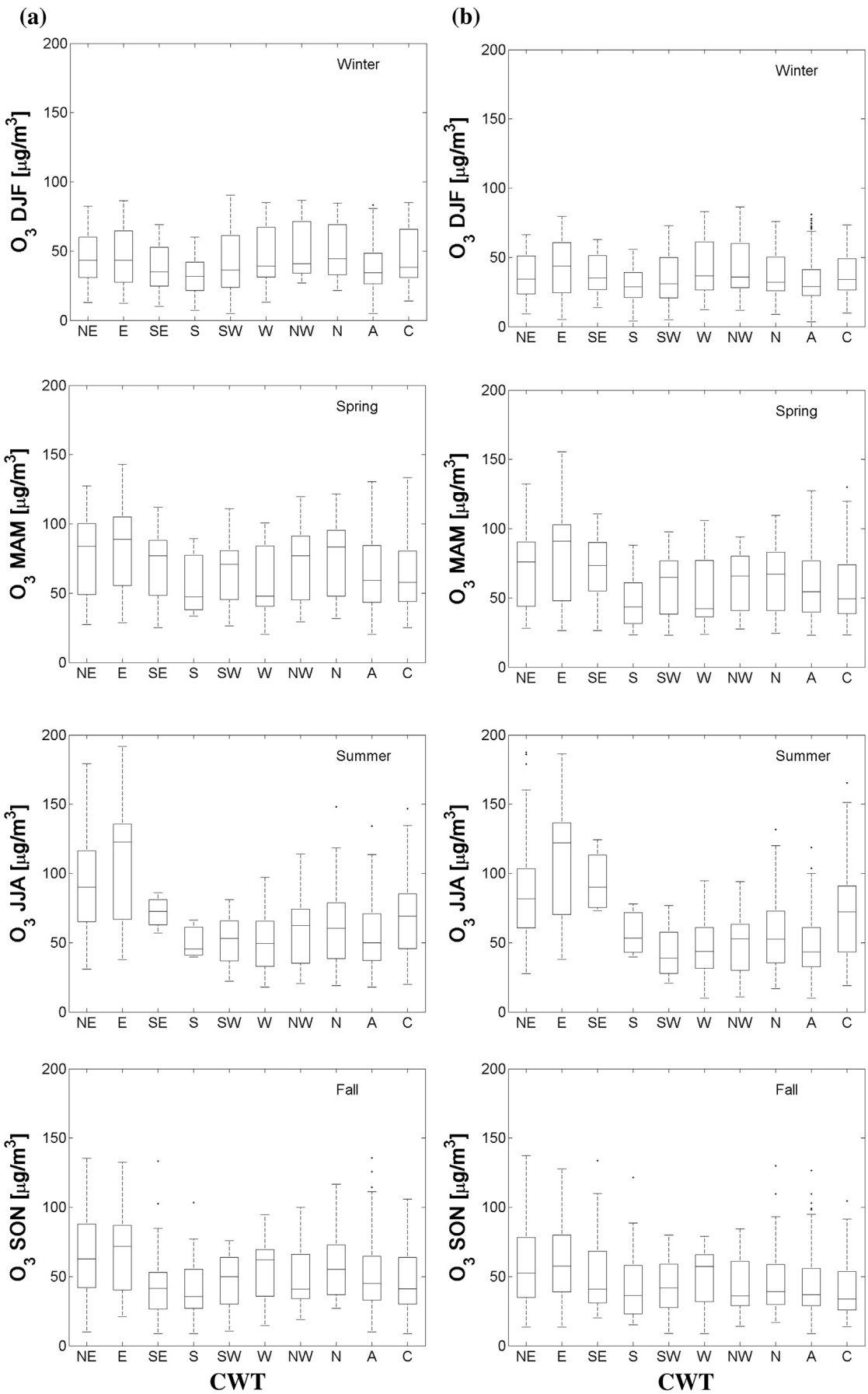

Fig. 6. As in Fig. 5, but for O_3.

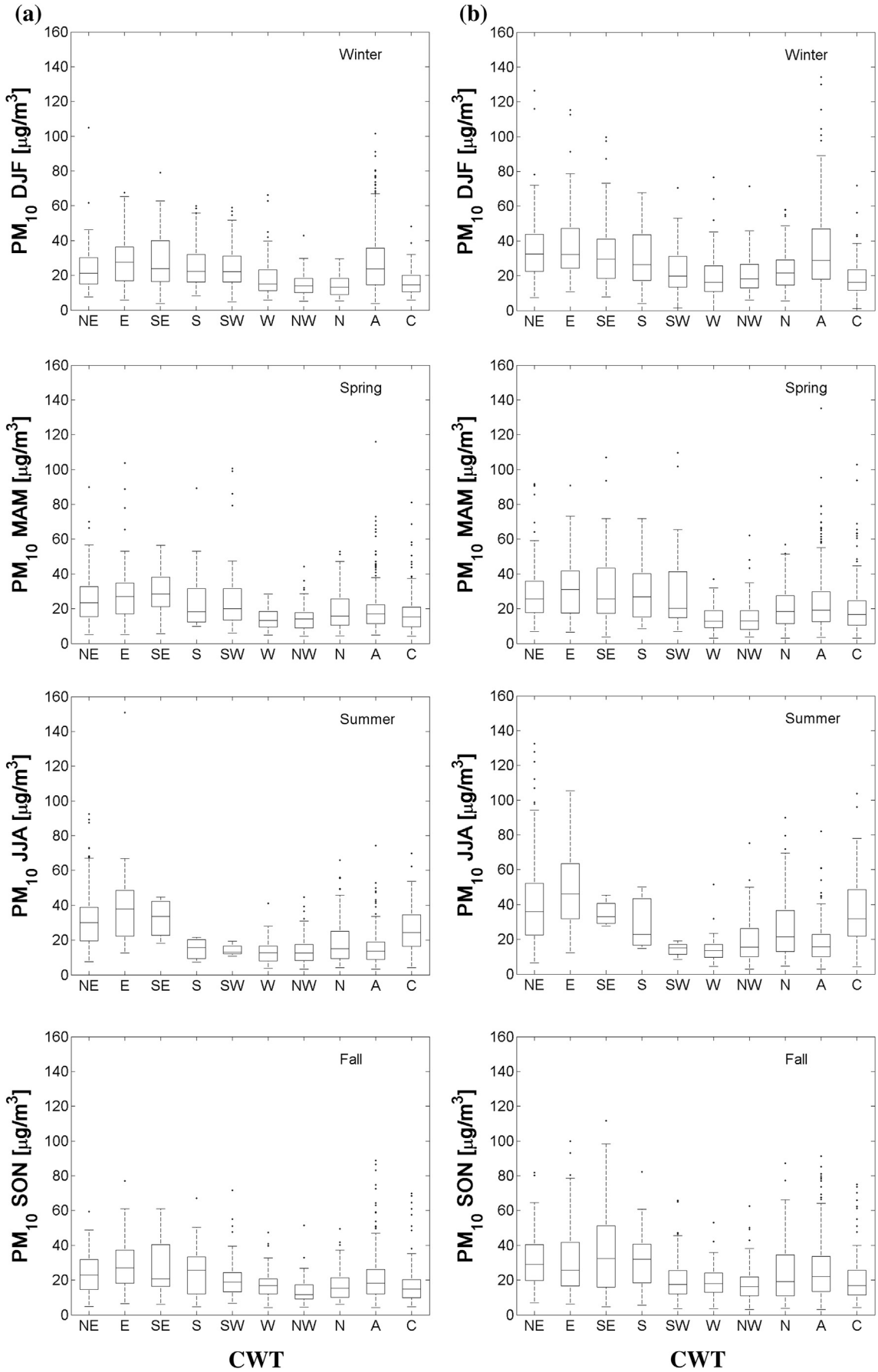

Fig. 7. As in Fig. 5, but for PM10.

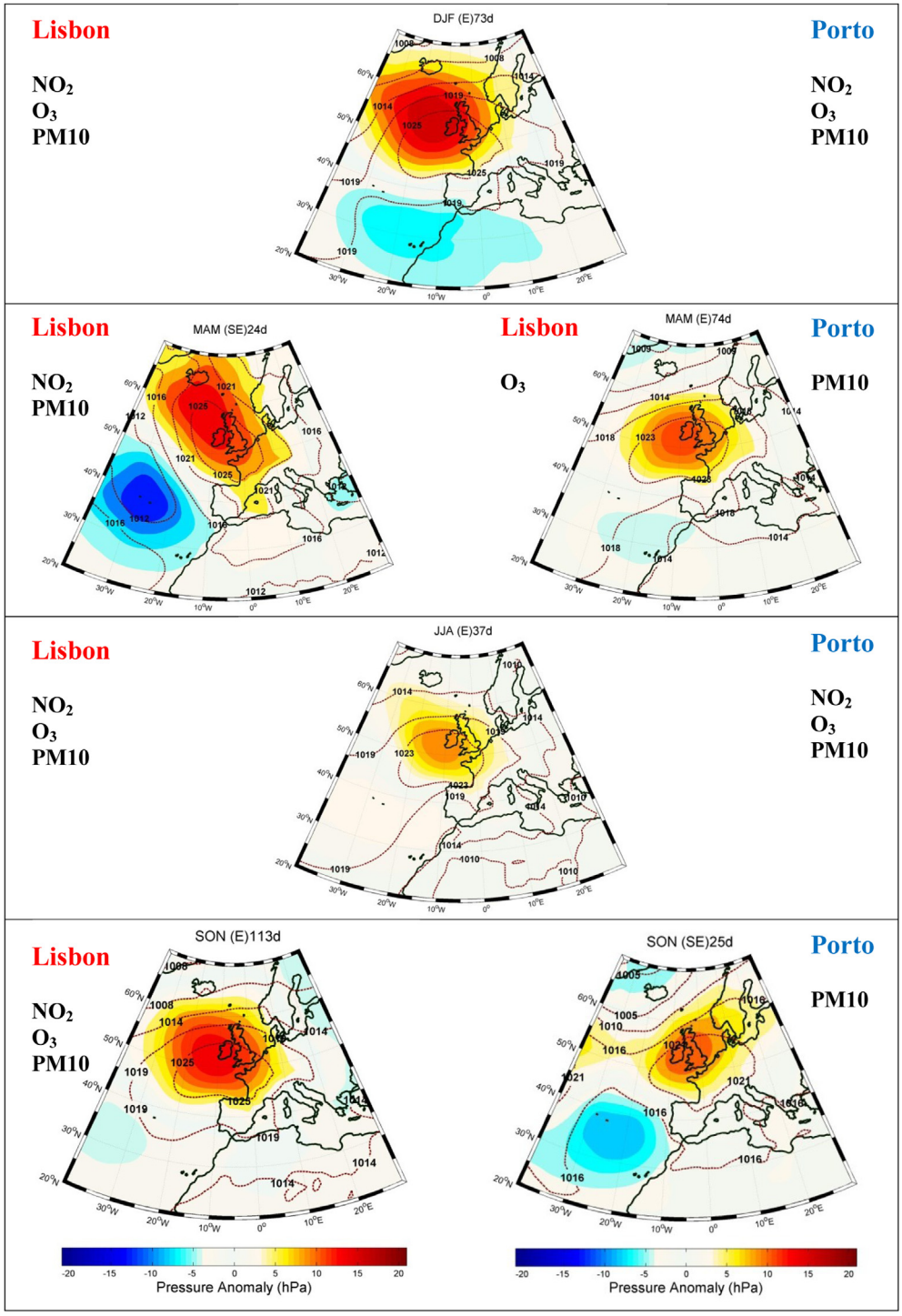

Fig. 8. MSLP and SLP anomaly fields (in hPa) for the circulation type in each season characterized by the highest median pollutants concentrations for the period 2002–2010. Colours show the pressure anomalies and dashed lines display MSLP levels. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

episodes, for example, for fine particles and ozone (Pohjola et al., 2004; Saavedra et al., 2012). Fundamentally, it is vital to understand the underlying processes that lead to episodes of air pollution at local, regional and continental scales.

PM10 episodes are often felt in many cities of various European countries (e.g. Poland, Italy, Slovakia, the Balkan region, Turkey and several other urban regions, namely, Lisbon, Porto, Paris). These episodes are more frequent in the winter and spring as a consequence of regional and long-range transport of pollution or associated to highly stable and windless atmospheric conditions (Pohjola et al., 2004; Bessagnet et al., 2005). In addition to primary PM emissions, rural PM concentrations are determined by contributions from secondary particles, i.e., by secondary inorganic aerosols and secondary organic aerosols (EEA, 2012). These contributions vary substantially across Europe and with season. Due to increased emissions from combustion in the cold season, the

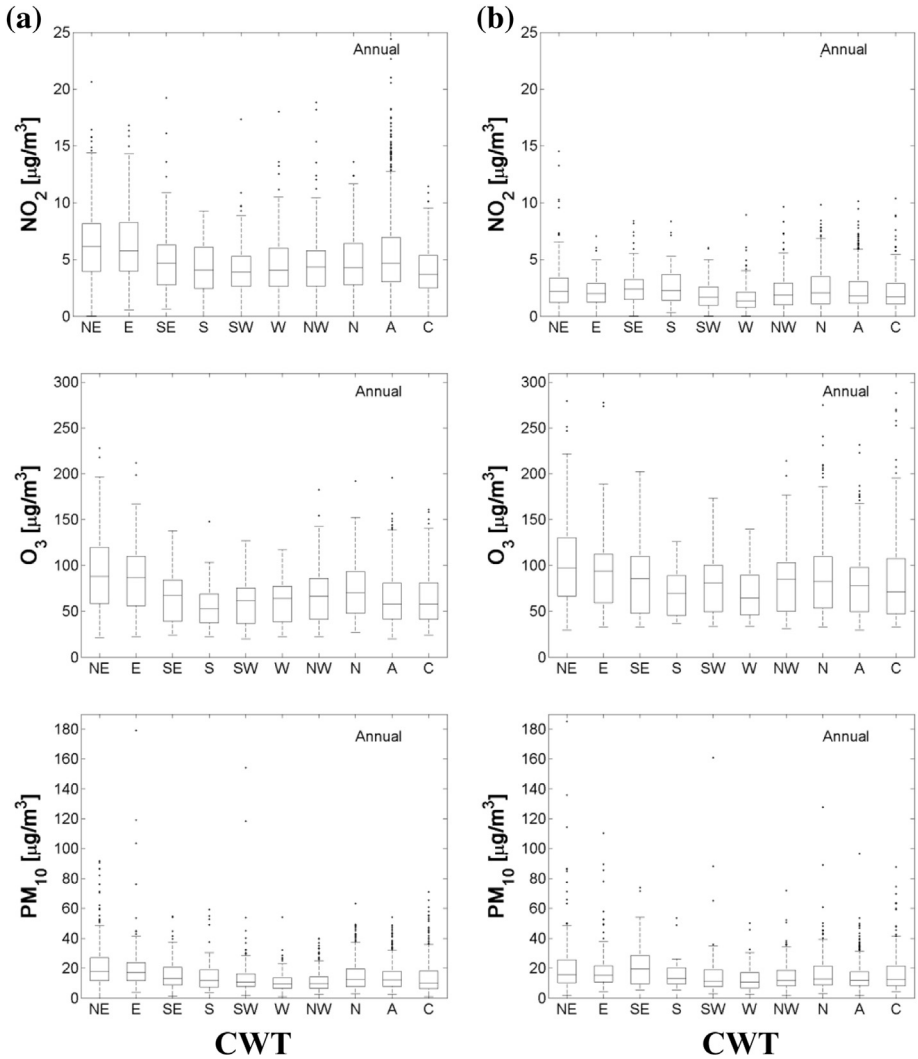

Fig. 9. Box–Whiskers plots with the concentrations of NO_2, O_3 and PM10 coherent with the results obtained through the CWT methodology for (a) Chamusca and (b) Lamas de Olo, according to the CWT type classes per season and year, averaged over the period 2002–2010.

inorganic contribution is higher in winter. The organic contribution is generally higher in summer and its mainly associated to the increase of the emissions from terrestrial vegetation (EEA, 2012). In Portugal, PM10 episodes with natural origin are related to the intrusion of air masses with African dust, although the impact of forests fire (in summer), the sea spray, or even the recirculation of aged air masses may have a high impact on PM levels (Querol et al., 2009; Carvalho et al., 2011). According to Martins et al. (2012b), the occurrence of summer forest fires in Portugal is considered to be of high importance in explaining the elevated measured PM10 values.

Episodes characterised by high levels of NO_2 can occur both in winter and summer, and the ozone levels are particularly high during summer (Pohjola et al., 2004) and spring. Thus, the relative importance of the referred factors is dependent on climatic characteristics, geographic region and time of year (Pohjola et al., 2004; Demuzere et al., 2009).

The occurrence of high-ozone episodes can be explained by the association of several factors, including: a) atmospheric stagnation due to stable boundary layer, b) the prevalence of winds that can favour the advection of ozone-rich air masses into a certain area from neighbouring regions; c) presence of clear skies and high temperatures; d) the influence of local winds (e.g., sea breezes and valley winds) which concentrate and recycle ozone and precursors in local areas (Saavedra et al., 2012; Evtyugina et al., 2006, 2009).

This section focuses on the analysis of two specific historical extreme episodes that have occurred in the Lisbon region, in order to complement the wider picture obtained so far (Fig. 10). In particular we intend to obtain a more clear insight into the meteorological factors that have influenced the formation and evolution of O_3 (August 1st, 2003) and PM10 (August 5th, 2005).

These two episodes correspond to the most notorious extreme events in terms of hourly concentrations measured in the majority of Lisbon's monitoring stations for a certain day or days for the period 2002–2010. The episode of August 2003 is coincident with one of the major heat waves affecting Portugal. The summer of 2003 was characterized by exceptional warm weather in Europe, particularly during the first two weeks of August (Trigo et al., 2006) In addition, there were extensive forest fires in Portugal (Trigo et al., 2006). Fig. 10 shows that in August 1st the population information threshold (180 μg m^{-3}) was exceeded in all the monitoring stations and the alert threshold (240 μg m^{-3}) was exceeded in three monitoring stations. The year 2005 was also affected by forest fires and, additionally, was stricken by an exceptional drought that affected more than one third of Portugal and part of southern Spain

(Gouveia et al., 2009). In the PM10 episode of August 5th, monitored concentrations exceed the daily limit value (50 μg m^{-3}) in all the stations of Lisbon, with hourly concentrations surpassing 400 μg m^{-3}. It was only one day later that high concentrations were measured by the CH station, which might be related with air mass transport to the CH location.

The composites of SLP (hPa) synoptic fields for these two events are shown in Fig. 11a, as well as the temperature and RH anomalies for the referred episodes (Fig. 11b,c). These daily anomalies were obtained after removing the daily long-term climatology for the summer months from the reference period (1981–2010). This representation confirms the outstanding magnitude of the heat-wave affecting western coastal Iberia in both episodes, with daily anomalies in the order of 10 °C (Fig 11b) and more than 30% reduction of RH (Fig. 11c). The associated CWTs are different in each case corresponding to the E (PM10) and to NE (O_3). Eastern CWT is one of the least relevant CWTs during summer (see Table 2), accounting for only 3.5% of the totality of days. On the contrary, NE is one of the most frequent CWTs during summer and winter, accounting for respectively 23% and 40.8%. This intense PM10 episode occurs associated to one of the least frequent CWT, with a strong positive temperature anomaly and a strong negative humidity anomaly (~40%). The O_3 episode occurs associated to a predominant pattern configuration, characterised by a strong positive temperature anomaly and a negative humidity anomaly (~40%). These results are coherent with previous results, i.e., pollutants' extreme events occur associated predominantly with situations characterized by an eastern component and advection of dry warmer air masses.

With the purpose of further validating this framework the NOAA HYSPLIT model was applied to the days when an extreme episode occurred (Fig. 12) and subsequent comparison of results was performed.

The main advection direction during the PM10 episode is from the central Iberian Peninsula, which is coherent with the E class obtained through the CWT methodology. It should be noted the relative concentration of trajectories arriving at Lisbon and originated well inside Spain. On the contrary, the O_3 extreme episode presents a much wider range of trajectories, with a cluster back-trajectories from NE, which are coherent with the results obtained through the CWT methodology, while other backtrajectories arrive in Lisbon from more eastern or southern sectors.

5. Conclusions and final remarks

Daily prevailing circulation patterns affecting Portugal were determined using the simple Geostrophic approximation according to the methodology proposed by Trigo and DaCamara (2000). The interannual variability of the ten resulting CWTs was determined for the period 2002–2010 and the number of days for each CWT and season for the same period was accounted for. The impact of each CWT on air quality regime was studied for the two largest urbanised areas of Portugal, namely Lisbon (South) and Porto (North) on a daily basis between 2002 and 2010. During this period, the most frequent CWTs were found to be the anticyclonic (A), the north (N) and the northeast (NE) types, accounting respectively for 34.7%, 10.9% and 14% of the days. However, higher-than-average concentrations occur associated predominantly with situations characterized by other-not so frequent-CWTs. Three of the less frequent CWTs, namely easterly (E), northeasterly (NE) and southeasterly (SE) types, that together contributed to less than one fourth of all observed days, are the classes which are associated to higher median and maximum concentrations of the three pollutants. An important outcome of this research is the fact that observed results for Lisbon and Porto between frequency of CWTs

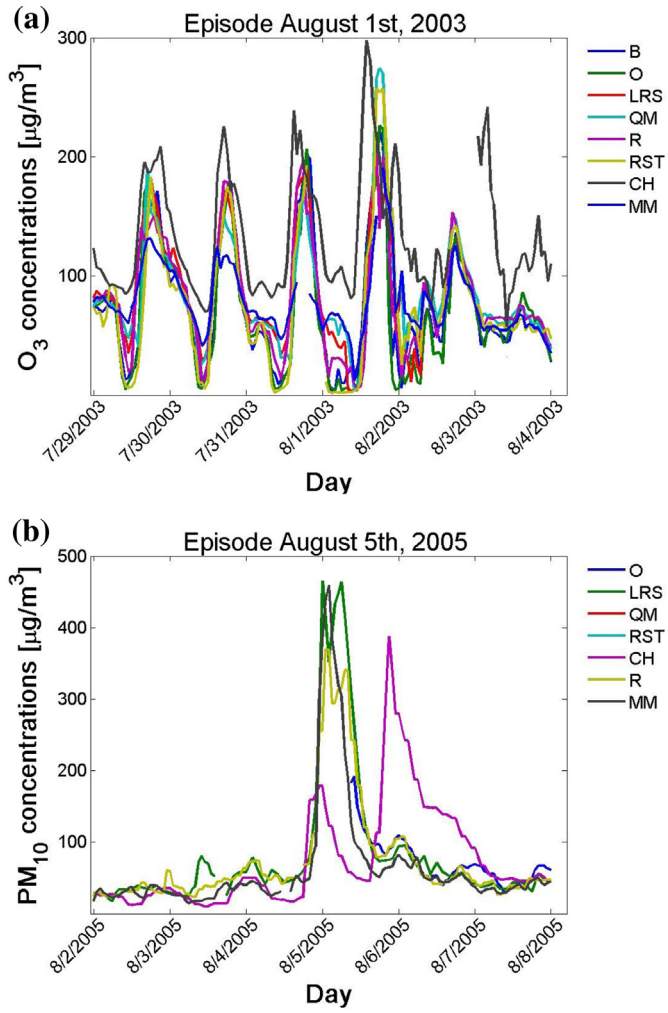

Fig. 10. Episodes hourly concentration time series (a) O_3 – August 1st, 2003; (b) PM10 – August 5th, 2005.

and air quality suggest that air quality regimes are quite similar in northern and southern regions of Portugal. Results obtained highlight the existence of strong links between the interannual variability of daily air quality and interannual variability of CWTs.

Additionally, two specific extreme episodes were evaluated in more detail in order to get a better insight into the meteorological factors that influence episodic events. These two episodes (O_3 (August 1st, 2003) and PM10 (August 5th, 2005)) represent the most outstanding cases in terms of hourly concentrations measured in the majority of the monitoring stations for a certain day or days for the period 2002–2010. The PM10 episode occurs associated to one of the least frequent CWT (E), and O_3 episode occurs associated with one of the most recurrent type (NE). The results are coherent with previous results, i.e., pollutants' extreme events occur associated predominantly with situations characterized by an eastern component and advection of dry air masses. At this stage one might wonder why similar synoptic configurations (i.e. CWT classes) can lead to either moderate or extreme pollution episodes. We are confident that this is to be expected as the atmospheric circulation, although important, corresponds to just one of the factors that modulate the amounts of pollutants observed at monitoring stations. Thus the level of anthropogenic and natural emissions varies significantly at all temporal scales (e.g. daily weekly seasonal, etc) due to different emission rates unrelated with the circulation. For example, it has been shown that some

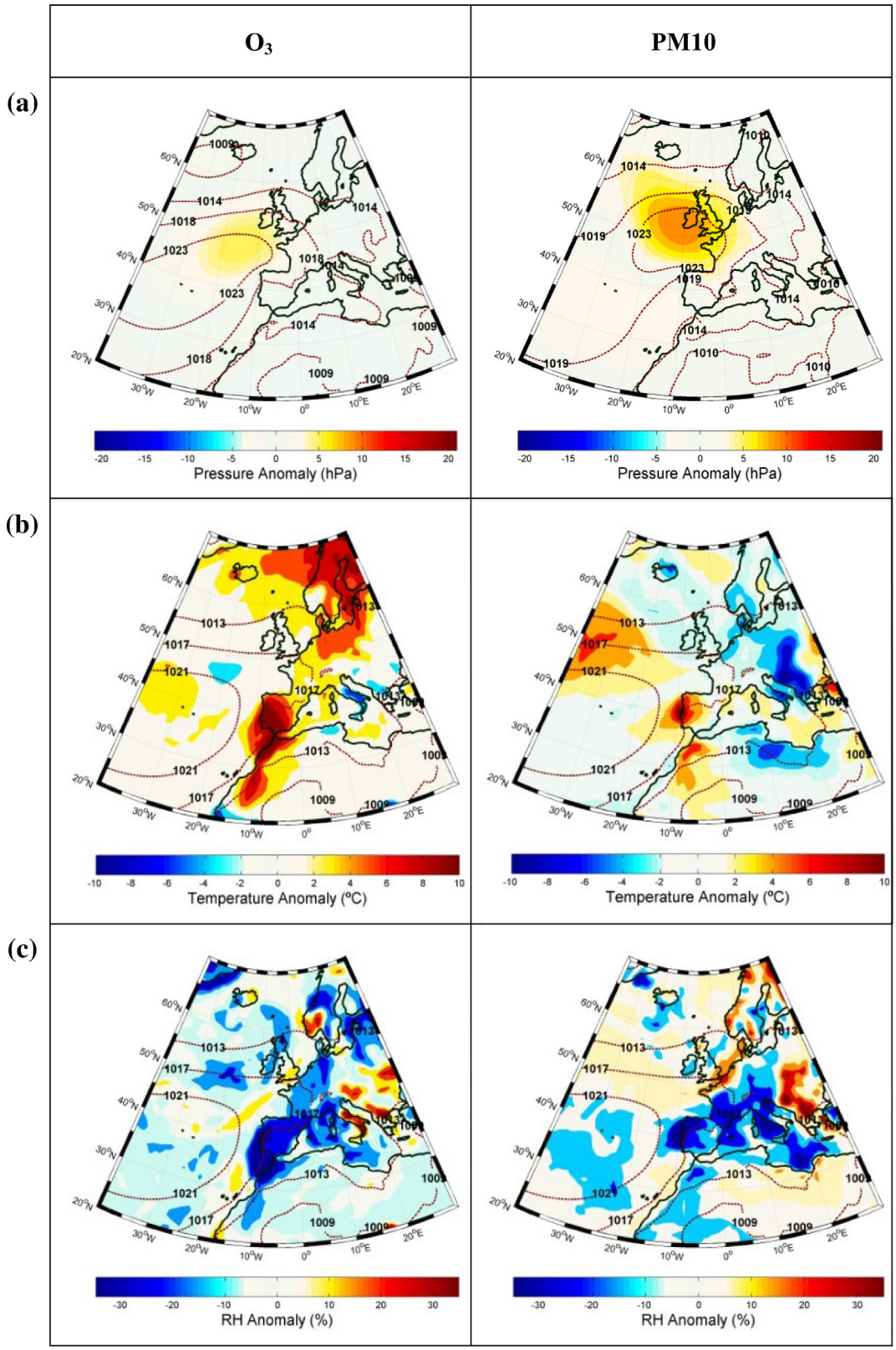

Fig. 11. (a) CWT of each episode; (b) Temperature anomaly fields and MSLP for each episode; (c) Relative humidity anomaly fields and MSLP for each episode. These daily anomalies were obtained after removing the daily long-term climatology of summer months for the reference period (1981–2010). The left panel refers to the O_3 (August 1st, 2003) episode and the right panel to the PM10 (August 5th, 2005) episode.

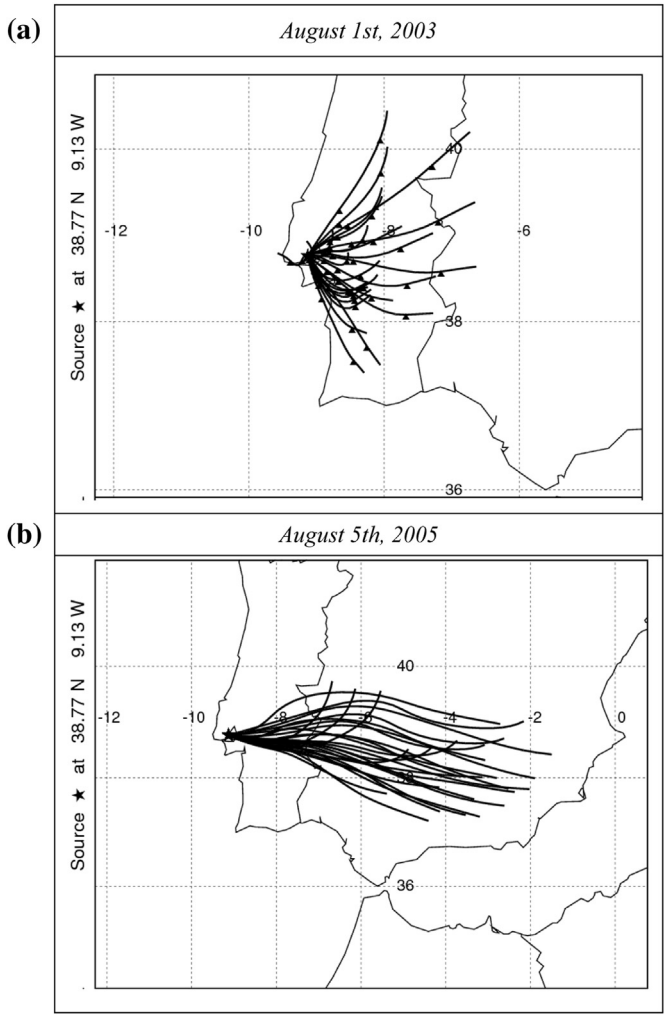

Fig. 12. HYSPLIT model results for the episodes of (a) O_3 – August 1st, 2003 and (b) PM10 – August 5th.

pollutants in Europe, associated with transportation and energy production (anthropogenic nature), present a weekly cycle with a minimum on Sundays, therefore an eastern type (E) day may produce milder levels on weekends when compared to weekdays. For example, it has been shown that some pollutants, associated with traffic such as PM, CO and NO_x present a weekly cycle with a minimum on Sundays (Morawska et al., 2002; Qin et al., 2004; Lonati et al., 2006) while tropospheric O_3, due to its photochemical origin, presents higher values on weekends (Qin et al., 2004). On the other hand, most Mediterranean countries suffer an additional burden during summer months related with the occurrence of forest fires (Pereira et al., 2011). In this regard it is worth mentioning that Portugal is the country with the largest amount of fires and burned area in relative terms within Europe (Pereira et al., 2011). As we showed in the previous section the ozone extreme event occurred during the 2003 heat wave in early August. The extensive forest fires in Portugal in the end of July/beginning of August 2003 (Trigo et al., 2006) were responsible for large emissions of atmospheric pollutants, which accordingly to the modelling studies by Martins et al. (2012b) led to the occurrence of high concentrations of O_3 and other atmospheric pollutants. Similarly, the summer of 2005 was characterized by the second highest burned area observed in summer months in Portugal (Pereira et al., 2011) that was amplified by the prolonged drought during that year (Gouveia et al., 2009). It is within this context that we should evaluate the other extreme episode with PM10 verified in early August 2005 (Martins et al., 2012b).

Although previous studies have related specific meteorological situations with air quality levels in Portugal, this study provides, to the best of our knowledge, the first long-term assessment (2002–2010) of air quality data for the two largest urban area of Portugal employing the most used CWT classification for this southern sector of Europe. Based on the results, the relationship between CWTs and poor air quality (at an annual and seasonal scale) allowed distinguishing between which types that are most frequently associated to episodes. In general, all the pollutants' extreme events occur associated predominantly with situations characterized by an eastern component and advection of dry air masses. These results are important as an aid to air quality forecasting since the high concentrations of atmospheric pollution are associated to less frequent CWTs and therefore the occurrence of these CWTs can be used as a warning signal of possible high concentrations of atmospheric pollutants.

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