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The role of aerosol in altering North Atlantic atmospheric circulation in winter and air-quality feedbacks

F. S. R. Pausata[1,2,*], **M. Gaetani**[1], **G. Messori**[2], **S. Kloster**[3], **and F. J. Dentener**[1]

[1]European Commission, Joint Research Center, Institute for Environment and Sustainability, Ispra (VA), Italy
[2]Department of Meteorology, Stockholm University and Bolin Centre for Climate Research, Stockholm, Sweden
[3]Land in the Earth System, Max Planck Institute for Meteorology, Hamburg, Germany
[*]now at: Department of Meteorology, Stockholm University and Bolin Centre for Climate Research, 10691, Stockholm, Sweden

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Correspondence to: F. J. Dentener (frank.dentener@jrc.ec.europa.eu)

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Abstract

Numerical model scenarios of future climate depict a global increase in temperatures and changing precipitation patterns, driven by increasing greenhouse gas (GHG) concentrations. Aerosol concentrations also play an important role in altering Earth's radiation budget and consequently surface temperature. Here, we use the general circulation aerosol model ECHAM5-HAM, coupled to a mixed layer ocean model, to investigate the impacts of future air pollution mitigation strategies in Europe on winter atmospheric circulation over the North Atlantic. We analyze the extreme case of a maximum feasible end-of-pipe reduction of aerosols in the near future (2030), in combination with increasing GHG concentrations. Our results show a more positive North Atlantic Oscillation (NAO) mean state in the near future, together with a significant eastward shift of the southern centre of action of the sea level pressure (SLP). Moreover, we show a significantly increased blocking frequency over the western Mediterranean. By separating the aerosol and GHG impacts, our study suggests that the aerosol abatement in the near future may be the primary driver of such circulation changes. All these concomitant modifications of the atmospheric circulation over the Euro-Atlantic sector lead to more stagnant weather conditions that favor air pollutant accumulation in the Mediterranean, especially in the western sector. These changes in atmospheric circulation should be included in future air pollution mitigation assessments. Our results suggest that an evaluation of NAO changes in individual climate model simulations will allow an objective assessment of the role of changes in wintertime circulation on future air quality.

1 Introduction

Future climate scenarios indicate a global increase in temperatures and changes in the hydrological cycle, mainly driven by increasing greenhouse gas (GHG) concentrations (IPCC, 2007). However, GHGs are not the only climate factor responsible for changing the Earth's radiation budget. Aerosols play a very important role in altering climate, both directly – by scattering and absorbing solar radiation – and indirectly – by influencing cloud radiative properties (cloud albedo effect; Twomey, 1977), and cloud formation and duration (cloud lifetime effect; Albrecht, 1989). The direct effect of non-absorbing aerosols – such as sulfates – produces an overall cooling of the atmosphere, while partly absorbing aerosols – such as black carbon and organic carbon – can lead to either a cooling or a warming, depending on the aerosols' properties and underlying albedo.

Global climate models can realistically reproduce the temperature trend of the last century only when the radiative impacts of both GHGs and aerosols are included (Gleckler et al., 2008; Nazarenko and Menon, 2005; Roeckner et al., 1999; IPCC, 2013). Therefore, increasing GHG concentrations as well as changes in aerosol abundance will control future climate and atmospheric circulation variations. High aerosol concentrations can have severe impacts on human health (Lim et al., 2012; WHO, 2013). Consequently, air quality standards have been introduced in many polluted regions to regulate harmful aerosol concentrations, and the current upward trends in aerosol emissions are expected to stabilize or reverse in the future. Hence, a realistic assessment of ongoing and future climate change relies on our ability to predict trends in both GHG and aerosol emissions, the resulting concentrations and their combined effect on climate.

While most of the GHGs are long-lived climate forcers, aerosols are controlled by a combination of direct or precursor emissions, chemical reactions and large-scale atmospheric circulation, and their impacts can have short-term repercussions on climate (Shindell et al., 2012). Furthermore, atmospheric circulation changes themselves can have feedbacks on air quality. Modeling and observational analyses suggest a warming climate degrades air quality, with increasing surface O_3 and particulate matter abundance in many populated regions (Fiore et al., 2012). Kloster et al. (2009), for example, used a coupled chemistry–atmosphere general circulation model to show that climate change alone would worsen the air pollution by aerosols.

Several other studies have demonstrated that local-to-regional scale pollutant concentrations can be influenced by large-scale atmospheric circulation patterns (Eckhardt et al., 2003; Christoudias et al., 2012; Barnes and Fiore, 2013; Pausata et al., 2012, 2013), such as the North Atlantic Oscillation (NAO). Pausata et al. (2013) have shown how positive shifts in the NAO in winter over the North Atlantic penalizes cities lying in the Mediterranean area, making it necessary for these countries to enforce more stringent emission reduction measures. This is of particular importance in view of a potential shift towards positive NAO regimes under future climate conditions.

The NAO commonly refers to swings in the atmospheric pressure difference between the subpolar and subtropical North Atlantic, and is the leading mode of winter atmospheric variability in the North Atlantic. The standard NAO index (NAOI) is defined as the difference in normalized mean sea-level pressure (SLP) between the Azores (or Portugal) and Iceland (Walker, 1932), and determines climate variability from the eastern seaboard of North America to Siberia and from the Arctic to the subtropical Atlantic. The NAO featured an upward trend of over 1 standard deviation in the 1980s and 1990s compared to the 1951–1970 winter mean (data available in http://www.cgd.ucar.edu/staff/jhurrell/naointro.html). Recent multi-model predictions confirm previous finding, reported in AR4 (e.g., Kuzmina, 2005; Stephenson et al., 2006), of a positive trend in future winter NAO (Gillett and Fyfe, 2013; Karpechko, 2010). However, there are substantial variations between NAO projections from different climate models. For example, Fischer-Bruns et al. (2008) have employed an atmosphere–ocean coupled model (ECHAM4-OPYC3) and used the Empirical Orthogonal Function (EOF) analysis to investigate potential future trends in the NAO. The study found no detectable shift in the leading mode of atmospheric variability under global warming scenarios. On the other hand, Müller and Roeckner (2008) have found a strong positive trend in the NAO in the ECHAM5/MPI-OM simulations. As a consequence of such uncertainties, the IPCC AR5, has expressed only medium confidence in near-term projections of NAO changes (IPCC, 2013).

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Recently, atmospheric variability in the North Atlantic and the NAO pattern have also been linked to Rossby wave-breaking in the upper troposphere and to atmospheric blocking (e.g., Croci-Maspoli et al., 2007; Woollings et al., 2008). The term atmospheric blocking is broadly used to describe situations in which the prevailing westerly flow is blocked, or distorted by a persistent, quasi-stationary anticyclone (e.g., Rex, 1950; Berrisford et al., 2007). However, the exact definition varies among studies. For example, Pelly and Hoskins (2003) pioneered the use of potential vorticity (PV) as an indicator for blocking, linking blocking occurrences to the meridional potential temperature gradient on a PV surface. In this framework, atmospheric blocking is therefore associated with Rossby wave breaking. For instance, it has been shown that different blocking patterns correspond to significantly different large-scale atmospheric circulations over the North Atlantic basin (Rex, 1950). Blocking situations are often responsible for stagnant atmospheric conditions that lead to the accumulation of pollutants at ground levels. This increases the likelihood of exceeding PM annual and daily limit concentrations, such as those imposed by European regulations (Directive 2008/50/EC).

The aim of this paper is to disentangle the role that future aerosol and GHG concentration changes may have in altering atmospheric circulation. We focus on the extreme case that by 2030 aerosol concentrations will be reduced to the maximum feasible extension by using all presently available end-of-pipe technology. In doing so, we use the results of an aerosol–atmosphere model coupled with a mixed-layer ocean. Finally, we evaluate the impact of such atmospheric circulation changes onto PM variability. The analysis includes simulations in which only GHG concentrations, only aerosol emissions or both are changed. The anthropogenic emission scenarios used to force the model are constant; hence, the changes in variability depicted by the model will be associated with changes in atmospheric circulation only. We investigate how GHG and/or aerosol forcings act on: (i) the structure of the SLP meridional dipole over the North Atlantic in terms of strength and location of its centers of action; (ii) changes in the NAO in the near future; (iii) the spatial structure and frequency of atmospheric blocking in the North Atlantic. Finally, we also examine how (iv) future changes in atmospheric circulation can influence air quality over Europe.

This work is structured as follows: Sect. 2 gives a description of the models used, the simulation set-up and the statistical tools adopted; Sect. 3 presents the GHG and aerosol-induced changes in the magnitude and spatial pattern of the meridional SLP dipole in the North Atlantic. We discuss the changes in the NAO and atmospheric blocking over the Atlantic, and the effects of such changes on PM variability. Discussions and conclusions are presented in Sect. 4.

2 Methods

2.1 Climate model

We have analyzed the climate simulations performed by Kloster et al. (2008, 2009) using the ECHAM5-HAM aerosol–climate model. In comparison to that work, we focus on the analysis of hitherto unexplored aspects of changes in NAO-patterns. The ECHAM5-HAM modeling system includes the atmospheric general circulation model ECHAM5 (Roeckner et al., 2003) coupled to a mixed layer ocean (Roeckner et al., 1995), and the microphysical aerosol model HAM (Stier et al., 2005). ECHAM5 was run on a T63 horizontal grid (about 1.8° on a Gaussian Grid), and on 31 vertical levels from the surface up to 10 hPa. A cloud scheme with a prognostic treatment of cloud droplet and ice crystal number concentration (Lohmann et al., 2007) provided fractional cloud cover prediction from relative humidity, according to Sundquist et al. (1989). The shortwave radiation scheme included 6 bands in the visible and ultraviolet spectra (Cagnazzo et al., 2007).

The microphysical aerosol module HAM treats the aerosol size distribution, mixing state, and composition as prognostic variables. It predicts the evolution of an ensemble of interacting aerosol modes and is composed of the microphysical core M7 (Vignati, 2004); an emission module for SO_2, black and organic carbon, and mineral dust particles; a sulfur oxidation chemistry scheme using prescribed oxidant concentrations for OH, NO_2, O_3 and H_2O_2 (Feichter et al., 1996); a deposition module; and a module defining the aerosol radiative properties. The aerosol optical properties were explicitly simulated within the framework of the Mie theory and provided as input for the radiation scheme in ECHAM5. Climate-sensitive natural emissions (dimethyl sulfide, sea salt and dust) were simulated interactively.

2.2 Simulation set-up

The GHG concentrations used in the numerical simulations were derived from the IMAGE 2.2 implementation of the SRES B2 scenario (IMAGE-team, 2001). The SRES B2 storyline describes a world with intermediate population and economic growth, in which the emphasis is on local solutions to economic, social, and environmental sustainability.

The anthropogenic emissions of carbonaceous aerosols, namely black carbon (BC) and organic carbon (OC), and sulfur dioxide (SO_2), the main precursor of sulfate aerosols, are extracted from an aerosol emission inventory developed by the International Institute for Applied System Analysis (IIASA). In this work, a Maximum Feasible Reduction (MFR) air pollutant emission scenario was explored for the year 2030 (Cofala et al., 2007). MFR assumes the full implementation of the most advanced available technologies for aerosol emissions abatement. It is built using projections of human activity levels (industrial production, fuel consumption, livestock numbers, crop farming, waste treatment and disposal) based on current national perspectives on the economic and energy development up to the year 2030. In regions where data were not available, the economic and energy future trends estimated in the IPCC SRES B2 MESSAGE scenario (IPCC, 2000; Riahi and Roehrl, 2000) were considered. Biomass burning emissions, both anthropogenic and natural, were assumed to stay constant at 2000 levels. Changes in land use were not taken into account.

In the present study the modifications of future North Atlantic atmospheric circulation are assessed by analyzing the differences between near future (year 2030) and present-day (year 2000) conditions reproduced in climate equilibrium simulations. A 60 yr control simulation was performed with GHG concentrations, aerosol and aerosol precursor emissions of the year 2000, and three 30 yr sensitivity experiments were performed, using three different combinations of GHG concentrations and aerosol emissions scenarios for the year 2000 and 2030. All simulation used a spin-up of 30 years, not used in the analysis.

- 2030GHG experiment: year 2030 GHG concentrations were assumed, and aerosol emissions were kept at the 2000 level;
- 2030AER experiment: GHG concentrations were kept at the 2000 level, and MFR was assumed for aerosol emissions;
- 2030MFR experiment: year 2030 GHG concentrations and MFR were assumed.

The 2030GHG and 2030AER experiments, in which aerosol emissions and GHG concentrations remained at the 2000 level, were performed to disentangle the effects of GHG concentrations and aerosols emissions, separately. The experimental setups are summarized in Table 1.

2.3 Statistical analysis methods

We evaluate three aspects of changing circulation patterns: (1) the SLP spatial structure (shift of centres of action), (2) the leading mode of atmospheric variability (NAO), and (3) the blocking frequency.

To investigate the impact of aerosol and GHG concentration changes on SLP spatial structure, we define the SLP centres of action for the winter season (January, February and December, DJF) by creating SLP coherence maps (Pausata et al., 2009). The coherence index value ($0 \leq CI \leq 1$) at each grid-point is the absolute value of the area-averaged correlation between the winter SLP time-series at that point and over the rest of the North Atlantic basin (20–85° N; 90° W–40° E). Higher values indicate that the SLP variability at that location has a higher *coherence* with variability throughout the

North Atlantic, either in-phase or anti-phase. The northern and southern SLP centres of action are identified as CI maxima over the North (north of 55° N) and subtropical Atlantic (south of 55° N), respectively. This method allows determining the spatial distribution and shifts of the SLP centers of action due to aerosol and GHG concentration changes, both in combination and separately.

In order to verify that the computed geographical shifts in the centers of action are outside the normal range of interannual variability, we use a statistical bootstrap approach to produce a set of 30 CI maps.

For each simulation, we randomly select 20-year subsamples, and then we identify the northern and southern SLP centres of action for each map. Consequently, we apply the Student's *t* test to determine whether the shift in the centers of actions between the 2000 control simulation and the sensitivity studies is significant.

Winter changes in the leading mode of atmospheric variability are investigated by using the monthly NAO Index (NAOI), defined as the difference in the normalized SLP anomalies between Ponte Delgada, Azores, and Stykkisholmur/Reykjavik, Iceland. The NAOI allows to look for shifts in the North Atlantic atmospheric circulation associated with future climate change (Hurrell, 1995).

The analysis of blocking frequency over the North Atlantic basin is performed as follows. An atmospheric blocking describes a situation in which the prevailing westerly flow is distorted or blocked by a persistent, quasi-stationary anticyclone. In order to define atmospheric blocking, the present paper utilizes a bi-dimensional index that identifies reversals in the meridional gradient of 500 hPa geopotential height (Davini and Cagnazzo, 2013; Davini et al., 2012a; Tibaldi and Molteni, 1990). For every model gridbox with coordinates (latitude = φ, longitude = λ), the following two quantities are defined:

$$\Delta_N(\varphi,\lambda) = \frac{Z_{500}(\varphi,\lambda) - Z_{500}(\varphi - 15^\circ,\lambda)}{15^\circ},$$

$$\Delta_S(\varphi,\lambda) = \frac{Z_{500}(\varphi + 15^\circ,\lambda) - Z_{500}(\varphi,\lambda)}{15^\circ},$$

over the domain where $30^\circ\,\mathrm{N} < \varphi < 72.5^\circ\,\mathrm{N}$, $180^\circ\,\mathrm{W} < \lambda \leq 180^\circ\,\mathrm{E}$. In order for a gridbox to be flagged as an atmospheric blocking, the following must hold:

$$\Delta_\mathrm{N} > 0; \qquad \Delta_\mathrm{S} < -10\,\mathrm{m}\,(^\circ\mathrm{latitude})^{-1}$$

A number of additional constraints are also enforced. Firstly, a model gridbox cannot be considered to be "blocked" in isolation. A gridbox is considered to be blocked only if it is part of a continuous cluster of blocked gridboxes spanning at least 15° longitude. The blocking must also last for at least five consecutive days. Persistence is calculated based on a 5° latitude by 10° longitude box. Therefore, a grid-box is considered blocked only if a blocking event is detected for 5 consecutive days in at least one point within an area of 5° latitude by 10° longitude centred around the gridbox itself. Note that this persistence requirement is imposed only on the gridboxes satisfying the longitudinal extent condition.

We analyze monthly means of model outputs, and all differences discussed in this study are investigated using the Student's *t* test. Significance is reported at the 95 % confidence level.

3 Results

The results presented here describe the effects of GHG and aerosol concentrations on the mean state and variability of the North Atlantic atmospheric circulation. The results are presented in three sections. In the first section, changes in the spatial structure of the SLP and its variability are investigated. In the second section, we extend the analysis to changes in the blocking frequency. Finally, in the third section, we quantify the impacts of such changes on PM variability.

3.1 Changes in SLP centers of actions and their variability

The 2030MFR and 2030AER simulations shows a north-eastward shift of the southern pole of the SLP centre of action compared to the 2000 control simulation (Fig. 1).

The area of highest SLP coherence in the 2000 simulation is located in the central-western part of the sub-tropical North Atlantic, whereas in the 2030MFR simulation it is shifted off the coast of northern Morocco. The northern pole instead remains located in central western Greenland. However, in the 2030MFR/2030AER simulation, a second maximum of the CI develops in the Norwegian Sea, and the areas with the CI maxima are broader. Secondary CI maxima develop at both high and low latitudes compared to the 2000 simulation (Fig. 1).

Both sensitivity simulations (2030GHG and 2030AER) show a tendency towards a north-eastward significant shift (see Sect. 2.3) of the southern center of action as well as broader areas of CI maxima compared to the 2000 simulation. Both these features are more pronounced in the 2030AER than in the 2030GHG simulation, in particular the shift towards the Mediterranean Sea of the CI southern maximum.

With regard to the SLP variability, the 2030MFR simulation shows a significant positive shift of the NAO mean state of 0.46 compared to the 2000 control period (Fig. 2). The probability of having an NAOI greater than +1 increases from 30 % to 40 % (Fig. 2). Both GHG increase (2030GHG) and aerosol reduction (2030AER) play a similar role in changing the NAO mean state and frequency of higher – in absolute terms – phases of the NAO relative to the control simulation (Fig. 2). Note, however, that only the change shown by the 2030MFR simulation is statistically significant.

In conclusion, whereas both GHG and aerosol changes have similar impacts on the NAOI frequency shift, the aerosol reduction alone plays the largest role in shifting the southern center of action of atmospheric circulation towards the Mediterranean.

3.2 Changes in blocking frequency

Blocking events can have a large impact on weather patterns and sometimes lead to the occurrence of extreme events (e.g., Yiou and Nogaj, 2004); hence, it is important to quantify the variability and possible changes in the preferred location of blocking occurrences.

The 2000 simulation shows a blocking frequency that peaks in the south over the sub-tropical North Atlantic (low-latitude blocking, LLB) and in the north over Greenland (Greenland blocking, GB), as shown in Fig. 3a. The LLB events are linked to a northward displacement of the subtropical high-pressure system. The GB events are characterized by long durations (on the order of 9 days), diverting the main flow southward (Davini et al., 2012a). The simulated 2000 blocking climatology is slightly different from the patterns seen in re-analysis data, which have a higher activity over the Nordic seas, but nevertheless shows a strong resemblance to the observed climatology (cf. Fig. 3a with Fig. 1 in Davini et al., 2012a).

The 2030MFR simulation shows a significant increase in LLB frequency, corresponding to a more invasive subtropical anticyclone (high-pressure system) over southern and central Europe in winter. On the other hand, high latitude blocking decreases (Fig. 3b). The reduction of high-latitude blocking agrees with the reduction in negative NAO phases shown in Sect. 3.1. This finding is also in agreement with Woollings et al. (2008) who showed that such high-latitude blocking events over Greenland are strongly anti-correlated with the NAO index. Furthermore, changes in the GB position (Wang and Magnusdottir, 2012) and frequency (Davini et al., 2012b) have been shown to influence not only the NAO index, but also its pattern.

Both the 2030GHG and 2030AER simulations show an increase in the frequency of LLB events over the mid-latitude North Atlantic and a decrease in the frequency of high-latitude blocking. The aerosol concentration reduction seems to be the main driver of the increase in LLB events over the Mediterranean (Fig. 3d).

This result strengthens the role of aerosols in affecting atmospheric dynamics in the North Atlantic, suggesting that they drive both (a) an eastward shift of the southern center of action of SLP and (b) an increased tendency of the sub-tropical anticyclone to expand towards the Mediterranean Sea.

3.3 Impacts on air-quality

The changes in atmospheric circulation shown in the above sections can affect the variability of PM over Europe. For example, an eastward shift of the SLP southern center of action together with an increased frequency of blocking events in the Mediterranean may lead to a higher frequency of stagnant weather conditions in southwestern Europe, thus worsening air quality. Hence, even though there will be an overall improvement in air quality conditions associated with an abatement of PM emissions, additional PM emission reduction measures may be necessary for those countries and cities lying in the Mediterranean area to reach similar levels of aerosol pollution. This hypothesis has already been suggested by Pausata et al. (2013) on the basis of an NAO analysis using the same model driven by ERA-40 re-analysis data. In this work, we further test the hypothesis by using climate simulations for the near future. To do so, we calculate the relative anomaly distributions of PM concentrations for four regions, to encompass the different areas of influence of the NAO in Europe:

- Western Mediterranean (WM): 34–43° N/0–10° W;
- Eastern Mediterranean (EM): 34–43° N/10° E–40° W
- Central Europe (CE): 44–53° N/0–15° E;
- Eastern Europe (EE): 46–60° N/20–40° E.

In the PM we have considered only the aerosol components included in ECHAM5-HAM that have a predominantly anthropogenic signature – namely black and organic carbon, and sulfates, thus disregarding aerosols of natural origin (e.g. sea-salt, SOA, mineral dust). Thus, the PM in this paper represents mostly $PM_{2.5}$, and is likely a lower bound on the “real” PM concentrations (for an evaluation of correspondence of model and measured $PM_{2.5}/PM_{10}$ see Supplement in Pausata et al., 2013).

Here, we analyze the skewness of the monthly PM relative anomaly distribution for the winter season. The skewness is the distribution’s third standardized moment, and is a measure of the asymmetry of the distribution. Positive skewness values typically indicate that the right side tail of the distribution becomes longer than the left side, and vice-versa for a negative value. PM relative anomaly distributions for all experiments and for all four regions show positive skewness values, meaning that extreme positive PM anomalies are more likely than negative ones (Fig. 4 and Table 2). Significance in the skewness differences is assessed by using a Kolmogorov–Smirnov (KS) test at 95 % confidence level. The KS test is a non-parametric tool that uses no assumptions on the shape of the data distribution. An "artificial" variability is introduced in the skewness values in each simulation through a bootstrap method. For each experiment, we calculate the skewness values of 100 distributions, generated by randomly selecting 100 different subsamples of half of the total number of years of the given experiment. The significance level is then identified based on this sample.

Our results show that in the 2030 experiments, the simulated PM distribution around the mean value changes significantly in all regions considered due to the altered atmospheric circulation (Fig. 4 and Table 2).

In the Western Mediterranean (WM), the area where the model simulates an increase in blocking frequency in the future (Fig. 3), the skewness experiences a remarkable increase from 0.26 in the 2000 to 1.02/1.05 in the MFR/AER simulations. This change is mainly led by the aerosol reduction, whereas the GHGs only drive a small contribution (Table 2). The large change in skewness in the MFR simulation is accompanied by a corresponding shift in the upper and lower percentiles of the distribution. The 5th and 95th percentiles rise relative to 2000 by 8 % and 4 % respectively, indicating a transition towards more positive PM anomalies. On the other hand, the median of the distribution decreases by almost 10 % (Table 3).

The Eastern Mediterranean (EM) also experiences an increased skewness in the MFR simulation relative to 2000. However, the changes are smaller compared to the WM, possibly because of the greater distance from the southern SLP center of action, which, in the MFR experiment, is located off the coast of the Iberian Peninsula.

On the other hand, Central (CE) and Eastern Europe (EE) show a decreased skewness in the MFR compared to the 2000 simulation. CE displays a shift from a skewness of 1.44 in the 2000 to 0.66 in the MFR simulation and EE from 1.70 to 1.18. Furthermore, CE also shows an increment in the number of negative extremes, with a 14 % decrease in the 5th percentile. However, CE also experiences an increase in positive extremes with a +7 % shift of the 95th percentile in the MFR simulation compared to the 2000 experiment (Table 3).

To conclude, the regions that will be more affected by a future NAO shift are the Western Mediterranean and Central Europe, both with increased high PM concentration episodes, but the latter with also a strong increment in low PM values relative to 2000.

The implications of these results for air quality policy are discussed in the following section.

4 Discussions and conclusions

The present study analyzes future scenarios of atmospheric circulation over the North Atlantic and possible impacts on air quality over Europe. The chemistry–atmosphere ECHAM5-HAM model, coupled to a mixed layer ocean, shows a change towards more positive NAO phases, together with an eastward shift of the southern SLP centre of action. This shift leads to an increased frequency of blocking events over the Western Mediterranean. Our results highlight how the decreased aerosol and aerosol precursor emissions, along with GHGs, are responsible for changes in radiative forcing that feedback onto the atmospheric circulation and alter the NAO mean state. These changes in atmospheric circulation in turn feedback significantly on air quality, leading to an increase in extreme pollution events over the Western Mediterranean.

Future shifts in the NAO phase have been discussed by several previous modeling studies (e.g., Gillett and Fyfe, 2013; Karpechko, 2010; Stephenson et al., 2006;

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Kuzmina, 2005; Hu and Wu, 2004); however, the driving mechanisms behind this shift are still debated.

Hori et al. (2007) have shown that NAO variability does not change substantially in the SRES-A1B and 20th century scenarios, and conclude that the trend in the NAO index is the result of an anthropogenic trend in the basic mean state, rather than being due to changes in NAO variability. Our results support Hori et al.'s (2007) findings by showing that anthropogenic changes in GHG and aerosols lead to a change in the NAO's mean state rather than its variability (Fig. 2).

The positive NAO shift comes along with a shift of the SLP centres of action. Hilmer and Jung (2000) have found an eastward shift in the SLP pattern associated with the interannual variability of the NAO from 1958–1977 to 1978–1997. Peterson et al. (2003) have suggested that this shift is simply a consequence of the trend towards a more positive NAO index in the last two decades of the 20th century. Hu and Wu (2004), using both data and a coupled general circulation model, have shown that a shift of both SLP centers of action took place in the second half of the last century, and will likely continue in the future. Our study has confirmed that this shift also occurs under a global warming scenario. However, while in our simulations the southern center undergoes a remarkable eastward shift, the northern one is fairly stable around southern Greenland – as demonstrated using the coherence index approach (Fig. 1). Nevertheless, the CI maps do show that in the 2030 simulations a secondary northern maximum – not present in the 2000 experiment – appears in the Norwegian Sea (Fig. 1). Furthermore, our simulations highlight how the abatement of the aerosol load is the leading factor for the eastward shift of the centres of action.

Our study also finds an increased blocking frequency over the Western Mediterranean. Such increase, together with an eastward displacement of the southern SLP center of action and a positive shift of the NAO mean state, leads to more frequent stagnant weather conditions that favor pollutant accumulation in the Mediterranean. This change in frequency of pollution events has also been described by Kloster et al. (2009), who showed that aerosol abundance is dependent on the climate state, as also highlighted in other modeling studies (e.g., Feichter et al., 2004; and overview in IPCC, 2013). Kloster et al. (2009) further found that aerosol burdens increase in the area due to less precipitation and reduced wet-deposition. Hence, they suggest that climate change alone would worsen air pollution by aerosols. Here we show that these findings are consistent with a straightforward NAO-behavior analysis, and that indeed a positive shift in future NAO may lead to more positive extreme pollution events over specific areas, such as in the western Mediterranean countries. This finding also supports the hypothesis of Pausata et al. (2013) that climate change will lead to more extreme pollution events over the Western Mediterranean, forcing Southern European countries to implement more stringent abatement measures to counteract adverse changes in PM variability. However, our study also highlights that the increase in the number of high PM episodes in the Western Mediterranean is partially counterbalanced by a lower median and a narrowing of the PM frequency distribution around the median itself (Fig. 4 and Table 3).

Current European legislation considers PM air quality thresholds of 25 $\mu g\,m^{-3}$ (annual average) for $PM_{2.5}$, and 50 $\mu g\,m^{-3}$ for PM_{10} (24 h, not to be exceeded for more than 35 days per year). Currently, between 20–31 % and 22–33 % of the urban population in Europe is exposed to $PM_{2.5}$ and PM_{10} levels above these thresholds (EEA, 2013). However, more stringent standards are currently in place in the USA (annual $PM_{2.5}$: 12 $\mu g\,m^{-3}$), or recommended by the World Health Organization (annual $PM_{2.5}/PM_{10}$: 10/20 $\mu g\,m^{-3}$), and may be adopted in Europe as well at some point in the future. Considering the more stringent WHO guidelines currently between 91–96 % ($PM_{2.5}$) and 85–88 % (PM_{10}) of the urban population is exposed to values above the thresholds (http://ec.europa.eu/environment/air/quality/standards.htm). Depending on threshold levels set by future EU air quality legislation, it is not a-priori clear how changes in PM frequency distributions will affect exceedance of these thresholds, and what levels of emission reductions are appropriate to reach these air quality objectives.

Unfortunately, our coarse resolution global model results allow only a qualitative assessment of the impact on air quality exceedance of future air pollution emissions and climate change. Therefore, we envision the need for more in-depth studies to further quantify the significance of our findings with respect to the relationship between future changes in atmospheric circulation and air-quality related issues. These studies should make use of both high vertically resolved coupled atmosphere–ocean general circulation models and regional air-quality models. The former models are needed to better quantify anthropogenic-induced changes in atmospheric circulation and their impacts on air quality, given the strong coupling between stratospheric and tropospheric circulation (e.g., Hoerling et al., 2001; Scaife et al., 2005; Omrani et al., 2013). The latter models can better constrain the effects of the altered atmospheric circulation on air-quality at regional scales. The aerosol 2030 simulations used in this study assumed MFR and the extent to which this will actually happen depends on the effectiveness of policies. Nevertheless, 60–70 % of the reduction assumed by the MFR scenario is not unrealistic and hence some of the feedbacks seen in this study are likely to be witnessed in the real world. Most of the EU estimates of benefits related to pollution reduction (e.g., a decrease in the number of premature deaths) are determined without taking into account the potential effect of a future atmospheric circulation change. Therefore, more quantitative studies in which high-resolution regional air quality models are coupled to global ocean–atmosphere–chemistry climate models are necessary to assess the climate feedback on aerosol abatements. Understanding and characterizing changes in the NAO in global models, thus providing meteorological and chemical boundary conditions to regional air quality models, will also allow a better analysis of exceedance rates of air quality standards associated with inter-annual variability of circulation patterns.

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Table 1. ECHAM5-HAM experiment design and number of years simulated for each experiment. The original denomination used by Kloster et al. (2009) is shown in the last column.

Experiment	GHG	Aerosols emissions	Years of simulation	Original names
2000	2000	2000	60	CONTROL
2030GHG	2030	2000	30	GHG
2030AER	2000	2030	30	AE
2030MFR	2030	2030	30	GHG + AE

Table 2. Skewness values for the four selected regions and for each experiment. All changes are significant at 95 % confidence level, except AER-MFR in Western Mediterranean.

	Western Mediterranean	Eastern Mediterranean	Central Europe	Eastern Europe
CNTRL	0.26	0.83	1.44	1.70
MFR	1.02	0.95	0.66	1.18
GHG	0.48	1.26	1.18	1.08
AER	1.05	1.17	0.94	1.03

Table 3. Percentiles for WM and CE regions in the 2000 experiment and their relative changes (in %) for the MFR simulation compared to 2000 values.

Region	Experiment	Percentile				
		5th	25th	50th	75th	95th
Western Mediterranean	2000 MFR	0.49 +8 %	0.73 +1 %	1.01 −9 %	1.25 −7 %	1.57 +4 %
Central Europe	2000 MFR	0.65 −14 %	0.80 −3 %	0.94 +2 %	1.13 +2 %	1.48 +7 %

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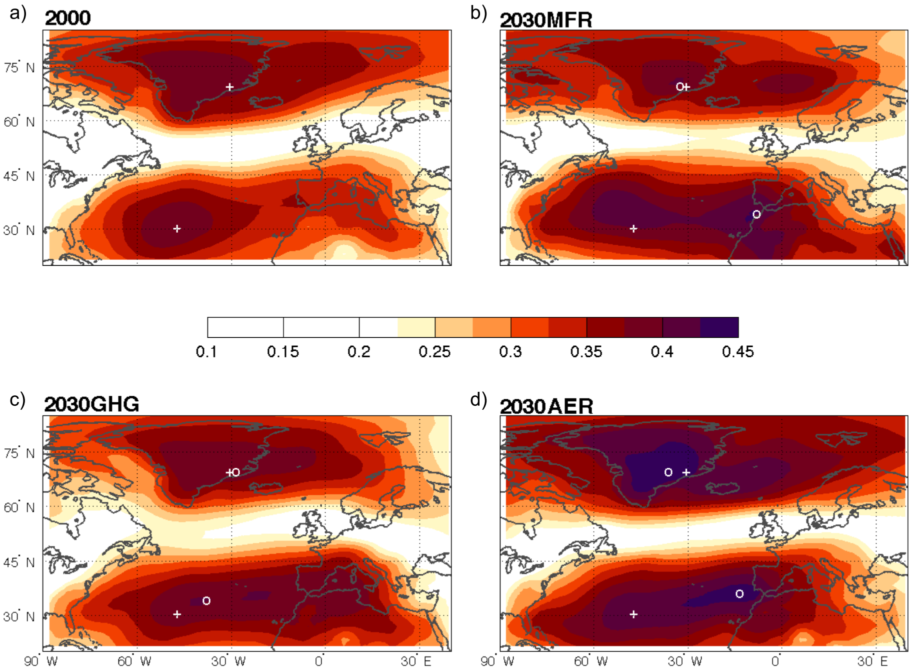

Figure 1. Coherence index maps of the North Atlantic sector for the 2000 **(a)** and 2030MFR **(b)** simulations and the two sensitivity studies (**c** and **d**) in winter (DJF). The SLP centres of action for the control run and for the 2030 simulations are shown by white crosses and white circles respectively.

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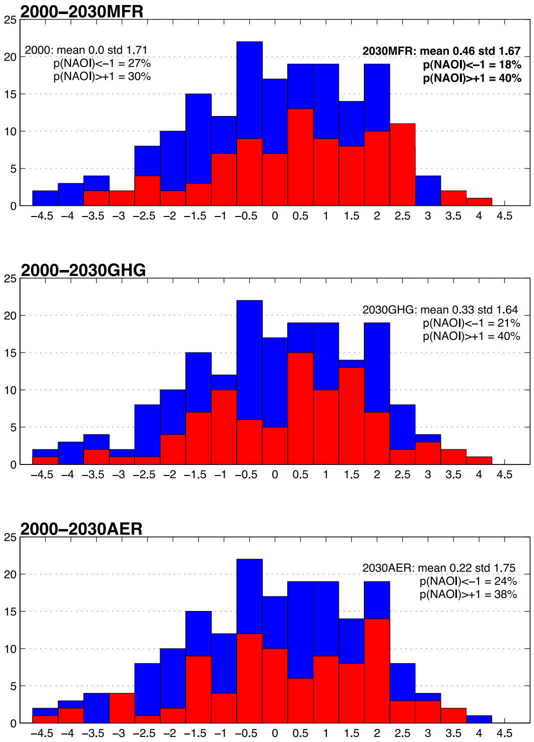

Figure 2. Frequency distributions (number of occurrences: y-axis) of the winter (DJF) NAOI (x-axis) for the 2000 control simulation (blue, all panels, 60 years), 2030MFR (red, upper panel, 30 years), 2030GHG (red, central panel, 30 years) and 2030AER (red, lower panel, 30 years). Numbers show the NAOI mean value, the standard deviation (std) and the probability of having a NAOI greater than +1 ($p(\text{NAOI}) > +1$) or smaller than −1 ($p(\text{NAOI}) < -1$). In bold values of the simulations having a NAOI mean significantly different from 2000 control mean at 95 % confidence level. The 2000s mean NAO is by definition equal to 0.

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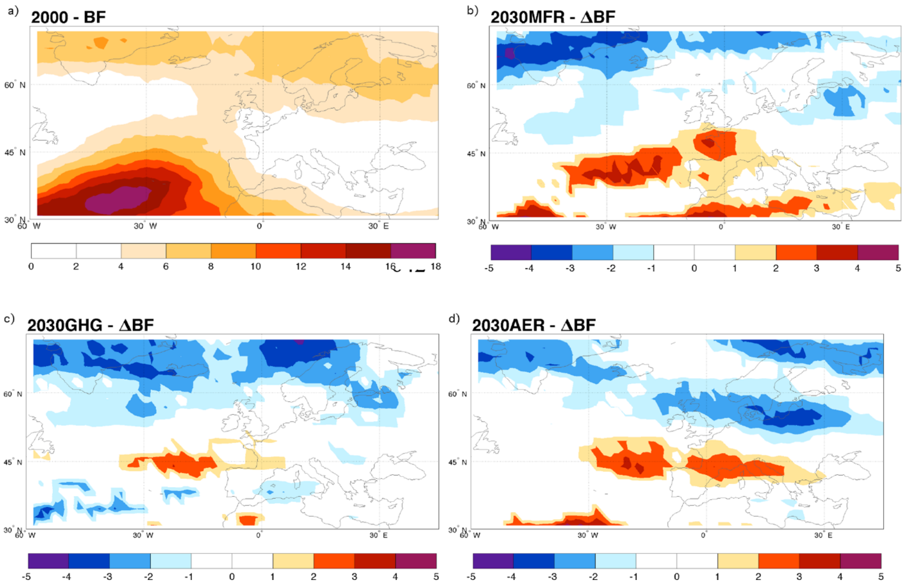

Figure 3. Blocking frequency (in %) over the Atlantic sector for the 2000 simulation **(a)**; changes in blocking frequency compared to the 2000 simulation for 2030MFR **(b)**, 2030GHG **(c)** and 2030AER **(d)** simulations in winter (DJF). Only areas in which the difference between the 2000 control and the sensitivity simulation is significant at 95 % confidence level are shaded.

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The role of aerosol in altering North Atlantic atmospheric circulation

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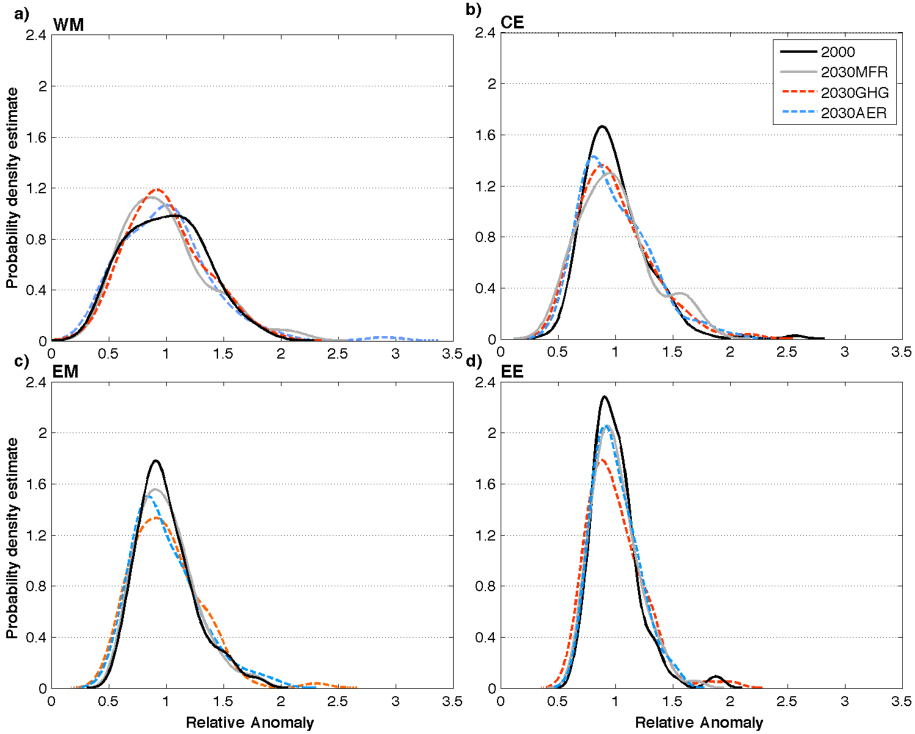

Figure 4. Probability density estimate (PDE) of PM relative anomalies for each region (Western and Eastern Mediterranean, Central and Eastern Europe) and for each experiment. Relative anomalies are computed as the ratio between winter (DJF) monthly timeseries and the winter (DJF) climatology of each experiment and region. The probability density estimates are based on a normal kernel function, which provide non-parametric PDE for random variables (Rosenblatt, 1956). The probability of a given relative anomaly to happen is obtained by the integral of the PDE in dx.