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Effects of teleconnection patterns on the atmospheric routes, precipitation and deposition amounts in the north-eastern Iberian Peninsula

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Abstract

The North Atlantic Oscillation (NAO) has been identified as one of the atmospheric patterns which mostly influence the temporal evolution of precipitation and temperature in the Mediterranean area. Recently, the Western Mediterranean Oscillation (WeMO) has also been proposed to describe the precipitation variability in the eastern Iberian Peninsula. This paper examines whether the chemical signature and/or the chemical deposition amounts recorded over NE Iberian Peninsula are influenced by these climatic variability patterns. Results show a more relevant role of the WeMO compared to NAO in the deposition of either marine (Cl^-, Na^+, Mg^{2+}) or anthropogenic pollutants (H^+, NH_4^+, NO_3^- and SO_4^{2-}). A cluster classification of provenances indicated that in winter (December to March) fast Atlantic air flows correspond to positive WeMO indices, while negative WeMOi are associated to Northeastern and Southwestern circulations. The negative phase of WeMO causes the entry of air masses from the Mediterranean into the Iberian Peninsula, that are enriched with marine ions and ions of anthropogenic origin (NH_4^+, NO_3^- and SO_4^{2-}). For these later, this suggests the advection over the Mediterranean of polluted air masses from southern Europe and the scavenging and deposition of this pollution by precipitation during the WeMO negative phases. This will carry transboundary pollutants to the NE Iberian Peninsula. However, local pollutants may also contribute, as precipitation events from the Mediterranean and the Atlantic (associated to both WeMO phases) may incorporate emissions that accumulate locally during the winter anticyclonic episodes typical of the region.

Key words: West Mediterranean, precipitation chemistry, deposition, back-trajectories, NAO, WeMO

WeMO index better describes winter precipitation in the NE Iberian Peninsula than NAO
Element deposition was correlated with NAO and WeMO depending on air provenance
Few studies have analyzed the relationship rain chemistry vs. NAO and WeMO

1. Introduction

The chemical composition of precipitation is strongly influenced by the predominant atmospheric transport patterns, which affect the scavenging of pollutants depending on the pollution climate encountered. Also, the precipitation amount is important as it influences the dilution and amount of pollutants. The Iberian Peninsula (IP) is located in the south-western corner of the European continent, with the Atlantic Ocean to the west and the Mediterranean to the east, the industrialized Europe to the north and the arid Africa to the south, thus it is in a crossroads influenced by pollutant sources differing strongly in strength and character, which will affect the precipitation chemistry. The levels of atmospheric particulate matter (PM) in this area and its chemical composition has been shown to depend on the origin of air masses (Pérez et al. 2008; Querol et al. 2009; Pey et al. 2009, 2010; Cusack et al. 2012) and the chemical composition of precipitation as well (Àvila and Alarcón 1999, 2003; Izquierdo et al. 2012). African events have been found to be related with the specific position of cyclonic lows in the western or southern flank of the IP or the permanence of high pressures over north Africa in summer (Escudero et al. 2005, 2011). A higher contribution of African precipitation events for particular years significantly affects the acidity/alkalinity balance of precipitation and the contribution of crustal components (Àvila et al. 1997, 2007; Àvila and Rodà 2002; Pulido-Villena et al. 2006; Morales-Baquero et al. 2013). Furthermore, the region is subject to a large load of anthropogenic emissions from the intense activity of large cities such as Marseille, Barcelona and Tarragona and their industrial surroundings and of the heavy traffic along the eastern coast of IP. The pollutant climate in this area is further modulated by a marked seasonality. In winter, the frequent entry of Atlantic relatively clean air fluxes induces the replacement of air masses, thus reducing the levels of atmospheric pollutants that may have accumulated during periods of anticyclonic stability (Rodríguez et al. 2003; Escudero et al. 2007). In contrast, the synoptic scenario in summer is characterized by very weak pressure gradients in the western Mediterranean which produce local circulations enhancing the regional accumulation of pollutants (Millán et al. 1991, 1997; Rodríguez et al. 2003).

In the Mediterranean area, a year to year variability in the amount of precipitation (Xoplaki 2002; Lionello et al. 2006; Mariotti and Dell'Aquilla 2012) and dust transport (Moulin et al. 1997; Ginoux et al. 2004; Pey et al. 2013) has been related to the atmosphere-ocean interaction defined as the North Atlantic Oscillation (NAO). The NAO strongly influences the atmospheric circulation and the hydrological cycle in the Northern hemisphere (Hurrell 1995; Wallace 2000; Hurrell et al. 2004; Hurrell and Deser 2010). An intensification of NAO (producing more westerly winds across the North Atlantic and into Eurasia) has been observed over the past few decades (Hurrell 1995). This intensification would bring an increase in precipitation in mid-latitude zones in Europe. Although many models suggest that such a change might be the result of anthropogenic greenhouse warming (Carnell and Senior, 2002; Zhang et al. 2007; Min et al. 2011), most models seem to underestimate the magnitude of this circulation change in central

Europe (Gillett et al. 2005). In the Mediterranean, Barkhordarian et al. (2013) indicate that changes in atmospheric greenhouse gas and sulphate concentrations are not the dominant forcing process affecting precipitation changes in this area. Additional anthropogenic forcing agents potentially have a larger effect on regional scale precipitation. The emission of aerosols related to traffic and industry and/or forcing from land-use changes such as deforestation are missing in current climate models and need to be incorporated.

Recently, the Western Mediterranean Oscillation (WeMO) climatic variability has been proposed to describe the synoptic framework of the western Mediterranean basin (Martín-Vide and López-Bustins, 2006). The WeMO index is calculated as the difference between the standardized surface pressure values recorded at Padua (45.40ºN, 11.48ºE) in northern Italy, an area with a relatively high barometric variability due to the influence of the central European anticyclone, and San Fernando (Cádiz) (36.28ºN, 6.12ºW) in south-western Spain, an area often influenced by the Azores anticyclone (Fig.1). This regional pattern strongly determines the variability of rainfall in the eastern façade of the IP (Martín-Vide and López-Bustins, 2006; González-Hidalgo et al. 2009). As in most of the variability patterns of the Northern Hemisphere, WeMO shows its most relevant dynamics during the winter (Martín-Vide and López-Bustins 2006). The WeMO positive phase has been shown to trigger air masses from the Atlantic to move into the IP, while its negative phase is associated to flows from the Mediterranean (Martin-Vide and López-Bustins 2006; López-Bustins et al. 2008). Thus, it can be foreseen that part of the precipitation variability and precipitation chemical signature in Northeastern Spain may be related to the WeMO pattern. The effects of the WeMO on precipitation amounts in NE Spain has been recently explored (López-Bustins et al. 2008, González-Hidalgo et al. 2009), but to our knowledge, no studies have been focused on the analysis of its influence on the precipitation chemical composition.

We explore here whether the chemical signature and/or the chemical deposition amounts reaching a site in Catalonia (NE Iberian Peninsula) is influenced by these climatic variability patterns. This is studied by comparing winter bulk precipitation chemistry data at a locality in Catalonia (La Castanya) and the NAO and WeMO indexes for a long-time series of weekly precipitation chemical data from 1984 to 2012. Thus, this study covers a 3-decade period where changes in emissions have taken place in Europe in response to emission abatement strategies agreed under the Convention on Long-range Transboundary Air Pollution (UNECE, available at http://www.unece.org/env/lrtap/). The aim here is to discern the influence of the main circulation patterns as represented by NAO and WeMO on precipitation amounts and the chemical loads of precipitation.

2. Material and Methods

2.1. Study site

La Castanya station (LC, 41º46'N, 2º21'E, 700 m) is located in the Montseny mountains of the Pre-litoral Catalan Range. Long-term biogeochemical studies have been undertaken since the 1970s in a forest plot close to the atmospheric sampling site (Rodà et al. 1999). The site is amidst extensive holm-oak (*Quercus ilex* L.) forests in the Montseny Natural Park, 40 km to the N-NE from Barcelona and 25 km from the Mediterranean coast (Fig.1). The climate in Montseny is meso-Mediterranean sub-humid, with high interannual precipitation variability (range: 503-1638 mm y^{-1}, mean: 840 mm y^{-1} at LC, from 1983-2009). Summer droughts are common and snow is sporadic. Mean air temperature was 9ºC during the period 1983-2000 at LC.

2.2. Sampling and chemical analysis

We analyze here the precipitation chemistry in the period 1984-2012. Weekly samples were obtained with 4 or 2 bulk deposition collectors (consisting of a polyethylene funnel connected to a 10L bottle) located in a clearing within the holm oak forest. Bulk deposition samples were obtained from 1983 to 2001; wet-only deposition was also collected in parallel with bulk deposition for 2008-2012. In order to obtain an homogeneous data record, regressions between bulk and wet weekly data were used to estimate bulk deposition for the whole period and very strong correlations were obtained ($R>0.8$, $p<0.001$). Measurements here considered correspond to bulk deposition weekly samples. Further details can be found in Izquierdo and Àvila (2012) and Izquierdo et al. (2012). Samples were processed in the CREAF laboratory according to previously described protocols (Àvila 1996; Àvila and Rodà 2002). Conductivity, alkalinity and pH were measured in unfiltered samples within 48h of sampling. Samples were filtered through 0.45μm membrane filters and stored at -20ºC. Ion chromatography was used to determine the concentrations of Na^+, K^+, Mg^{2+}, Ca^{2+}, NH_4^+, Cl^-, NO_3^- and SO_4^{2-}. Quality control included the repeated inclusion of certified samples and the checking of ratios (measured conductivity against the calculated conductivity and the sum of cations against the sum of anions) and samples were reanalyzed or discarded for differences > 20% (Àvila 1996; Àvila and Rodà 2002).

2.3. Cluster analysis

A daily meteorological analysis was undertaken based on 96-h isosigma back-trajectories at 12:00h UTC and 1500m asl by using the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) 4.0 dispersion model from the Air Resources Laboratory (ARL, available at http://www.arl.noaa.gov/ready/hysplit4.html, Draxler and Rolph 2003). This height can be taken as representative of the mean transport wind at a synoptic scale within the upper boundary layer. The meteorological input was obtained from the ARL (Air Resources Laboratory) reanalysis database for the 1984-1996 period, and from FNL for the 1997-2005 period and from GDAS (Global Data Assimilation System) for 2006-2012, from the NCEP (National Center for Environmental Prediction). Even though the WeMO index corresponds to surface level pressures, a height of 1500 m, corresponding to 850 hPa standard pressure level, was selected for trajectory computation because both levels can be considered comparable for synoptic scale circulation features. Indeed, 850 hPa is the most representative level for transport in the lower troposphere. This layer is typically sensitive to cyclonic-wave features and is the approximate boundary between the surface-wind regime and the free troposphere (Artz et al. 1985). Moreover, a relationship between the 850 hPa wind direction and the prevailing weather patterns associated with the passage of cyclonic waves is well established (Dayan and Lamb 2003).

Cluster analysis statistically aggregates observations into clusters so that each of them is as homogeneous as possible with respect to the clustering variables (Sharma 1996). To compose each cluster, HYSPLIT has a grouping module based on variations in the Total Spatial Variance (TSV) between different clusters which is compared to the spatial variance (SPVAR) within each cluster component. The final number of clusters is determined by a change in TSV as clusters are iteratively paired (Draxler et al. 2009). This statistical methodology was applied to daily trajectories for the period from 1984 to 2012. Because most of the variability patterns of the Northern Hemisphere show its most relevant dynamics during the winter (Goodess and Jones, 2002; Martín-Vide and López-Bustins, 2006), a subset corresponding to the boreal winter period (December to March) was also analyzed.

The precipitation chemistry database consisted of weekly observations but trajectories were obtained daily. To overcome this mismatch we estimated a daily chemical concentration for the days with precipitation by proportionally correcting weekly chemical concentrations by the precipitation contribution of the rainy days to the weekly amount. The rainy days within each week and their precipitation amount were obtained from records at LC, and from the AEMET stations (Spanish Meteorological Service) of Turó de l'Home and Tagamanent which are 7 and 8 Km distant from LC, respectively. Precipitation events of <3 mm were not included, and only the days with precipitation amount of >0.2 mm were considered for the determination of precipitation days within a week. The precipitation chemical concentrations were weighted by the precipitation amount to obtain ion volume weighted means (VWM, in µeq L^{-1}). Wet deposition amounts (kg $ha^{-1}y^{-1}$) were calculated as the product of VWM precipitation concentrations times the precipitation amount (L m^{-2}).

We analyze here the atmospheric circulation patterns and the amount and chemistry of precipitation for:

1) An annual database, where the clustering methodology was applied to the daily trajectories for three periods encompassing a decade each: an early period (1984-1993), an intermediate period (1994-2003) and a recent one (2004-2012) period. This splitting was forced by the impossibility to deal with the whole dataset due to computing limitations. A

unique annual classification of air masses was obtained by combining the clusters of the three decades.

2) Winter database: Cluster analysis was applied to winter (DJFM) daily trajectories from 1984 to 2012 in a single run.

The NAO (available at https://climatedataguide.ucar.edu/climate-data/hurrell-north-atlantic-oscillation-nao-index-station-based, Hurrell, James and National Center for Atmospheric Research Staff 2013) and WeMO (available at http://www.ub.edu/gc/English/wemo.htm, Group of Climatology, University of Barcelona) indexes for the annual and winter period (DJFM) were regressed (least-square linear regressions) with: 1) the frequency of provenances, 2) the number of rainy days in each provenance and 3) the amount of precipitation amount (L m^{-2}) in each provenance from both (annual and winter) datasets. Regressions were also explored for these climatic indices (winter period) and winter wet deposition amount for the main precipitation components calculated for each provenance. The WeMOi value for 2012 was unavailable, therefore regressions considered the 1984-2011 period. In addition years 2001, 2002, 2004 and 2005 were not included due to fragmentary precipitation chemistry sampling. Correlation coefficients of the regressions were considered significant when $p<0.05$.

3. Results

3.1. Annual atmospheric circulation patterns and precipitation regime

Back-trajectory clusters obtained from the annual database were classified in seven main provenances: 1) Northern flows, 2) North-Western, 3) Western, 4) North-Eastern, 5) Mediterranean, 6) Iberian Peninsula and 7) Regional/Local recirculation.

The annual NAOi was negatively correlated with Northern ($R= -0.40$ $p<0.05$) and positively with Mediterranean ($R=0.39$ $p<0.05$) annual precipitation amount, but no correlation was found between annual NAOi and the frequency, the number of rainy days, and annual precipitation amounts of provenances other than the Mediterranean and Northern ones (Table 1). The annual WeMOi was positively correlated with the frequency and the number of rainy days of NW, W, Mediterranean and Regional provenances, and with the NW precipitation amount (Table 1). Conversely, annual WeMOi showed a negative significant relationship with Northern frequency and number of rainy days ($R= -0.54$ an -0.35; Table 1).

3.2. Winter atmospheric routes and precipitation regime

The cluster analysis of winter (DJFM) daily trajectories from 1984 to 2012 showed seven main winter transport routes (Fig.2): Northern flows (cluster1), North-Western (cluster 2 and 3), Western (cluster 4 and 5), South-Western (cluster 6) and North-Eastern (cluster 7). Figure 2 shows winter frequencies for each cluster. This classification broadly matched the annual one, except that the Iberian and Regional/Local clusters did not appear. Instead, a SW cluster was obtained.

The winter NAOi was negatively correlated with the number of rainy days and the precipitation amounts when considering all provenances together (R= -0.57 and -0.48 respectively; Table 2)

When split by clusters, winter NAOi showed negative correlations with frequency, number of rainy days and precipitation amount in the NW moderate cluster (R=-0.52, -0.69 and -0.68; $p<0.05$; Table 2). Also a negative significant correlation was observed for NAOi and precipitation amount in SW flows (R= -0.42).

When considering all provenances together, the winter WeMOi was only significantly (and negatively) correlated with precipitation amount (R= -0.38; Table 2). When split by clusters, the winter WeMOi was negatively correlated with the frequency and number of rainy days in SW and NE provenances; for the former one, also the precipitation amount was inversely correlated (Table 2). On the other hand, winter WeMOi was positively correlated with the provenance frequency and number of precipitation days in NW fast and W fast clusters (Table 2). These fast Atlantic trajectories accounted for 20% of total winter days while provenances from SW and NE about doubled this frequency (37%) and represented about 60% of winter precipitation amount (Fig. 2). The time trends of winter frequencies by cluster were also analysed. Frequencies of all clusters showed non-significant trends, except for the W fast cluster that showed a significant decreasing trend (R= -0.40; p=0.03).

3.3. Winter precipitation deposition fluxes

Winter NAOi was inversely correlated with deposition fluxes of anthropogenic compounds (H^+, NH_4^+, NO_3^- and SO_4^{2-}) in W moderate flows (Table 4).

Winter WeMOi was negatively correlated with the winter deposition of marine ions (Na^+, Mg^{2+} and Cl^-) and anthropogenic compounds (NH_4^+, NO_3^- and SO_4^{2-}) for the SW cluster (Table 4). On the other hand, a positive relationship was found between winter WeMOi and the deposition of Ca^{2+}, NH_4^+, NO_3^- and SO_4^{2-} from W fast fluxes (Table 4). When considering the whole data irrespective of provenance distinction, only a negative correlation was found between WeMOi and NO_3^- (Table 4).

Winter SW deposition fluxes accounted for 28-34% of total winter deposition for all chemical compounds (Table 5). In fact SW plus NE deposition fluxes accounted for between 50% and 65%, whereas W moderate and fast deposition fluxes only represented 9-17% and 5-10% of deposition respectively (Table 5).

4. Discussion

4.1. The influence of NAO and WeMO on the annual atmospheric circulation patterns and precipitation amount

There is a general interest regarding the effects of the atmospheric dynamics, especially in a context of climate change. We analyzed the influence of NAO and WeMO on the atmospheric transport routes and precipitation amount. The NAO is a large-scale oscillation in atmospheric mass, with centres of action near Iceland and over subtropical Atlantic (Visbeck et al. 2001). WeMO is a pattern of low-frequency variability defined as an alternative to the NAO for explaining the precipitation behavior on the East coast of the IP (Martín-Vide and Lopez-Bustins 2006). Consequently, the use of the WeMOi is justified here by its clear influence on Mediterranean coastal precipitation (Gonzalez-Hidalgo et al. 2009), which is very weakly correlated to the NAO (Rodó et al. 1997).

Analysis of the data at an annual timeframe (Table 1) showed the lack of correlation between NAOi and the frequency of provenance, the number of rainy days and the precipitation amount, whereas WeMOi was significantly correlated with several of these variables. This indicated a stronger influence of WeMO in defining the precipitation in the Mediterranean coast (Table 1). Indeed, the positive correlations between WeMOi and Atlantic flows (NW and W provenances) was in accordance with what is expected in the WeMO positive phase (Martin-Vide and López-Bustins 2006; López-Bustins et al. 2008). However, the positive relationship between WeMOi and the Mediterranean provenance was unexpected considering the air fluxes, which head eastwards, in WeMO positive phase (Martin-Vide and López-Bustins 2006; López-Bustins et al. 2008). Because of these contradictory results, cluster analysis was further explored for winter (December to March) since it is also the period where the influence of WeMO is most exacerbated in the Mediterranean coastal fringe (González-Hidalgo et al. 2009). Also the relationship between NAOi and precipitation is stronger in winter (Visbeck et al. 2001; Goodess and Jones 2002).

4.2. The influence of NAO and WeMO on the winter atmospheric transport routes and precipitation amount

Non significant correlations were observed between provenances and the positive NAO phase in winter (Table 2). In this mode, the pressure gradient between the Icelandic low and the subtropical high pressure centre is more intense than normal, so that westerly winds are stronger across northern Europe. This brings Atlantic air masses over the northern continent (associated with mild temperatures and higher precipitation) and dryer conditions across southern Europe. The negative relationship between NAO and the frequency and precipitation amounts from the W moderate cluster (Table 2) was in agreement with the fact that when the pressure gradient is low, cold and dry air masses often dominate over northern Europe and the Atlantic weather systems and storm tracks tend toward a more southerly trajectory, bringing higher than normal precipitation levels to the IP (Vicente-Serrano et al. 2011). The increase of total precipitation amount in the IP during negative winter NAO detected in previous studies (Goodess and Jones 2002; Rodríguez-Puebla and Nieto 2010; Vicente-Serrano et al. 2011), was also observed at LC.

In addition, results of the winter dataset showed that the changes in the frequency of circulations patterns were more closely associated to the influence of WeMO over the northeastern IP. The positive WeMO phase which corresponds to the anticyclone over the Azores enclosing the south-west Iberian quadrant and low-pressures in the Liguria Gulf (Martin-Vide and López-Bustins 2006) would agree with the occurrence of fast Atlantic flows. This was effectively observed for winter and for the whole year (Tables 2 and 3). The negative phase, which coincides with a central European anticyclone located north of the Italian peninsula and a low-pressure centre in the Iberian south-west (Martin-Vide and López-Bustins 2006) was well-correlated with the SW and NE clusters in winter. This corroborates that the most consistent results and the best correlations for circulation patterns were found during the winter period, as expected.

The decreasing trend of winter W fast cluster frequency and the low winter fast Atlantic frequencies observed (9%, Table 3) could be related with a major frequency of high pressures over Iberia at the end of 20th century, which prevents the arrival of Atlantic storms to Mediterranean latitudes. In fact, this could be the main cause of a significant decrease in winter precipitation registered in western and central IP, while no variations or even slight precipitation increase are detected on the eastern fringe as some north-easterly winds convey moisture to the south-eastern region (Lopez-Bustins et al. 2008). A non-significant time trend was observed for rainy days and precipitation amount in winter at LC, therefore no changes in winter precipitation behaviour were detected. Conversely, other studies showed an increase of the winter precipitation amount (Gonzalez-Hidalgo et al. 2009) as well as the torrential precipitation episodes (Goodess and Jones 2002; Martín-Vide et al. 2008) in the Mediterranean fringe of the IP.

The negative correlation (Table 2) between total winter precipitation amount at LC and the winter WeMOi indicates that higher precipitation amounts were registered from the Mediterranean than from the Atlantic provenance. This is consistent with the negative correlation between winter SW precipitation amount and the winter WeMOi at Montseny and that reported by other studies on the role of WeMO on precipitation in the eastern IP (Martín-Vide and López-Bustins, 2006; González-Hidalgo et al. 2009). It is important to note the high occurrence of rainy days from SW and NE flows, which accounted for 44% compared to 19% from fast Atlantic air masses. Both clusters represented ~ 60% of winter precipitation amount, indicating the dominant role of the negative WeMO phases in precipitation. The high SW and NE precipitation amounts (Table 3) may be related with the significant correlation between negative values of WeMOi and torrential precipitation in north-eastern IP, despite that, according to Martín-Vide et al. (2008), only 22% of torrential precipitation cases occurred in winter.

4.3. The influence of winter WeMOi and NAOi on deposition fluxes

In Europe, the effects of NAO on air temperature, storminess and winds have been thoroughly studied (Hurrell and Deser 2010, and references therein), but very few studies have approached the links between the NAO variability and the chemistry of precipitation and wet deposition. One exception to this are the works of Evans et al. (2001) and Fowler et al. (2005) for the British Islands. The former study took into account the winter NAO variability to interpret the variation of lake concentrations in the UK and concluded that a cycle of high deposition of marine ions in the early 1990s was in phase with the winter NAO indices and suggested the need for a further exploration of this relationship (Evans et al. 2001). Fowler et al. (2005) studied the effect of annual and winter NAO índices on wet deposition in the UK, but did not find significant relationships for any of the studied ions (Na^+, Cl^-, NH_4^+, NO_3^-, SO_4^{+2}, H^+); thus the NAO variability appeared to have a negligible effect for acidifying components regarding the changes in emissions in the UK.

Other studies have investigated the effect of NAO on atmospheric mineral dust and anthropogenic aerosols in the Mediterranean, and have found a correspondence between African particulate matter and NAO, particularly when considering the summer period (Moulin et al. 1997; Ginoux et al. 2004; Pey et al. 2013). However, wet deposition of dissolved African dusts did not show significant relationships with NAO in NE Spain (Àvila and Rodà, 2002). This was interpreted as resulting from two effects that vary in opposite directions: high NAO indices correspond to an increase of atmospheric dust (Cusack et al. 2012) but in years of high NAO there is a decrease in the precipitation amount in the IP, two processes that cancel each other in providing the deposition amounts (the product of concentrations times precipitation amount).

In Montseny, winter precipitation amount (Table 2) and deposition (Table 4) of NO_3^- were negatively correlated with winter WeMOi, indicating that the Catalan fringe receives higher precipitation and NO_3^--N deposition when WeMO is in its negative phase during winter. The relationships between the trajectory frequencies and precipitation parameters with the studied indices were weak or counterintuitive in the annual dataset. The winter dataset produced more consistent results. However, for the NAO teleconnection, no significant relationships were found between the NAOi and the analysed precipitation variables and, for ion deposition, only when decomposing the data for provenances, a significant inverse relationship appeared with anthropogenic ions for the Western (moderate) provenance (Table 4). Several studies in the Iberian Peninsula have shown that the influence of NAOi is lower than the indices describing the Mediterranean variability such as the WeMOi or the MOi (Mediterranean Oscillation index) either at the annual scale (Gonzàlez Hidalgo et al. 2009) or specifically for winter (López-Bustins et al. 2008; Gonzàlez Hidalgo et al. 2009).

When decomposed by provenances, winter WeMOi showed negative correlations with the deposition of marine ions (Na^+, Mg^{2+} and Cl^-) and anthropogenic species (NH_4^+, NO_3^- and SO_4^{2-}) in SW fluxes (Table 4). This implies that, during years characterized by negative winter WeMOi, SW air masses would convey higher deposition of the mentioned ions. Since the WeMO negative phase is associated to flows from the Mediterranean (Martin-Vide and López-Bustins 2006; López-Bustins et al. 2008), a higher deposition of marine ions with negative WeMOi is an expected result. However, for NO_3^- , NH_4^+ and SO_4^{2-}, which are emitted mostly from continental traffic, power generation, industrial activities and agriculture, this result suggests the presence over the Mediterranean of polluted air masses that would have been advected from highly industrialised areas in southern Europe, and then they will be delivered by Mediterranean precipitation events. In fact, Millán et al. (1997) have described a recirculation process of air masses that occurs over the Western Mediterranean and favours the accumulation of atmospheric anthropogenic pollutants across the area. These pollutants are probably incorporated into the precipitation during negative WeMO phases producing the observed enhanced deposition observed for NH_4^+, NO_3^- and SO_4^{2-}. The study of more anthropogenic-derived components in the precipitation (e.g. trace metals) could help validate this statement. On the other hand, the growing industrialisation in North Africa and the intensification of ship traffic on the Mediterranean has also been suggested contribute to anthropogenic deposition associated with Mediterranean provenances (Izquierdo et al. 2012).

In Table 4 it is seen that the NAOi has a negative significant correlation with anthropogenic pollutants but only in the West moderate cluster. Thus it is seen that both indices explain different parts of the variation, with the WeMOi influencing SW and W fast flows and the NAO influencing Western flows of shorter spatial range.

Furthermore, local emissions may also have a role. Especially, extremely intense pollution episodes associated with the transport by sea-mountain breezes of aged air masses from industrial/urban areas around major cities such as Barcelona during winter anticyclonic episodes (WAE) (Pérez et al. 2008; Pey et al. 2010). Usually, the meteorological conditions during these episodes enhance the local anthropogenic resuspension processes in urban areas, but also favour the transport of aerosols to nearby mountains such as Montseny. These specific features and the intense emissions of ammonia in the northeastern IP give rise to the formation of high amounts of secondary inorganic aerosols, mainly ammonium nitrate, and in minor proportion ammonium sulphate, recorded at LC during these meteorological scenarios (Pey et al. 2010). At the end of the WAE, stagnant polluted air masses are replaced by air masses coming from Mediterranean or Atlantic (Pey et al. 2010). As Mediterranean episodes are related with the occurrence of precipitation (Pérez et al. 2008), NH_4^+, NO_3^-, and SO_4^{2-} from local anthropogenic emissions may be incorporated into precipitation and contribute to the correlation with the WeMOi. The WAE in wintertime was recurrent with a mean frequency from 15% to 25% of the days (Cusack et al. 2012).

A positive correlation was observed between winter WeMOi and the deposition of Ca^{2+}, NH_4^+, NO_3^- and SO_4^{2-} in W fast fluxes, indicating higher deposition of the mentioned ions associated to the positive phase of WeMO. This is consistent with the description of the WeMO positive phase as triggering air fluxes from the Atlantic into the IP (Martin-Vide and López-Bustins 2006) that would arrive to the Mediterranean coast. Previous studies showed that Atlantic advection episodes could have a cleaning effect on aerosols at LC (Cusack et al. 2012), therefore the winter WeMOi relationship with the anthropogenic-derived species may be attributed to WAE as has been explained above. However, this provenance only contributed to 9% (Table 3) of total precipitation at our site, compared to the SW provenance which accounted for 33% total precipitation. In terms of deposition, the SW provenance accounted for 29%, 28% and 32% of NO_3^--N, NH_4^+-N and SO_4^{2-}-S fluxes (Table 5) compared to 7%, 10% and 8%, of NO_3^--N, NH_4^+-N and SO_4^{2-}-S fluxes for the fast West provenance. This suggests that most of the anthropogenic pollutants deposited at Montseny can be attributed to the arrival of Mediterranean air masses. Thus the WeMO index, which in its negative phase triggers the transport of air masses from the Mediterranean to the Iberian Peninsula, can be partially used to describe the precipitation amounts and the anthropogenic pollutant deposition variability at the northeast fringe of the Iberian Peninsula.

5. Conclusions

This study has shown that the influence of the NAO index is lower than an index describing the Mediterranean variability, the WeMOi, at a site in the northeast of the Iberian Peninsula. Consistently with other works in the west Mediterranean, winter precipitation amount was inversely correlated with winter WeMOi and NAOi. The relationship between element deposition and indices of climatic variability has not been usually explored in the literature for Europe, other than a few studies in the British Isles in which the NAO variability had a negligible effect for acidifying components albeit a probable influence for marine ions.

Our results indicated the relevant role of the WeMO and the NAO in the deposition of anthropogenic pollutants (H^+, NH_4^+, NO_3^- and SO_4^{2-}). For marine ions (Cl^-, Na^+, Mg^{2+}) only the WeMOi had a significant effect. The cluster classification of provenances indicated that fast Atlantic air flows corresponded to positive winter WeMO indices, while negative winter WeMOi were associated to NE and SW circulations in winter. Most of the ion deposition was conveyed by air masses from the Mediterranean and was significantly correlated with the negative phase of the WeMO. These Mediterranean episodes can bring pollution from aged air masses recirculated in the Western Mediterranean and also incorporate local anthropogenic emissions from winter anticyclonic episodes.

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TABLES

Table 1. Annual dataset: significant correlation coefficients ($p<0.05$) between the studied climatic variability indices (NAOi and WeMOi) and the provenance frequency, number of rainy days and the precipitation amount distinguished by provenances from the cluster analysis, for the period 1984-2012.

Annual provenances	Provenance frequency (%)		Number of rainy days		Precipitation amount (mm)	
	NAOi	WeMOi	NAOi	WeMOi	NAOi	WeMOi
1. N		-0.54		-0.35	-0.40	
2. NW		0.63		0.46		0.41
3. W		0.49		0.35		
4. NE						
5. MED		0.49		0.46	0.39	
6. IP						
7. REG		0.52		0.59		
TOTAL						

Table 2. Winter (DJFM) dataset: significant correlation coefficients ($p<0.05$) between the studied climatic variability indices (NAOi and WeMOi) and the provenance frequency, number of rainy days and the precipitation amount distinguished by provenances from the cluster analysis for the period 1984-2012.

Winter provenances	Provenance frequency (%)		n rainy days		Precipitation amount (mm)	
	NAOi	WeMOi	NAOi	WeMOi	NAOi	WeMOi
C1.N fast						
C2. NW fast		0.59		0.45		
C3. NW slow						
C4. W fast		0.69		0.48		
C5.W moderate	-0.52		-0.69		-0.68	
C6. SW		-0.57		-0.59	-0.42	-0.58
C7. NE		-0.66	-0.39	-0.60		
Total			-0.57		-0.48	-0.38

Table 3.. Frequency of rainy days and precipitation amount for each cluster in winter (DJFM) from 1984 to 2012.

	n rainy days	Rainy days vs. total rainy days	Precipitation amount (mm)	% Total precipitation amount
C1.N fast	69	7%	145	2%
C2. NW fast	82	8%	332	5%
C3. NW slow	155	15%	878	12%
C4. W fast	107	11%	649	9%
C5.W moderate	152	15%	976	13%
C6. SW	208	21%	2405	33%
C7. NE	238	23%	1928	26%
Total	1011	100%	7313	100%

Table 4. Significant correlation coefficients ($p<0.05$) between winter (DJFM) NAOi/WeMOi and winter ion deposition in precipitation for each cluster in the 1984-2012 period.

Winter	Winter NAOi							Winter WeMOi						
deposition	H^+	Na^+/Cl^-	Mg^{2+}	Ca^{2+}	NH_4^+	NO_3^-	SO_4^{2-}	H^+	Na^+/Cl^-	Mg^{2+}	Ca^{2+}	NH_4^+	NO_3^-	SO_4^{2-}
C1.N fast														
C2. NW fast														
C3. NW slow														
C4. W fast											0.45	0.52	0.43	0.55
C5.W moderate	-0.46				-0.54	-0.46	-0.46							
C6. SW									-0.47/-0.45	-0.48		-0.50	-0.67	-0.42
C7. NE														
Total													-0.45	

Table 5. Winter element deposition (in kg ha^{-1} y^{-1}) for each cluster and the percentage accounted by each one for the period 1984-2012.

Winter ion deposition (DJFM)	C1.N fast	C2. NW fast	C3. NW slow	C4. W fast	C5.W moderate	C6. SW	C7. NE	Total
Na^+	0.019	0.064	0.180	0.120	0.171	0.556	0.518	1.63
	1.2%	3.9%	11.1%	7.3%	10.5%	34.2%	31.8%	100%
Ca^{2+}	0.031	0.088	0.213	0.098	0.353	0.709	0.610	2.10
	1.5%	4.2%	10.1%	4.7%	16.8%	33.7%	29.0%	100%
Mg^{2+}	0.004	0.011	0.033	0.022	0.031	0.088	0.082	0.272
	1.6%	4.2%	12.3%	8.0%	11.3%	32.3%	30.3%	100%
NH_4^+-N	0.015	0.056	0.082	0.062	0.086	0.177	0.146	0.626
	2.5%	9.0%	13.2%	10.0%	13.8%	28.3%	23.3%	100%
NO_3^- -N	0.017	0.044	0.091	0.048	0.085	0.190	0.170	0.645
	2.6%	6.8%	14.1%	7.4%	13.2%	29.4%	26.4%	100%
SO_4^{2-} -S	0.022	0.070	0.163	0.094	0.150	0.398	0.358	1.26
	1.8%	5.5%	13.0%	7.5%	12.0%	31.7%	28.5%	100%
Cl^-	0.035	0.121	0.344	0.236	0.330	1.03	0.959	3.05
	1.2%	4.0%	11.3%	7.7%	10.8%	33.7%	31.4%	100%
H^+	0.0006	0.0009	0.005	0.002	0.003	0.011	0.009	0.031
	1.9%	3.0%	16.7%	7.0%	8.6%	34.5%	28.4%	100%

FIGURES

Figure 1. Location of La Castanya study site (LC) and WeMO phases

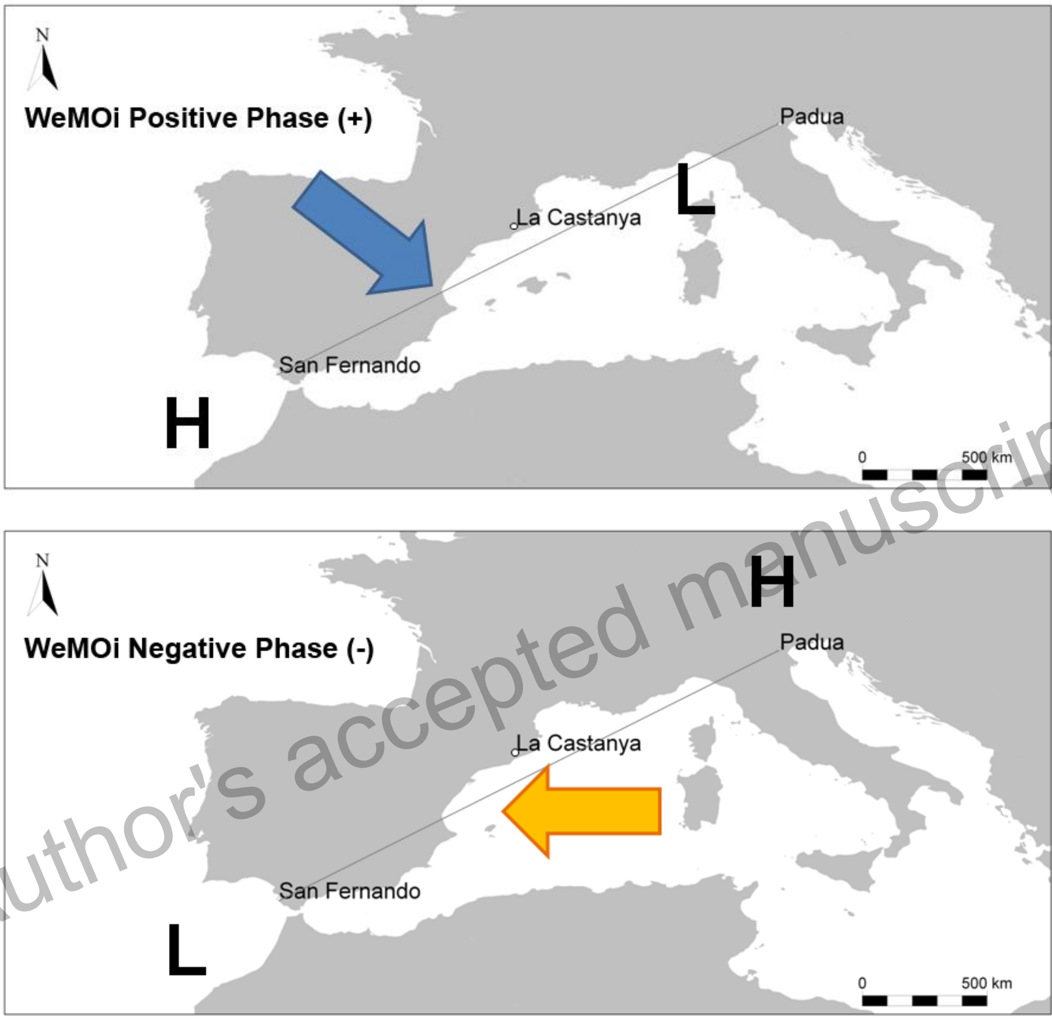

Figure 2. Back-trajectory centroids and frequency associated to each cluster in winter (DJFM) for 1984-2012 periods. Back-trajectories (72h) from LC calculated at 1500m asl.