



## Impact of the North Atlantic oscillation

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# Impact of the North Atlantic Oscillation on the variations of aerosol ground levels through local processes over Europe

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## Abstract

This contribution assesses non-anthropogenic variations in ground-level aerosol concentrations over Europe associated to changes in the phase of the North Atlantic Oscillation (NAO). The NAO controls a large amount of the European climate variability with asymmetric impacts in both time and space. Based on simulated data and focusing on how the local atmospheric processes (without considering large-scale mechanisms) governed by the NAO affect the levels of various aerosol species, this study highlights that positive NAO phases favor increased aerosols levels in southern (northern) regions in winter (summer), while negative NAO phases enhance them in northern (southern) regions in winter (summer). Variations are up to and over 100 % for most aerosols, being clearly related to the NAO-impact on precipitation and wind, as they act to clean the atmosphere through removal and dispersion processes, and to the NAO-impact on the radiation balance (i.e. cloudiness) as it rebounds on the biogenic emitting activity and on the oxidative capacity of the atmosphere. Beyond deepening on the understanding of fundamental interactions between climate and air quality, these results provide a basis for improving the potential predictability of this later since much work is being done in order to gain accuracy in the NAO predictions.

## 1 Introduction

Air pollution is a major environmental and health problem affecting industrialized and developing countries around the world. Main detrimental consequences of the exposure of humans to photochemical and particulate matter pollution include respiratory difficulties, especially for sensitive people (Pope et al., 2006). The results of the APHEIS study (Ballester et al., 2008) indicate that atmospheric pollution causes the premature death of over 310 000 dwellers in the European Union each year. The most serious problems in Europe are related with particulate matter with a diameter of less than 10 micrometers ( $PM_{10}$ ), nitrogen dioxide and ozone. In this sense, worldwide epi-

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demological studies show a consistent increase in cardiac and respiratory morbidity and mortality from exposure to air pollution (e.g. Pope et al., 2009). Besides, ecosystems are also affected, with losses of agricultural crops and damages in aquatic and terrestrial ecosystems having been reported (Van Dingenen et al., 2009).

5 In order to anticipate when and why episodes of air pollution arise and how they can be abated, reliable estimations of air pollution levels and a better understanding of the chemico-physical processes behind them are of paramount importance.

Air pollution levels depend on both emissions (either natural or anthropogenic) and the atmospheric conditions steering and transforming them through processes related to chemistry, transport and removal (Jacob and Winner, 2009). Precipitation provokes wet deposition, hence favoring the removal of airborne pollutants, and wind favors their transport and dispersion, although enhanced winds also promote the formation of marine aerosols over the water mass areas. On the other hand, radiation levels and temperature play a major role on gas-phase chemistry through the photolysis of primary and secondary pollutants (Katragkou et al., 2010), determining also the vegetation activity and thereby modifying natural emissions.

10 Despite the great climate heterogeneity and variability that characterizes Europe, just a few large-scale teleconnection modes control a large amount of it (Trigo et al., 2008). In particular, many studies establish the fundamental role of the North Atlantic Oscillation (NAO) at this regard, affecting especially western Europe with an asymmetric impact between northern and southern areas (Osborn et al., 1999; Wanner et al., 2001; Trigo et al., 2002, 2008). The NAO pattern consists of a meridional gradient in Sea Level Pressure (SLP) over the North Atlantic with centers roughly around the Azores Islands (high pressures) and near Iceland (low pressures). During its positive phases (NAO<sup>+</sup>), such a dipole in SLP is enhanced with the consequent intensification of westerly winds in northern Europe associated with an intensified jet stream. This configuration promotes also the steering of most storms over northern regions, advecting humid air from the ocean, hence favoring cloudiness and precipitation. Contrary and consistently, NAO<sup>+</sup> reduces cloudiness and precipitation in southern Europe. Con-

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versely, the intense zonal circulation promoted by the negative NAO phases (NAO<sup>-</sup>) enhances the westerly flow penetrating in southern Europe through the Iberian Peninsula (Jerez et al., 2013b) and extending until eastern Europe and allows cyclones to follow more southern paths within the European continent (Trigo, 2006). Most studies have focused their analysis on NAO-climate links during the winter season, where the SLP gradient is stronger and the climatic impact of this large-scale mode of variability is extensive to Eurasia (Wanner et al., 2001; Trigo et al., 2002) but also North America, Greenland, the Arctic and North Atlantic oceans (Hurrell and van Loon, 1997). Readers looking for comprehensive reviews of the dynamics of the NAO and associated impacts should consult the books by Hurrell et al. (2003); Vicente-Serrano and Trigo (2011). However, recent works have shown that the summer NAO still plays an important role on shaping the climate of northern and central Europe (albeit less relevant than in winter) including parts of the Mediterranean basin (Folland et al., 2009; Bladé et al., 2012).

These NAO-related impacts on the atmospheric fields are bound to inflict an important impact on air pollution levels (Dayan et al., 2008; Sanchez-Lorenzo et al., 2008; Chiacchio and Wild, 2010), particularly on aerosols concentrations. This influence should arise through both local processes and large-scale air pollutants transport, either from Europe to Arctic regions (Eckhardt et al., 2003) or from North America and Africa towards Europe (Moulin et al., 1997; Dayan et al., 2011; Christoudias et al., 2012). However, since few studies are devoted to explore air pollution levels from a climatic perspective, the impact of the NAO in such a framework is still hardly established. Hence, the objective of the present study is to elucidate the signature of the NAO in terms of mean concentration of aerosols in a region covering the entire Mediterranean basin from north Africa to north Europe, as well as the associated underlying mechanisms. To achieve this goal, we use a numerical simulation of the atmospheric chemical composition that spans three decades of the recent past without considering variations in the anthropogenic emissions, thus allowing to isolate the natural variations in the aerosol levels. Besides, the simulation was designed to disregard the contribution from

large scale transport. Hence, we focus on the role played by local processes as they are governed by the NAO, restricting the evaluation to the ground-level.

The structure of this work is as follows. Section 2 describes the modeling system and the experimental set-up. Section 3 provides the methodology. Section 4 presents the results. Finally, Section 5 summarizes and discusses the main conclusions.

## 2 Data

### 2.1 Air-quality modeling system

The modeling system consists of a climatic version of the Fifth-Generation Pennsylvania State University – National Center for Atmospheric Research Mesoscale Model (MM5) (Grell et al., 1994) driven by ERA40 reanalysis (Uppala et al., 2005), when available, or ECMWF analysis data when not, coupled off-line to the CHIMERE chemistry transport model (Bessagnet et al., 2004; Rouil et al., 2009). MELCHIOR2 gas-phase mechanism is implemented within CHIMERE (Derognat et al., 2003). The chemistry transport model includes aerosol and heterogeneous chemistry, distinguishing among different chemical aerosol components, namely nitrate, sulfate, ammonium, elemental and organic carbon with three subcomponents (primary, secondary anthropogenic and secondary biogenic) and marine aerosols. Unspecified primary anthropogenic aerosols and aerosol water are additionally kept as separate components. The model considers the thermodynamic equilibrium using the ISORROPIA model (Nenes et al., 1998). Last, the aerosol microphysical description for CHIMERE is based on a sectional aerosol module including 6 bins from 10 nm to 40  $\mu\text{m}$  using a geometrical progression.

The CHIMERE domain considered in the modeling system covers all the Mediterranean basin extending to northern Europe having a spatial resolution of  $0.2^\circ$  in the horizontal, which is around 25 km at the European latitudes considered, and eight vertical levels unevenly spaced up to 550 hPa. This resolution enhances from previous

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works (e.g. Carvalho et al., 2010; Katragkou et al., 2010; Jiménez-Guerrero et al., 2011; Juda-Rezler et al., 2012; Manders et al., 2012).

The fields from MM5 (provided with a spatial resolution of 90 km) are bilinearly interpolated to the CHIMERE working grid. Detailed descriptions of the climate modeling system used and its skill to realistically reproduce the main regional features of the climate in the target domain, such as the temperature and precipitation annual cycles or the interaction between the large-scale circulation and the orography, which largely modulates the rainfall patterns having an important influence on air quality, can be found in Gomez-Navarro et al. (2011) and Jerez et al. (2013a).

Boundary conditions for the chemistry transport model are based on the global chemistry model LMDz-INCA2 (Szopa et al., 2009) developed by the Laboratoire des Sciences du Climat et l'Environnement (LSCE). A detailed description of the Interactive Chemistry and Aerosol (INCA) model is presented in Hauglustaine et al. (2004) and Folberth et al. (2006). For aerosols, boundary conditions are taken from the GOCART model (Chin et al., 2002). Monthly mean data are interpolated in the horizontal and vertical dimensions to force the major chemical concentrations at the boundaries of the CHIMERE domain. Although the influence of using climatological boundary conditions on ground level concentrations is largely overwhelmed by local processes (Jiménez-Guerrero et al., 2012), it should be acknowledged that, in particular, this hampers to capture the interannual variability of the NAO-impact on the aerosol concentration levels at the domain boundaries, thus avoiding the evaluation of large-scale transport mechanism related to the NAO phase. On the contrary, this experimental design allows to better isolate and understand the role of the local processes, including the pollutants transport between different areas within our domain.

Year-to-year varying anthropogenic emissions are derived from the EMEP database on a monthly basis (Vestreng et al., 2009). Natural emissions depend on climate conditions, and consequently they are modeled according to the MM5 meteorological outputs. However, the levels of air pollutants are estimated without considering possible changes on vegetation, land use or any feedback from the chemical compounds to the

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meteorological fields. Biogenic emissions were generated dynamically using MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006) with the parametrization form of the canopy environment model. The model estimates hourly isoprene, monoterpene, and other BVOC emissions based on plant functional type and as a function of temperature and ground level shortwave radiation.

This modeling system has been validated by comparing a simulation covering the period 1990–2010 with EMEP observations (Tørseth et al., 2012). A thorough evaluation of the modeling approach is not included in the main objectives of this work, and that task constitutes the focus of an on-going paper. In any case, we acknowledge that it is opportune to show shortly that the simulated series of aerosols concentration correlates acceptably and captures a large fraction of the variability of the observational series. These two aspects are the most relevant for the assessment performed below, while systematic biases would not represent a major concern as they should be largely canceled when computing the composites of the NAO-impact (see next Section). For the sake of brevity, we provide here the results for the simulated concentration of aerosols under several size-thresholds, namely  $PM_{10}$  and  $PM_{2.5}$  (Fig. 1). For both cases, correlations between simulated and observational series are overwhelmingly above 0.8, in most cases above 0.9, both in winter and summer. As well, it is possible to verify that biases in the standard deviation of the simulated series are largely negligible in comparison to their mean values (being orders of magnitude smaller). It is also worth mentioning that normalized biases are always found to stay below 30 % the worst (not shown), which is in the top range expected (Pay et al., 2010). This guarantees the phase accordance (timing) between the simulated and observational series, their similar amplitude and, also, the quantitative accuracy of the simulated climatologies, hence making us confident on the validity of the modeling system for the purpose of this study.

## 2.2 Experimental set-up

In order to isolate the influence of climate variability on air quality, it arises mandatory to avoid the signals derived from human policies. During the last decades, these policies

have committed European countries to strongly reduce the emissions of contaminants to the atmosphere, which has caused strong non-natural trends in the observational series of almost all aerosols species (Vestreng et al., 2009; Tørseth et al., 2012). Hence, the impact of the NAO has been evaluated here based on a 30 yr long MM5-CHIMERE simulation for which emissions were fixed at their 2005 mean levels. The simulated period (1970–1999) is not particularly significant in itself, but its length is relevant as it supports the robustness of our results from a climatic perspective.

### 3 Methodology

This assessment focuses on several aerosol families and species, namely natural inert aerosols (sea salt aerosols, SALT, and wind-blown and resuspended dust, DUST), secondary inorganic aerosols (sulfate,  $\text{SO}_4^{2-}$ , nitrate,  $\text{NO}_3^-$ , and ammonium,  $\text{NH}_4^+$ ), organic matter (OM), with particular attention to secondary organic aerosols (SOA), and elemental carbon (EC). The total concentrations of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  are also studied.

The analysis is performed at the seasonal timescale for winter (December-to-March averages) and summer (June-to-September averages). This decision is based on previous tests performed at the monthly timescale showing the strongest responses in those months, with very similar structures within each season considered but very different in each one of them. The remaining months from the two transitional seasons depicted intermediate and softer signals being disregarded from the rest of the analysis.

In order to assess the NAO-impact, we have followed a common approach adopted in previous studies (e.g. Suski and Ridgway, 2007; Gouveia and Trigo, 2008) and defined classes of positive ( $\text{NAO}^+ \geq 70\text{th percentile}$ ) and negative ( $\text{NAO}^- \leq 30\text{th percentile}$ ) NAO phases using the seasonal means of the monthly NAO index provided by the Climate Prediction Center (CPC) from the National Ocean and Atmospheric Administration (NOAA). This procedure ensures the balance between the number of events of each class that will be considered. Within the 30 yr simulated period (1970–1999), we obtain 9 winters and summers of each NAO class (Fig. 2). The NAO-impact on the

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assessed magnitudes is then evaluated through composites showing the differences in the mean fields between positive and negative NAO phases. Several significance controls are applied to these differences ensuring both their statistical robustness and physical consistence, so that they are just considered when fitting the following criteria: (1) being statistically significant above the 90 % confidence level, and (2) being supported by statistically significant temporal correlations (above the 90 % confidence level) between the NAO and the corresponding chemical component series. Statistical significance is evaluated by performing two-tailed  $t$  tests for the null hypothesis of equal means or zero correlation respectively (Snedecor and Cochran, 1989). Moreover, the signal-to-noise ratio, defined as the ratio between the absolute value of the NAO<sup>+</sup> minus NAO<sup>-</sup> differences in a specific magnitude and the standard deviation of the whole seasonal series of such a magnitude, is considered as a relative measure of the importance of the NAO-impact. In particular we have blurred the areas where the signal-to-noise ratio is below the unit. In the areas not blurred, the NAO-impact exceeds one standard deviation of the series and hence we are likely moving from one tail to the other of the probability density function describing the variability in the levels of the assessed magnitude when the NAO phase changes.

## 4 Results

### 4.1 Revisiting the NAO-impact on atmospheric conditions

Several works have been devoted to explore the role and signature of the NAO in the European climate (e.g. Hurrell, 1995; Hurrell and van Loon, 1997; Hurrell et al., 2003; Trigo et al., 2002, 2008). Indeed, the reported NAO-impacts on the climatic variables motivated the present study. Other works have shown a good capacity of the general circulation models to reproduce the large-scale patterns of this climatic impact over Europe particularly for winter (Osborn et al., 1999; Osborn, 2011; Hurrell et al., 2003) but also for summer (Bladé et al., 2012). Hence, this Section does not intend to provide

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novel insights, but essentially to evaluate the ability of our climate simulation (that is driving the CHIMERE run) to simulate the NAO-impact on the atmospheric conditions. This fulfills two relevant requirements, namely to (1) guarantee that it is effectively able to reproduce the known NAO-impacts on the European climate, and (2) provide an appropriate context for the interpretation of the following results of the NAO-impact on aerosols concentrations, since the analysis is specifically focused on those variables driving air pollution levels.

Figure 3 further confirms the expected NAO-impact on the wind field, showing asymmetric responses in winter and summer. In winter,  $\text{NAO}^+$  ( $\text{NAO}^-$ ) promotes a windy westerly flow in northern (southern) Europe (Fig. 3a, c), with the largest significant differences in the wind speed (above 10%) appearing northward in the western sector of the domain (Fig. 3c). Conversely, in summer,  $\text{NAO}^-$  phases have associated a stronger westerly flow in northern Europe than  $\text{NAO}^+$  phases (Fig. 3b, d), with differences in wind module exceeding 5% and extending more eastward than in winter. Windier conditions associated to the  $\text{NAO}^-$  phase prevail also in the south-west of the domain in summer, mainly in the surroundings of the Iberian Peninsula (differences about 5%) (Fig. 3d).

Consequences in precipitation, cloudiness and temperature derived from the former conditions are also provided in Fig. 3, as these variables play a key role on the levels of air pollutants in general, and aerosols in particular. Obtained patterns are in good agreement with previous works for winter (e.g. Osborn, 2011) or summer (Bladé et al., 2012) relating them to the westerly winds advection of humid air from the Atlantic, which favors the formation of clouds and enhances precipitation.  $\text{NAO}^+$  minus  $\text{NAO}^-$  differences in precipitation (Fig. 3e, f) are larger in winter than in summer, when they are overall negative and affect mainly southern Europe ranging west-to-east from 40% ( $50 \text{ mm month}^{-1}$ ) to 20% ( $20 \text{ mm month}^{-1}$ ). In summer, these differences reach 20% (up to  $30 \text{ mm month}^{-1}$ ) over large sectors of Europe, being negative in northern Europe while positive, and less important, in the southern affected areas. Consistently, the composites for the column integrated cloud water (a variable representative of cloudi-

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ness) (Fig. 3g, h) show negative values in southern Europe in winter (resembling the west-to-east gradient of the precipitation signal, with differences ranging from 30 to 10 %) and northern areas in summer (differences around 10 %), and positive values in north-western areas in winter (i.e. NAO<sup>+</sup> enhancing cloudiness there, around 20 %).

Regarding the NAO-impact on mean 2-meter temperature, the most important signal consists of positive NAO<sup>+</sup> minus NAO<sup>-</sup> differences (up to 3 K) largely spread over northern Europe in winter (Fig. 3i). Smaller negative (positive) differences (up just to 1 K) appear also over some southernmost (northern) areas in winter (summer) (Fig. 3i, j).

These results support the ability of the climate simulation to reproduce the expected responses to the NAO phase, providing a meaningful representation of the atmospheric conditions governing during positive and negative NAO events that will be used to understand the NAO-impact on aerosol concentration levels assessed in the next Section.

## 4.2 NAO-impact on mean ground-level aerosols concentrations

NAO<sup>+</sup> minus NAO<sup>-</sup> composites of mean ground-level concentration of the various aerosol species are provided in Figs. 4 and 5. In general, the positive phase of NAO enhances aerosol concentrations in southern Europe in winter and in northern Europe in summer. These positive NAO<sup>+</sup> minus NAO<sup>-</sup> differences match generally well the sectors characterized by negative signals in wind speed, precipitation and cloudiness (Fig. 3). Thus such patterns can be well explained by the diluting effect of stronger winds and scavenging processes by an enhanced precipitation and by the inhibitory effect of enhanced cloudiness as it implies reduced shortwave solar radiation (a main precursor for photochemistry and biogenic emitting activity).

### 4.2.1 Winter signals

As a general rule, in winter, the regions holding the largest signals are predominantly located in the Mediterranean basin, particularly over the Iberian Peninsula, northern

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Italy and the Balkans (Figs. 4 and 5, left column). There, results show increases in the mean concentration of all aerosols species during positive NAO phases exceeding the 50 % of the mean levels during negative NAO phases, even doubling these latter in some occasions. In absolute values (although these should be taken with care due to our experimental design not accounting for real emission levels), the largest variations between NAO phases reach  $5 \mu\text{g m}^{-3}$  and affect the inorganic compounds ( $\text{DUST}$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^-$  and  $\text{NO}_3^-$ ), while the change in the concentration of carbonaceous material is just about  $1 \mu\text{g m}^{-3}$ . It is worth stressing the slight but still significant negative patches of  $\text{NAO}^+$  minus  $\text{NAO}^-$  differences in the British Islands and in north-eastern Europe that appear in the patterns of OM and EC, indicating a distinct response of this compounds to the same NAO phase depending on the latitude.

The NAO-impact on  $\text{NO}_3^-$  concentration levels is negligible over most of Europe. However, the removal effect of the  $\text{NAO}^-$  phase associated with enhanced precipitation in northern Italy (not counteracting by enhanced temperatures) seems to play a key role at this regard in this  $\text{NO}_3^-$ -strong emitting area.

Contrary to the rest of aerosol species, enhanced winds promote the formation of SALT (sea salt aerosol emissions depends on the cube of the wind speed), thus leading to higher concentrations. This is mainly observed over the water mass areas, with  $\text{NAO}^+$  leading to 40 % higher SALT concentrations in the surroundings of the British Islands, which matches with the  $\text{NAO}^+$  enhanced winds observed in this area (Fig. 3c), but diminishing them in the southern Mediterranean (Fig. 4a). Singular positive signals appear also in the Gulf of Genoa (north-westward of Italy) and the strait of Gibraltar (southward of Spain). Regarding these three latter cases, it can be roughly appreciated in Fig. 3a that  $\text{NAO}^+$  promotes windier conditions over the mentioned areas (longer arrows) than  $\text{NAO}^-$ , although these differences in wind speed do not remain after applying the significance controls as they are not reflected in Fig. 3c.

The NAO-impact on winter air quality is appreciable in the various size-classes of particulate matter. Figure 5g, h shows that  $\text{NAO}^+$  enhances 20–40 % the concentrations of both  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  in the southern European regions (where the PM clima-

tologies present the higher values and the number of exceedances of the limit values for the protection to human health are more frequent), and also, but to a smaller extent, in some northernmost areas. Therefore, the winter NAO phase has a clear impact on the air quality-related human healthy risk in this season.

#### 4.2.2 Summer signals

In summer, the highest signals within our domain appear in the British Islands, northern France, Belgium, Netherlands, northern Germany and northern Poland (Figs. 4 and 5, right column). It is also interesting to see the recurrent positive signals appearing over Italy also in this season, although it should be acknowledged the patchy nature of them.

As in winter, differences in the aerosol concentrations between NAO phases are up to 100 %, demonstrating the profound impact of the NAO in this area/season. Likewise, the obtained NAO<sup>+</sup> related increments are, in absolute values, about (even over) 5 µg m<sup>-3</sup> for each inorganic species but SALT (which shows smaller signals, Fig. 4b). The NAO<sup>+</sup>-related increases in the concentration of the organic and carbonaceous aerosols are also a bit smaller (when they are expressed in µg m<sup>-3</sup>).

It is worth stressing the signal in mean SOA levels (Fig. 5d), which can be doubly related to the reducing effect of NAO<sup>+</sup> in precipitation (Fig. 3f), as it prevents wet deposition, and to the increased temperatures during NAO<sup>+</sup> enhancing the biogenic emissions (such as isoprene and monoterpenes), which leads to higher levels of biogenic SOA.

The NO<sub>3</sub><sup>-</sup> signal appears again mainly restricted to the stronger emitting-areas, being located in this season between northern France, Belgium and Germany (Fig. 4j). There, the reduced precipitation during NAO<sup>+</sup> events doubles the concentrations of nitrate in comparison to the levels during NAO<sup>-</sup> phases.

SALT depicts slight variations associated to the NAO phase in summer (around 0.5 µg m<sup>-3</sup>, Fig. 4b). However, contrary to the winter analysis, significant impact areas are now located over land areas and not over the sea, namely in eastern Iberia

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and Italy and in northeastern Europe, where the results show positive NAO<sup>+</sup> minus NAO<sup>-</sup> differences.

Last, PM<sub>2.5</sub> and PM<sub>10</sub> show differences up to 10 and 20 µg m<sup>-3</sup> respectively (higher concentrations during NAO<sup>+</sup>), mainly concentrated in the northernmost areas of the domain and representing variations up to 20–40 % between NAO phases (Fig. 5h, j).

## 5 Conclusions

This study establishes the strong impact that the NAO-related local atmospheric processes have on mean ground-level aerosol concentrations over Europe. For that we use a 30 yr long air quality simulation with a spatial resolution of 25 km over the target region in which the masking influence of human policies aimed at reducing emissions has been intentionally omitted. Moreover, this simulation allows isolating the influence of the local processes, i.e. those taking place within the boundaries of the domain, as the boundary conditions for the aerosols concentration levels did not vary from year to year in our experimental design. In this sense, it must be underlined that our results should not be considered deterministic.

The results show impacts with asymmetries in both time (i.e. between seasons) and space (i.e. between northern and southern areas). In winter, higher ground-level concentrations of all aerosol species (except for sea salt) are observed around the Mediterranean basin during the positive NAO phases, while these signals are northward shifted in the summer season. These differences involve variations up to and over 100 %. Eventually, softer signals of opposite sign (i.e. NAO<sup>-</sup> enhancing the ground-level concentration of aerosols) are observed in northern (southern) areas in winter (summer).

The causes for these NAO-related variations in the levels of aerosols have to be sought in a multiplicity of factors varying between NAO positive/negative phases. The main climate-related mechanisms include (1) increased temperature (particularly in northern Europe), (2) different distribution of precipitation patterns across Europe, (3) increased photolysis of primary and secondary pollutants due to lower cloudiness, and

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(4) the cleaning effect of enhanced winds (Katragkou et al., 2010). According to the results of this work, precipitation drives the modification in the concentration of most aerosol components (in both northern and southern Europe), since a decrease in the precipitation modeled leads to a regional increase in the levels of secondary inorganic aerosols and mineral matter (e.g. Jiménez-Guerrero et al., 2012; Manders et al., 2012). As well, weaker winds favor the increase of particulate matter in polluted regions such as large cities or entire industrial regions (e.g. Po valley in northern Italy and the Rhine-Ruhr area in northern Germany and Holland). Also, the enhanced oxidative capacity of the atmosphere with high temperatures causes SO<sub>2</sub> gas-phase emissions to turn into the particulate phase, thus increasing sulphate concentrations. Last, the levels of secondary organic aerosols (SOA) are conditioned by the dependence of biogenic emissions on the climatological patterns of variability. In this sense, SOA over Europe is mainly driven by the warming-induced increase in biogenic emitting activity. Although vegetation is kept invariable in the simulation analyzed here, MEGAN estimations of these emissions strongly depends on shortwave radiation and temperature (Guenther et al., 2006), which are substantially conditioned by the cloudiness and thereby by the NAO. Accordingly, the lower cloudiness associated to NAO<sup>+</sup> phases mostly in southern Europe arises as a main driver for the secondary conversion of aerosols.

These results deepen on the knowledge between the climatic conditions and air quality levels, highlighting that the great dependence of the European climate on the NAO phase has associated strong natural variations in the aerosols concentration levels. Additionally, we are confident that these results can provide the basis for inferring future air quality scenarios from either future projections or short-to-medium range forecasts of the NAO. Although the potential predictability of the NAO or other large-scale climatic indexes is still moderate (Gámiz-Fortis et al., 2002; Saunders and Quian, 2002) and future projections of the NAO differ much from one experiment to the other (Hurrell et al., 2003), there are realistic expectations that this would be largely improved in the near future (e.g. Brands et al., 2012).



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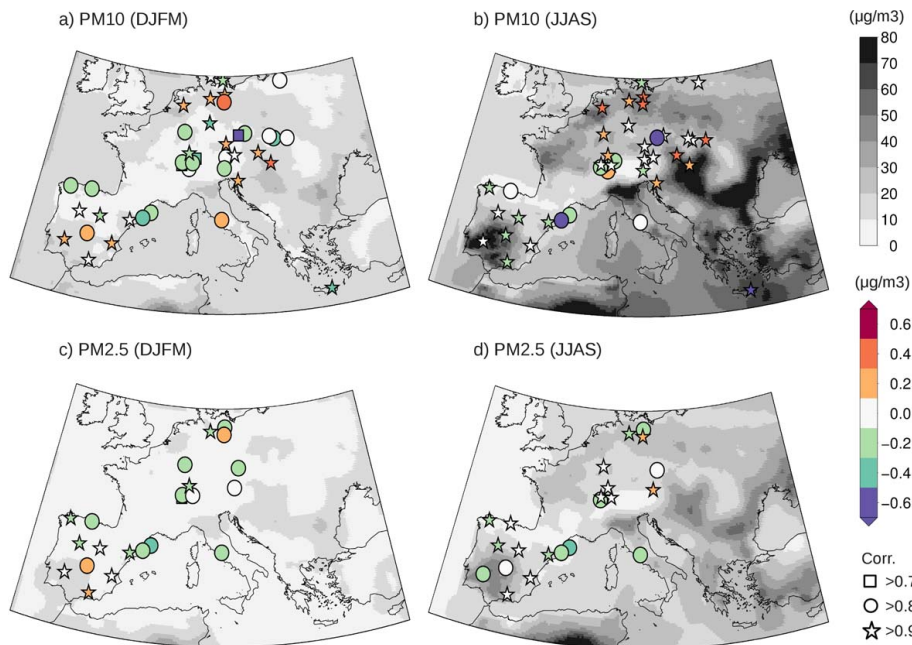
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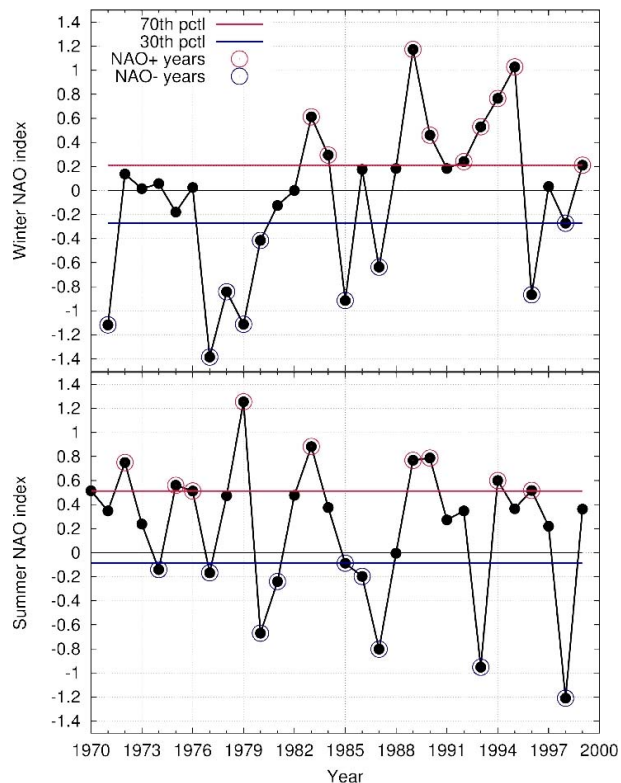




**Fig. 1.** Evaluation of the MM5-CHIMERE air quality modeling system described in Sect. 2.1. Gray shaded colors depict the simulated climatologies in  $\mu\text{g}\text{m}^{-3}$  of  $\text{PM}_{10}$  (up) and  $\text{PM}_{2.5}$  (bottom) in winter (DJFM averages, left) and summer (JJAS averages, right). The comparison of the simulated series with EMEP observations is provided by symbols: their color shows the difference in the standard deviation of the series between the simulation and the observations (in  $\mu\text{g}\text{m}^{-3}$ ), and their shape informs on the magnitude of the temporal correlation between the simulated and the observed series.

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**Fig. 2.** Winter (up) and summer (bottom) NAO series for the period 1970–1999. The former are DJFM averages (therefore resulting in 29 values) and the latter are JJAS averages (resulting in 30 values). The red (blue) lines denote the 70th (30th) percentile value of each series, having been highlighted the years/values with a NAO index above (below) that percentiles, i.e. those selected as NAO<sup>+</sup> (NAO<sup>-</sup>) events for the composites analysis.

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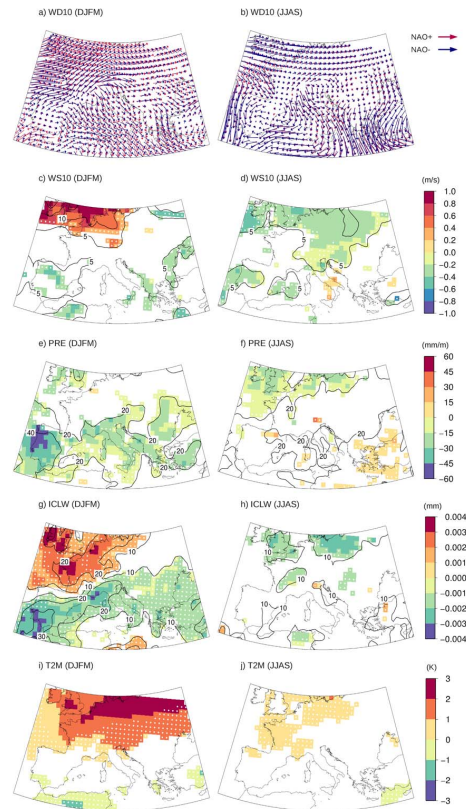
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**Fig. 3.** NAO-impact in winter (left) and summer (right) on the atmospheric conditions. **(a, b)** depict mean 10 m wind direction during NAO<sup>+</sup> (red) and NAO<sup>-</sup> (blue) phases (being the arrows length proportional to the wind speed). The rest of panels provide the NAO<sup>+</sup> minus NAO<sup>-</sup> composites for mean **(c, d)** 10 m wind module (in m/s), **(e, f)** precipitation (in mm/month), **(g, h)** integrated cloud water (in mm) and **(e, f)** 2 m temperature (in K). Differences are represented only if they are statistically significant at the 90 % level, dots blur the values not representing a signal-to-noise ratio above 1, and contours depict differences expressed as percentage.

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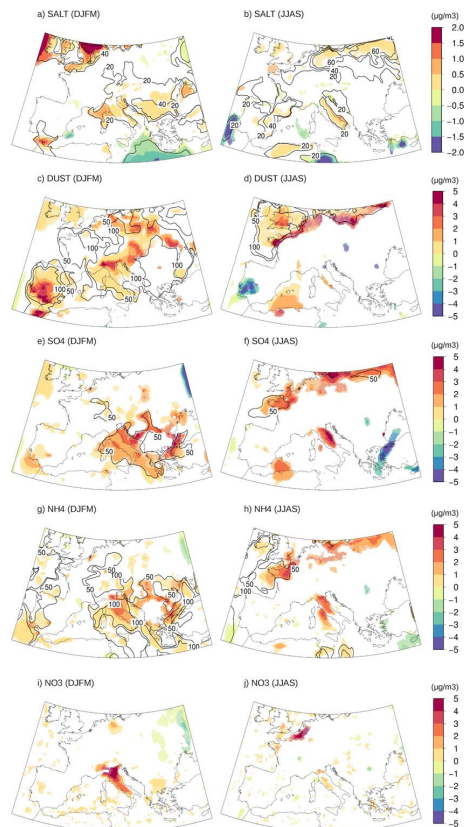
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**Fig. 4.** NAO-impact on mean ground-level aerosols concentrations: NAO<sup>+</sup> minus NAO<sup>-</sup> composites for (a, b) SALT, (c, d) DUST, (e, f) SO<sub>4</sub><sup>2-</sup>, (g, h) NH<sub>4</sub><sup>+</sup> and (i, j) NO<sub>3</sub><sup>-</sup> in winter (left) and summer (right). Differences in  $\mu\text{g m}^{-3}$  are represented only if they are statistically significant at the 90% level, dots blur the values not representing a signal-to-noise ratio above 1, and contours depict differences expressed as percentage.

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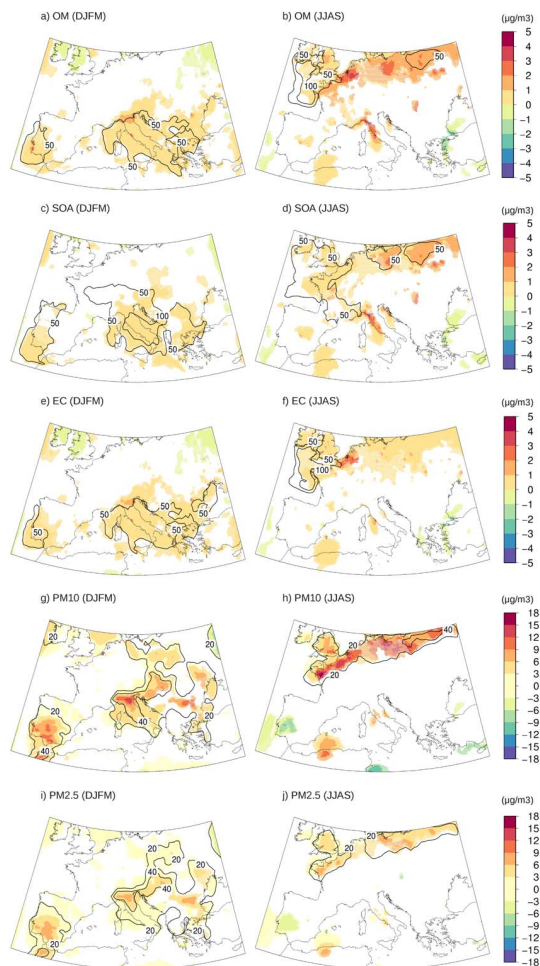


Fig. 5. As Fig. 4 for (a, b) OM, (c, d) SOA, (e, f) EC, (g, h) PM<sub>10</sub> and (i, j) PM<sub>2.5</sub>.

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