

## 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 #2

Received and published: 31 July 2013

Referee Comments on the manuscript “Impact of the North Atlantic Oscillation on the variations of aerosol ground levels through local processes over Europe” by S. Jerez et al. submitted to ACPD.

### Author's response

#### General comments:

This work presents an attempt to characterize the variations of aerosol ground levels over Europe caused by local processes and NAO patterns. In terms of scientific quality and significance, the subject of this paper is of scientific interest but the presentation of the work needs a number of revisions to make it suitable for publication with ACP. My main concern is focused on the following issues: the evaluation of the model results that the authors state it will be part of another paper, the use of NAO index from CPC (NOAA) without describing how it relates to the ECMWF data used for the MM5 that drive the air quality simulations and the lack of specific and detailed information in several parts of the text. For example, how can the authors discuss about the NAO patterns without including the Atlantic Ocean in their modeling domain?

**We thank the reviewer for the time devoted to revise the manuscript and acknowledge the concerns raised. In particular, we have made an effort to argument better the model validation, to discuss the robustness of our results despite the data source and method employed for computing the NAO index at the time-scale analyzed here, and to amend the lack of required details. Please, find below the answers to every single comment.**

The title of the manuscript reflects part of the contents of the paper, but since the results are based on non-realistic model simulations, I would suggest a slight change in the title: “Impact of the North Atlantic Oscillation on the variations of aerosol ground levels through multiannual model simulations over Europe using fixed anthropogenic emissions”

**We agree with the reviewer that the original title was not entirely faithful to the manuscript contents and, after considering the reviewer's suggestion, decided to change to the following title: 'Impact of the North Atlantic Oscillation on European aerosol ground levels through local processes: a seasonal model-based assessment using fixed anthropogenic emissions'.**

The specific comments that follow will help to clarify important issues and suggest a number of changes to be made in the text. My suggestion for publishing this work with Atmospheric Chemistry and Physics is to reconsider after the major revision comments have been addressed.

**All the comments below have been carefully considered in the revision of the manuscript. We sincerely appreciate the feedback provided.**

#### Specific comments:

##### Abstract:

I cannot see how the objectives described in the last sentence are met in the work presented here. How can we improve the predictability of climate-air quality interactions based on the findings of this work? This is not supported by the conclusions. I suggest that the abstract follows the conclusions more carefully and avoid using statements that cannot be substantiated.

**We have removed the last sentence of the abstract. Its content is now only discussed in the conclusion section.**

Introduction:

1. Page 13891, line15: The influence of radiation and temperature on gas-phase chemistry has been studied by numerous researchers worldwide. The authors should cite here more publications besides Katragkou et al. (2010).

**Now we cited also the works by Katragkou et al. (2010); Forkel et al. (2012); Jiménez-Guerrero et al. (2012); Meier et al. (2012); Colette et al. (2013) and Hedegaard et al. (2013). Please find the detailed references in the revised version of the manuscript.**

2. Page13892, line 29: How did the authors disregard the contribution from the large scale transport mechanisms? This is not adequately explained in the text. And why would they want to disregard a physical mechanism that strengthens the realistic representation of the atmospheric conditions?

**We understand the reviewer's incredulity on this option and have now made an effort to further clarify these two points in the introduction:**

**1. We disregard the impact of the large-scale transport mechanisms by using constant climatological boundary conditions for the aerosol concentrations.**

**2. The rationale for that is to allow focusing/isolating on the role of the local processes, unlike previous works that have been focused on the role of the large-scale transport processes. Each contribution can go in one different direction, which makes the problem difficult to be faced from an integrated approach without having trying before to disentangle the role of each mechanism. For example, according to other authors, negative NAO phases would promote the large-scale advection of pollutants from North America into Europe, preventing at the same time the African dust intrusions in south-western Europe, and also, according to our results, favoring cleaner atmospheres in southern Europe in winter by enhancing precipitation and wind there. Hence, in view of the complexity of the problem raising by the mix of scales and mechanisms, we find it interesting to isolate and assess each contribution separately. Naturally, we acknowledge that such option does not provide the most realistic framework (as we noted in the text) but helps improving our understanding of the underlying processes.**

Section 2:

1. Page 13893: How can the authors discuss the NAO patterns without including the Atlantic Ocean in the modelling domain (only a small part)? Is the MM5 domain different from the one we see in Figs3-5? This should have been explained in the text.

**Opposite to the CHIMERE air quality simulation, the MM5 climate simulation uses varying boundary conditions. They are obtain from ERA40 and updated every 6 hours. Hence, although the points/areas where the NAO index is defined are mostly out of the model domain, all the information about the circulation in the Atlantic ocean is given to the model through the domain boundaries every 6 hours. This way, the influence of the NAO on the climatic conditions in and, thereby, within the borders of the MM5 domain is actually captured (even though the MM5 domain does not include most of the Atlantic ocean). In fact, this can be observed in the patterns of Figures 3 and 4. We have now clarified this issue at the end of the 4th paragraph of section 2.1.**

2. Page 13894, lines 5-9: The authors have used meteorological fields with coarse spatial resolution of 90km and these meteo fields have been interpolated to 0.2deg for the air quality model simulations. Even though the present work covers the European continent and most part of the Mediterranean Sea, the authors refer to publications on the Iberian Peninsula for the discussion on the skills of the modelling systems. This is quite misleading, as the basic question that arises from this part is how a 90km horizontal resolution can give reasonable results on rainfall patterns for the entire European

continent. In addition, there is no mention in the text if the model setup was exactly the same as in the 2 cited publications. I believe that all the above present a very weak point of the presented work. The authors have not thoroughly evaluated any part of their simulations (meteorological or air quality fields) and they state that this is part of an on-going paper. Yet, the results and conclusions of this paper depend entirely on the performance of the modelling systems, both for the NAO pattern and the atmospheric pollutants concentration.

**We understand this complaint as it is certainly true that the two references provided to support the good performance of the climate modeling system focus only on the Iberian Peninsula. Partially because of that, we revise the patterns of the NAO-impact on the climate conditions obtained from the MM5 simulation in section 4.1. Since they are fairly in good agreement with the extensive literature on this regard (e.g. Osborn et al., 1999; Trigo and Palutikof, 2000; Trigo et al., 2002, 2004, 2008), where some of the authors have contributed significantly in the last 15 years by assessing NAO-climate links relative to the European continent, we are confident that our climate simulation captures well the signature of the NAO on the European climate, and as so it should be properly provided to the air-quality modeling system. Moreover, the ability of MM5 (MM5-CHIMERE) with identical or similar setups for reproducing the main meteorological (and air quality) features over different parts of Europe has been demonstrated in many works (e.g. Kotlarski et al., 2005; Monteiro et al., 2007; Flaounas et al., 2009; Renfrew et al., 2009; Péré et al., 2010; Pfeiffer and Zängl, 2010). These references have been now introduced in the revised manuscript. Besides, we do provide a partial validation of the air quality modeling system in Figure 1. This is not a general validation, but it does include the most relevant aspects for the purpose of this study, as argued in the text. A complete validation of the quality modeling system requires an additional long analysis, and corresponds to a separate independent paper that is still being elaborated.**

**Overall, the above mentioned arguments together with the coherence of the found signals, raises our confidence on the suitability of our modeling system for our experimental purpose.**

**As required, we now specify in the text that the MM5 model setup used here is the same as in Gomez et al., 2011. It is also used and validated (among others) in Jerez et al., 2013a. Please, see changes in the 3rd paragraph of section 2.1.**

3. Page 13894, lines 17-22: What is the exact meaning of this sentence? That the long range transport is disregarded because of the climatological boundary conditions? If this is the case, I have to express my disagreement with this statement. Long-range transport occurs when atmospheric pollutants travel thousands of km away from their sources, i.e from N-Europe to N-Africa, and the domain shown in the figures can include part of such transport mechanisms. Especially, the last sentence has to be considered erroneous "the experimental design allows to better isolate and understand the role of the local processes, including the pollutants transport between different areas within our domain". I would suggest to clarify what is the meaning of "local processes" when simulating atmospheric pollutants in such coarse domain, since pollutant transport and transformation is included by default.

**We use constant boundary conditions for all pollutants concentrations (and these can be different in each month). Hence, at the domain boundaries, the model sees every year the same concentrations regardless the NAO phase. Therefore, we are not taking into account the influence of changes in these concentrations due to changes in the NAO phase, i.e. the influence of the large-scale (planetary) transport mechanisms as they are governed by the NAO, as argued above (comment #2 of the introduction). We do model (by default, exactly) transports within the domain boundaries, which certainly involves a wide area where transports along large distances may occur, as pointed out by the reviewer. We have now carefully explained these features in the text, including a more precise definition (by giving an example) of what we call 'local processes'. Please see changes in the 4th paragraph of section 2.1.**

4. Page 13895, lines 1-5: How does the model produces wind-blown dust and resuspended dust? This part is missing from the text. Especially since the African deserts are outside the modeling domain, I am not sure of the origin of the wind-blown dust that is shown in the results. A proper discussion should be included in the text.

**For a detailed description of how the model produces wind-blown dust and resuspended dust, the reader is referred to Vautard et al. (2005); a paper devoted to the description of these processes. The developers of CHIMERE assume that the available concentration of dust (both wind-blown and resuspended) only depends on the wetness of the surface and the module of the 10-m wind. In this empirical view, the resuspension flux is governed by:**

$$F = P f(w) u^{*1.43}$$

**where  $f(w)$  is a function of the soil water content and  $P$  is a constant tuned in order to approximately close the PM10 mass.  $u^*$  stands for the module of the 10-m wind.**

**On the other hand, regarding the fact that the modeling domain does not include the African deserts, note that the influence from outer regions is provided through the domain boundaries to the modeling system in the form of boundary conditions. Recall also the arguments to the comment #1 of section 2.**

5. Page 13895, lines 6-7: The model-observation comparison has been performed using monthly, weekly, daily or hourly data? This information is not included in the text and it is important to understand how the correlation is above 0.7 most of the times. The authors should also consider giving the bias (not the standard deviation as in Fig.1). If the results are based on monthly PM10 and PM2.5 data, then there is no information on whether the model can capture the variability of the observations, since the lifetime of most aerosol species is a few days up to maximum one week. In that case, it should be made clear in the abstract and introduction that this is a seasonal analysis of the NAO impact. The 20 years simulation gives an overwhelming amount of data to handle, but I believe a more proper comparison should be included in the paper to convince the readers of the validity of the approach. Including the evaluation of the modelling approach in another publication when the model results support the findings in this work, is not appropriate.

**We fully agree with the reviewer's suggestion to better clarify the information on the time-scale at which the validation was performed, as it was missed in the text (although it was specified in the caption of Figure 1). As the assessment focuses on interannual variations (note that our simulation is 30-year long, not 20-year long!), differentiating between winter (DJFM averages) and summer (JJAS averages), the validation presented here was done at the same time-scale, i.e. using separately DJFM-averaged series and JJAS-averaged series of the PM10 and PM2.5 concentration series. Although this validation considered temporal correlation, standard deviation ratio and bias, we chose the two former statistics for Figure 1 as they are the most relevant for our assessment: we need to ensure above all the reliability of the temporal evolution and the amplitude of our simulated series. Biases are always important, but they are likely canceled in our NAO-impact analysis based on composites, as pointed out in the text. Nonetheless, please note that the order of the biases obtained in the validation is also briefly commented in the text.**

**Hence, following the suggestions of the reviewer and taking into account the above arguments, we have specified (1) in the abstract, that this is a seasonal analysis aimed at capturing the NAO-impact on the interannual variations of the aerosol concentrations; and (2) in the text, that the validation presented here of the modeling system was thus performed at that time-scale.**

**Regarding the apprehension raised concerning the appropriateness of the validation provided, please recall the arguments provided above.**

6. Section 2.2 is quite small in length and it does not need to be a separate sub-section. This can be part of section 2.1 (which will be named section 2). In this paragraph the authors say that they isolate the influence of climate variability on air quality by keeping the anthropogenic emissions fixed for 2005 during the 30-year run. Please explain the reason for choosing the year 2005 in the text. Since the climate variability is affected to a large extent by the feedback mechanisms between air quality and atmospheric conditions, it seems that this setup does not correspond to the real atmospheric conditions. It is rather a sensitivity model experiment that tests the model response (fixed anthropogenic influence) to NAO patterns. This should be clearly described in the text.

**First, regarding the advisability of merging sections 2.1 and 2.2, we should acknowledge our previous own doubts. However, we do prefer to differentiate between the modeling system itself (whose reliability can be validated as it considers year-to-year varying emissions; section 2.1), and the way in which we used it for the purpose of this work (whose reliability can not be validated as it fixes emissions; section 2.2), although it can be certainly summarized in just one paragraph.**

**We agree with the reviewer that there is not a special reason for choosing the year 2005 emissions over Europe. Emissions are kept constant through the simulations in order to clarify all the analysis related to NAO patterns, as stated by the reviewer, and then the election of the year for emissions does not condition the simulations. The final reason for selecting 2005 emissions is that previous simulations by the group having 2005 emissions as reference (e.g. Jiménez-Guerrero et al., 2001; Jiménez-Guerrero et al., 2012; Jiménez-Guerrero et al., 2013) showed a good performance. Those results indicate a reliable behavior of the model for reproducing air quality climatologies over Europe, and therefore this year was also selected in this work.**

### Section 3:

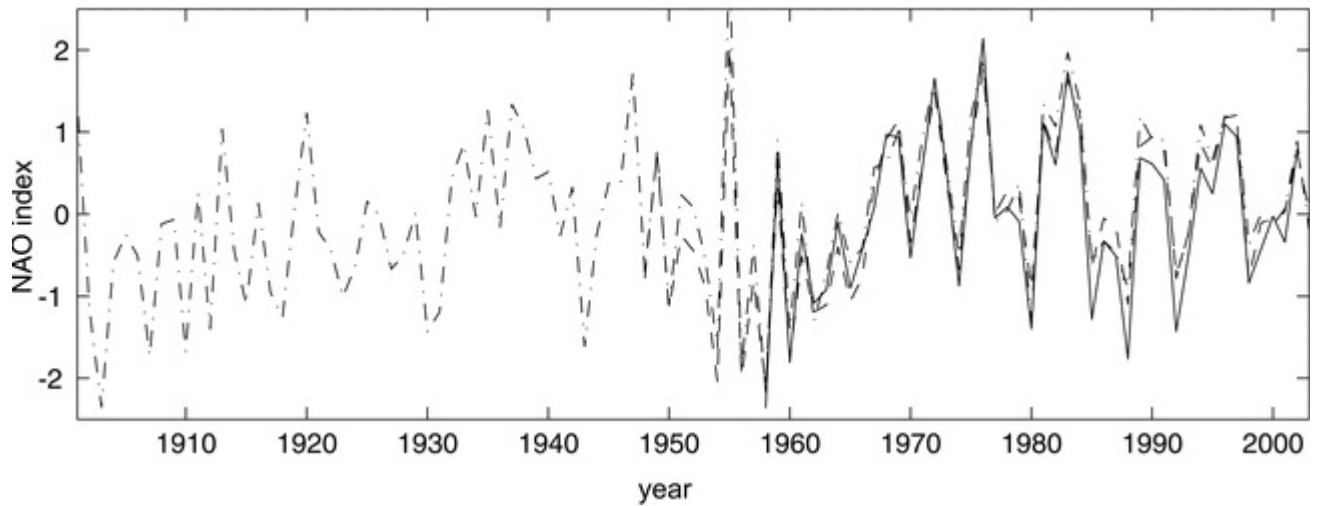
Page13896, lines 23-25: The authors are using the NAO index provided by CPC (NOAA) and the use ERA40 or ECMWF analysis fields for the MM5 simulations. How well the 2 systems relate when it comes to the calculation of the NAO index? If the two datasets give very different indices then the results from this work cannot be justified as they compare NAO patterns that do not relate to the air quality simulations. This is a very important part of this work and should be handled with caution in section 3.

**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.**

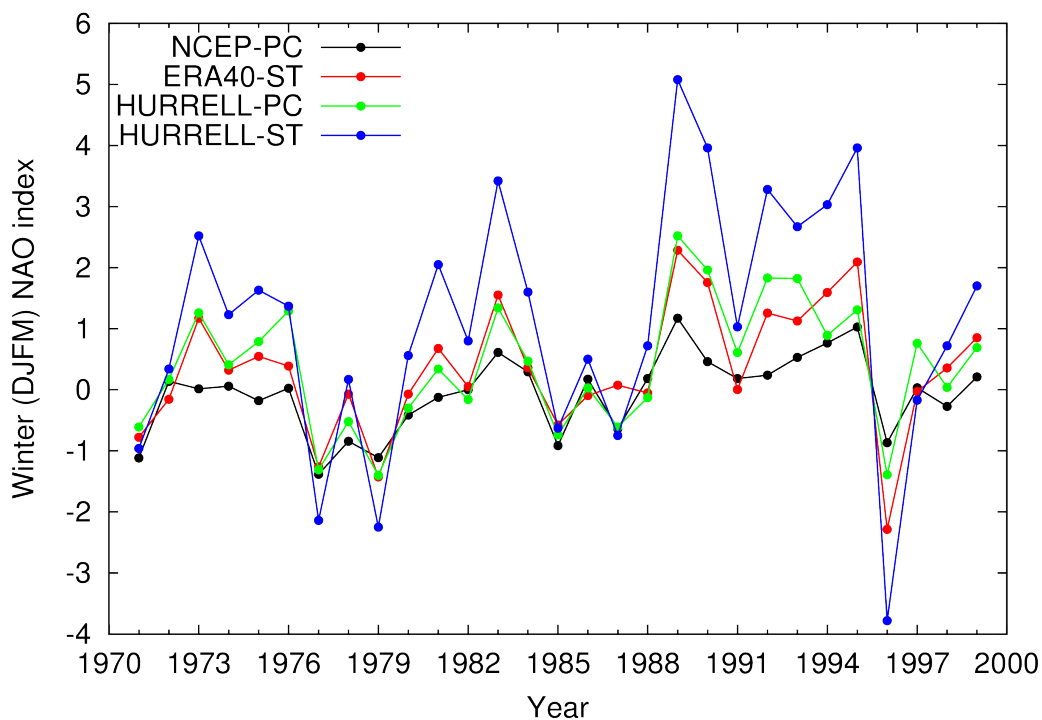
**Since we did not find a similar plot for the winter NAO time series in the literature, we provide in Figure R2 the time series of different winter (DJFM-averaged) NAO indexes. Despite the apparent differences among them (due to the different methodologies and data sources employed for computing the 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<sup>+</sup> or NAO<sup>-</sup> 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_pc_djfm_1.txt)

(\*\*) [http://climatedataguide.ucar.edu/sites/default/files/cas\\_data\\_files/asphilli/nao\\_station\\_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).**

#### Section 4:

1. Page 13898, line 1: The phrase “essentially to evaluate the ability of our climate simulation” is not supported anywhere in this section. How is this evaluation performed? There is no comparison with observed or measured values and the above statement is not appropriate. There is no evidence that the differences shown in Fig.3 are representative of the actual atmospheric conditions. This is also where one of the main questions arise again: the NAO phases are calculated with the NOAA index but the atmospheric simulation is driven by ECMWF data. Are these comparable?

**The NAO-impact on the European climatic conditions is well known and established elsewhere, as it is also proved the capacity of climate models to reproduce it. Hence, in section 4.1 we just want to certify that our simulation does reproduce the well-known NAO-impact patterns on the European climate, so that we can trust on it as provider of these climatic differences between NAO phases to CHIMERE. And, in fact, the patterns of Figures 3 and 4 agree with those available in the literature, as pointed out in the text. Hence, this proves "the ability of our climate simulation (that is driving the CHIMERE run) to simulate the well-known NAO-impact on the European atmospheric conditions". The alluded sentence is actually preceded and followed by these clarifications. Hopefully they help to avoid misunderstandings.**

**Please, recall also the arguments provided on comment #2 of section 2 and on the comment on section 3 related to the negligible influence of the choice of the NAO index in the framework of this study.**

2. Page 13900, lines5-7: How is the enhanced DUST concentration in the Iberian peninsula (Fig.4c) related to the precipitation in the same area (Fig. 3e)? Please be more descriptive on the analysis of the results in cases like this one.

**In a nutshell we explain the general 'rule' in the first paragraph of section 4.2 (before sections 4.2.1 and 4.2.2). In particular, it is said there:**

**'... 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).'**

**We try to avoid repeating this arguing for the sake of space. Besides, these general arguments are later recalled in the conclusions section.**

3. Page 13901, line16: The SOA levels are shown in Fig.5f not 5d.

**This has been updated with the new figures numbering.**

## Conclusions:

The results from this work, as discussed in this section, state that the aerosol concentrations are influenced by the changes in precipitation, temperature and wind fields. This is a result already known from the physics and chemistry of the underlying processes, without the need to perform a 30-year model simulation. I suggest that the authors focus their conclusions on the new findings of their work that are associated with the NAO phases and impacts on the European continent and the differences therein.

**The 3rd paragraph of the conclusions section (the one alluded by the reviewer's comment) is devoted to explain the causes for the NAO-related variations in the levels of aerosols (which are summarized in the 2nd paragraph). These are primarily related to the NAO-impact on the precipitation, temperature and wind fields. As the influence of these fields on the aerosol concentrations is certainly well-known, we just recall it here in the framework of the links between the NAO-signals obtained for the climate conditions with the NAO-signals obtained for the aerosol concentrations. We agree, however, that this paragraph needed to be improved as it often lacked to mention the role of the NAO explicitly. This could certainly provoke misunderstandings, since the objective of this paragraph is not to elucidate the impact of climate on aerosol concentrations but to link the NAO-impact on climate to the NAO-impact on aerosol concentrations. Hence, we have emphasized it better in the new version of the manuscript, but still conserving the explanations provided for the link between climate (as it is controlled by the NAO) and aerosol concentrations.**

**On the other hand, the 30-year long simulations allowed us to assess variations at the interannual time-scale (i.e. changes in the seasonal means between years with opposite NAO phases) with statistical robustness, as pointed out at the end of section 2.2, instead of focusing on a small number of events. We thus think that the length of the simulation is really valuable.**

## Figures 3 to 5:

The quality of these figures that present the main findings of this work is not acceptable. It is very difficult to see the details in each plot and see how the text is supported by the figures. The authors should leave 4-6 panels (maximum) in each figure and make sure that the details are easily discernible.

**Following this requirement, the former Figure 3 is now split into the current Figures 3 and 4, and the former Figures 4 and 5 are now split into the current Figures 5, 6, 7 and 8.**

## Technical corrections:

1. Please replace “non-antropogenic: with “non-anthropogenic” everywhere in the text.

**Replaced.**

2. Abstract, line 13: replace the word “rebounds” with “influences” or “affects”.

**Replaced.**

3. Abstract, line 16: please rephrase the part that reads “of this later” as the meaning is not clear. What is this later?

**The sentence has been removed.**

4. Introduction, p13891, line 7: please replace the word “paramount” with a more modest one.

**Changed.**

5. Section 2.1, page 13893: The sentence “This resolution enhances from previous works. . .” must be rephrased as the verb “enhance” is not appropriate.



**Amended.**

6. Page13895, line26: Please rephrase the “it arises mandatory” with “it becomes mandatory”.

**Done.**

**REFERENCES (not included in the manuscript):**

**Vautard, R., Honoré, C., Beekmann, M., and Rouil, L., 2005: Simulation of ozone during the august 2003 heat wave and emission control scenarios. *Atmos. Environ.*, 39(16):2957-2967).**

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

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7 S. Jerez<sup>(1)</sup>, P. Jimenez-Guerrero<sup>(2)</sup>, J.P. Montáñez<sup>(2)</sup> and R.M. Trigo<sup>(1)</sup>

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15 **Keywords:** North Atlantic Oscillation, air quality simulation, aerosols, Europe

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19 Manuscript submitted to: Atmospheric Chemistry and Physics

20 Date: **May September** 2013

## 21 Abstract

22

23 ~~This contribution assesses non-anthropogenic variations in ground-level aerosol~~  
24 ~~concentrations over Europe associated to changes in the phase of the North~~  
25 ~~Atlantic Oscillation (NAO).~~ The North Atlantic Oscillation (NAO) controls a large  
26 amount of the European climate variability with asymmetric impacts in both time and  
27 space. Here we investigate how the local atmospheric processes (without  
28 considering large inter-continental transport mechanisms), as they are governed by  
29 the NAO, affect the levels of various aerosol species using simulated data under  
30 constant emissions, which are fixed to the 2005 levels in order to avoid  
31 anthropogenic-induced signals. In particular, we analyze interannual variations at  
32 the seasonal time-scale and focus on the ground-level. The results show ~~Based on~~  
33 ~~simulated data and focusing on how the local atmospheric processes (without~~  
34 ~~considering large-scale mechanisms) governed by the NAO affect the levels of~~  
35 ~~various aerosol species, this study highlights~~ that positive NAO phases favor  
36 increased aerosols concentrations levels in southern (northern) regions in winter  
37 (summer), while negative NAO phases enhance them in northern (southern) regions in  
38 winter (summer). The underlying processes are ~~Variations are up to and over 100%~~  
39 ~~for most aerosols, being~~ clearly related to the NAO -impact on precipitation and wind, as  
40 they act to clean the atmosphere through removal and dispersion processes, and to the  
41 NAO -impact on the radiation balance (i.e. cloudiness) as it ~~rebounds on~~ affects the  
42 biogenic emitting activity and on the oxidative capacity of the atmosphere. Differences for  
43 all the species studied (natural inert, secondary inorganic and organic aerosols) are  
44 up to  $5 \mu\text{g m}^{-3}$ , reaching 10 and  $20 \mu\text{g m}^{-3}$  for PM<sub>10</sub> and PM<sub>2.5</sub> respectively, which  
45 represents variations about 20-40% in their mean levels between opposite NAO  
46 phases. ~~Beyond deepening on the understanding of fundamental interactions~~  
47 ~~between climate and air quality, these results provide a basis for improving the~~  
48 ~~potential predictability of this later since much work is being done in order to gain~~  
49 ~~accuracy in the NAO predictions.~~

## 50 **1 Introduction**

51

52 Air pollution is a major environmental and health problem affecting industrialized and  
53 developing countries around the world. Main detrimental consequences of the exposure of  
54 humans to photochemical and particulate matter pollution include respiratory difficulties,  
55 especially for sensitive people (Pope et al., 2006). The results of the APHEIS study  
56 (Ballester et al., 2008) indicate that atmospheric pollution causes the premature death of  
57 over 310.000 dwellers in the European Union each year. The most serious problems in  
58 Europe are related with particulate matter with a diameter of less than 10 micrometers  
59 (PM10), nitrogen dioxide and ozone. In this sense, worldwide epidemiological studies  
60 show a consistent increase in cardiac and respiratory morbidity and mortality from  
61 exposure to air pollution (e.g. Pope et al., 2009). Besides, ecosystems are also affected,  
62 with losses of agricultural crops and damages in aquatic and terrestrial ecosystems having  
63 been reported (Van Dingenen et al., 2009). In order to anticipate when and why episodes  
64 of air pollution arise and how they can be abated, reliable estimations of air pollution levels  
65 and a better understanding of the chemico-physical processes behind them are **crucial of**  
66 **paramount importance.**

67

68 Air pollution levels depend on both emissions (either natural or anthropogenic) and the  
69 atmospheric conditions steering and transforming them through processes related to  
70 chemistry, transport and removal (Jacob and Winner, 2009). Precipitation provokes wet  
71 deposition, hence favoring the removal of airborne pollutants, and wind favors their  
72 transport and dispersion, although enhanced winds also promote the formation of marine  
73 aerosols over the water mass areas. On the other hand, radiation levels and temperature  
74 play a major role on gas-phase chemistry through the photolysis of primary and secondary  
75 pollutants (Katrakou et al., 2010; **Forkel et al., 2012; Jiménez-Guerrero et al., 2012;**  
76 **Meier et al., 2012; Colette et al., 2013; Hedegaard et al., 2013, just to mention some**  
77 **recent works on the topic**), determining also the vegetation activity and thereby  
78 modifying natural emissions.

79

80 Despite the great climate heterogeneity and variability that characterizes Europe, just a  
81 few large-scale teleconnection modes control a large amount of it (Trigo et al., 2008). In  
82 particular, many studies establish the fundamental role of the North Atlantic Oscillation  
83 (NAO) at this regard, affecting especially western Europe with an asymmetric impact

84 between northern and southern areas (Osborn et al., 1999; Wanner et al., 2001; Trigo et  
85 al., 2002, 2008). The NAO pattern consists of a meridional gradient in Sea Level Pressure  
86 (SLP) over the North Atlantic with centers roughly around the Azores Islands (high  
87 pressures) and near Iceland (low pressures). During its positive phases (NAO<sup>+</sup>), such a  
88 dipole in SLP is enhanced with the consequent intensification of westerly winds in northern  
89 Europe associated with an intensified jet stream. This configuration promotes also the  
90 steering of most storms over northern regions, advecting humid air from the ocean, hence  
91 favoring cloudiness and precipitation. Contrary and consistently, NAO<sup>+</sup> reduces cloudiness  
92 and precipitation in southern Europe. Conversely, the intense zonal circulation promoted  
93 by the negative NAO phases (NAO<sup>-</sup>) enhances the westerly flow penetrating in southern  
94 Europe through the Iberian Peninsula (Jerez et al., 2013b) and extending until eastern  
95 Europe and allows cyclones to follow more southern paths within the European continent  
96 (Trigo, 2006). Most studies have focused their analysis on NAO-climate links during the  
97 winter season, where the SLP gradient is stronger and the climatic impact of this large-  
98 scale mode of variability ~~is extensive~~ extends to Eurasia (Wanner et al., 2001; Trigo et  
99 al., 2002) but also North America, Greenland, the Arctic and North Atlantic oceans (Hurrell  
100 and van Loon, 1997). Readers looking for comprehensive reviews of the dynamics of the  
101 NAO and associated impacts should consult the books by Hurrell et al. (2003); Vicente-  
102 Serrano and Trigo (2011). However, recent works have shown that the summer NAO still  
103 plays an important role on shaping the climate of northern and central Europe (albeit less  
104 relevant than in winter) including parts of the Mediterranean basin (Folland et al., 2009;  
105 Bladé et al., 2012).

106

107 These NAO-related impacts on the atmospheric fields are bound to inflict an important  
108 impact on air pollution levels (Dayan et al., 2008; Sanchez-Lorenzo et al., 2008; Chiacchio  
109 and Wild, 2010), particularly on aerosols concentrations. This influence should arise  
110 through both local processes and large-scale air pollutants transport. **For instance,**  
111 **Eckhardt et al. (2003) showed that NAO<sup>+</sup> enhances northward transport of pollutants**  
112 **from Europe to Arctic regions, mainly in winter and spring, compared to NAO<sup>-</sup>**  
113 **phases. At the same time, NAO<sup>+</sup> promotes African dust intrusions into south-**  
114 **western European regions (which occurs mostly in the summertime) as the westerly**  
115 **winds associated to NAO<sup>-</sup> events prevent subtropical air masses to reach the**  
116 **European mid latitudes; although, on the other hand, this NAO<sup>-</sup> related large-scale**  
117 **zonal winds also favor the transport of pollutant from North America into Europe**

118 (Moulin et al., 1997; Dayan et al., 2011; Christoudias et al., 2012; Cusack et al., 2012;  
119 Pey et al., 2013). But the signature of the NAO in the climatic conditions (and  
120 thereby on air pollution levels) does not restrict to changes in the large-scale  
121 circulation patterns, as these have also repercussions on other atmospheric  
122 variables such as precipitation and temperature as commented above, with a  
123 potential subsequent impact on air quality through local processes such as aerosol  
124 wet deposition. However, few studies have been devoted to explore air pollution  
125 levels from a climatic perspective, even less disentangling between local and large-  
126 scale mechanisms, therefore the contribution of the local NAO-controlled processes  
127 on the climatology of air pollution levels is still hardly established., ~~either from~~  
128 ~~Europe to Arctic regions (Eckhardt et al., 2003) or from North America and Africa~~  
129 ~~towards Europe (Moulin et al., 1997; Dayan et al., 2011; Christoudias et al., 2012).~~  
130 ~~However, since few studies are devoted to explore air pollution levels from a~~  
131 ~~climatic perspective, the impact of the NAO in such a framework is still hardly~~  
132 ~~established.~~

133  
134 Hence, the objective of the present study is to **deepen on elucidate** the signature of the  
135 NAO in terms of mean concentration of aerosols in a region covering the entire  
136 Mediterranean basin from north Africa to north Europe **with the focus on elucidating the**  
137 **influence of the small scale processes, as well as the associated underlying**  
138 **mechanisms**. To achieve this goal, we use a numerical simulation of the atmospheric  
139 chemical composition that spans three decades of the recent past without considering  
140 variations in the anthropogenic emissions, thus allowing to isolate the natural variations in  
141 the aerosol levels. ~~Besides, t~~The simulation was designed to disregard the contribution  
142 from **the long-range large-scale** transport **by using constant climatological boundary**  
143 **conditions for the aerosol concentrations. This design does not provide a realistic**  
144 **picture, but allows to improve our understanding on the role of the local underlying**  
145 **mechanisms, as it remains unmasked by the large-scale inter-continental advective**  
146 **phenomena**. Hence, we focus on the role played by local processes as they are governed  
147 by the NAO, restricting the evaluation to the ground-level.

148  
149 The structure of this work is as follows. Section 2 describes the modeling system and the  
150 experimental set-up. Section 3 provides the methodology. Section 4 presents the results.  
151 Finally, Section 5 summarizes **and discusses** the main conclusions.

152

153

## 154 **2 Data**

155

### 156 **2.1 Air-quality modeling system**

157

158 The modeling system consists of a climatic version of the Fifth-Generation Pennsylvania  
159 State University - National Center for Atmospheric Research Mesoscale Model (MM5)  
160 (Grell et al., 1994) driven by ERA40 reanalysis (Uppala et al., 2005), when available, or  
161 ECMWF analysis data when not **(in both cases without nudging)**, coupled off-line to the  
162 CHIMERE chemistry transport model (Bessagnet et al., 2004; Rouil et al., 2009).  
163 MELCHIOR2 gas-phase mechanism is implemented within CHIMERE (Derognat et al.,  
164 2003). The chemistry transport model includes aerosol and heterogeneous chemistry,  
165 distinguishing among different chemical aerosol components, namely nitrate, sulfate,  
166 ammonium, elemental and organic carbon with three subcomponents (primary, secondary  
167 anthropogenic and secondary biogenic) and marine aerosols. Unspecified primary  
168 anthropogenic aerosols and aerosol water are additionally kept as separate components.  
169 The model considers the thermodynamic equilibrium using the ISORROPIA model (Nenes  
170 et al., 1998). Last, the aerosol microphysical description for CHIMERE is based on a  
171 sectional aerosol module including 6 bins from 10 nm to 40  $\mu\text{m}$  using a geometrical  
172 progression.

173

174 The CHIMERE domain considered in the modeling system covers all the Mediterranean  
175 basin extending to northern Europe having a spatial resolution of 0.2 degrees in the  
176 horizontal, which is around 25 km at the European latitudes considered, and eight vertical  
177 levels unevenly spaced up to 550 hPa. This resolution **is higher than usual in climate**  
178 **runs enhances from previous works (e.g.** Carvalho et al., 2010; Katragkou et al., 2010;  
179 Jiménez-Guerrero et al., 2011; Juda-Rezler et al., 2012; Manders et al., 2012).

180

181 The fields from MM5 (provided with a spatial resolution of 90 km) are bilinearly interpolated  
182 to the CHIMERE working grid. Detailed descriptions of the climate modeling system used  
183 and its skill to realistically reproduce **the main** regional **climatic** features **of the climate in**  
184 **the target domain**, such as **the** temperature and precipitation annual cycles or the

185 interaction between the large-scale circulation and the orography, which largely modulates  
186 the rainfall patterns having an important influence on air quality, can be found in Gomez-  
187 Navarro et al. (2011) and Jerez et al. (2013a). **The model setup used here is the same**  
188 **as in the former reference; in the second, it is evaluated in the context of a multi-**  
189 **physics ensemble of present-day climate simulations. Although both works focus**  
190 **on the Iberian Peninsula, there are extensive literature demonstrating the ability of**  
191 **MM5 for reproducing diverse meteorological features along Europe (e.g. Kotlarski et**  
192 **al., 2005; Renfrew et al., 2009; Pfeiffer and Zängl, 2010). Moreover, Section 4.1**  
193 **provides a revision of the accuracy of the patterns of the NAO impact on climate as**  
194 **they are obtained from the climate simulation driven CHIMERE.**

195  
196 Boundary conditions **of gas-phase pollutants concentration** for **CHIMERE the-**  
197 **chemistry-transport-model** are based on the global chemistry model LMDz-INCA2  
198 (Szopa et al., 2009) developed by the Laboratoire des Sciences du Climat et  
199 l'Environnement (LSCE). A detailed description of the Interactive Chemistry and Aerosol  
200 (INCA) model is presented in Hauglustaine et al. (2004) and Folberth et al. (2006). For  
201 aerosols, boundary conditions are taken from the GOCART model (Chin et al., 2002).  
202 **These boundary conditions consist of constant m**Monthly mean data **that** are  
203 interpolated in the horizontal and vertical dimensions to force the major chemical  
204 concentrations at the boundaries of the CHIMERE domain. ~~Although the influence of~~  
205 ~~using~~ **The use of constant** climatological boundary conditions **prevents on-ground-level**  
206 ~~concentrations is largely overwhelmed by local processes (Jiménez-Guerrero et al.,~~  
207 ~~2012), it should be acknowledged that, in particular, this hampers~~ to capture the  
208 interannual variability of the NAO -impact on the aerosol concentration levels at the  
209 domain boundaries, thus avoiding the evaluation of large-scale **inter-continental** transport  
210 mechanisms related to the NAO phase. **On the contrary, and despite the pollutants**  
211 **transport between different areas within our extensive domain is modeled by**  
212 **default, this experimental design allows to largely isolate the role of more local**  
213 **processes (for instance, how the NAO influences the concentration of aerosols**  
214 **through its impact on local precipitation patterns). ~~On the contrary, this~~**  
215 ~~experimental design allows to better isolate and understand the role of the local~~  
216 ~~processes, including the pollutants transport between different areas within our~~  
217 ~~domain.~~ **At this regard, it should be noted that the design of the driving MM5 climate**  
218 **simulation does not follow this approach. The climate boundary conditions at the**



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

223

224 Year-to-year varying anthropogenic emissions are derived from the EMEP database on a  
225 monthly basis (Vestreng et al., 2009). Natural emissions depend on climate conditions,  
226 and consequently they are modeled according to the MM5 meteorological outputs.  
227 However, the levels of air pollutants are estimated without considering possible changes  
228 on vegetation, land use or any feedback from the chemical compounds to the  
229 meteorological fields. Biogenic emissions were generated dynamically using MEGAN  
230 (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006) with the  
231 parametrization form of the canopy environment model. The model estimates hourly  
232 isoprene, monoterpene, and other BVOC emissions based on plant functional type and as  
233 a function of temperature and ground level shortwave radiation.

234

235 **Beyond the several works supporting the ability of similar MM5-CHIMERE systems**  
236 **to reproduce the main air quality features over Europe (e.g. Monteiro et al., 2007;**  
237 **Flaounas et al., 2009; Péré et al., 2010), our This** modeling system has been validated  
238 by comparing a simulation covering the period 1990-2010 with EMEP observations  
239 (Tørseth et al., 2012). **A Although a** thorough evaluation of the modeling approach is not  
240 included in the main objectives of this work, ~~and that task constitutes the focus of an~~  
241 ~~on-going paper. In any case, we acknowledge that it is opportune to here we~~ show  
242 shortly that the simulated series of aerosols concentrations correlates acceptably and  
243 captures a large fraction of the **interannual** variability of the observational series **at the**  
244 **working time-scale (i.e. seasonal)**. These two aspects are the most relevant for the  
245 assessment performed below, while systematic biases would not represent a major  
246 concern as they should be largely canceled when computing the composites of the NAO  
247 ~~-impact~~ (see next Section). For ~~the sake of~~ brevity, we provide here the results for the  
248 simulated concentration of aerosols under **several two** size-thresholds, namely PM10 and  
249 PM2.5 (Figure 1). For both cases, correlations between simulated and observational  
250 **seasonal-averaged** series are overwhelmingly above 0.8, in most cases above 0.9, both  
251 in winter and summer. ~~As well~~ **Also**, it is possible to verify that biases in the standard  
252 deviation of the simulated series are largely negligible in comparison to their mean values

253 (being orders of magnitude smaller). It is also worth mentioning that normalized biases are  
254 always found to stay below 30% ~~the-worst~~ (not shown), which is in the top range expected  
255 (Pay et al., 2010). ~~This~~ **These results** guarantees the phase accordance (timing)  
256 between the simulated and observational series, their similar amplitude and, also, the  
257 quantitative accuracy of the simulated climatologies, hence making us confident on the  
258 **accuracy suitability** of the modeling system for the purpose of this study.

259

## 260 **2.2 Experimental set-up**

261

262 In order to isolate the influence of climate variability on air quality, it ~~arises~~ **becomes**  
263 mandatory to avoid the signals derived from human policies. During the last decades,  
264 these policies have committed European countries to strongly reduce the emissions of  
265 contaminants to the atmosphere, which has caused strong non-natural trends in the  
266 observational series of almost all aerosols species (Vestreng et al., 2009; Tørseth et al.,  
267 2012). Hence, the impact of the NAO has been evaluated here based on a 30-year long  
268 MM5-CHIMERE simulation for which emissions were fixed at their 2005 mean levels. The  
269 simulated period (1970-1999) is not particularly significant in itself, but its length ~~is-~~  
270 **relevant as it** supports the robustness of our results from a climatic perspective.

271

272

## 273 **3 Methodology**

274

275 This assessment focuses on several aerosol families and species, namely natural inert  
276 aerosols (sea salt aerosols, SALT, and wind-blown and resuspended dust, DUST),  
277 secondary inorganic aerosols (sulfate,  $\text{SO}_4^{2-}$ , nitrate,  $\text{NO}_3^-$ , and ammonium,  $\text{NH}_4^+$ ), organic  
278 matter (OM), with particular attention to secondary organic aerosols (SOA), and elemental  
279 carbon (EC). The total concentrations of PM10 and PM2.5 are also studied.

280

281 The analysis is performed at the seasonal timescale for winter (December-to-March  
282 averages) and summer (June-to-September averages). This decision is based on previous  
283 tests performed at the monthly timescale showing the strongest responses in those  
284 months, with very similar structures ~~within in~~ each season ~~considered-but-very-different~~  
285 ~~in-each-one-of-them~~. The remaining months from the two transitional seasons depicted  
286 intermediate and softer signals **and thus are not included in being disregarded from**

287 the rest of the analysis.

288

289 **Seasonal means (DJFM and JJAS averages) of the monthly NAO index provided by**  
290 **the Climate Prediction Center (CPC) from the National Ocean and Atmospheric**  
291 **Administration (NOAA) are used to define classes of positive and negative NAO**  
292 **phases (NAO<sup>+</sup> and NAO<sup>-</sup> respectively) throughout the study period. Following a a**  
293 **common approach adopted in previous studies (e.g. Suski and Ridgway, 2007;**  
294 **Gouveia and Trigo, 2008), NAO<sup>+</sup> (NAO<sup>-</sup>) phases are defined as those with the NAO**  
295 **index above (below) the 70<sup>th</sup> (30<sup>th</sup>) percentile of the NAO index time series**  
296 **considered (Figure 2). The use of these non-subjective thresholds in the definition**  
297 **of NAO<sup>+</sup> and NAO<sup>-</sup> phases ensures the evenness between the number of events of**  
298 **each class (within the 30-year simulated period, 1970-1999, we retain 9 events of**  
299 **each class in each season) and prevents the unbalancing influence of potential**  
300 **long-term trends in the NAO index time series. On the other hand, it should be**  
301 **noticed that different data sources and/or methods for computing the NAO index**  
302 **may provided different values of it. In particular, we are aware that the CPC/NOAA**  
303 **NAO index is not based on the ECMWF data used to drive MM5 in our modeling**  
304 **system. However, we confirmed that, at the time-scale assessed here, this influence**  
305 **is negligible (Greatbatch and Rong, 2006). Hence, the choice of the CPC/NOAA NAO**  
306 **index for the present study is based on its easy access and its up-to-date**  
307 **computation approach, which in fact makes it widely used nowadays in studies**  
308 **focused on the recent past (e.g. Bladé et al., 2012; Jerez et al., 2013b; Pey et al.,**  
309 **2013).**

310

311 ~~In order to assess the NAO impact, we have followed a common approach adopted~~  
312 ~~in previous studies (e.g. Suski and Ridgway, 2007; Gouveia and Trigo, 2008) and~~  
313 ~~defined classes of positive (NAO<sup>+</sup> ≥ 70<sup>th</sup> percentile) and negative (NAO<sup>-</sup> ≤ 30<sup>th</sup>~~  
314 ~~percentile) NAO phases using the seasonal means of the monthly NAO index~~  
315 ~~provided by the Climate Prediction Center (CPC) from the National Ocean and~~  
316 ~~Atmospheric Administration (NOAA). This procedure ensures the balance between~~  
317 ~~the number of events of each class that will be considered. Within the 30-year~~  
318 ~~simulated period (1970-1999), we obtain 9 winters and summers of each NAO class~~  
319 ~~(Figure 2).~~ The NAO impact on the assessed magnitudes is then evaluated through  
320 composites showing the differences in the mean fields between positive and negative NAO

321 phases. Several significance controls are applied to these differences ensuring both their  
322 statistical robustness and physical consistence, so that they are just considered when  
323 fitting the following criteria: (1) being statistically significant above the 90% confidence  
324 level, and (2) being supported by statistically significant temporal correlations (above the  
325 90% confidence level) between the NAO and the corresponding chemical component  
326 series. Statistical significance is evaluated by performing two-tailed t-tests for the null  
327 hypothesis of equal means or zero correlation respectively (Snedecor and Cochran, 1989).  
328 Moreover, the signal-to-noise ratio, defined as the ratio between the absolute value of the  
329 NAO<sup>+</sup> minus NAO<sup>-</sup> differences in a specific magnitude and the standard deviation of the  
330 whole seasonal series of such a magnitude, is considered as a relative measure of the  
331 importance of the NAO -impact. In particular we have blurred the areas where the signal-  
332 to-noise ratio is below the unit. In the areas not blurred, the NAO -impact exceeds one  
333 standard deviation of the series and hence we are likely moving from one tail to the other  
334 of the probability density function describing the variability in the levels of the assessed  
335 magnitude when the NAO phase changes.

336

337

## 338 **4 Results**

339

### 340 **4.1 Revisiting the NAO -impact on atmospheric conditions**

341

342 Several works have been devoted to explore the role and signature of the NAO in the  
343 European climate (e.g. Hurrell, 1995; Hurrell and van Loon., 1997; Hurrell et al., 2003;  
344 Trigo et al., 2002, 2008). Indeed, the reported NAO -impacts on the climatic variables  
345 motivated the present study. Other works have shown a good capacity of the general  
346 circulation models to reproduce the large-scale patterns of this climatic impact over Europe  
347 particularly for winter (Osborn et al., 1999; Osborn, 2011; Hurrell et al., 2003) but also for  
348 summer (Bladé et al., 2012). Hence, this Section does not intend to provide novel insights,  
349 but essentially to evaluate the ability of our climate simulation (that is driving the CHIMERE  
350 run) to simulate the **extensively reported** NAO -impact on the **European** atmospheric  
351 conditions. This fulfills two relevant requirements, namely to (1) guarantee that it is  
352 effectively able to reproduce the known NAO -impacts on the European climate, and (2)  
353 provide an appropriate context for the interpretation of the following results of the NAO  
354 -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<sup>+</sup> (NAO<sup>-</sup>) 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<sup>-</sup> phases **have are** associated **with** a stronger westerly  
362 flow in northern Europe than NAO<sup>+</sup> 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<sup>-</sup> 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 34, 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<sup>+</sup> minus NAO<sup>-</sup> 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<sup>+</sup> enhancing cloudiness there, around 20%). Regarding the NAO -impact on  
383 mean 2-meter temperature, the most important signal consists of positive NAO<sup>+</sup> minus  
384 NAO<sup>-</sup> 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

389 responses to the NAO phase, providing a meaningful representation of the atmospheric  
390 conditions governing during positive and negative NAO events that will be used to  
391 understand the NAO -impact on aerosol concentration levels assessed in the next Section.  
392

## 393 **4.2 NAO -impact on mean ground-level aerosols concentrations**

394  
395 NAO<sup>+</sup> minus NAO<sup>-</sup> composites of mean ground-level concentration of the various aerosol  
396 species are provided in Figures **4-and 5 to 8**. In general, the positive phase of NAO  
397 enhances aerosol concentrations in southern Europe in winter and in northern Europe in  
398 summer. These positive NAO<sup>+</sup> minus NAO<sup>-</sup> differences match generally well the sectors  
399 characterized by negative signals in wind speed, precipitation and cloudiness (Figures **3**  
400 **and 4**). Thus such patterns can be well explained by the diluting effect of stronger winds  
401 and scavenging processes by an enhanced precipitation and by the inhibitory effect of  
402 enhanced cloudiness as it implies reduced shortwave solar radiation (a main precursor for  
403 photochemistry and biogenic emitting activity).  
404

### 405 **4.2.1 Winter signals**

406  
407 As a general rule, in winter, the regions holding the largest signals are predominantly  
408 located in the Mediterranean basin, particularly over the Iberian Peninsula, northern Italy  
409 and the Balkans (Figures **4-and 5 to 8**, left column). There, results show increases in the  
410 mean concentration of all aerosols species during positive NAO phases exceeding the  
411 50% of the mean levels during negative NAO phases, even doubling these latter in some  
412 occasions. In absolute values (although these should be taken with care due to our  
413 experimental design not accounting for real emission levels **neither for long-range**  
414 **transports**), the largest variations between NAO phases reach  $5 \mu\text{g m}^{-3}$  and affect the  
415 inorganic compounds (DUST,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^-$  and  $\text{NO}_3^-$ ), while the change in the concentration  
416 of carbonaceous material is just about  $1 \mu\text{g m}^{-3}$ . It is worth stressing the slight but still  
417 significant negative patches of NAO<sup>+</sup> minus NAO<sup>-</sup> differences in the British Islands and in  
418 north-eastern Europe that appear in the patterns of OM and EC, indicating a distinct  
419 response of this compounds to the same NAO phase depending on the latitude.  
420

421 The NAO -impact on  $\text{NO}_3^-$  concentration levels is negligible over most of Europe. However,  
422 the removal effect of **the NAO<sup>-</sup> phase associated with** enhanced precipitation in northern

423 Italy during NAO<sup>-</sup> phases (~~not counteracting by enhanced temperatures~~) seems to  
424 play a key cleaning role at this regard in this NO<sub>3</sub><sup>-</sup>-strong emitting area.

425

426 Contrary to the rest of aerosol species, enhanced winds promote the formation of SALT  
427 (sea salt aerosol emissions depends on the cube of the wind speed), thus leading to  
428 higher concentrations. This is mainly observed over the water mass areas, with NAO<sup>+</sup>  
429 leading to 40% higher SALT concentrations in the surroundings of the British Islands,  
430 which matches with the NAO<sup>+</sup> enhanced winds observed in this area (Figure 3c), but  
431 diminishing them in the southern Mediterranean (Figure 45a). Singular positive signals  
432 appear also in the Gulf of Genoa (north-westward of Italy) and the strait of Gibraltar  
433 (southward of Spain). Regarding these three latter cases, it can be roughly appreciated in  
434 Figure 3a that NAO<sup>+</sup> promotes windier conditions over the mentioned areas (longer  
435 arrows) than NAO<sup>-</sup>, although these differences in wind speed do not remain after applying  
436 the significance controls as they are not reflected in Figure 3c.

437

438 The NAO<sup>-</sup> impact on winter air quality is appreciable in the various size-classes of  
439 particulate matter. Figure 5g,h 8a,c shows that NAO<sup>+</sup> enhances 20-40% the  
440 concentrations of both PM<sub>10</sub> and PM<sub>2.5</sub> in the southern European regions (where the PM  
441 climatologies present the higher values and the number of exceedances of the limit values  
442 for the protection to human health are more frequent), and also, but to a smaller extent, in  
443 some northernmost areas. Therefore, the winter NAO phase has a clear impact on the air  
444 quality-related human healthy risk in this season.

445

#### 446 4.2.2 Summer signals

447

448 In summer, the highest signals within our domain appear in the British Islands, northern  
449 France, Belgium, Netherlands, northern Germany and northern Poland (Figures 4 and 5 to  
450 8, right column). It is also interesting to see the recurrent positive signals appearing over  
451 Italy also in this season, although it should be acknowledged the patchy nature of them.

452

453 As in winter, differences in the aerosol concentrations between NAO phases are up to  
454 100%, demonstrating the profound impact of the NAO in this area/season. Likewise, the  
455 obtained NAO<sup>+</sup> related increments are, in absolute values, about (even over) 5 μg m<sup>-3</sup> for  
456 each inorganic species but SALT (which shows smaller signals, Figure 45b). The NAO<sup>+</sup>-

457 related increases in the concentration of the organic and carbonaceous aerosols are also  
458 a bit smaller (when they are expressed in  $\mu\text{g m}^{-3}$ ).

459

460 It is worth stressing the signal in mean SOA levels (Figure 57d), which can be doubly  
461 related to the reducing effect of NAO<sup>+</sup> in precipitation (Figure 3f 4b), as it prevents wet  
462 deposition, and to the increased temperatures during NAO<sup>+</sup> enhancing the biogenic  
463 emissions (such as isoprene and monoterpenes), which leads to higher levels of biogenic  
464 SOA.

465

466 The NO<sub>3</sub><sup>-</sup> signal appears again mainly restricted to the stronger emitting-areas, being  
467 located in this season between northern France, Belgium and Germany (Figure 4j 6d).  
468 There, the reduced precipitation during NAO<sup>+</sup> events doubles the concentrations of nitrate  
469 in comparison to the levels during NAO<sup>-</sup> phases.

470

471 SALT depicts slight variations associated to the NAO phase in summer (around  $0.5 \mu\text{g m}^{-3}$ ,  
472 Figure 45b). However, contrary to the winter analysis, significant impact areas are now  
473 located over land areas and not over the sea, namely in eastern Iberia and Italy and in  
474 northeastern Europe, where the results show positive NAO<sup>+</sup> minus NAO<sup>-</sup> differences.

475

476 Last, PM<sub>2.5</sub> and PM<sub>10</sub> show differences up to 10 and 20  $\mu\text{g m}^{-3}$  respectively (higher  
477 concentrations during NAO<sup>+</sup>), mainly concentrated in the northernmost areas of the  
478 domain and representing variations up to 20-40% between NAO phases (Figure 5h,j 8b,d)  
479 **as observed for winter.**

480

481

## 482 **5 Conclusions**

483

484 This study establishes the strong impact that the NAO-related local atmospheric processes  
485 have on mean ground-level aerosol concentrations over Europe. For that we use a 30-year  
486 long air quality simulation with a spatial resolution of 25 km over the target region in which  
487 the masking influence of human policies aimed at reducing emissions has been  
488 intentionally omitted. Moreover, this simulation allows isolating the influence of the local  
489 processes, i.e. those taking place within the boundaries of the domain, as the boundary



490 conditions for the aerosols concentration levels did not vary from year to year in our  
491 experimental design. In this sense, ~~it must be underlined that~~ our results should not be  
492 considered deterministic, **i.e. while the whole real picture does include varying**  
493 **emissions and the effect of long-range pollutant transport, our study, aimed at**  
494 **getting a better understanding of the natural and local processes, is narrowly**  
495 **focused on them.**

496

497 The results show impacts with asymmetries in both time (i.e. between seasons) and space  
498 (i.e. between northern and southern areas). In winter, higher ground-level concentrations  
499 of all aerosol species (except for sea salt) are observed around the Mediterranean basin  
500 during the positive NAO phases, while these signals are northward shifted in the summer  
501 season. These differences involve variations up to and over 100% **in the mean levels of**  
502 **each species; about 20-40% for PM10 and PM2.5.** Eventually, softer signals of opposite  
503 sign (i.e. NAO<sup>-</sup> enhancing the ground-level concentration of aerosols) are observed in  
504 northern (southern) areas in winter (summer).

505

506 The causes for these NAO-related variations in the levels of aerosols have to be sought in  
507 a multiplicity of **climatic** factors varying between NAO positive/negative phases, **namely:** ~~;~~  
508 ~~The main climate-related mechanisms include~~ (1) increased/reduced temperature  
509 (particularly in northern Europe), (2) different distribution of **the** precipitation patterns  
510 across Europe, (3) **increased changes in the** photolysis of primary and secondary  
511 pollutants due to **changes in lower** cloudiness, and (4) the cleaning effect of enhanced  
512 winds (~~Katragkou et al., 2010~~). According to the results of this work **and based on the**  
513 **established relationship between meteorological fields and air-quality (e.g. Wu et al.,**  
514 **2008; Katragkou et al., 2010; Jiménez-Guerrero et al., 2012; Manders et al., 2012), the**  
515 **NAO impact on climate supports the NAO impact on aerosol concentrations as**  
516 **follows. Changes in** precipitation drives the **NAO impact on modification-in** the  
517 concentration of most aerosol components (in both northern and southern Europe), **since**  
518 **a with the** decrease in the precipitation modeled **during NAO<sup>+</sup> phases** leadings to a  
519 regional increase in the levels of secondary inorganic aerosols and mineral matter (e.g.  
520 Jiménez-Guerrero et al., 2012; Manders et al., 2012). As well, **the** weaker winds  
521 **associated to the NAO<sup>+</sup> events** favor the increase of particulate matter in polluted  
522 regions such as large cities or entire industrial regions (e.g. Po valey in northern Italy and  
523 the Rhine-Ruhr area in northern Germany and Holland). Also, the enhanced oxidative

524 capacity of the atmosphere with high temperatures causes SO<sub>2</sub> gas-phase emissions to  
525 turn into the particulate phase, thus increasing sulphate concentrations **as observed**  
526 **during NAO<sup>+</sup> phases in southern (northern) regions in winter (summer)**. Last, the  
527 levels of secondary organic aerosols (SOA) are conditioned by the dependence of  
528 biogenic emissions on the climatological patterns of variability. In this sense, SOA over  
529 Europe is mainly driven by the warming-induced increase in biogenic emitting activity.  
530 Although vegetation is kept invariable in the simulation analyzed here, MEGAN  
531 estimations of these emissions strongly depends on shortwave radiation and temperature  
532 (Guenther et al., 2006), which are substantially conditioned by the cloudiness and thereby  
533 by the NAO. Accordingly, the lower cloudiness associated to NAO<sup>+</sup> phases mostly in  
534 southern Europe arises as a main driver for the secondary conversion of aerosols.

535

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

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547

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857 **List of captions**

858

859 **Figure 1.** Evaluation of the MM5-CHIMERE air quality modeling system described in  
860 Section 2.1. Gray shaded colors depict the simulated climatologies in  $\mu\text{g m}^{-3}$  of PM10 (up)  
861 and PM2.5 (bottom) in winter (DJFM averages, left) and summer (JJAS averages, right).  
862 The comparison of the simulated series with EMEP observations is provided by symbols:  
863 their color shows the difference in the standard deviation of the series between the  
864 simulation and the observations (in  $\mu\text{g m}^{-3}$ ), and their shape informs on the magnitude of  
865 the temporal correlation between the simulated and the observed series.

866

867 **Figure 2.** Winter (~~up~~ top) and summer (bottom) NAO series for the period 1970-1999. The  
868 former are DJFM averages (therefore resulting in 29 values) and the latter are JJAS  
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871 index above (below) that percentiles, i.e. those selected as NAO<sup>+</sup> (NAO<sup>-</sup>) events for the  
872 composites analysis.

873

874 **Figure 3.** NAO impact in winter (left) and summer (right) on atmospheric conditions.  
875 (a,b) depict mean 10m-wind direction during NAO<sup>+</sup> (red) and NAO<sup>-</sup> (blue) phases  
876 (being the arrows length proportional to the wind speed). (c,d) provide the NAO<sup>+</sup>  
877 minus NAO<sup>-</sup> composites for the mean 10m-wind module (in m/s). These differences  
878 are represented only if they are statistically significant at the 90% level, dots blur  
879 the values not representing a signal-to-noise ratio above 1, and contours depict  
880 differences expressed as percentage.

881

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

884

885 **Figure 5.** NAO impact in winter (left) and summer (right) on mean ground levels of  
886 natural inert aerosols: NAO<sup>+</sup> minus NAO<sup>-</sup> composites for (a,b) SALT and (c,d) DUST.  
887 Differences in  $\mu\text{g m}^{-3}$  are represented only if they are statistically significant at the  
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889 contours depict differences expressed as percentage. Period considered: 1970-

890 **1999.**

891

892 **Figure 6. As in Figure 5, here for secondary inorganic aerosols: (a,b)  $\text{SO}_4^{2-}$ , (c,d)  $\text{NH}_4^+$**   
893 **and (e,f)  $\text{NO}_3^-$ .**

894

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896

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

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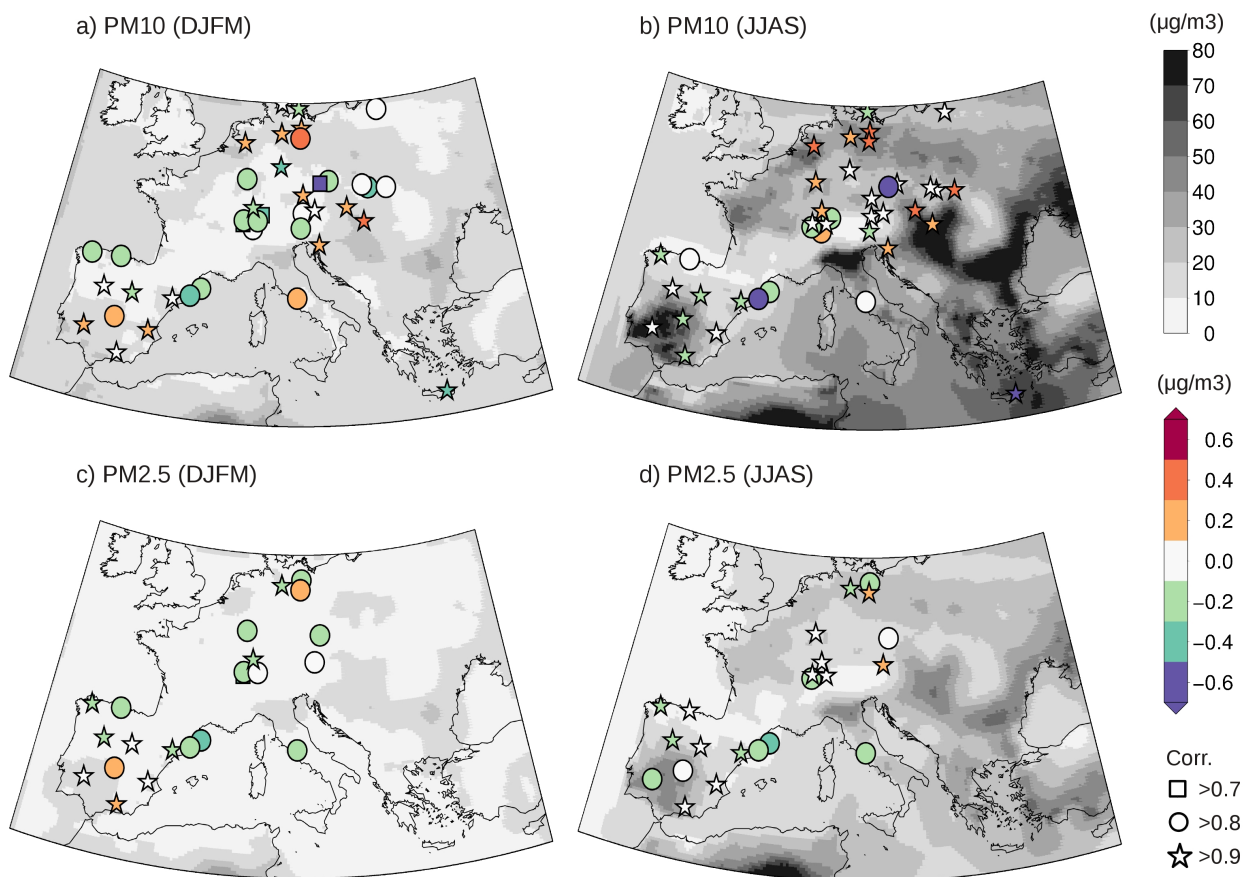
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900 ~~**conditions. (a,b) depict mean 10m-wind direction during NAO<sup>+</sup> (red) and NAO<sup>-</sup> (blue)**~~  
901 ~~**phases (being the arrows length proportional to the wind speed). The rest of panels**~~  
902 ~~**provide the NAO<sup>+</sup> minus NAO<sup>-</sup> composites for mean (c,d) 10m-wind module (in m/s),**~~  
903 ~~**(e,f) precipitation (in mm/month), (g,h) integrated cloud water (in mm) and (e,f) 2m-**~~  
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905 ~~**significant at the 90% level, dots blur the values not representing a signal-to-noise**~~  
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907

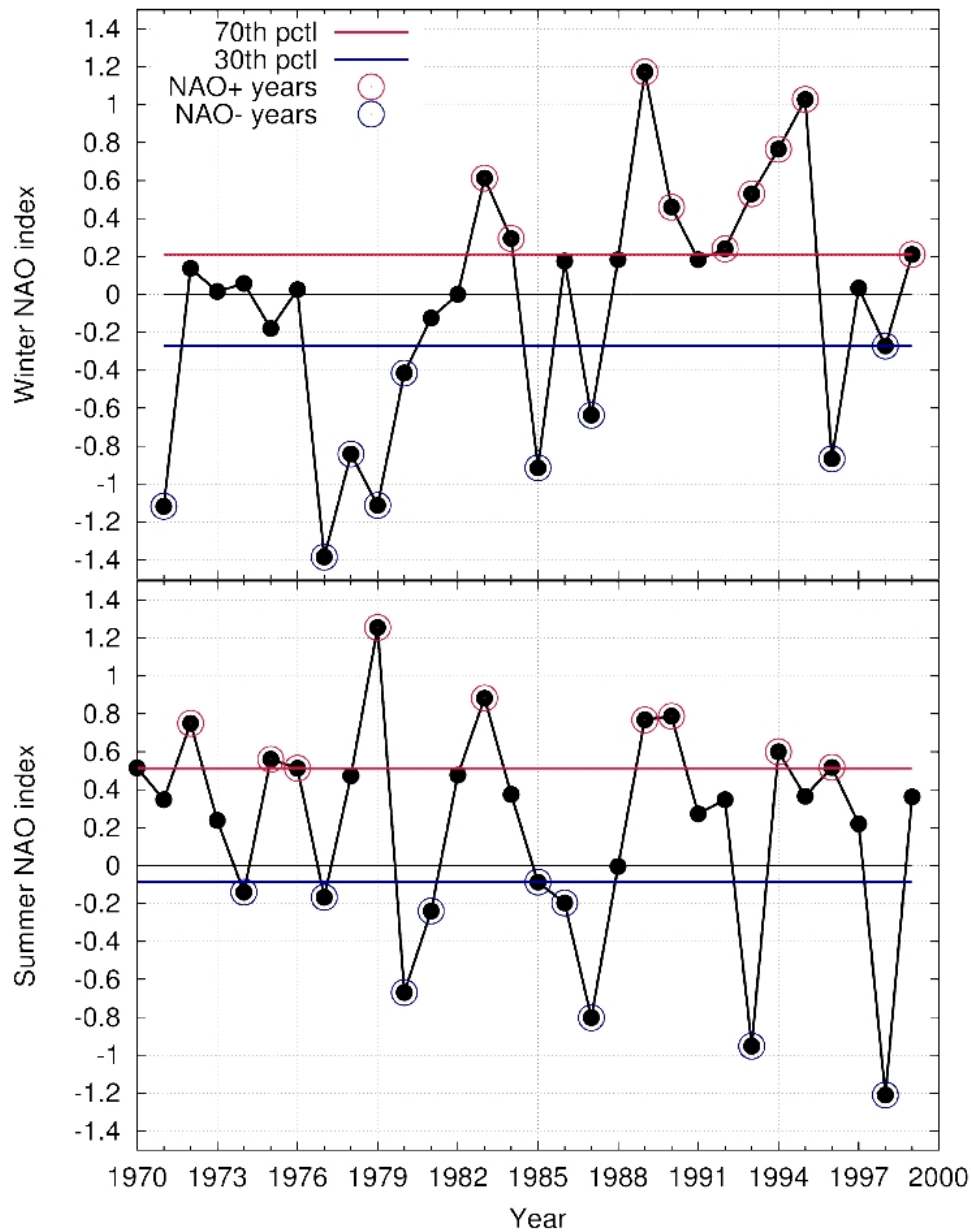
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909 ~~**NAO<sup>-</sup> composites for (a,b) SALT, (c,d) DUST, (e,f)  $\text{SO}_4^{2-}$ , (g,h)  $\text{NH}_4^+$  and (i,j)  $\text{NO}_3^-$  in**~~  
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914

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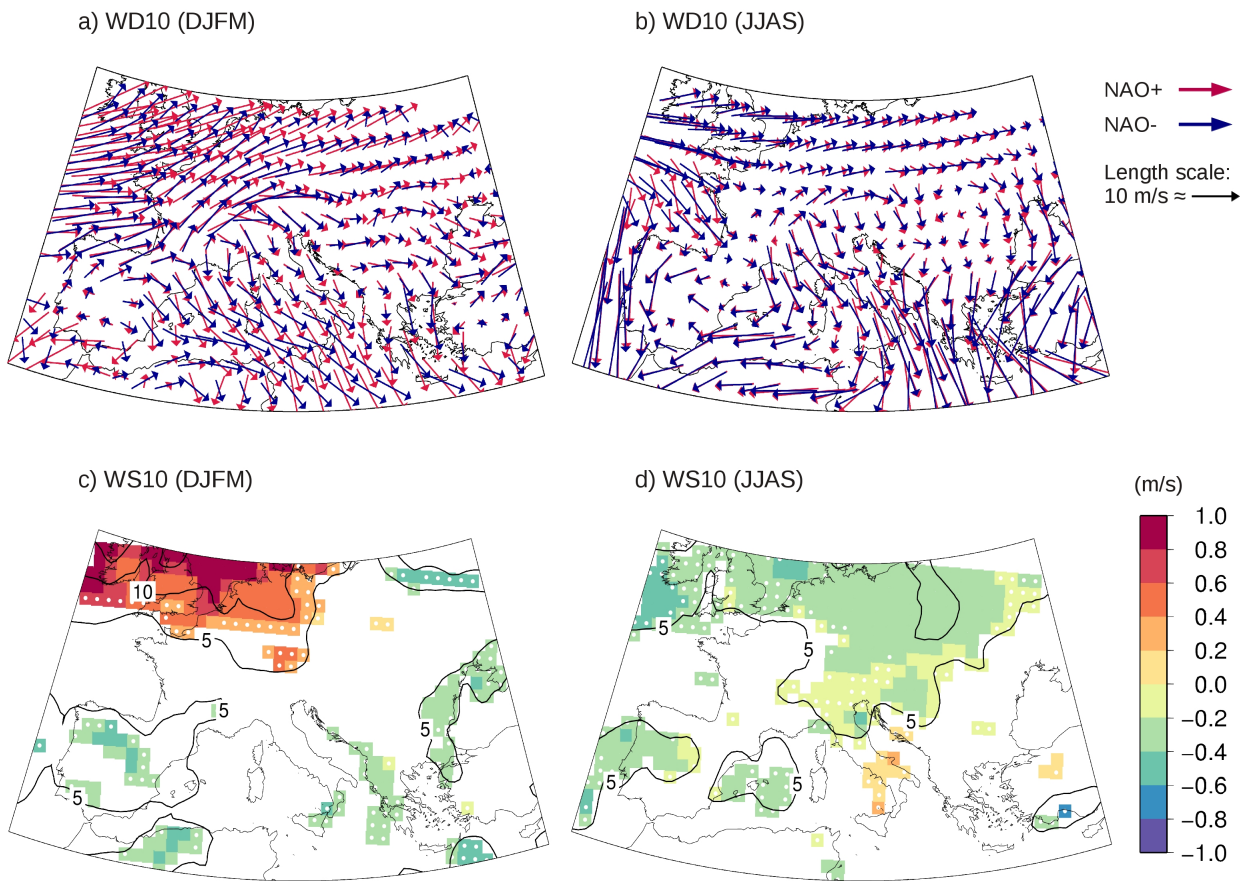


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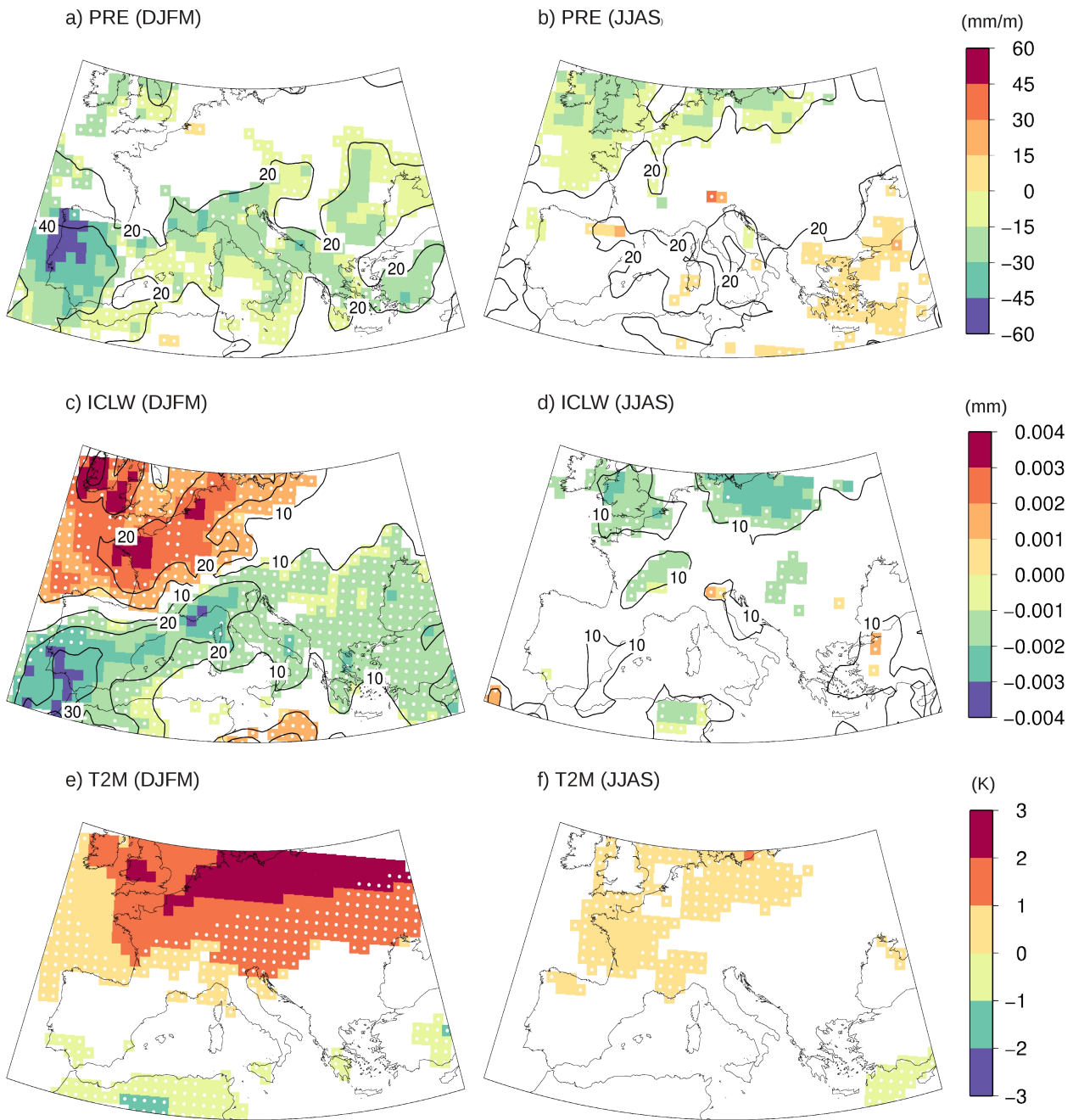


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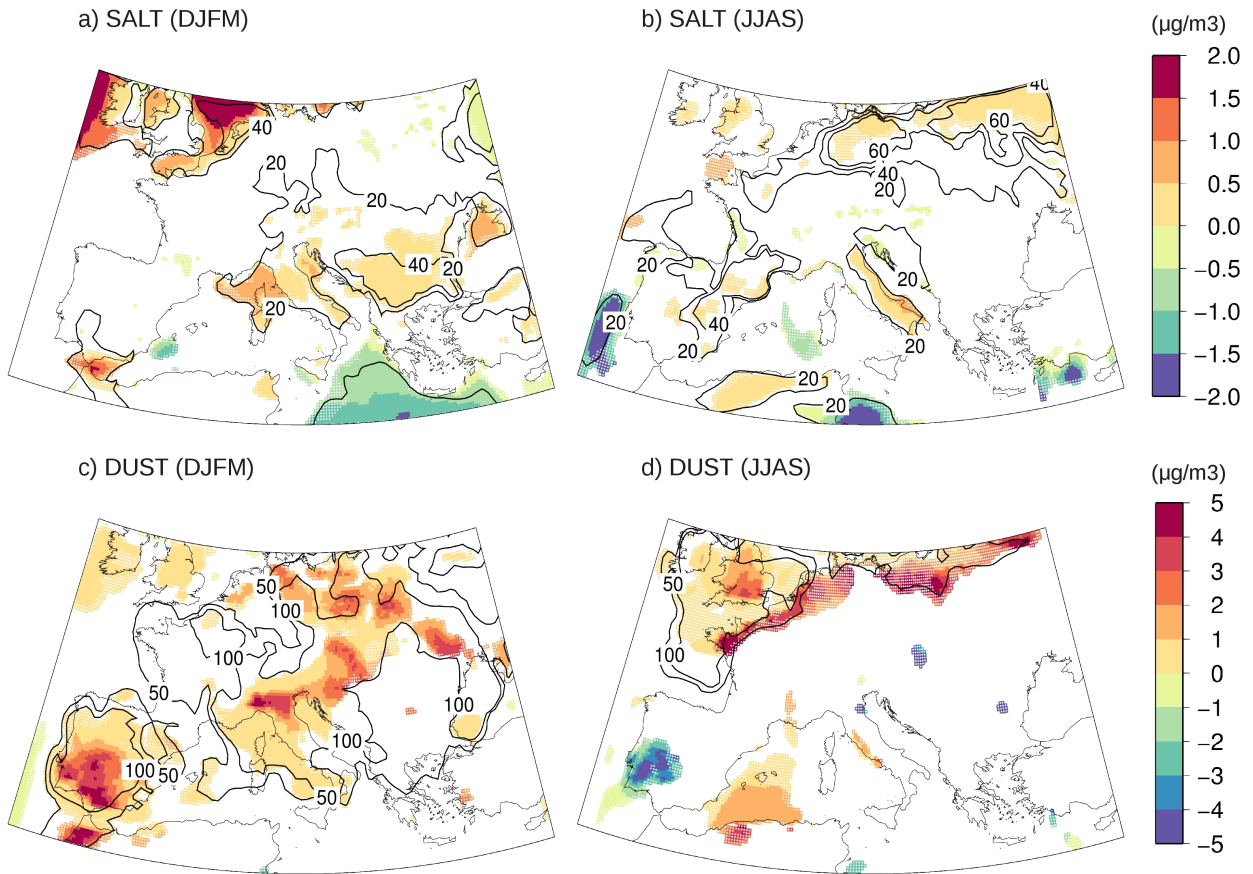




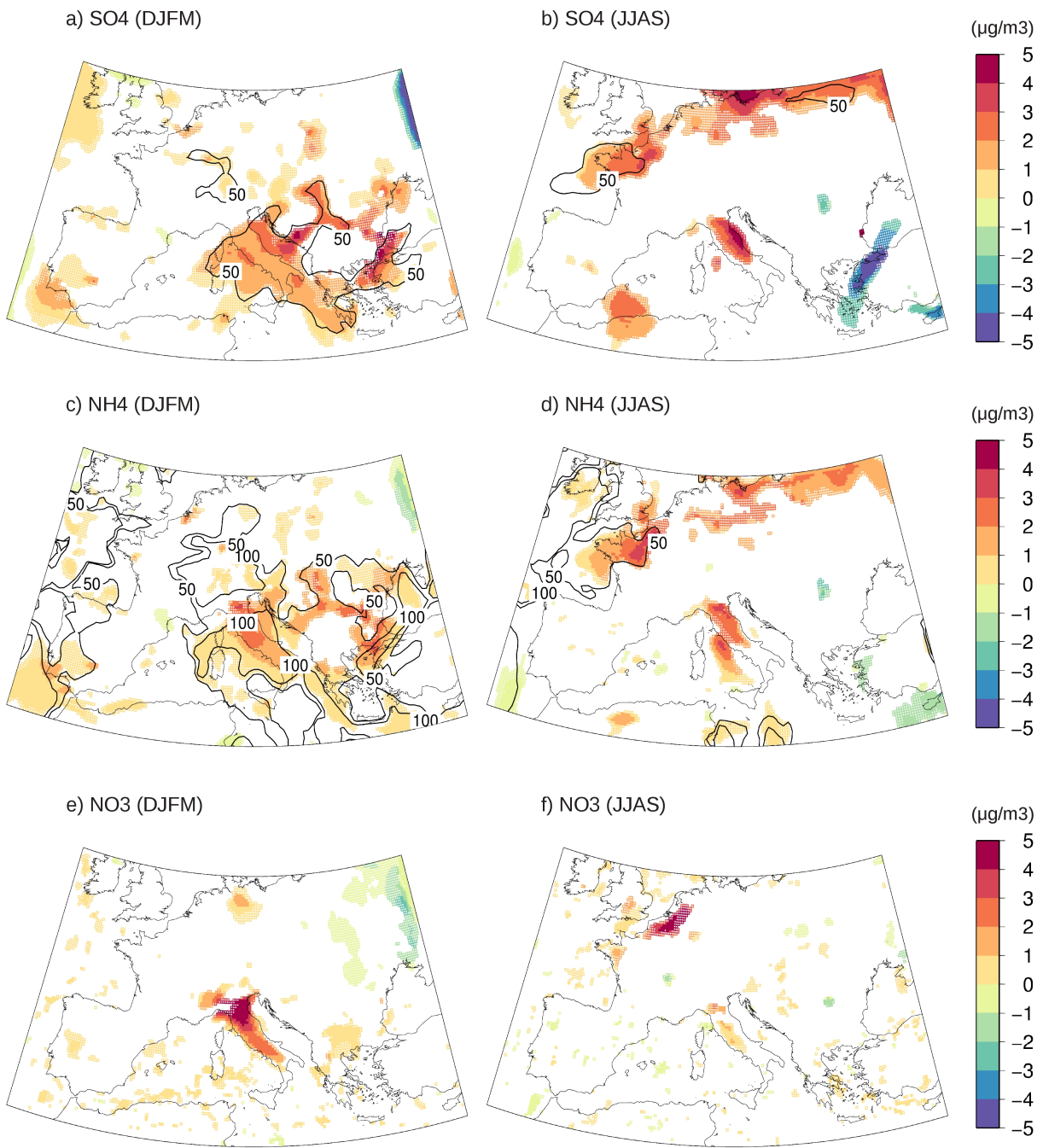
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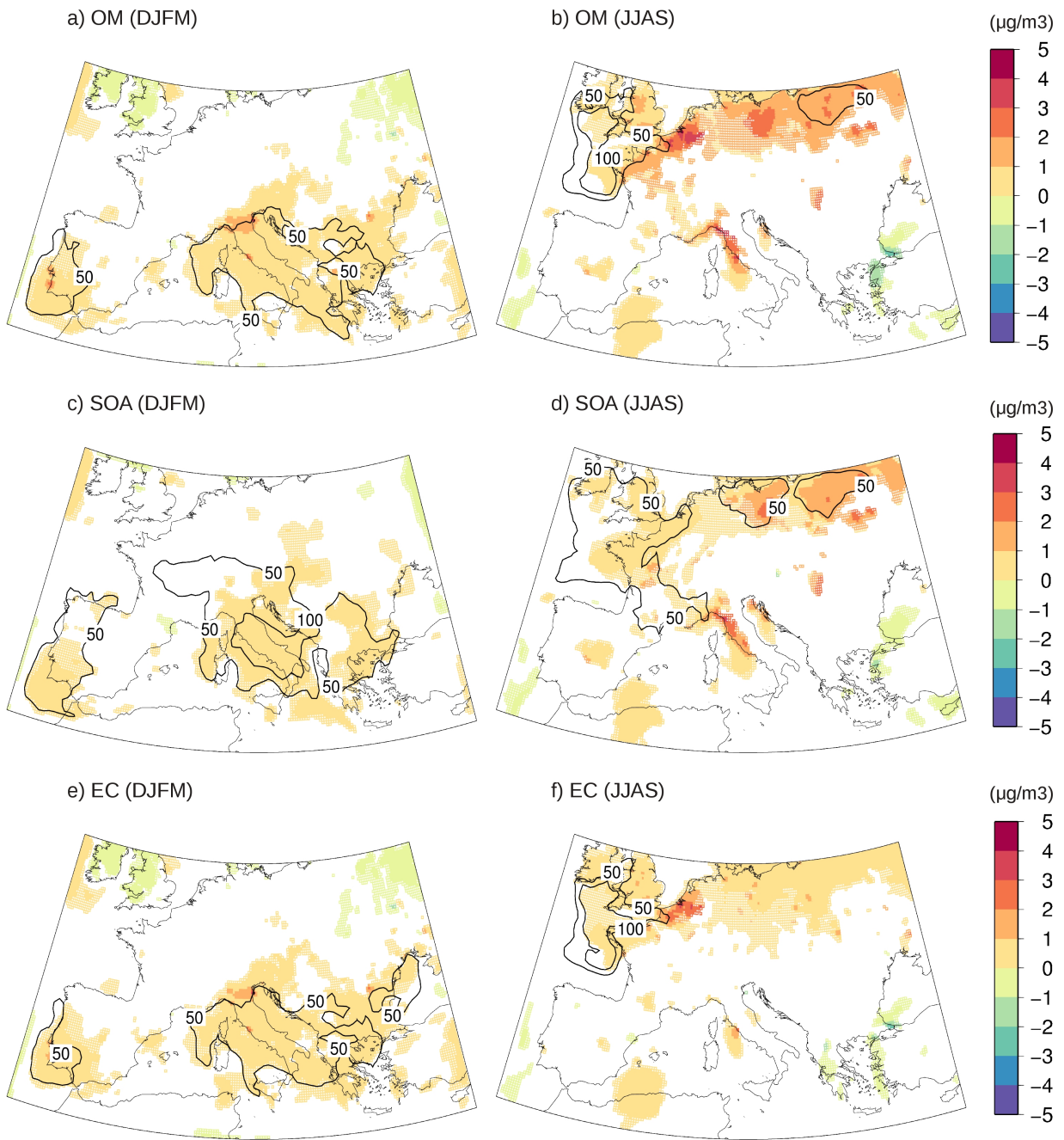
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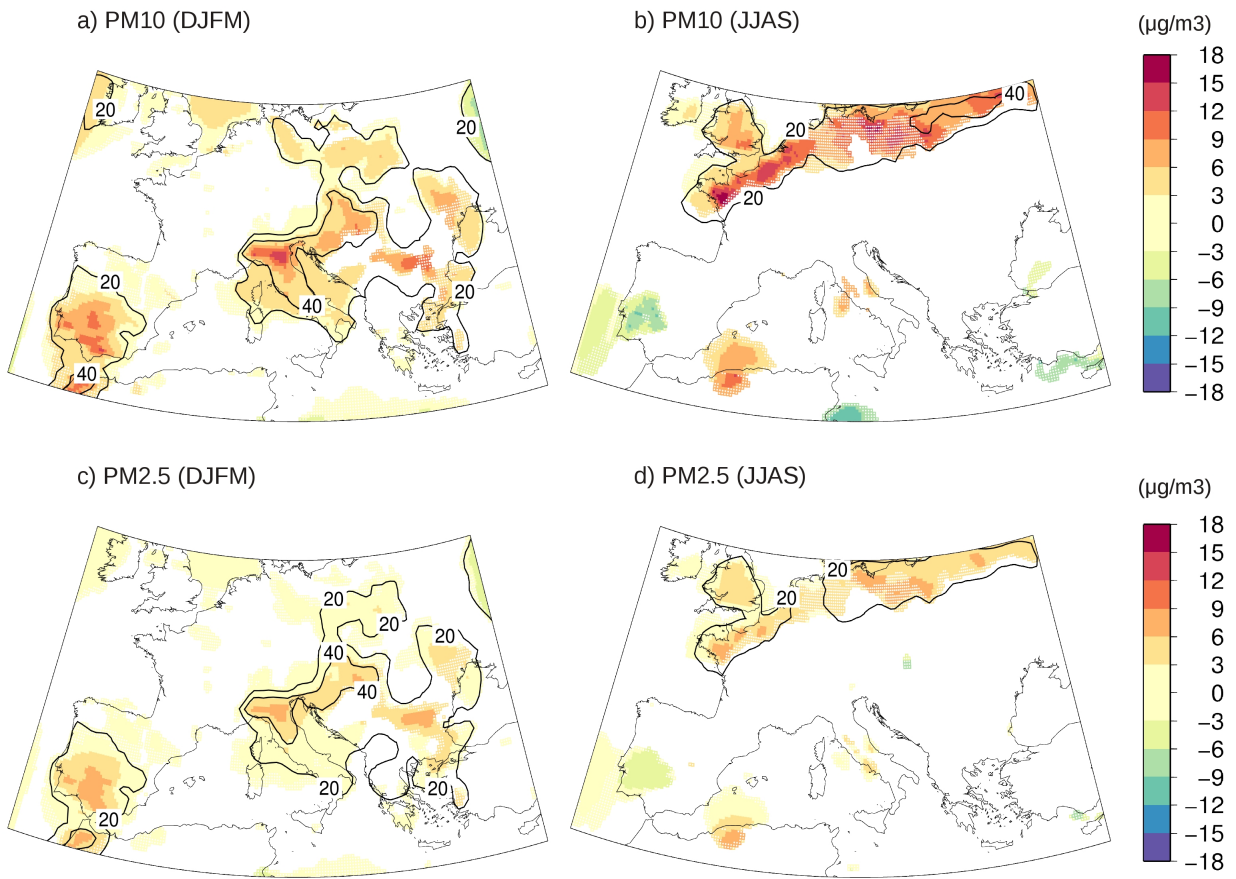
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