Interactive comment on “Impact of the North Atlantic Oscillation on the variations of aerosol ground levels through local processes over Europe” by S. Jerez et al.

Anonymous Referee #1
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Author's response

The paper aims to investigate the impact of the North Atlantic Oscillation (NAO) on the variation of ground level concentration of locally emitted aerosols during the winter and summer seasons over Europe. The climatic impact of the NAO is reviewed in the context of the modeling system to provide for model validation. The impact of the NAO on aerosol concentrations is investigated as the difference of high/low NAO index composites, with the aim to link the large-scale teleconnection pattern to natural local pollution variability. Overall the paper is relevant, focused and concise. Some changes are needed for the standard required by the journal.

We appreciate the positive view on the relevance of the paper and acknowledge the feedback provided by the reviewer. We have carefully considered all the comments, concerns and suggestions. We hope that the revised manuscript looks suitable for publication.

General Comments:

The original time-series of winter monthly values of the NAO index presents a positive trend over the last 30 years. Therefore its distribution is dominated by positive values, leading to late winter averages above zero. It’s my understanding that this is taken into account for the present study but needs to be elaborated upon. How were the 30/70 percentiles defined? Was the distribution normalised to zero mean, unit standard deviation over the period studied? This information should be included in the text.

This appreciation is right, i.e. the winter NAO index time series presents a slight positive trend over the last 30 years (and also the summer NAO index time series presents a slight negative trend). The related problem is taken into account in our approach by defining positive (negative) NAO events through the use of the 70th (30th) percentiles of the whole NAO index time series. In this way, we avoid the use of subjective thresholds (let's say 0.5 and -0.5 for the NAO^+ and NAO^- events respectively), and thereby we prevent unbalances between the number of NAO^+ and NAO^- events retained. Thus our positive NAO events represent situations with higher than 'average' NAO phases in the recent past (and similarly for the negative phases), although, in order to obtain clearer signals by avoiding 'neutral' situations, we do not use the average but the 70th (30th) percentile as the non-subjective threshold in our definition of NAO^+ (NAO^-) phases. The 30th (70th) percentile defines the value below (above) which the 30% of the values remain. This is applied separately to the winter and summer NAO index time series. We think that this is clearly depicted in Figure 2, but now we have made it clearer in the text as well.

As the NAO index from NCEP/NCAR Reanalysis Project (CDAS) is used in the study, but ECMWF (re)analysis data are used to drive the model, how well do the two datasets agree on the NAO index? What is the uncertainty of this onz the results? Why not calculate/use the corresponding NAO index from the ECMWF data instead?

We acknowledge the convenience of this warning, which was also raised by the other reviewer. Indeed, the NAO index is not only affected by the use of different data sources, but also of different methods for its computing (i.e. station or PC based, location of the stations and fields from which the PCs are computed). However, at the seasonal time-scale analyzed here, these differences are fairly negligible. In particular, the dependence of the NAO on the reanalysis used for its computing is minimal, as these datasets are very similar

at the monthly/seasonal timescales. This is clearly visible in the work by Greatbatch and Rong (2006). They show the summer NAO index computed from both the NCEP and the ERA40 reanalysis through the same methodology (PCA applied to the SLP field) (Figure R1). It can be observed that both series greatly agree in the overlapping period.

Since we did not find a similar plot for the winter NAO time series in the literature, we provide in Figure R2 the time series of different winter (DJFM-averaged) NAO indexes. Despite the apparent differences among them (due to the different methodologies and data sources employed for computing the NAO index in each case; we believe that the former cause plays the most important role), the correlations between all the series pairs is above 0.85 (Table R1). Besides, the years that would have been selected in each case as winter NAO^+ or NAO^- years following our methodology mostly coincide (Table R2), which is even more important from a practical point of view, i.e. in order to support the validity of our approach/results.

Therefore, our results, at the time-scale analyzed here, are not really affected by the specific NAO index used. Hence, we have maintained the use of the NAO index provided by the CPC of the NOAA as it is the most commonly used nowadays.

Nonetheless, as the former conclusion may not work at other time-scales, we have included a comment in the manuscript reflecting the caveat raised by both reviewers in this regard.

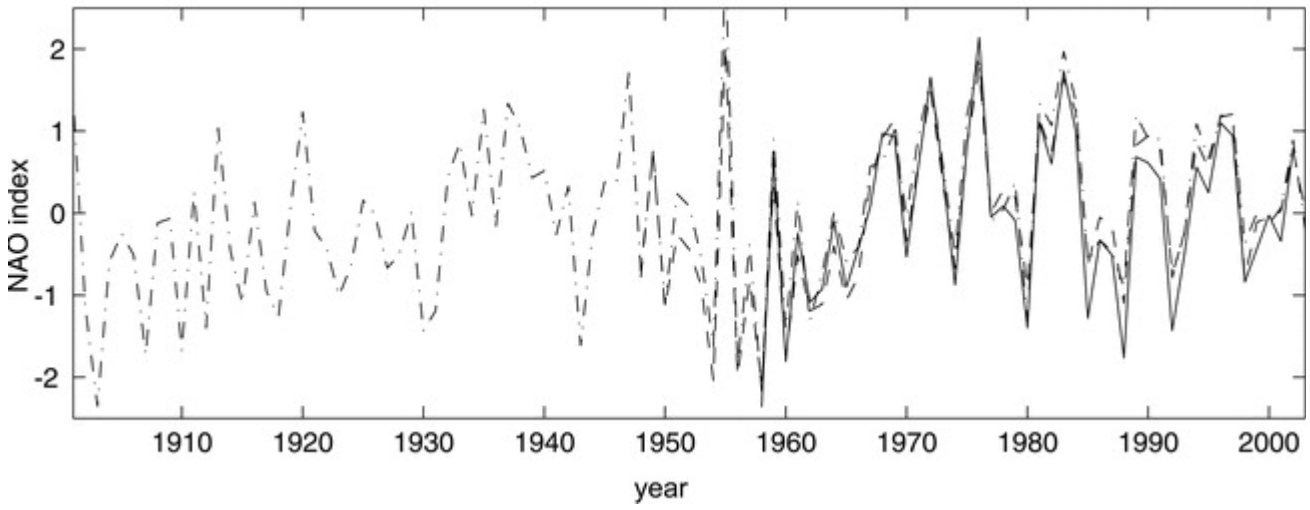

Figure R1. Adapted from Greatbatch and Ping-ping Rong (2006). Principal component (PC) time series for the leading EOF computed from SLP averaged over July and August in each year (i.e. summer NAO index) using the NCEP–NCAR (dashed line) and the ERA-40 (solid line) reanalysis.

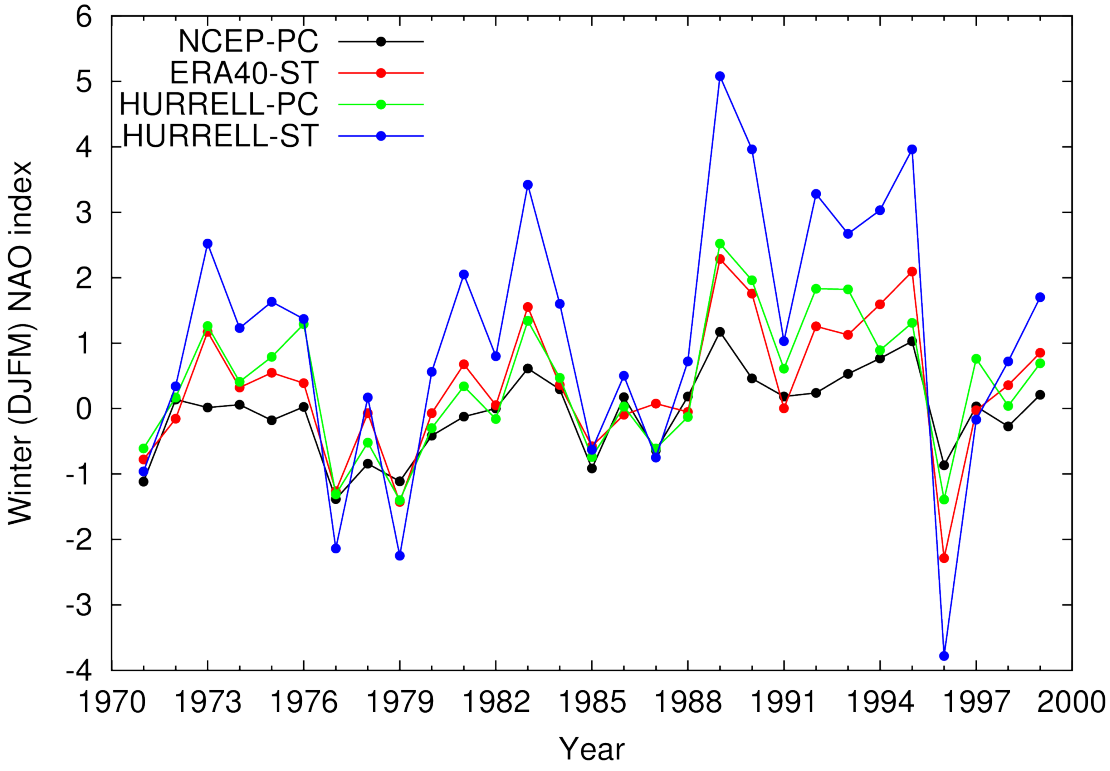

Figure R2. Winter (DJFM-averaged) NAO index time series obtained using different methods and datasets. In black: NAO index from the CPC of the NOAA (this is the NAO index series used in the manuscript). In green: PC-based Hurrell NAO index(*). In blue: station-based Hurrell NAO index(**). In red: as the station-based Hurrell NAO index but computed using ERA40 data. Note that the different methodologies seem also to imply differences in the pre and post-processing of the data. In particular, the black series looks to be normalized and varying between -1 and 1 while the others are not.

(*) http://climatedataguide.ucar.edu/sites/default/files/cas_data_files/asphilli/nao_pc_djfm_1.txt
(**) http://climatedataguide.ucar.edu/sites/default/files/cas_data_files/asphilli/nao_station_djfm_1.txt

	NCEP-PC	HURRELL-PC	HURRELL-ST	ERA40-ST
NCEP-PC				
HURRELL-PC	0.86			
HURRELL-ST	0.88	0.93		
ERA40-ST	0.86	0.90	0.98	

Table R1. Temporal correlation between the different DJFM-averaged NAO index time series depicted in Figure R2.

Year	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99
NCEP-PC		■						■	■	■	■			■	■	■		■		■	■		■	■	■	■	■		■	■
HURRELL-PC		■		■			■	■	■	■	■		■	■		■		■		■	■		■	■	■	■	■			
HURRELL-ST		■	■	■				■	■	■		■		■		■		■		■	■		■	■	■	■	■	■		
ERA40-ST		■	■	■				■	■	■	■			■		■	■			■	■		■	■	■	■	■			■

NAO ≥ 70th pctl ■ NAO ≤ 30th pctl ■

Table R2. Years with winter (DJFM-averaged) NAO index above the 70th percentile (in pink) and below the 30th percentile (in blue) in the series depicted in Figure R2.

Though it is mentioned, it should be made more clear in the text and abstract that varying emissions are used for the validation of the model, but emissions fixed to the year 2005 are used to draw conclusions on the NAO impact on ground aerosol concnetrations.

We agree with the reviewer's suggestion and have thus emphasized it more in both the abstract and the body of the manuscript (data and conclusion sections).

Individual comments:

We do agree on the convenience of the changes proposed below. All have been considered in the new version of the manuscript. We provide some additional specific clarifications for some of them below.

Throughout the paper "NAO-impact" → "NAO impact" (Remove hyphen).

Comments below are prefixed with the page and line number:

Abstract:

Emphasis needs to be added in the abstract that the study is on the seasonal time scale with climatological monthly-averaged emissions based on the year 2005.

l.1: This contribution assesses → We assess

l.1: non-anthropogenic → non anthropogenic-induced

l.10: "Variations are up to and over 100% for most aerosols": Needs rephrasing, to be made more specific

Introduction:

p13891 l.7: remove paragraph break

p13892 l.7: is extesive to → extends to

p13892 l.17: aerosols → aerosol

p13892 l.29: Remove "Besides,"

Sec 2:

p13893 l.10: driven by ECMWF data: Are (re)analysis data used as boundary conditions or is the model nudged towards the data throughout the domain? It should be made clear in the text that the model produces its own meteorology.

p13893 l.12,13: Why is gas-phase chemistry included/mentioned? The paper deals with aerosols only. This is confusing for the reader.

Although the paper focuses only on aerosols, this section aims at providing a general description of the modeling system.

p13894 l.17-22: Sentence beginning "Although the influence...", ending "... related to the NAO phase." is too long-winded and very hard to understand. Needs to be rephrased to clarify the meaning.

p13894 l.19: "hampers" is erroneously used. Should be replaced with eg. "is unable"

p13895 l.8: Remove "and that task constitutes the focus of an on-going paper"

p13895 l.10: "it is opportune to show shortly" → "it remains to be demonstrated"

p13895 l.15: For the sake of brevity → For brevity

p13895 l.16: under several → under two

p13895 l.18: As well → Also

p13895 l.21: Remove "the worst"

p13895 l.26: arises → is

p13896 l.6: Remove "is relevant as it"

Section 3:

p13896 l.17: "with very similar structures within each season considered but very different in each one of them" needs rephrasing for clarity

p13896 l.19: being disregarded → are not included

Section 4:

p13898 l.12: have associated → are associated with

p13900 l.14: "not counteracting by enhanced temperatures" is unclear

It was actually erroneous and has been now removed.

Conclusions:

p13902 l.15: "Should not be considered deterministic". In what context is deterministic used here? Please consider rephrasing.

Figures:

Fig 2 caption: up → top having been highlighted → highlighting

Fig 3: a,b) Panels are too busy; please consider reducing vector density. Vector magnitude scale missing from the label.

Impact of the North Atlantic Oscillation on ~~the variations of~~ European aerosol ground levels through local processes ~~over Europe~~: a seasonal model-based assessment using fixed anthropogenic emissions

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Keywords: North Atlantic Oscillation, air quality simulation, aerosols, 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 **North Atlantic Oscillation (**NAO**)** controls a large amount of the European climate variability with asymmetric impacts in both time and space. **Here we investigate how the local atmospheric processes (without considering large inter-continental transport mechanisms), as they are governed by the NAO, affect the levels of various aerosol species using simulated data under constant emissions, which are fixed to the 2005 levels in order to avoid anthropogenic-induced signals. In particular, we analyze interannual variations at the seasonal time-scale and focus on the ground-level. The results show** ~~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 aerosol~~s~~ **concentrations** ~~levels~~ in southern (northern) regions in winter (summer), while negative NAO phases enhance them in northern (southern) regions in winter (summer). **The underlying processes are** ~~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~~ **affects** the biogenic emitting activity and on the oxidative capacity of the atmosphere. **Differences for all the species studied (natural inert, secondary inorganic and organic aerosols) are up to 5 µg m^{-3}, reaching 10 and 20 µg m^{-3} for PM10 and PM2.5 respectively, which represents variations about 20-40% in their mean levels between opposite NAO phases.** ~~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 (PM10), nitrogen dioxide and ozone. In this sense, worldwide epidemiological 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). 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 **crucial** ~~**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**; Forkel et al., 2012; Jiménez-Guerrero et al., 2012; Meier et al., 2012; Colette et al., 2013; Hedegaard et al., 2013, just to mention some recent works on the topic**), determining also the vegetation activity and thereby modifying natural emissions.

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^+), 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^+ reduces cloudiness and precipitation in southern Europe. Conversely, the intense zonal circulation promoted by the negative NAO phases (NAO^-) 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~~ **extends** to Eurasia (Wanner et al., 2001; Trigo et al., 2002) but also North America, Greenland, the Ar**c**tic 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 aerosol~~s~~ concentrations. This influence should arise through both local processes and large-scale air pollutants transport**. For instance, Eckhardt et al. (2003) showed that NAO^+ enhances northward transport of pollutants from Europe to Arctic regions, mainly in winter and spring, compared to NAO^- phases. At the same time, NAO^+ promotes African dust intrusions into south-western European regions (which occurs mostly in the summertime) as the westerly winds associated to NAO^- events prevent subtropical air masses to reach the European mid latitudes; although, on the other hand, this NAO^- related large-scale zonal winds also favor the transport of pollutant from North America into Europe**

(Moulin et al., 1997; Dayan et al., 2011; Christoudias et al., 2012; Cusack et al., 2012; Pey et al., 2013). But the signature of the NAO in the climatic conditions (and thereby on air pollution levels) does not restrict to changes in the large-scale circulation patterns, as these have also repercussions on other atmospheric variables such as precipitation and temperature as commented above, with a potential subsequent impact on air quality through local processes such as aerosol wet deposition. However, few studies have been devoted to explore air pollution levels from a climatic perspective, even less disentangling between local and large-scale mechanisms, therefore the contribution of the local NAO-controlled processes on the climatology of air pollution levels is still hardly established. ~~**, either from Europe to Artic 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 **deepen on** ~~**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 **with the focus on elucidating the influence of the small scale processes** ~~**, 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, t**~~**T**he simulation was designed to disregard the contribution from **the long-range** ~~**large scale**~~ transport **by using constant climatological boundary conditions for the aerosol concentrations. This design does not provide a realistic picture, but allows to improve our understanding on the role of the local underlying mechanisms, as it remains unmasked by the large-scale inter-continental advective phenomena.** 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 **(in both cases without nudging)**, 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 μm 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 degrees 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 **is higher than usual in climate runs** ~~**enhances from previous 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 **climatic** 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). **The model setup used here is the same as in the former reference; in the second, it is evaluated in the context of a multi-physics ensemble of present-day climate simulations. Although both works focus on the Iberian Peninsula, there are extensive literature demonstrating the ability of MM5 for reproducing diverse meteorological features along Europe (e.g. Kotlarski et al., 2005; Renfrewet al., 2009; Pfeiffer and Zängl, 2010). Moreover, Section 4.1 provides a revision of the accuracy of the patterns of the NAO impact on climate as they are obtained from the climate simulation driven CHIMERE.**

Boundary conditions **of gas-phase pollutants concentration** for **CHIMERE** ~~**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). **These boundary conditions consist of constant m**~~**M**~~onthly mean data **that** 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**~~ **The use of constant** climatological boundary conditions **prevents** ~~**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 **inter-continental** transport mechanism**s** related to the NAO phase. **On the contrary, and despite the pollutants transport between different areas within our extensive domain is modeled by default, this experimental design allows to largely isolate the role of more local processes (for instance, how the NAO influences the concentration of aerosols through its impact on local precipitation patterns).** ~~**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.**~~ **At this regard, it should be noted that the design of the driving MM5 climate simulation does not follow this approach. The climate boundary conditions at the**

MM5 domain boundaries are updated every 6 hours according to the ECMWF data specified above. Hence, although the MM5 domain does not cover, in particular, most of the Atlantic ocean, the influence of the NAO on the climate conditions simulated within its borders should be actually captured.

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 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.

Beyond the several works supporting the ability of similar MM5-CHIMERE systems to reproduce the main air quality features over Europe (e.g. Monteiro et al., 2007; Flaounas et al., 2009; Péré et al., 2010), our ~~**This**~~ modeling system has been validated by comparing a simulation covering the period 1990-2010 with EMEP observations (Tørseth et al., 2012). ~~**A**~~ **Although 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**~~ **here we** show shortly that the simulated series of aerosol**s** concentration**s** correlate**s** acceptably and captures a large fraction of the **interannual** variability of the observational series **at the working time-scale (i.e. seasonal)**. 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**~~ **two** size-thresholds, namely PM10 and PM2.5 (Figure 1). For both cases, correlations between simulated and observational **seasonal-averaged** series are overwhelmingly above 0.8, in most cases above 0.9, both in winter and summer. ~~**As well**~~ **Also**, 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~~ **These results** guarantee~~s~~ 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 ~~accuracy~~ **suitability** 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~~ **becomes** 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-year 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, SO_4^{2-}, nitrate, NO_3^-, and ammonium, NH_4^+), organic matter (OM), with particular attention to secondary organic aerosols (SOA), and elemental carbon (EC). The total concentrations of PM10 and PM2.5 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~~ **in** each season ~~considered but very different in each one of them~~. The remaining months from the two transitional seasons depicted intermediate and softer signals **and thus are not included in** ~~being disregarded from~~

the rest of the analysis.

Seasonal means (DJFM and JJAS averages) of the monthly NAO index provided by the Climate Prediction Center (CPC) from the National Ocean and Atmospheric Administration (NOAA) are used to define classes of positive and negative NAO phases (NAO^+ and NAO^- respectively) throughout the study period. Following a a common approach adopted in previous studies (e.g. Suski and Ridgway, 2007; Gouveia and Trigo, 2008), NAO^+ (NAO^-) phases are defined as those with the NAO index above (below) the 70^{th} (30^{th}) percentile of the NAO index time series considered (Figure 2). The use of these non-subjective thresholds in the definition of NAO^+ and NAO^- phases ensures the evenness between the number of events of each class (within the 30-year simulated period, 1970-1999, we retain 9 events of each class in each season) and prevents the unbalancing influence of potential long-term trends in the NAO index time series. On the other hand, it should be noticed that different data sources and/or methods for computing the NAO index may provided different values of it. In particular, we are aware that the CPC/NOAA NAO index is not based on the ECMWF data used to drive MM5 in our modeling system. However, we confirmed that, at the time-scale assessed here, this influence is negligible (Greatbatch and Rong, 2006). Hence, the choice of the CPC/NOAA NAO index for the present study is based on its easy access and its up-to-date computation approach, which in fact makes it widely used nowadays in studies focused on the recent past (e.g. Bladé et al., 2012; Jerez et al., 2013b; Pey et al., 2013).

~~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 ($NAO^+ \geq 70^{th}$ percentile) and negative ($NAO^- \leq 30^{th}$ 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-year simulated period (1970-1999), we obtain 9 winters and summers of each NAO class (Figure 2).~~ The NAO -impact on the 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^+ minus NAO^- 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 -impact~~s~~ 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 novel insights, but essentially to evaluate the ability of our climate simulation (that is driving the CHIMERE run) to simulate the **extensively reported** NAO -impact on the **European** atmospheric conditions. This fulfills two relevant requirements, namely to (1) guarantee that it is effectively able to reproduce the known NAO -impact~~s~~ 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

355 variables driving air pollution levels.
356
357 Figure 3 further confirms the expected NAO -impact on the wind field, showing asymmetric
358 responses in winter and summer. In winter, NAO^+ (NAO^-) promotes a windy westerly flow
359 in northern (southern) Europe (Figures 3a,c), with the largest significant differences in the
360 wind speed (above 10%) appearing northward in the western sector of the domain (Figure
361 3c). Conversely, in summer, NAO^- phases ~~have~~ **are** associated **with** a stronger westerly
362 flow in northern Europe than NAO^+ phases (Figure 3b,d), with differences in wind module
363 exceeding 5% and extending more eastward than in winter. Windier conditions associated
364 to the NAO^- phase prevail also in the south-west of the domain in summer, mainly in the
365 surroundings of the Iberian Peninsula (differences about 5%) (Figure 3d).
366
367 Consequences in precipitation, cloudiness and temperature derived from the former
368 conditions are ~~also~~ provided in Figure ~~3~~**4**, as these variables play a key role on the levels
369 of air pollutants in general, and aerosols in particular. Obtained patterns are in good
370 agreement with previous works for winter (e.g. Osborn, 2011) or summer (Bladé et al.,
371 2012) relating them to the westerly winds advection of humid air from the Atlantic, which
372 favors the formation of clouds and enhances precipitation. NAO^+ minus NAO^- differences
373 in precipitation (Figure ~~3e,f~~ **4a,b**) are larger in winter than in summer, when they are
374 overall negative and affect mainly southern Europe ranging west-to-east from 40% (50
375 mm/month) to 20% (20 mm/month). In summer, these differences reach 20% (up to 30
376 mm/month) over large sectors of Europe, being negative in northern Europe while positive,
377 and less important, in the southern affected areas. Consistently, the composites for the
378 column integrated cloud water (a variable representative of cloudiness) (Figure ~~3g,h~~ **4c,d**)
379 show negative values in southern Europe in winter (resembling the west-to-east gradient
380 of the precipitation signal, with differences ranging from 30 to 10%) and northern areas in
381 summer (differences around 10%), and positive values in north-western areas in winter
382 (i.e. NAO^+ enhancing cloudiness there, around 20%). Regarding the NAO -impact on
383 mean 2-meter temperature, the most important signal consists of positive NAO^+ minus
384 NAO^- differences (up to 3 K) largely spread over northern Europe in winter (Figure ~~3i~~ **4e**).
385 Smaller negative (positive) differences (up just to 1 K) appear also over some
386 southernmost (northern) areas in winter (summer) (Figure ~~3i,j~~ **4e,f**).
387
388 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^+ minus NAO^- composites of mean ground-level concentration of the various aerosol species are provided in Figures ~~4 and~~ 5 **to 8**. In general, the positive phase of NAO enhances aerosol concentrations in southern Europe in winter and in northern Europe in summer. These positive NAO^+ minus NAO^- differences match generally well the sectors characterized by negative signals in wind speed, precipitation and cloudiness (Figure**s** 3 **and 4**). 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 Italy and the Balkans (Figures ~~4 and~~ 5 **to 8**, 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 **neither for long-range transports**), the largest variations between NAO phases reach 5 μg m^{-3} and affect the inorganic compounds (DUST, SO_4^{2-}, NH_4^- and NO_3^-), while the change in the concentration of carbonaceous material is just about 1 μg m^{-3}. It is worth stressing the slight but still significant negative patches of NAO^+ minus 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 NO_3^- concentration levels is negligible over most of Europe. However, the removal effect of ~~the NAO^- phase associated with~~ enhanced precipitation in northern

Italy **during NAO^- phases** ~~(not counteracting by enhanced temperatures)~~ seems to play a key **cleaning** role at this regard in this 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 NAO^+ leading to 40% higher SALT concentrations in the surroundings of the British Islands, which matches with the NAO^+ enhanced winds observed in this area (Figure 3c), but diminishing them in the southern Mediterranean (Figure ~~4~~5a). 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 Figure 3a that NAO^+ promotes windier conditions over the mentioned areas (longer arrows) than NAO^-, although these differences in wind speed do not remain after applying the significativity controls as they are not reflected in Figure 3c.

The NAO -impact on winter air quality is appreciable in the various size-classes of particulate matter. Figure ~~5g,h~~ **8a,c** shows that NAO^+ enhances 20-40% the concentrations of both PM10 and PM2.5 in the southern European regions (where the PM climatologies 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 (Figures ~~4 and~~ 5 **to 8**, 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^+ related increments are, in absolute values, about (even over) 5 μg m^{-3} for each inorganic species but SALT (which shows smaller signals, Figure ~~4~~5b). The NAO^+-

related increases in the concentration of the organic and carbonaceous aerosols are also a bit smaller (when they are expressed in μg m^{-3}).

It is worth stressing the signal in mean SOA levels (Figure ~~5~~7d), which can be doubly related to the reducing effect of NAO^+ in precipitation (Figure ~~3f~~ 4b), as it prevents wet deposition, and to the increased temperatures during NAO^+ enhancing the biogenic emissions (such as isoprene and monoterpenes), which leads to higher levels of biogenic SOA.

The NO_3^- signal appears again mainly restricted to the stronger emitting-areas, being located in this season between northern France, Belgium and Germany (Figure ~~4j~~ 6d). There, the reduced precipitation during NAO^+ events doubles the concentrations of nitrate in comparison to the levels during NAO^- phases.

SALT depicts slight variations associated to the NAO phase in summer (around 0.5 μg m^{-3}, Figure ~~4~~5b). However, contrary to the winter analysis, significant impact areas are now located over land areas and not over the sea, namely in eastern Iberia and Italy and in northeastern Europe, where the results show positive NAO^+ minus NAO^- differences.

Last, PM2.5 and PM10 show differences up to 10 and 20 μg m^{-3} respectively (higher concentrations during NAO^+), mainly concentrated in the northernmost areas of the domain and representing variations up to 20-40% between NAO phases (Figure ~~5h,j~~ 8b,d) **as observed for winter**.

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-year 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**, i.e. while the whole real picture does include varying emissions and the effect of long-range pollutant transport, our study, aimed at getting a better understanding of the natural and local processes, is narrowly focused on them**.

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% **in the mean levels of each species; about 20-40% for PM10 and PM2.5**. Eventually, softer signals of opposite sign (i.e. NAO^- 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 **climatic** factors varying between NAO positive/negative phases**, namely:** ~~.~~ ~~The main climate-related mechanisms include~~ (1) increased**/reduced** temperature (particularly in northern Europe), (2) different distribution of **the** precipitation patterns across Europe, (3) ~~increased~~ **changes in the** photolysis of primary and secondary pollutants due to **changes in** ~~lower~~ cloudiness, and (4) the cleaning effect of enhanced winds ~~(Katragkou et al., 2010)~~. According to the results of this work **and based on the established relationship between meteorological fields and air-quality (e.g. Wu et al., 2008; Katragkou et al., 2010; Jiménez-Guerrero et al., 2012; Manders et al., 2012)**, **the NAO impact on climate supports the NAO impact on aerosol concentrations as follows. Changes in** precipitation drive~~s~~ the **NAO impact on** ~~modification in~~ the concentration of most aerosol components (in both northern and southern Europe), ~~since a~~ **with the** decrease in the precipitation modeled **during NAO^+ phases** lead**ing**~~s~~ 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, **the** weaker winds **associated to the NAO^+ events** favor the increase of particulate matter in polluted regions such as large cities or entire industrial regions (e.g. Po valey 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_2 gas-phase emissions to turn into the particulate phase, thus increasing sulphate concentrations **as observed during NAO^+ phases in southern (northern) regions in winter (summer)**. 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^+ 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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List of captions

Figure 1. Evaluation of the MM5-CHIMERE air quality modeling system described in Section 2.1. Gray shaded colors depict the simulated climatologies in µg m^{-3} of PM10 (up) and PM2.5 (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 µg m^{-3}), and their shape informs on the magnitude of the temporal correlation between the simulated and the observed series.

Figure 2. Winter (~~**up**~~ **top**) 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**~~ **highlighting** the years/values with a NAO index above (bellow) that percentiles, i.e. those selected as NAO^+ (NAO^-) events for the composites analysis.

Figure 3. NAO impact in winter (left) and summer (right) on atmospheric conditions. (a,b) depict mean 10m-wind direction during NAO^+ (red) and NAO^- (blue) phases (being the arrows length proportional to the wind speed). (c,d) provide the NAO^+ minus NAO^- composites for the mean 10m-wind module (in m/s). These 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.

Figure 4. As in Figure 3c,d, here for (a,b) precipitation (in mm/month), (c,d) integrated cloud water (in mm) and (e,f) 2m-temperature (in K).

Figure 5. NAO impact in winter (left) and summer (right) on mean ground levels of natural inert aerosols: NAO^+ minus NAO^- composites for (a,b) SALT and (c,d) DUST. Differences in µg 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. Period considered: 1970-

1999.

Figure 6. As in Figure 5, here for secondary inorganic aerosols: (a,b) SO_4^{2-}, (c,d) NH_4^+ and (e,f) NO_3^-.

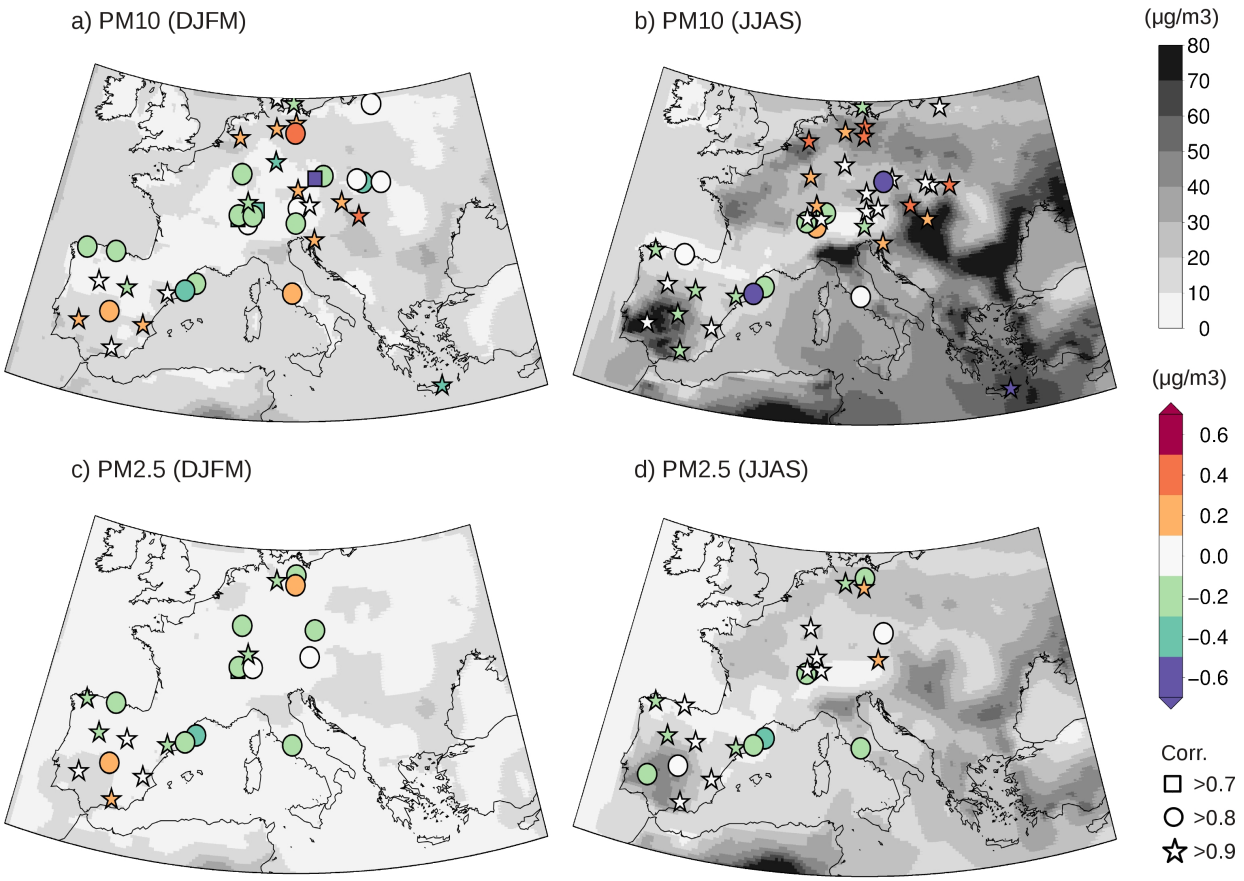
Figure 7. As in Figure 5, here for organic materials: (a,b) OM, (c,d) SOA and (e,f) EC.

Figure 8. As in Figure 5, here for particulate matter: (a,b) PM10 and (c,d) PM2.5.

~~**Figure 3. NAO-impact in winter (left) and summer (right) on the atmospheric conditions. (a,b) depict mean 10m-wind direction during NAO^+ (red) and NAO^- (blue) phases (being the arrows length proportional to the wind speed). The rest of panels provide the NAO^+ minus NAO^- composites for mean (c,d) 10m-wind module (in m/s), (e,f) precipitation (in mm/month), (g,h) integrated cloud water (in mm) and (e,f) 2m-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.**~~

~~**Figure 4. NAO-impact on mean ground-level aerosols concentrations: NAO^+ minus NAO^- composites for (a,b) SALT, (c,d) DUST, (e,f) SO_4^{2-}, (g,h) NH_4^+ and (i,j) NO_3^- in winter (left) and summer (right). Differences in µg 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.**~~

~~**Figure 5. As Figure 4 for (a,b) OM, (c,d) SOA, (e,f) EC, (g,h) PM10 and (i,j) PM2.5.**~~



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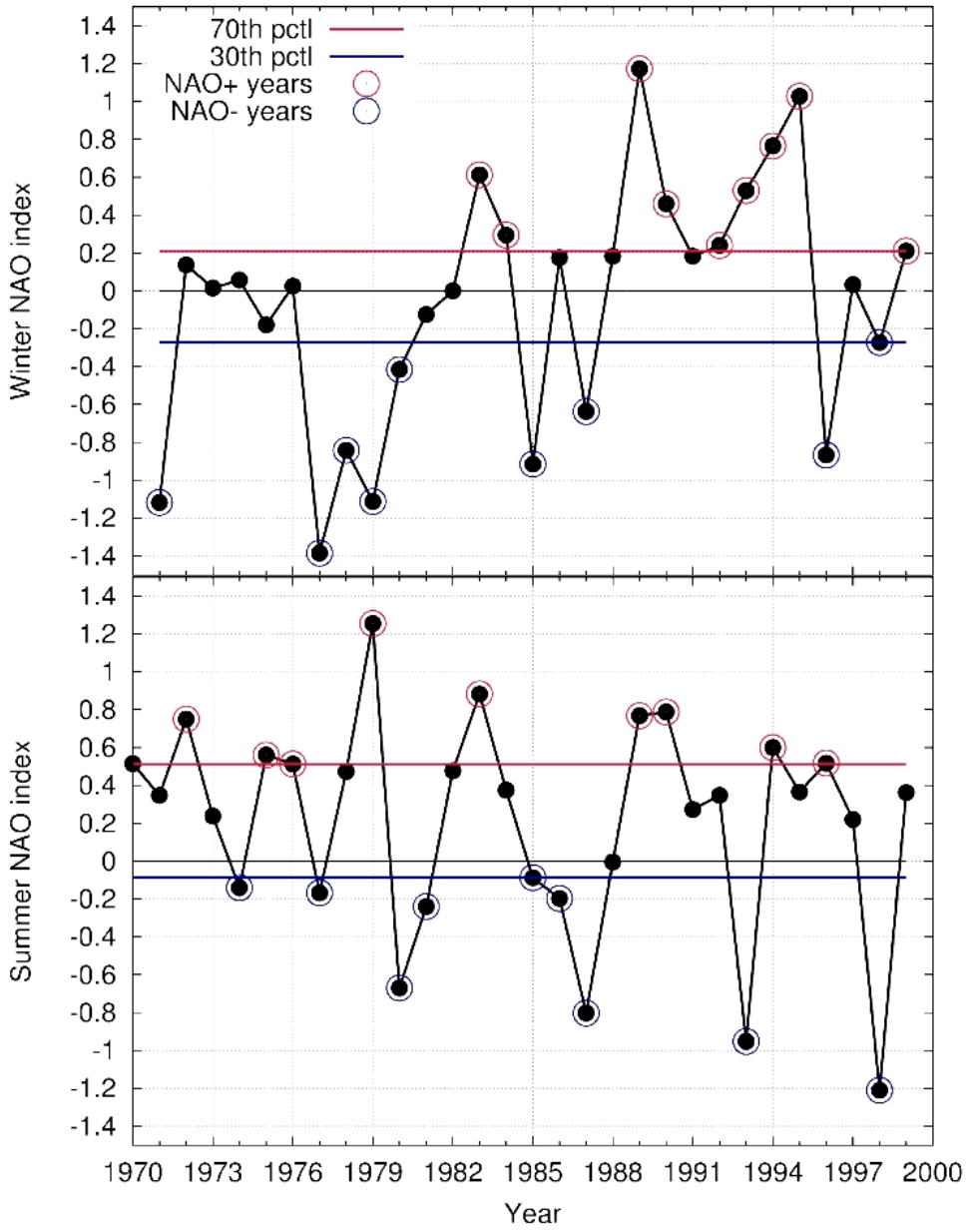

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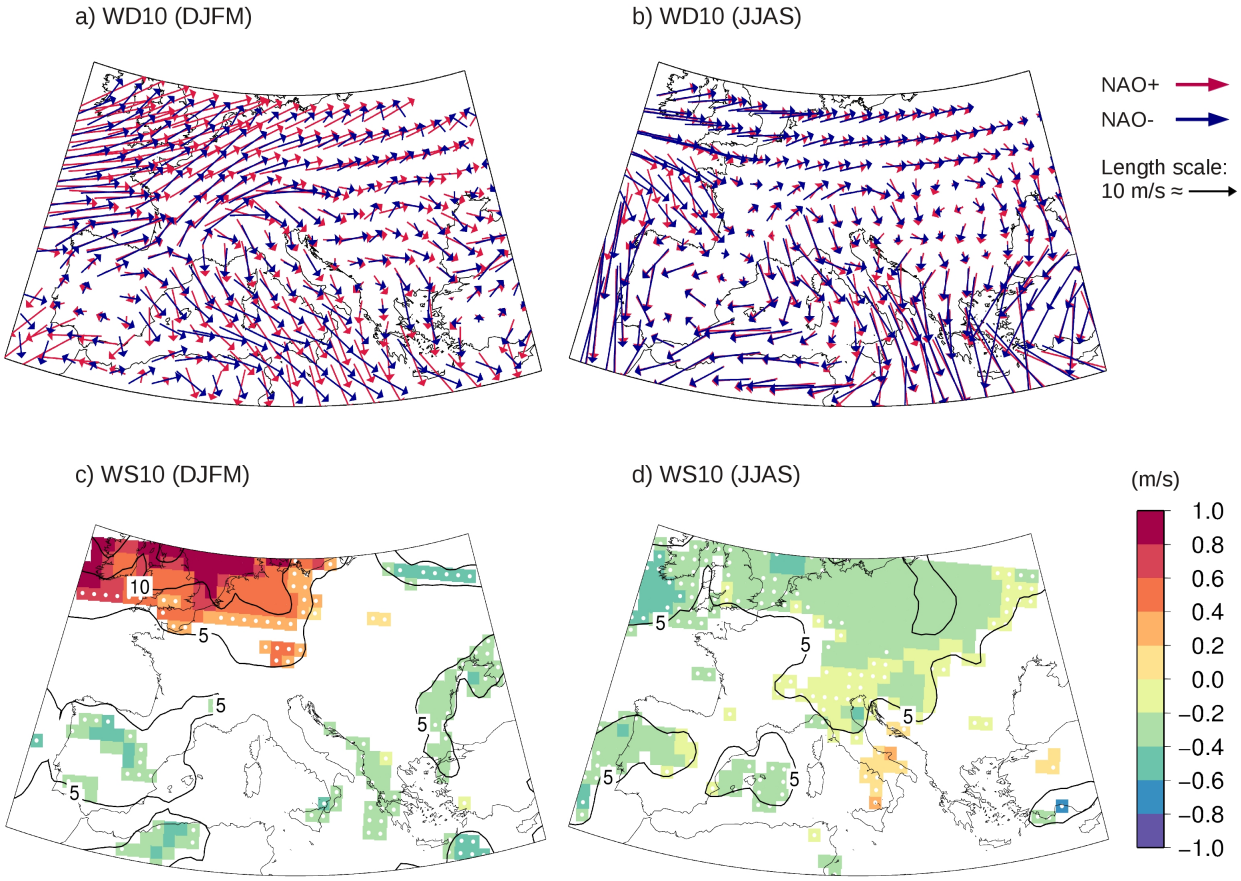

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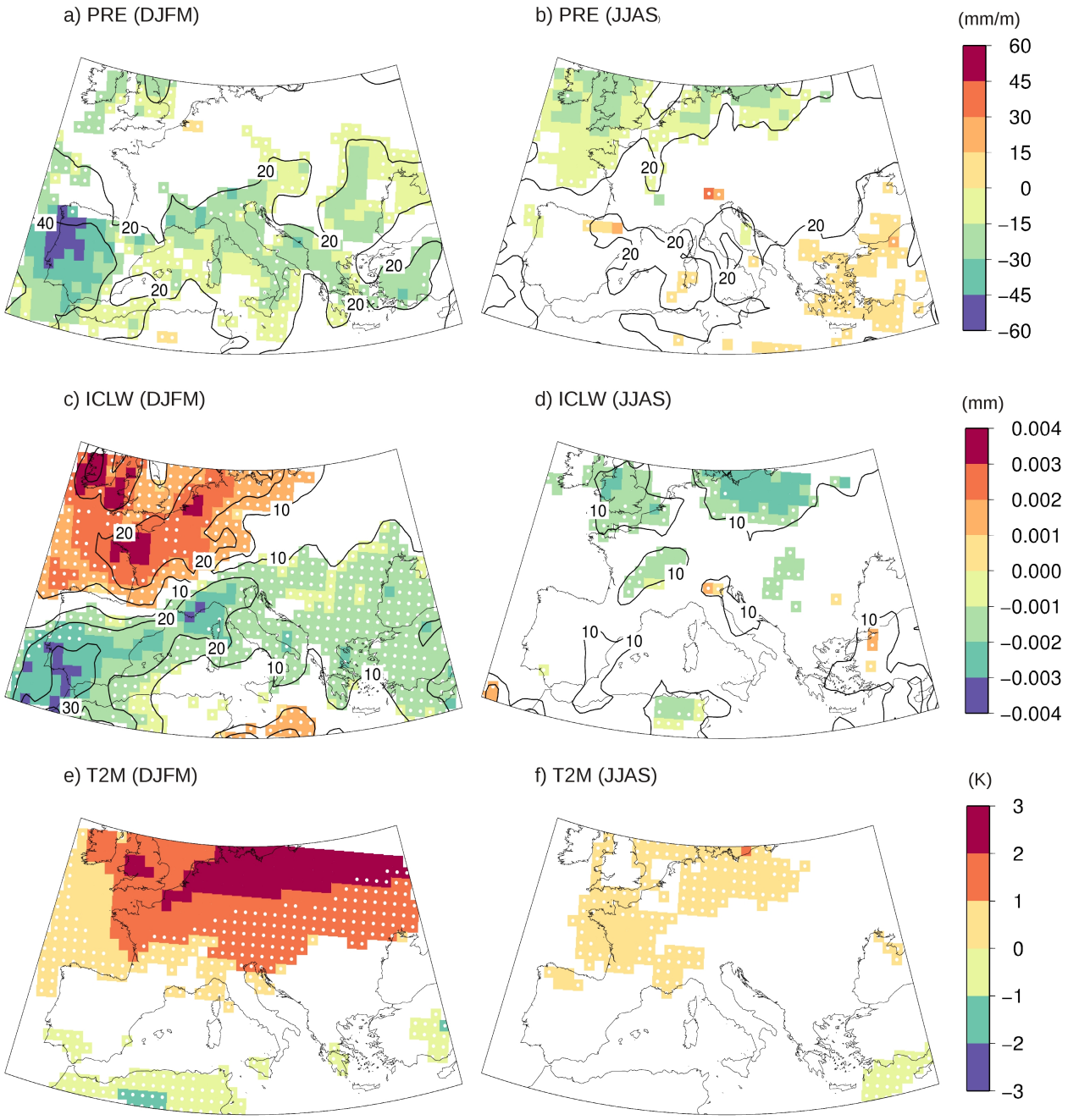

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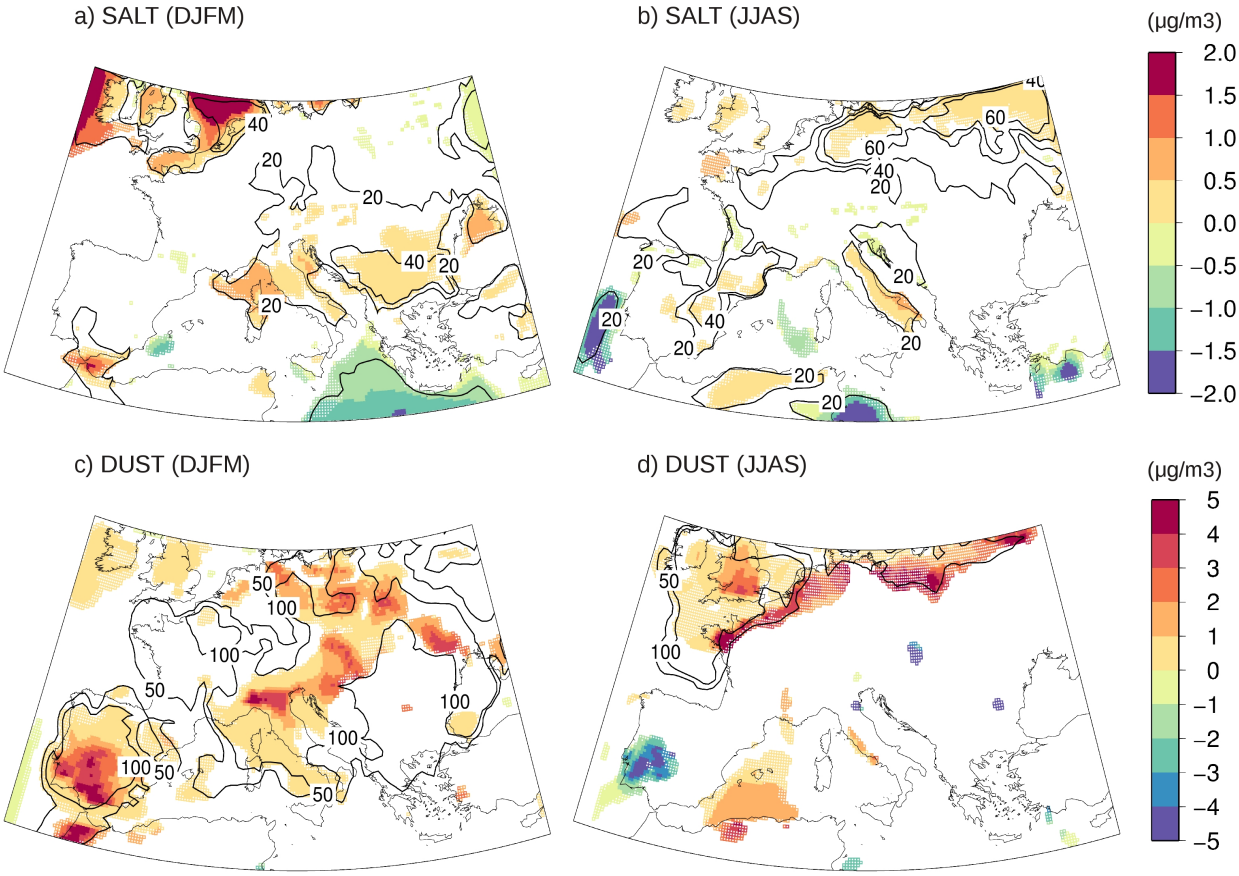

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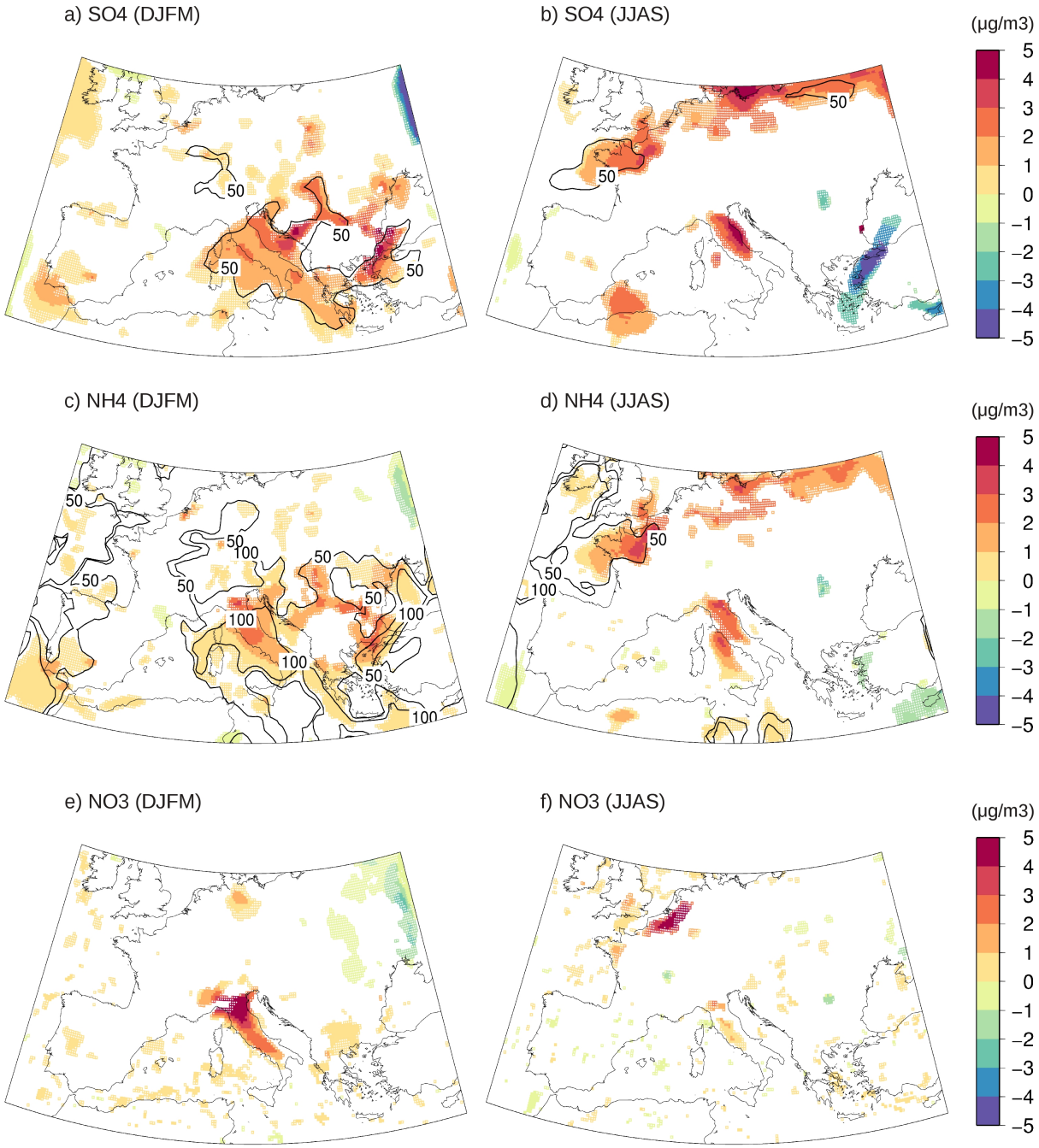

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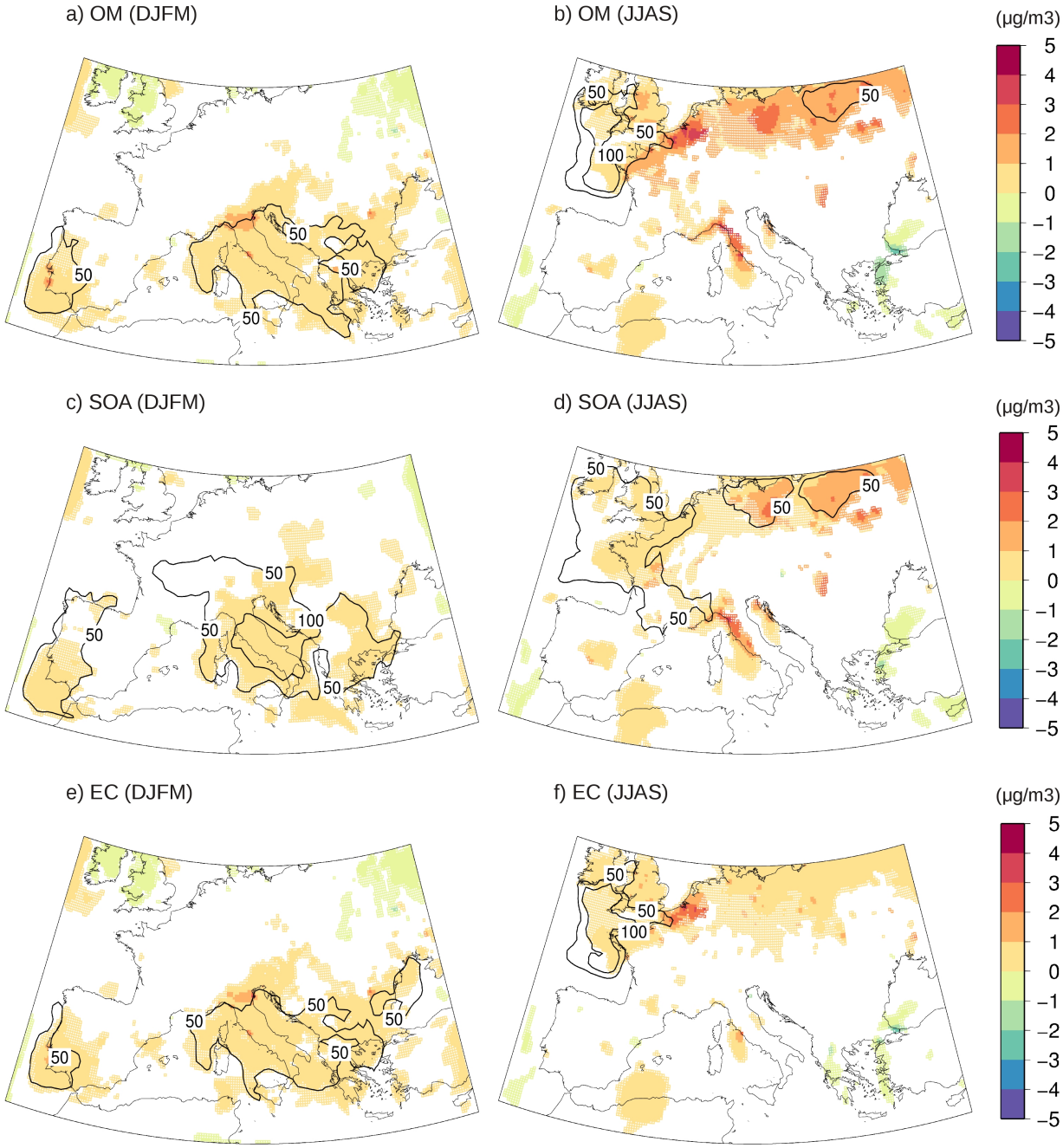

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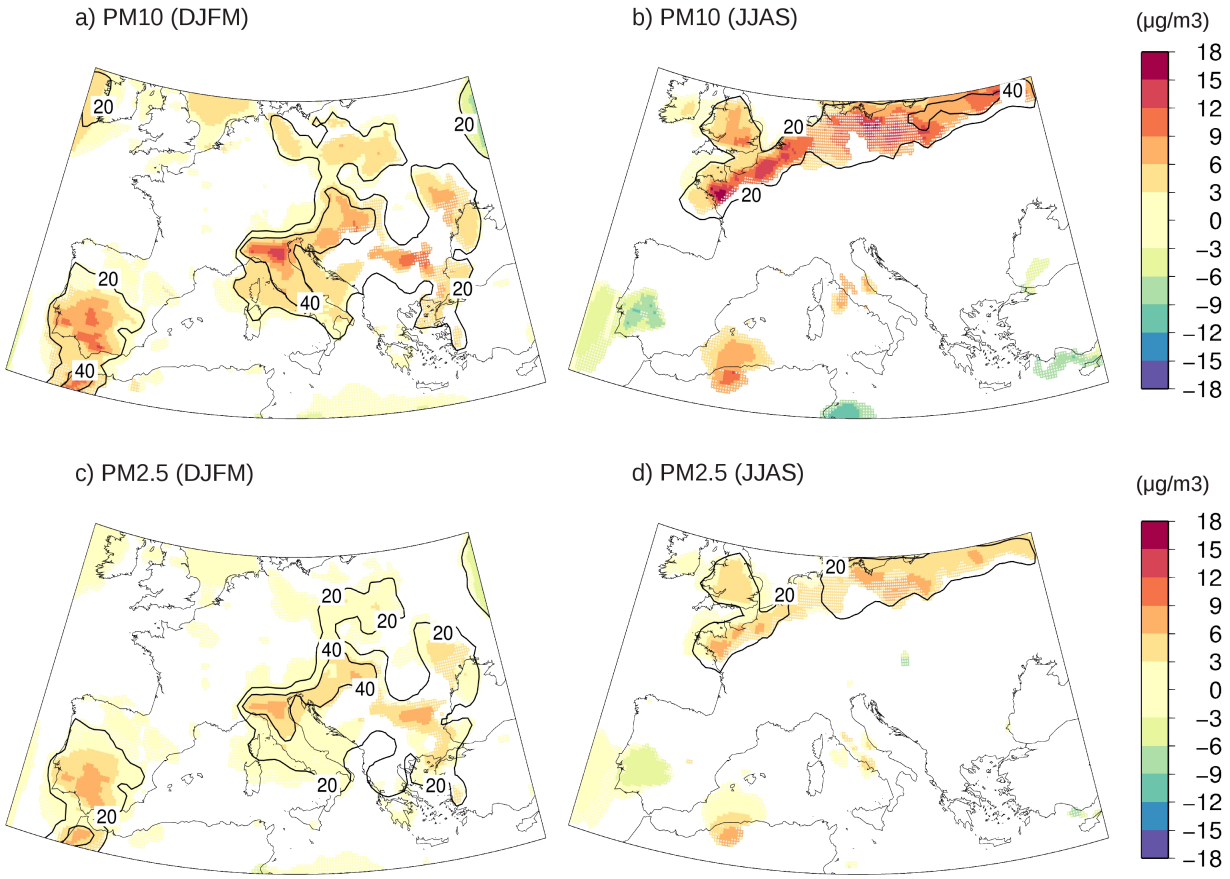

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