CHARACTERIZATION OF OZONE AND PARTICULATE MATTER LEVELS IN A COASTAL SITE WITH THE APPLICATION OF A TRAJECTORY CLUSTERING CORRELATION METHODOLOGY

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INTRODUCTION

The relationship between synoptic-scale atmospheric transport patterns and the surface levels of air pollutants observed at a coastal urban site, the city of Barcelona (BCN; 41.4N, 2.11E), and a rural site in Cap de Creus (CC; 42'31N, 3'31E), both at the northeastern Iberian Peninsula (IP), is analyzed by the application of a trajectory clustering correlation methodology. Trajectory and cluster analysis have been increasingly employed to study the movement of air parcels carrying pollutants from sources situated at long distances (e.g. Moody, 1986; Brankov et al., 1998). Cluster analysis is a statistical tool that can be used to find out relationships between the large-scale weather regime patterns and the pollution climatology of a site. This method classifies a large set of trajectories into dominant groups called clusters.

METHODOLOGY

A spectral decomposition is applied to the air quality database in order to separate the short-term component (meteorological-related) from the seasonal component (long-term-climatic variation) embedded in the time-series. Due to the interest in studying the relation between the transport of pollutants with meteorological situations, the Kolmogorov-Zurbenko filter (KZ) is used for this purpose. The KZ filter is a low-pass filter produced by repeated iterations of a simple moving average (Zurbenko, 1996). Following Rao et al. (1997) and Brankov et al. (1998) the window length of the filter is set to 15 days and the number of iterations to 5. In order to reduce the variance of the sample, a log-transform of the data-series is applied before the KZ filter. Figure 1 shows the spectral decomposition of surface O_3 concentrations in 2000-2003 8-hour-daily-mean time-series on BCN and CC. After this data treatment, the short-term component of the pollutant time-series is analyzed related to the meteorological situation by means of backtrajectory cluster analysis.

A cluster algorithm (Dorling et al., 1992; Jorba et al., 2004) was implemented to analyze the backward trajectories arriving at BCN at 18 UTC. Four years (2000-2003) of 2-day 500m-kinematic back trajectories, computed with the HYbrid Single-Particle Lagrangian Integrated Trajectory model version 4 (HYSPLIT) (Draxler and Hess, 1998) with FNL meteorological data, were clustered and classified in groups of similar length and curvature. Ozone (O_3) and PM10 concentration levels measured by air quality stations in BCN are segregated according to the backtrajectory clusters. A statistical analysis is then applied in order to determine significant differences associated to each cluster. An annual and seasonal description of the main features of each cluster related to pollutant concentrations are presented and discussed.

BACKTRAJECTORY CLUSTER ANALYSIS: SYNOPTIC METEOROLOGY

BCN, the urban site, is located at the northeastern coast of IP within the Western Mediterranean Basin (WMB), and CC is located 150 km northeasterly of BCN. The IP in winter is characterized by an increase of extratropical cyclone activity, and it is the wet season with strongest winds. On the other hand, during the summer season the subtropical highs increase their influence and, due to subsidence and the resulting stable lapse rate, produce characteristically hot dry summers. The mean summer wind pattern is similar to that of winter but with much lower speeds. The complex orography of the eastern IP contributes to the development of typical local winds as Tramuntana (northern wind) and Cierzo (northwestern wind). The former directly influences the rural site, and with less intensity the urban site, the latter doesn't affect directly the rural site. The development of sea breeze and mountain-valley winds, and the typical Iberian thermal low (ITL) are characteristic features of the region under summer situations.

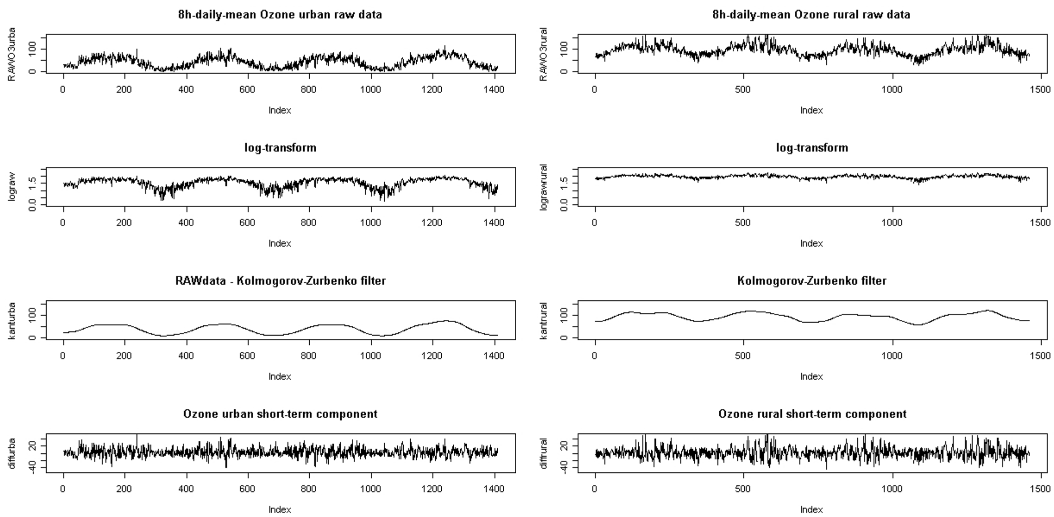
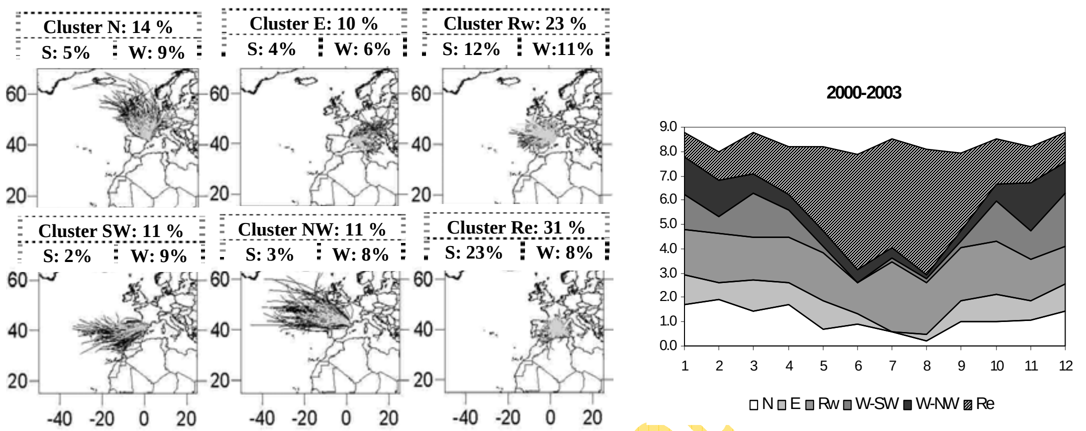


Figure 1. Spectral decomposition of surface ozone concentrations time-series for BCN urban site (left) and CC rural site (right).

Six principal clusters are obtained after applying cluster analysis to the 4-year backtrajectory database (Figure 2a). The region of study is mainly influenced by low-pressure gradient situations with a 31% of occurrence in the 4-years analyzed (cluster Re), while zonal advective situations represent up to a 22% of the situations (cluster SW and NW). Northern, eastern and northwestern advections are also significant for the region of study but with lower occurrence, rising up to 24% (cluster N and E). An important cluster (cluster Rw) groups the meteorological situations dominated by the Azores anticyclone wedge, inducing slow motions of air masses over the IP, particularly over the southwest in summer, and the influence of the cold-European-anticyclone affecting the region. Wintertime is especially characterized by advective situations associated to front traveling over the IP and anticyclonic situations with elevated subsidence. Summertime is mainly characterized by low-pressure gradient situations with slow motions clearly observed with the short backtrajectories and important subsidence associated to the Azores anticyclone that shifts northwards during this period. Figure 2b clearly shows the major frequency of low baric gradient situations during summertime, and a major dominance of advective scenarios during wintertime.

Western recirculations include very slow westerly flows, some recirculations over the IP, and some southern flows advecting warm air from the African continent to the area of study. The cluster of eastern flows contains trajectories recirculating over the Mediterranean Sea and Eastern Europe. In summer, regional recirculations are frequent at low levels due to the influence of the Azores high-pressure system, which produces several days of similar low-pressure gradient situations at low levels. Meanwhile, weak westerlies and northwesterlies are common in the middle troposphere. Hence, the development of mesoscalar phenomena, induced by the particular topography of the region, is dominant. These phenomena are mainly sea breezes, up-slope and down-slope winds and valley channeled winds, and the development of a large mesoscale circulation with marked diurnal cycles: the ITL. The strong heating of the surface air over the Spanish plateau, and the Guadalquivir valley causes low-level convergence and ascent of the air masses, contributing to develop the ITL. Some of these situations are associated with local episodes of air pollution in the region that result in higher levels of O_3 and an increase of particulate matter within the boundary layer in the area of study during summer. Also, these recirculations are associated with a multilayer arrangement of aerosols observed over BCN under typical summertime situations in the absence of large scale forcing. In winter, the influence of the European anticyclone can also produce several days of stagnant high pressure over the area with a poor development of the boundary layer. Under this situation, recirculations of air masses are common, but with less intensity than during summer.



(a) *(b)*

Figure 2. (a) The six cluster membership plots for four-day back trajectories arriving at 500 m for the 2000-2003 period [Percentage of total, summer and winter back trajectories of each cluster are shown in the left-bottom panel of each illustration. Black: winter back trajectories; gray: summer back trajectories]; (b) Monthly distribution of clusters in percentage [Percentage values are calculated in base to the total number of back trajectories used in the cluster analysis] for the 2000-2003 period.

INFLUENCE OF SYNOPTIC METEOROLOGY ON SURFACE POLLUTANTS CONCENTRATION

The short-term component of the O_3 concentration has been related to cluster analysis results. Figure 3 shows the box-whisker plots of this relation for the urban and the rural site. Several particularities of the region can be outlined from these results. The situations embedded in cluster E present the highest increase in O_3 levels in the urban site and in the rural site especially in summer. Attending to the direction of air masses, eastern advections, and the simultaneous high levels in both sites, one could relate these O_3 concentrations to a regional transport of air masses with higher O_3 concentrations or primary pollutants from the WMB and the European continent to the eastern coast of the IP. Also, situations characterized by recirculation of air masses for several days over the region, grouped in cluster Re, present high O_3 levels, with a higher increase in the concentrations for CC (rural site) than in BCN (urban site). This may be explained as follows: the stagnant meteorological conditions during summertime and the local transport of polluted air masses mainly from the emitter urban area of BCN provokes the accumulation of O_3 in the NO_x-limited rural site of CC. However, the VOCs-limited area of BCN presents a more marked daily cycle of formation and depletion of O_3 (Jiménez and Baldasano, 2004).

The advective westerly situations (cluster SW and NW) are characterized by a decrease of pollutants in both regions. The results show an important decrease in cluster SW during wintertime for both sites. However, western-northwestern advections (cluster NW) just present an important decrease in O_3 levels in the rural stations during summertime. The Ebro valley canalization and the possible development of the Tramuntana near CC may contribute to the decrease in O_3 concentrations. One should remark that the BCN urban site is not affected by the Tramuntana wind; under northwestern flows, the region is characterized by slow winds.

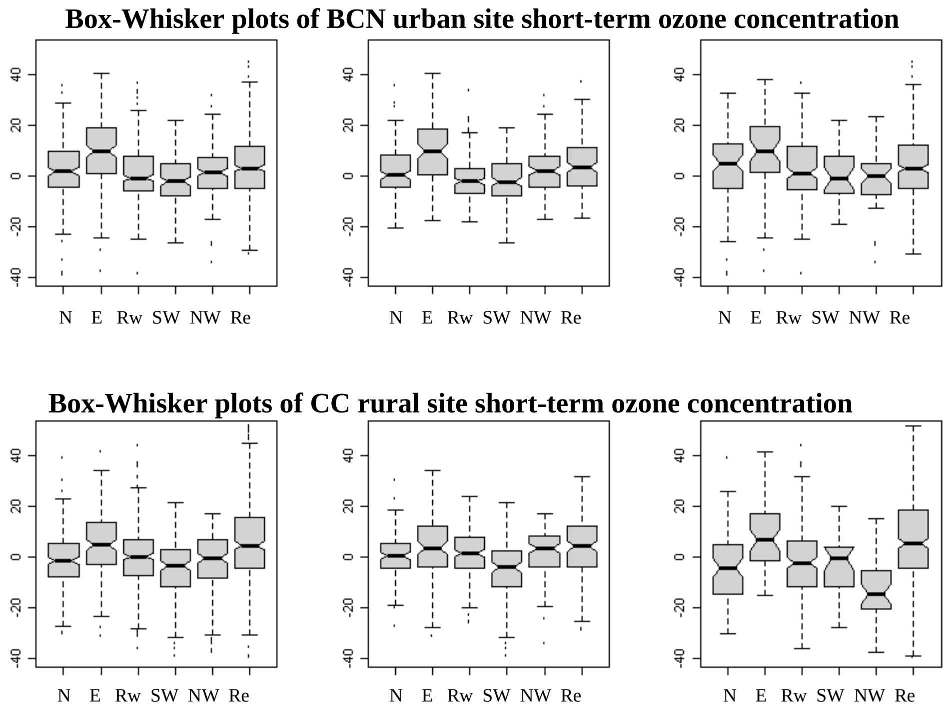


Figure 3. Box-Whisker plots of urban (top) and rural (bottom) site of short-term surface O_3 concentration ($\mu g/m^3$) for (left) annual, (center) winter, and (right) summertime [Percentiles 5, 25, 50, 75 and 95 are drawn].

A similar procedure is followed with the PM10 measurements. In this case, KZ filtering technique is not applied due to the inability of the method to discern the occurrence of episodes with aerosol increase from the climatic behavior of this pollutant. Thus, PM10 concentrations have been related to the cluster results directly (Figure 4). Advective situations, as northern, eastern and western flows, are characterized by lower PM10 mean concentrations. It is important to remark that eastern situations are characterized by low winter levels of PM10. This behavior is opposed to the observed with O_3, which presents a clear increase on concentration compared with the climatic compound. Eastern flows are maritime air masses that present low concentration of PM10. Cluster Rw presents more scenarios with higher concentrations related to re-suspension of aerosols due to the recirculation of air masses over the region of study. This behavior is observed also in cluster Re, which presents the higher mean values of PM10. This cluster includes marked episodes of re-suspension of aerosols with high concentrations of PM10 related to the accumulation of aerosols within the region for several days due to the low baric gradient situation with slow motions. The significance of the Saharan dust outbreaks over the IP is also observed in cluster SW, with higher levels of PM10. This cluster groups the back trajectories with a marked southwestern component, which may be related with advective situations of warm air masses from the north Africa to the IP with high concentrations of aerosols. On the other hand, cluster NW presents the lower levels of PM10. The advective characteristics of this cluster and the association with the front pass situations with development of rain contribute to the decrease of PM10 concentrations.

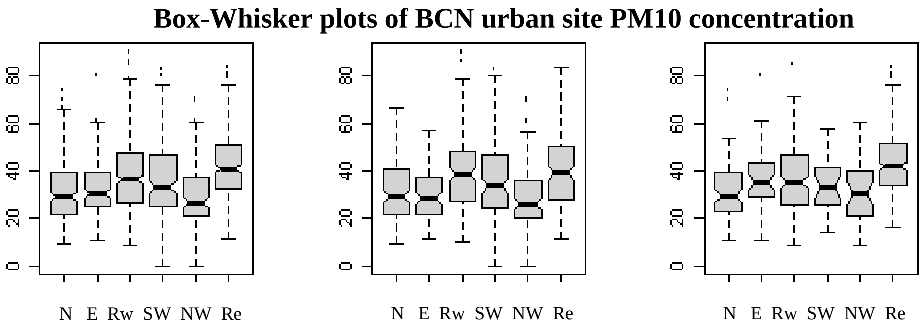


Figure 4. Box-Whisker plots of urban site of surface PM10 concentration for (left) annual, (center) winter, and (right) summertime [Percentiles 5, 25, 50, 75 and 95 are drawn].

SUMMARY

Results show a clear decrease in O_3 levels during wintertime, with higher concentrations associated to eastern regional transport. During summertime, O_3 levels increase, especially in meteorological situations characterized by low-pressure gradient with recirculations of air masses within the WMB. PM10 concentrations present a smooth behavior between clusters, with higher levels associated to anticyclonic and low-pressure gradient situations characterized by important atmospheric subsidence over the zone and slow flows.

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