We would like to thank Reviewer #1 for the insightful comments, which we believe, can substantially improve our manuscript. We provide this rather detailed preliminary reply to clarify some of the aspects that did not fully convince the Reviewer. We feel that we are able to address all of the Reviewer's major concerns, and we think this reply may provide useful information for other reviewers as well, who may have had similar concerns. This reply is not meant to be the full reply to Reviewer #1, which will be submitted after receiving reviewers' comments.

Reviewer #1's major concern was the lack of a clear connection between the different analyses presented in the paper: (1) the impact of aerosols on the NAO, the coherence index (CI) and the blocking frequency over the Atlantic basin; (2) the mutual relationships among these atmospheric indices; and (3) the mechanisms connecting changes in atmospheric circulation to changes in the local pollutant distribution over Europe. In this reply we would like to provide some clarifications about the relationship between the CI analysis, the NAOI and the blocking events (Reviewer's major comments 2 and 3) and their relationship with air pollution (Reviewer's major comment 1). We also show a number of new plots, for possible inclusion in the revised version of the manuscript.

Coherence index – NAO relationship

The CI analysis of the SLP field indicates the areas that best correlate with the SLP variability over a given basin. In other words, the maxima in the CI represent the points that capture SLP variability in a given domain. On the other hand, the NAOI is a measure of the wintertime SLP swings between two specific points in the North Atlantic, located in the "eye" of the two stable pressure areas: the Azores high and Icelandic low. Therefore, these two locations should capture a good amount of SLP variability in the basin, also providing information on the strength and direction of the westerly mid-latitude flow. To give a recent example of how the CI and the NAO are connected to each other, we refer to an earlier study by Pausata et al. (2009). The authors have shown that, in present climate the CI patterns of surface temperature (precipitation) closely resemble the correlation patterns between surface temperature (precipitation) and the leading Principal Component (PC) of the SLP field (which is an index similar to the canonical NAOI; Figure R1, panels (a), (c): this figure is a modified version of figure 7 in Pausata et al. 2009; see also figure 8 in Pausata et al. (2009) for the precipitation).

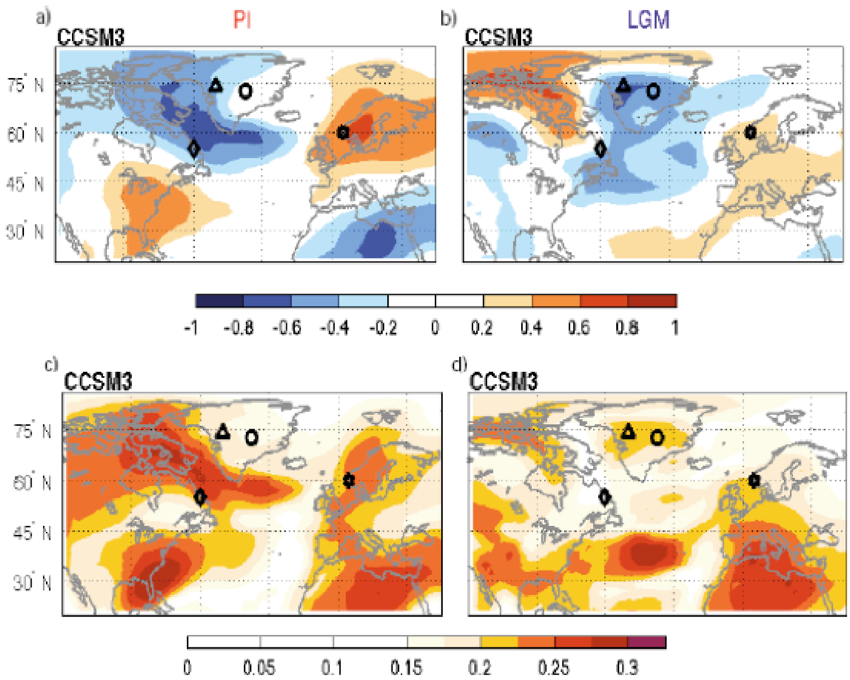

Figure R1: Pre-industrial and Last Glacial Maximum correlations between North Atlantic winter surface air temperature (November to April) and PC1 (NAO-like index) for CCSM3 **(a)**, **(b)**. An indicator of temperature coherence in the sector **(c)**, **(d)**: the value at each point is the absolute value of the area-averaged correlation between temperature at that point and the rest of the North Atlantic basin. Only the winter months are included, as this is when the NAO-like signal is strongest. When including all months the result is the same, but with slightly weaker correlation patterns. Markers indicate the locations used in Table 4 in Pausata et al. (2009).

To further demonstrate the link between CI and NAOI, we have calculated the correlation between SLP and the leading PC of the SLP field, following Pausata et al. 2009. For simplicity, in the manuscript and later on in the reply, we have used the canonical definition of the NAOI, since PC1 and NAOI in winter are highly correlated ($r > 0.90$, see also Hurrell, 1995).

Figure R2 shows that the correlation between PC1 and SLP is very similar to the CI pattern and the correlation maxima of both analyses are quite close to each other. The advantage of the CI analysis compared to the PC/SLP (or temperature or precipitation) correlation analysis is that the CI analysis does not depend only on the leading mode of variability (PC1) but directly integrates all other modes that directly affect the fluctuations of the analyzed variable.

Pausata et al. (2009) have shown that in climates different from the present one, the CI and the PC correlation patterns can be completely different (Figure R1, panels (b), (d)).

We have therefore decided to adopt the CI in addition to the canonical NAOI as a further metric to better understand and interpret large-scale circulation changes: while the NAOI gives a good picture of some features of the mid-latitude atmospheric circulation (strength and direction of the westerly mid-latitude flow), the CI provides additional information on atmospheric variability at a domain scale (shifts in the centres of action).

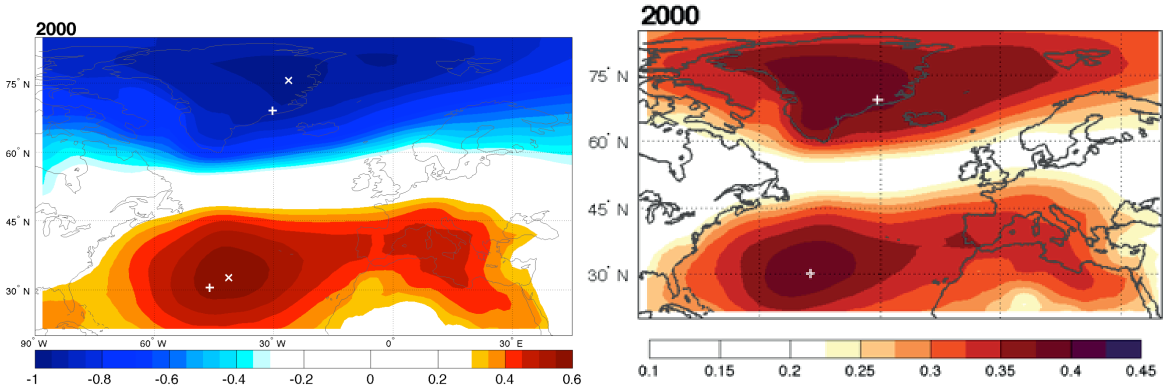

Figure R2: Correlations between North Atlantic winter SLP (December to February) and PC1 of SLP (left) and coherence index of winter SLP (right). The markers indicate the maxima in CI (+ sign) and in the SLP/PC1 (x sign) correlation.

Blocking events and NAO relationship

Blocking events are strongly tied to pressure variability and gradients and hence to the NAO phase. In general terms, positive phases of the NAO lead to a deepening of the Icelandic low pressure and a strengthening of the Azores high-pressure as shown by the leading EOF of the SLP (Figure R3).

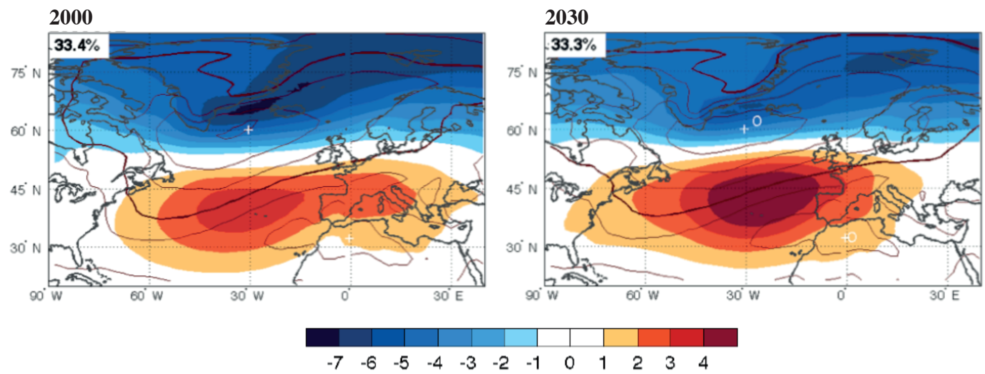

Figure R3: Leading EOF of monthly SLP anomalies (colored shading: hPa/standard deviation of PC) and SLP climatology (contours: 4 hPa interval; bold contour denotes 1016 hPa) in the North Atlantic sector (DJF) for the 2000 and 2030MFR simulations. Numbers show the amount of variance explained by the first mode as a percentage of the total variance. The crosses and circles indicated the minima and maxima of the SLP climatology for 2000 and 2030 respectively (note that their positions do not change).

Positive phases of the NAO are consequently related to a decrease in high-latitude blocking/Greenland blocking events and instead favor low latitude blocking (LLB) events due to a stronger Azores anticyclone. In figure R4 we show changes in LLB events between positive (>1) and negative (<-1) NAOIS in the 2000 simulation. During positive phases of the NAO, the number of high latitude blocking events decreases (deeper Icelandic low) whereas the LLB increases especially over Western Mediterranean Sea (stronger Azores/sub tropical High).

A relationship between positive NAO phases and increased blocking frequency over Europe has been also described in details by Yao and Luo (2014).

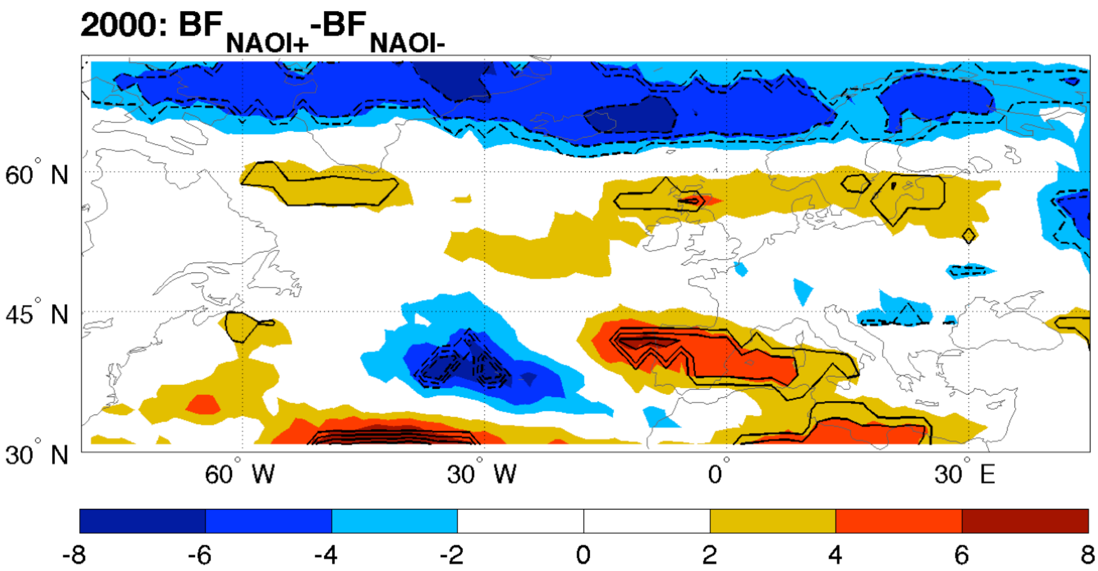

Figure R4: Differences in blocking event frequency (%) between positive (>1) and negative (<-1) NAOI for the 2000 simulation. Contour intervals follow the color-scale (solid positive and dashed negative) and represent the areas with differences significant at 90% confidence level.

CI and Blocking events relationship

As the positions of CI maxima are connected to the NAOI, we expect that changes in the position of the southern CI maximum have an influence on the LLB event frequency. In the manuscript, we mentioned that an eastward shift of the southern center of action is also associated to a higher blocking frequency. To prove this point, we generated – through a bootstrap technique – 10000 subsamples of 15 years from the 60-year control simulation and we then computed the CI for each subsample. We then compared the blocking frequency of 100 subsamples with the southern CI maximum most shifted towards the eastern Atlantic (i.e. towards the Mediterranean Sea) to that of the subsamples with the southern CI maximum most shifted towards the western Atlantic (Fig. R5). Our analysis clearly shows the increased blocking frequency in the central-eastern Atlantic and western Mediterranean when the CI southern maximum shifts eastward (Fig. R5).

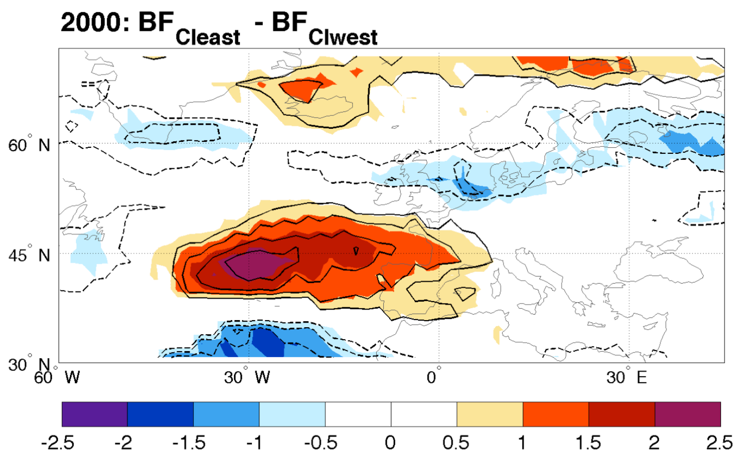

Figure R5: Differences in blocking event frequency (in % of days in which a blocking event occur at a given grid box) between subsamples of the 2000 control simulation having the southern CI maximum with the most pronounced shift towards the eastern and western Atlantic. Contour intervals follow the color-scale (solid positive and dashed negative) and represent the areas with differences significant at 95% confidence level. We have chosen a higher confidence level compared to figure 4 because of the larger number of data used in this analysis.

Looking at changes in the frequency of blocking events therefore provides additional information on the regions where the mean zonal flow will be most affected by the high pressure/low pressure changes, as measured by the CI and NAOI.

Large-scale atmospheric dynamics changes and aerosol distribution

A further concern of Reviewer #1 was that "while changes in blocking may impact aerosol distributions there could be a multitude of reasons for the change in shape of the simulated aerosol distributions. For example changes in the mean precipitation, changes in the structure of the boundary layer etc may be responsible for the change in the skewness of the aerosol distribution."

We agree with the reviewer that also many other factors – such as changes in the mean precipitation or the structure of the boundary layer – could influence the change in shape of the simulated aerosol distributions. However, we argue that these factors are implicitly included in our analysis, since changes in the CI, NAOI and blocking events impact precipitation amounts/distributions and the structure of the boundary layer. Hence, we consider the latter to be the more localized expressions of the large-scale circulation pattern changes, which are the subject of our paper. In a previous paper, Pausata et al. (2013) already demonstrated the significant link between PM and NAOI in winter (Fig. 2 in Pausata et al. 2013): a more positive NAOI leads to more stable conditions over central-western Mediterranean basin (e.g., Walker and Bliss (1932)) and consequently increased positive PM anomalies. Moreover, the linkage between large scale and synoptic atmospheric circulation and PM has also been discussed in other studies, highlighting how stagnant atmospheric conditions are favorable to a deterioration in air quality (e.g. Horton et al. (2014); Jacob and Winner (2009)). Here, we make a further step by investigating changes in future large-scale atmospheric dynamics, which can favor these stagnant conditions over the Mediterranean basin, and hence affect PM concentrations. The main aim of this part of the paper is therefore to provide a general coherent overview of the impacts of large-scale circulation changes on air-quality. We are planning a more detailed study to investigate specific aspects more systematically.

Finally, we do not claim that the increased frequency of LLB events is the sole driver of changes in the aerosol distribution. Rather, the concomitant changes in CI/ NAOI and blocking events lead to more stagnant conditions over the Western Mediterranean, and hence to a change in aerosol distribution. We will clarify this aspect in the revised version of the manuscript.

We plan to include some of the above analysis, together with a more thorough discussion of the various dynamical features and their link to pollution, in the revised version of the manuscript. We will include a discussion on the other factors that can have an impact on PM distributions.

Again, we appreciate the comments of the Reviewer #1, which provided this opportunity to clarify some aspects of our paper.

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