

Response to Reviewers

We wish to thank the reviewers for their insightful comments which we feel have substantially improved our manuscript. We believe we have addressed all major and minor comments that were raised by the reviewers and, in doing so, have crafted a paper that is more rigorous in content. First, we copy below the reviewers' comment (in bold), and then we describe how we have addressed each of the issues.

Reviewer #1

- a) **Page 3137, lines 26-27: The authors mention a substantial overestimate of the modelled ozone compared to measurements by up to 15 ppbv in some regions. Please clarify which regions and possible reasons for this large overestimate. For example clarify the possible role of stratosphere-troposphere exchange or photochemistry in this overestimate.**

We have extended the section related to modelled ozone overestimation. We have referred to the MPI-report (Rast et al., 2012), that shows that the best agreement between modelled and observed surface ozone concentrations is reached in the Eastern European region, "the worst results are obtained for the Mediterranean region, where the model overestimates ozone concentrations by more than 40% on average. The correlation between observations and model results is excellent for Central Europe (for all 18 sites r is greater than 0.65), it is reasonable for Northern, Western and Eastern Europe (about half of the sites have r greater 0.65), and it is poor for the Mediterranean (only 3 sites with $r > 0.65$) and the mountain sites".

"The high bias observed in the preceding analysis may be a consequence of advection of ozone-rich air from aloft and trapping of polluted air masses in the boundary layer (see analysis of ozone sonde and MOZAIC data in sections 4.4, 4.5)" (Rast et al., 2012).

- b) **The authors mention that the NAOI timeseries have been calculated as the difference in the normalized SLP anomalies between the model grid boxes corresponding to the location of Ponte Delgada, Azores and Stykkisholmur/Reykjavik, Iceland. It might worth looking how the modelled NAOI compares to the NAOI based on observations.**

We use an hindcast simulation based on ERA-40 reanalysis, therefore, by construction the modelled NAOI has a really high correlation with the station based observation. Moreover we have mentioned at the beginning of section 3.1 that "The simulated SLP climatology, interannual variability and the NAO faithfully reproduce the observation (<http://www.cgd.ucar.edu/cas/jhurrell/indices.info.html#n>) given that the simulated meteorology fields are relaxed towards the ERA-40 reanalyzed data." As a consequence the modelled NAOI and the station based NAOI are highly correlated ($r=0.94$, see also Fig. R1). We have included in the text the correlation between the modelled NAOI and the station based NAOI.

- c) **Page 3139, lines 20-21: It is mentioned that the NAOI and PC1 monthly timeseries are strongly correlated in winter (0.86) and less correlated in summer (0.59). Please**

specify if these correlations are based on daily values, monthly values or seasonal values.

Thank you for pointing this out. The correlations are based on monthly data as pointed out in section 2: "The model data analyzed here are restricted to the Atlantic sector (20°–90°N, 90°W–40°E) and the results are based on monthly anomalies from the climatological monthly mean." In any case, we have specified also in the result section that the EOF analysis has been calculated using monthly SLP field: *"The EOF analysis of the monthly SLP field shows that in winter, ... "*

- d) **Pages 3142-3143: It is mentioned that the negative phase of NAO leads to a southward shift towards the Mediterranean Sea of the storm track and consequently, the O3 enriched air masses from the Atlantic Ocean, increase of few ppbv the surface O3 concentrations in the western part of the Iberian Peninsula (Fig. 6). Is this small and spatially limited increase of ozone over the Iberian Peninsula statistically significant to make the authors speculating for the reasons of its presence in their model results? Also I am not sure why the authors claim that the air masses originating from the Atlantic Ocean are enriched in O3? If we exclude cases of intercontinental pollution transport from US then usually the Atlantic ozone baseline ozone is lower than the European continental ozone levels.**

The small and spatially limited increase of ozone over the Iberian Peninsula during negative phases of the NAO are not statistically significant in winter (Fig. R2); however, the correlation between PC1 and surface ozone over this area is statistically significant (Fig. 5), therefore, we felt it was worth mentioning it.

We claim that the air masses originating from the Atlantic Ocean are enriched in O3 at the surface because the model shows it (Fig. 2 in the manuscript: ozone concentration over the continents are lower than over the ocean in three out of four seasons: winter, spring and fall). Figure 3 in the manuscript shows similar results for tropospheric ozone.

- e) **Page 3143, lines 3-6: The authors state that 'More storms in the Mediterranean Sea enhance southerly flow towards Italy and northern Africa leading to a slightly increased surface O3 concentrations over the Italian peninsula and decreased surface O3 values in north-eastern Africa.' I find this statement rather speculative and I would ask the authors to clarify and explain what do they mean. Furthermore, what the authors claim as an increase over the Italian Peninsula in the negative phase of NAO is actually small response limited over a small area in Central Italy. Is this increase statistically significant at 95% or 90%. Similarly the negative ozone anomalies over the Adriatic Sea during positive NAO phases in summer are small and limited and I am wondering if they are also statistically significant.**

We mean that an increased number of storms in the Mediterranean Sea is connected to more southerly winds blowing from areas with higher surface ozone concentrations (the Mediterranean Sea) towards the Italian peninsula (see figure 2 in the manuscript). The same line of reasoning can be applied for north-eastern Africa, where with positive NAO it experiences northerly flow

(enriched O₃ from the sea), whereas with negative NAO it experiences southerly O₃ depleted air mass advection. However, as shown in the new Figure 6 (see answer f) the anomalies are not significant at 90% confidence level. The same holds true for summer. Nevertheless, we feel like mentioning it since as shown in figure 5 the correlation between O₃ and PC1 is actually significant in those areas.

- f) **Following my previous comments, I would suggest for clarity reasons to indicate in Figure 6 the areas where the anomalies are statistically significant at 95% or 90% (e.g. by using a t-test). This is the most common way to point the statistically significant differences between two groups of data.**

We have plotted in Figure 6 the areas that have anomalies statistically significant at 90% using a t-test (see fig. R2).

- g) **Page 3145, lines 1-4: The authors state that in summer 'The negative NAO phase leads to surface O₃ negative anomalies that are shifted south compared to positive anomalies, affecting western Africa, part of the Sahara desert and great part of the Mediterranean Sea, whereas mainly no anomalies are displayed in the Nordic Sea and Baltic Region.' The authors attribute these negative ozone anomalies over southwestern Europe to less ozone enriched European continental flow. Could it be also attributed to a cyclonic anomaly introducing more clouds, less solar radiation and hence less photochemical ozone production?**

We believe that the cyclonic anomaly suggested by the reviewer may have an influence on surface ozone concentrations over southwestern Europe (Iberian Peninsula), however, the strongest cyclonic anomaly influence is over western Europe (northern France, British Isles) as it may be inferred by the correlation between PC1 and precipitation (Fig. R3). We have included this suggestion in the manuscript.

- h) **Page 3146, lines 9-12: The authors state that 'Unfortunately, since in our study we do not include a diagnostic for transport of stratospheric air into the troposphere, we could not quantify neither the amount of surface O₃ variability associated with the STT, nor the frequency of STT events associated with the NAO phases.' Although I understand that the authors do not have stratospheric ozone tracer as diagnostic in their simulations, they could possibly use other common parameters from their simulations as diagnostics such as potential vorticity or water vapour mixing ratio. These diagnostics may not assist to quantify the ozone anomalies but at least would give a hint and stronger evidence for interpretation of their results.**

Thank you for the suggestion. We have looked at CO and performed the same analysis done for the ozone (Figs. 7 and 8 in the manuscript). We have preferred CO rather than H₂O mixing ratio, because the latter is more influenced by contingent atmospheric conditions. The CO anomalies pattern (Figs. R4 and R5, see also section 3.3) resembles the O₃ pattern (sign reversed) for the mid-to-high latitudes. However, the relative anomalies are less strong for CO than for O₃. This may indicate that STT does play a role in affecting O₃ concentration at the surface at mid-to-high latitudes, but O₃ tropospheric production and advection are also an important component.

At mid-to-low latitudes, the CO and O3 anomaly patterns differ suggesting a weaker influence of STT at the surface. These views are consistent with a vast body of literature.

- i) **Finally in the discussion section (page 3147), the authors state that their approach of PC1/O3 relation can be used to identify areas at risk of high O3 concentrations, without performing costly chemical weather forecasting at monthly timescale. I find this statement about chemical weather forecasting rather obscure and unnecessary. The use of statistical indices and relations may assist chemical weather forecasting but cannot substitute it. I suggest rephrase of this sentence in order to give the right message.**

We have changed 'without performing costly chemical weather forecasting at monthly timescale' to read as 'supporting chemical weather forecasting at monthly timescale'.

Reviewer #2 (Anonymous)

General Comments:

1. **Throughout the text, there are attributions of enhanced or depleted surface O3 during NAO phases to specific transport pathways. The basis for these attributions is not always obvious. For example, why is the enhanced wintertime surface ozone during positive NAO phases necessarily advected from the marine lower troposphere as opposed to subsiding from aloft around the anticyclone?**

Thank you for pointing this out. We believe that the subsidence could sometimes play a role, but with this line of reasoning one would not be able to explain the negative correlation/anomalies during positive NAO phase (Figs. 5 and 6) in the Iberian peninsula and Western Africa, where the enhanced subsidence is at the strongest (Fig. 1): enhanced subsidence does not lead to increased O3, but actually a decrease.

2. **How do the findings here differ from those in Hess and Lamarque (2007, JGR, doi:10.1029/2006JD007557) who examined contributions of regional emissions and stratospheric ozone using sensitivity simulations and tracers in their model (similar to the approach recommended by the authors on p 3149)? A clear description of the advances made here is needed.**

In the introduction we have mentioned Hess and Lamarque study, pointing out that they focused on ozone variability only in February and March over the entire Northern Hemisphere using the broader AO index, whereas we look at the ozone variability in all four seasons focusing on the North Atlantic (using therefore the NAOI). We have inserted a comment on this study in the discussion section when we recommend the use of tracers in future works. Now it reads:

Our study highlights the importance of the NAO in driving surface ozone variability in large parts of Europe, not only affecting low level transport but also middle and upper troposphere and stratosphere-to-troposphere transport. However, a quantitative analysis on the relative impact of STTs and low level transport associated to the NAO phases on surface ozone concentrations

was not possible using our chemical re-analysis model simulation, due to a lack of appropriate diagnostic tools. Therefore, We strongly recommend that future studies using climate-chemistry models are supplemented with appropriate diagnostics (e.g., tracers) to investigate more in depth the effects of each NAO phase on surface ozone interannual and decadal variability and trend, as done for example by Hess and Lamarque (2007). However, new studies should not only be performed in spring, but also in other seasons.

3. **The paper should be strengthened by including more evaluation of the model with observations (e.g., in Figure 2). P3137 mentions an ozone bias in some regions. Is this year-round? It would be better to show how the model compares with the observations at the sites in Table 1. Statistics should be included to support statements like, 'model captures the observed range of interannual o3 variability' and those on top of P3138, on P3140 (seasonal and spatial variability in surface and tropospheric column ozone), etc.**

The model has been already extensively evaluated with regards to ozone. The ozone biases are mainly year-round as it also could be inferred by figure 3 in the manuscript and in figure R6 (from Pozzoli et al. (2011)).

For more details see answer a) to Reviewer #1

Specific Comments:

- a) **The abstract could be clearer in places. Some questions: Over what region is surface ozone examined (lines 6-7)? Is the NAO much weaker in fall and thus does not have much impact on surface ozone in this season? What kind of lead time is possible for using the SLP PC1 to predict surface ozone? What period is the 1990s increase relative to (1980s?) or is there an increase over the 1990s? How does the NAO affect this (e.g., a shift from negative to positive over the period) ?**

Thanks for pointing this out, we have included the region where surface ozone is examined and we have clarified the other points. However, we feel that going onto the details why the NAO does not have much impact on surface ozone during fall would add too many details for an abstract. Finally we refer to the surface ozone increase during/over the 1990s.

Now the abstract reads:

Ozone pollution represents a serious health and environmental problem. While ozone pollution is mostly produced by photochemistry in summer, elevated ozone concentrations can also be influenced by long range transport driven by the atmospheric circulation and stratospheric ozone intrusions. We analyze the role of large scale atmospheric circulation variability in the North Atlantic basin in determining surface ozone concentrations over Europe. Here, we show, using ground station measurements and a coupled atmosphere-chemistry model simulation for the period 1980–2005, that the North Atlantic Oscillation (NAO) does affect surface ozone concentrations – on a monthly timescale, over 10 ppbv in southwestern, central and northern Europe – during all seasons except fall. The commonly used NAO index is able to capture the link existing between atmospheric dynamics and surface ozone concentrations in winter and spring but it

fails in summer. We find that the first Principal Component, computed from the time variation of the sea level pressure (SLP) field, detects the atmosphere circulation/ozone relationship not only in winter and spring but also during summer, when the atmospheric circulation weakens and regional photochemical processes peak. Given the NAO forecasting skill at intraseasonal time scale, the first Principal Component of the SLP field could be used as an indicator to identify areas more exposed to forthcoming ozone pollution events. Finally, our results suggest that the increasing baseline ozone in western and northern Europe during the 1990s could be related to the prevailing positive phase of the NAO in that period.

- b) **P3133 L4-5 Some early papers could be included – Wild et al. 2001, Yienger et al., 2000, both in JGR.**

Thank you, we have included them in the manuscript.

- c) **P3133 L10-17 The point here is not clear as to whether the authors are suggesting that STT is important for the local-to-regional elevated ozone (e.g., specific events) or baseline levels. In general, it could be clearer that the majority of O3 production, at least in summer, is produced from local-to-regional precursor emissions.**

We refer to baseline levels and it is now specified in the manuscript . We did point out that the majority of O3 production, at least in summer, is produced from local-to-regional precursor emissions: pag 3132 lines 23-27 and page 3133 line 1:

In many modern cities and surrounding areas, ozone concentrations are often above thresholds for plant damage and negative health effects especially during late spring and summer (e.g., (e.g. Akimoto, 2003; Lovblad et al., 2004; Dentener et al., 2006; EEA report, 2010). Elevated O₃ concentrations are local-to-regional phenomena, mainly associated with photochemical reactions favored by a combination of intense solar radiation and emissions of air pollutants.

- d) **P3134 L5 The intercontinental transport is occurring in the westerly flow, correct? L18- 19. Are the droughts and smog associated with positive or negative NAO phases? Elsewhere, please specify sign of NAO and associated impact wherever possible. L20- 24. Is this the same as the northern annular mode definition? If so, there is a lot of literature on these modes that should be cited for context here.**

P3134 L5 Yes, that's correct.

L18-19 Yes, droughts can be associated to NAO phases as well as photochemical smog: on P3134 L3-11 we mentioned the effects of the different phases of the NAO: prolonged positive NAO anomalies may lead to droughts in the Mediterranean and droughts in the nordic countries during prolonged negative NAO phases.

L20-24 The northern annular mode (NAM) is defined over the entire northern hemisphere, whereas the NAO is defined only in the Atlantic sector. We have added some references regarding the NAO definition using the EOF analysis (see also answer 1 to Reviewer #3).

- e) **P3136 L8-10 Hess and Lamarque, JGR, 2007 study 1980-2001 so there is some precedent for analysis of AO, O3 variability over multiple decades.**

We have mentioned this study on P3135 L22-25. We have now included that this study looks at multiple decades. See also answer 2 on general comments.

- f) **P3136 L13-16. Stronger support for stratospheric influence could be made by using a stratospheric tracer in the model, a commonly used approach [e.g., Wang et al. JGR 1998, doi:10.1029/98JD00156. In the absence of such a tracer, water vapor or CO distributions should provide additional evidence.**

We have added a paragraph discussing the CO anomalies associated to the NAO phases (see also answer h to Reviewer #1)

- g) **P3137 L12-15. Which meteorological variables are nudged? How is stratospheric ozone simulated in the model?**

In 'ECHAM5 vorticity, divergence, sea surface temperature, and surface pressure are relaxed towards the re-analysis data every time step with a relaxation time scale of 1 day for surface pressure and temperature, 2 days for divergence, and 6 h for vorticity (Jeuken et al., 1996). The relaxation technique forces the large scale dynamic state of the atmosphere as close as possible to the re-analysis data, thus the model is in a consistent physical state at each time step but it calculates its own physics, e.g. for aerosols and clouds ' Pozzoli et al., 2011.

"Stratospheric O3 concentrations are prescribed as monthly mean zonal climatology derived from observations (Logan, 1999; Randel et al., 1998). These concentrations are fixed at the topmost two model levels (pressures of 30 hPa and above). At other model levels above the tropopause, the concentrations are relaxed towards these values with a relaxation time of 10 days following Horowitz et al. (2003)" Pozzoli et al., 2011. The model is able to reflect the total ozone variability associated to the NAO (for example compare figure R7 with figure 3 in Appenzeller et al., 2000).

We have inserted in the paper a brief summary of these two paragraphs.

- h) **P3137 L20. It would help to briefly summarize the emissions used. For example, is biomass burning varying inter-annually in these simulations? Do fires vary with NAO (e.g., drier regions) and thus affect O3? Same for natural emissions (lightning, biogenic) –is NAO-driven variability in these included?**

Emissions of O3 precursors from natural processes are calculated on-line considering the meteorological variability, therefore, also in the model NAO may well play a role in affecting fires. "The emissions from vegetation of CO and VOCs (isoprene and terpenes) were calculated interactively using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006). Lightning NOx emissions (Fig. 3b) are calculated following the parameterization of Grewe et al. (2001). We use the compilation of interannual varying biomass burning emissions published by Schultz et al. (2008), who used literature data, satellite observations and a dynamical vegetation model in order to obtain continental-scale emission estimates and a geographical distribution of fire occurrence. Other natural emissions are kept constant during the entire simulation period. CO emissions from soil and ocean are based on the Global Emission Inventory Activity (GEIA) as in Horowitz et al. (2003), amounting to 160 and 20 Tg yr-1, respectively.

Natural soil NO_x emissions are taken from the ORCHIDEE model (Lathiere et al., 2006), resulting in 9 Tg(N) yr⁻¹. See Figure 3 of Pozzoli et al. 2011 for the interannual variability of natural emissions.

In view of the length of the paper, we have not gone in the details of the emissions used but we have pointed to the comparison paper by Pozzoli et al. (2011).

- i) **P3139. Clarify in text that Figure 1 is the EOF of the SLP.**

We have now clarified in the manuscript that Figure 1 refers to the EOF of SLP.

- j) **P3143 L3-5. 'More storms in the Mediterranean sea': from observations or model? L11-16 is out of place in a 'winter' section. 'Spring' section discussion could be sharpened to clarify the main point, I found the discussion here confusing.**

P3143 L3-5: The model is nudged and so it closely resembles the observations. In any case, the negative phase of the NAO leads on average to more storms in the Mediterranean by construction.

L11-16 We have moved the paragraph at the end of the section as a concluding paragraph.

The spring section has been sharpened and reads as follow: *In spring (MAM), the PC1/O₃ spatial correlation coefficients are similar but somewhat weaker than winter, with positive values over the British Isles and Scandinavia in the north and north Africa in the south. The NAOI/O₃ correlation pattern also shows significant values over the British Isles and part of Scandinavia, but the overall pattern starts becoming different compared to the PC1/O₃ correlation pattern. The weakening of the relationship between NAO and O₃ concentrations is in agreement with the data that show a reduced number of ground stations significantly correlated with the NAO (Fig. 4). Over central-eastern Europe the measurements and the model show correlations of opposite signs between NAO and O₃ concentrations, but in both cases the correlations are not significant.*

- k) **P3145 The case for stratospheric influence is not particularly compelling in Figure 7 and 8 and could perhaps be strengthened by considering the three-dimensional nature of intrusions (see for example Langford et al., 2012, JGR, doi:10.1029/2011JD016766 who also show that STT events can impact surface air on a short-term basis).**

In order to better understand the stratospheric influence we have analyzed the CO anomalies associated to the NAO phases. We have considered latitudinal and longitudinal transects in Figure 7 and 8 at specific locations, such as Mace Head. In the new figures related to NAO/CO anomalies we have also considered such transects (Figs. R4 and R5) and this provides new insights on the STT events (see also answer h) to Reviewer #1).

- l) **P3146. The discussion of long-term trends being associated with NAO phasing is confusing. Has there been a trend in NAO phasing or is variability driven by NAO mistakenly being interpreted as a long-term trend?**

The NAO phase showed constant positive anomalies in the late 80s and 90s, influencing surface O₃. Therefore, the long-term trend in O₃ seen in Northern Europe might have been influenced by the variability driven by the NAO. We have rewritten the paragraph as follow:

The O_3 anomalies that extend from the lower stratosphere to the surface seem to be due to the interplay between two main processes: i) low level O_3 transport both in winter and summer and enhanced photochemistry in summer; and ii) STT events that are able to affect O_3 concentration at the surface especially in the winter months and at mid-to-high latitudes (Figs. 9 and 10). Lamarque and Hess (2004) and Hess and Lamarque (2007), focusing mainly on spring months, have shown that STT events have a contribution to the O_3 tropospheric anomaly pattern, dominating the ozone variability signal throughout the depth of the troposphere over Northern Canada and the Arctic region. On the other hand, they have shown tropospheric production of ozone and tropospheric ozone transport are also very important for the lower tropospheric ozone over the Atlantic basin and Europe. Therefore, SST events do play a role for the overall surface baseline ozone budget (see also Hocking, 2007). Hence, when a given NAO phase persists for several subsequent years, STTs may also contribute to long term trends in surface O_3 concentrations.

m) **P3147 Can a new pair of sites be proposed to better capture the centers of action for NAO in summer or is it always necessary to do the full PC analysis?**

Thank you for the suggestion. We did consider other locations for summer and even though we found two locations ($43^\circ\text{N } 25^\circ\text{W}$ and $82^\circ\text{N } 8^\circ\text{W}$ which are better correlated to the summer PC1 (0.85 versus 0.59 of the standard locations) and are able to capture the anomalies over the Iberian peninsula, it fails over the British Isles (see fig. R8). So eventually we believe the PC1 analysis is the best atmospheric circulation predictor for surface ozone concentrations in summer over Europe. We have added a brief discussion about it in the discussion section together with figure R8:

In order to evaluate the possibility of accounting for the summer northward shift of atmospheric circulation, using simply two alternative locations rather than the canonical ones (northern location: $65^\circ\text{N } 23^\circ\text{W}$ and southern location: $38^\circ\text{N } 26^\circ\text{W}$) or the EOF analysis, we have looked for the two model grid boxes that maximizes the correlation between the new summer-NAOI (SNAOI) and the PC1. We have found that the northern location shifts at $82^\circ\text{N } 8^\circ\text{W}$ and the southern center of action at $43^\circ\text{N } 25^\circ\text{W}$. The SNAOI calculated using the above-mentioned centers of action has a correlation of 0.85 with the summer PC1 (instead of 0.59 of the standard locations). However, while the SNAOI is able to capture ozone positive anomalies over the Iberian Peninsula, it fails in capturing the correlation over the British Isles (cf. Figs. 11 and 5). Therefore, the PC1 timeseries remains the best atmospheric predictor for surface ozone concentrations in summer over Europe as a whole.

n) **P3148 The model and observations look like they are trending different directions in MAM in Figure 9. JJA does not look well captured either.**

We agree that modeled and observed MAM anomalies trend in different directions even though in the first years the increasing trend is in common as well as the decrease in 1995-1997 and finally the flattening in the last 8 years. The overall trend of JJA seems fairly captured. Anyway, the purpose of this figure (now figure 12) is just to help the reader comparing the Mace Head ozone variability with the NAOI/PC1 variability without looking for papers showing Mace Head ozone trends. In fact, the positive trends in surface ozone at Mace Head and northern-western Europe

have been already shown in several studies (references now included in the manuscript) and it is not a result of our paper. The seasonal PC1 variability over the analyzed period has been now included in the figure.

- o) **Figure 2. Do observations support the simulated higher ozone levels over the Atlantic than over the continents?**

Figure 3 shows the retrieved tropospheric O₃ concentrations that also present higher ozone concentrations over the ocean compared to the European continent. We are not aware of any ozone station off the coast of Ireland or over the North Atlantic.

- p) **It would help to place modeled and observed correlations in Figures 4 and 5 together for easier comparison, perhaps by superimposing the circles in Figure 4 on the modeled correlations.**

We have tried to superimpose the circles in Figure 4 but it look too crowded and therefore we have preferred to improve the linkage between the figures in the text.

Reviewer #3 (Anonymous)

Major Comments:

1. **The authors state that the PC1 index is an alternative to the NAOI index (p 3134, l 22). This is not clear to me. In fact the AO is usually identified with the leading component of the 1000-hPa height anomaly variability. Thus it is not clear that the PC1 index as defined by the authors isn't actually an index of the AO oscillation. I don't think the authors really need to get into the potential differences between the AO and NAO, but I do think it is mis-leading to state the PC1 index is an index of the NAO. A better procedure would be to simply use the established AO index instead of the PC1 index in their analysis. This is unlikely to change their results but is more consistent with the literature.**

The Northern Annular Mode (NAM) or Arctic Oscillation is defined as the first EOF of NH (0/20-90 N) winter SLP data (<http://www.cgd.ucar.edu/cas/jhurrell/indices.html>). Whereas when the domain is restricted to the extratropical North Atlantic, the PC1 time series better represents the NAOI (Hurrell, 1995). AO and the NAO are highly correlated, however, since we are focusing on Europe, we strongly feel it is better to use the NAO domain avoiding disturbances from the North Pacific Oscillation. Moreover, several studies have been done referring to the EOF1 of the SLP field defined over the North Atlantic basin as the leading mode of the North Atlantic atmospheric variability, i.e. the NAO (e.g., Hurrell (1995); Hurrell et al. (2003); Pausata et al. (2009, 2011))

2. **The authors show a nice correlation plot of the NAOI and PC1 with ozone over Europe (Fig. 4), but they do not show the magnitude of the measured ozone anomalies associated with the correlation. The authors simulate very large ozone anomalies**

(e.g., Figure 6) associated with the NAO, up to 10 ppbv. They should additionally analyze the NAO ozone anomalies from station data. Does the station data support the large simulated anomalies?

Thanks for pointing this out. Several studies (e.g., Pozzoli et al, 2011, Rast et al., 2012) have already shown that the model is able to capture observed ozone variability as mentioned before. Therefore the model is also able to capture the variability due to the NAO, which is enclosed in the total variability. In our study we are not trying to separate the amount of variability due to the NAO from the variability due to other factors. We point out that the modeled and observed ozone variability are influenced by the NAO, meaning that months with strong NAO anomalies are likely to lead to strong modeled and observed ozone concentration anomalies as well. In any case, we have also shown seasonal ozone anomalies for Mace Head ozone station that does show anomalies associated to the NAO up to 7-8 ppbv (seasonal average) in good agreement with the model data (Fig.12 in the revised manuscript).

- 3. The authors claim that the NAO may help to explain the ozone trend over Mace Head, and show measured and simulated ozone over Mace Head in Fig. 9. I think if the authors want to show this figure they need to, at a minimum, examine the correlation between the NAO/PC1 and Mace Head ozone. To what extent does the NAO explain the measured/modeled ozone anomalies at this site? I think more analysis is necessary with regards to this figure.**

We did show the correlation between NAO/PC1 and Mace Head ozone station in figure 4 and modeled ozone in figure 5 on monthly time scales. We have now added in the text the correlation values in section 3.2.1 in order to give a better idea on the influence of the NAO on measured ozone anomalies. Anyway, the purpose of this figure is just to help the reader comparing the Mace Head ozone variability with the NAO/PC1 variability without looking for papers showing Mace Head ozone trends. In fact, the positive trends in surface ozone at Mace Head and northern-western Europe have been already shown in several studies (references now included in the manuscript) and it is not a result of the paper. Nevertheless, we have added the seasonal averaged PC1 timeseries and the correlation between the seasonal PC1 and the O3 (modeled and observed) timeseries (Fig. 12 in the revised manuscript).

- 4. I think the references in the paper could be improved somewhat. There are a numerous locations where a statement needs additional references. -p 3133, l 5 and 6. This needs some references. Some relevant papers of which I am aware: Brownsteiner and Hess (2011), Liang et al. (2004), Fiore et al , 2002 ... -p 3133, l 14 'specific STT events': see study by Lin et al., (2012) -p 3134, l 19: references needed with regard to the importance of NAO during summer months -Hess and Lamarque (2007) and Lamarque and Hess (2004) examined the response of STE to the AO/NAO. This previous work seems relevant to the current paper (i.e., in section 3.3)**

Thank you for pointing this out. -p 3133, l 5 and 6 we have included the reference to the study of Guerova et al. 2006 that focus on the Atlantic basin and Europe and show that in summer

long range O₃ transport mainly occurs during perturbed weather conditions that are generally not associated with highest surface O₃ concentrations. We have also added Brownsteiner and Hess (2011) when we refer to studies that show that tropospheric ozone can be transported over long distances, affecting O₃ concentrations at the surface; - p 3133 l14 we have included Lin et al (2012) in the text; -p 3134 l 19 we have provided further references (besides the Barnston and Livezey (1987)); - We have included a brief discussion of Hess and Lamarque (2007) and Lamarque and Hess (2004) studies in section 3.3, that reads:

The O₃ anomalies that extend from the lower stratosphere to the surface seem to be due to the interplay between two main processes: i) low level O₃ transport both in winter and summer and enhanced photochemistry in summer; and ii) STT events that are able to affect O₃ concentration at the surface especially in the winter months and at mid-to-high latitudes (Figs. 9 and 10). Lamarque and Hess (2004) and Hess and Lamarque (2007), focusing mainly on spring months, have shown that STT events have a contribution to the O₃ tropospheric anomaly pattern, dominating the ozone variability signal throughout the depth of the troposphere over Northern Canada and the Arctic region. On the other hand, they have shown tropospheric production of ozone and tropospheric ozone transport are also very important for the lower tropospheric ozone over the Atlantic basin and Europe. Therefore, SST events do play a role in the overall surface baseline ozone budget (see also Hocking (2007)). Hence, when a given NAO phase persists for several subsequent years, STTs may also contribute to long term trends in surface O₃ concentrations.

Minor Comments:

1. **p 3135, l 24 Hess and Lamarque (2007) analyzed ozone variability due to the AO, not the variability in general.**

Thanks. We have corrected it. Now it reads:

Hess and Lamarque (2007) have used a chemistry transport model to analyze tropospheric ozone variability in February and March in the period 1980–2001 due to the Arctic Oscillation. The Arctic Oscillation and the NAO are highly correlated, however, the Arctic Oscillation also includes the dominant mode of variability of the North Pacific (the North Pacific Oscillation). Therefore, when focusing exclusively over the Atlantic basin the NAO is more appropriate (e.g., Hurrell (1995); Hurrell et al. (2003)).

2. **p 3136, l 23: I think the authors should say something more about how these particular stations were picked.**

We have clarified in the manuscript the reasons why those particular stations were picked. Now it reads:

In this study we selected a total of 17 stations from the 150 stations belonging to the European Monitoring and Evaluation Programme (EMEP, <http://www.emep.int/>) for which monthly mean O₃ concentrations are available between 1990 and 2005 (Table 1). We have chosen a subset of relatively unpolluted stations with sufficiently long timeseries in order to have a representative coverage of the areas most influenced by the NAO.

3. **p 3137, l 19: What do the authors mean by a 'consistent physical state'? Does the model calculate surface heat and moisture fluxes or are these input?**

In 'ECHAM5 vorticity, divergence, sea surface temperature, and surface pressure are relaxed towards the re-analysis data every time step with a relaxation time scale of 1 day for surface pressure and temperature, 2 days for divergence, and 6 h for vorticity (Jeuken et al., 1996). The relaxation technique forces the large scale dynamic state of the atmosphere as close as possible to the re-analysis data, thus the model is in a consistent physical state (with the re-analysis) at each time step but it calculates its own physics, e.g. for aerosols and clouds' Pozzoli et al., 2011. The model does calculate the surface heat and moisture fluxes.

4. **p 3137: Please give more information on the specification of biogenic emissions. Also please give more information on the specification of the stratosphere and the relevant boundary conditions used. Does the simulated stratosphere reflect variability due to the NAO? If not, I think the authors need to make some caveats about the impact of the NAO on middle and upper tropospheric ozone.**

See answer h) to Reviewer # 2 above and figure R7.

5. **p 3138, l 6-7: What do the authors mean by monthly anomalies from the climatological seasonal cycle?**

We meant monthly anomalies from the climatological monthly mean, not for example the annual mean. We have rephrase it to avoid misunderstanding as follow:

The model data analyzed here are restricted to the Atlantic sector ... and the results are based on monthly anomalies from the climatological monthly mean.

6. **p 3139: 'investigates' should be investigate, 'trough' should be through.**

Thank you.

7. **p 3139, l 13: Please specify which index is referred to here.**

Thank for pointing this out. We have rephrased the sentence in order to make clear that the EOF analysis shows that the NAO controls 40% of SLP variance in winter. However, we are referring to the NAO as a phenomenon rather to a specific index in that paragraph.

Now it reads as follow: *The EOF analysis of the SLP field shows that in winter, the NAO controls a significant fraction of the SLP variability in the North Atlantic accounting for more than 40% of its total variance (Fig. 1).*

8. **Figure 3: The authors should really use the same color scale for both the model and the measurements. Otherwise, it is very difficult to compare the two. However, I think it is reasonable to change this scale between the different seasons.**

The point of figure 3 is not to show the absolute values but rather how the patterns are similar between retrieved and modeled tropospheric ozone. We strongly feel like changing the color scale would lead to a misinterpretation of the meaning of the figure.

9. **p 3140, l 11: Does the model overestimate the measurements everywhere?**

We have add more detailed information (see answer a) to Reviewer #1).

10. **p 3140, l 18: 'does not present an issue'. I would say this a bit differently: it is not critical to the study.**

Thank you for the suggestion. We have implemented it in the text.

11. **Figure 4: this is a nice figure, but it is hard to discern the stations with significant correlations. One solution might be to make the size proportional to the significance and the color proportional to correlation. However, the authors might have a better idea.**

Thank you for the suggestion. We have noticed that the stations that have ozone values significantly correlated with PC1/NAOI are difficult to distinguish and since the significancy is just a threshold value we have decided to thicken the contours in order to better visualize the stations with significant correlation. We feel that since the plot shows single point locations and not smooth contiguous areas using the smooth color scale used in Figure 5 rather than dots size proportional to the correlation may well not help.

12. **p 3141, l 27-29: I think the authors need a reference here. While ozone lifetime is longer in winter photochemistry is also important. One example is the NO tritration of ozone in the European boundary layer. Probably want to reword this somewhat.**

Thanks for pointing this out. The NO titration of ozone for sure plays a role in winter reducing O₃ concentration in polluted area, however we wanted to point out that in winter the photochemical reactions do not have a main role in affecting O₃ concentration at the surface and atmospheric dynamic is instead dominant. Now the paragraph reads as:

in winter, the limited photochemical production of ozone, leads to a larger role played by the atmospheric dynamics in determining the O₃ variability, and in summer both dynamical and photochemical processes come into play, leading to elevated O₃ concentrations.

13. **p 3142. The springtime pattern here looks remarkably similar to Hess and Lamarque (2007). This is probably worth mentioning.**

Thanks, we have included it in the discussion.

14. **p 3142, l 18: by 'down' due the authors mean southward? It is not clear what is happening in the vertical.**

Thanks for pointing it out. We have corrected it.

15. **Figure 6: please indicate the regions where the correlation is significant (from the previous figure) instead of where the standard deviation is larger than 0.5**

Done. See also figure R2

16. **p 3144, l 4: "the different correlations could be" – the authors should be able to easily check this hypothesis.**

We have shorten that paragraph as suggested by Reviewer #2 (see answer j). Anyway, we have checked that hypothesis, but the different length in the time series seems not to be the reason of the different correlation sign between observation and model. We have, therefore, removed that sentence in the revised manuscript.

17. p 3145, l 24: it is not clear why the authors use 'extreme' here.

We mean "extreme NAO" in the sense of both pronounced positive and negative phases of the NAO (absolute NAOI value greater than 1). We have changed the word 'extreme' to read 'pronounced'.

18. p 3146, l 14-16: I find that the points the authors are making could be stated better. I find point (ii) somewhat misleading as stated and in fact the authors seem to contradict it in the next few sentences. Many studies show STT has an appreciable effect at the surface. This point could be clarified somewhat. In addition, in regard to point (i) the 'smog' photochemical reactions are likely to influence the ozone throughout the depth of the troposphere. Please clarify point (i).

Thank you for the comments. In light of the new CO analysis, we are rewritten the paragraph as shown to answer 1) to Reviewer #2.

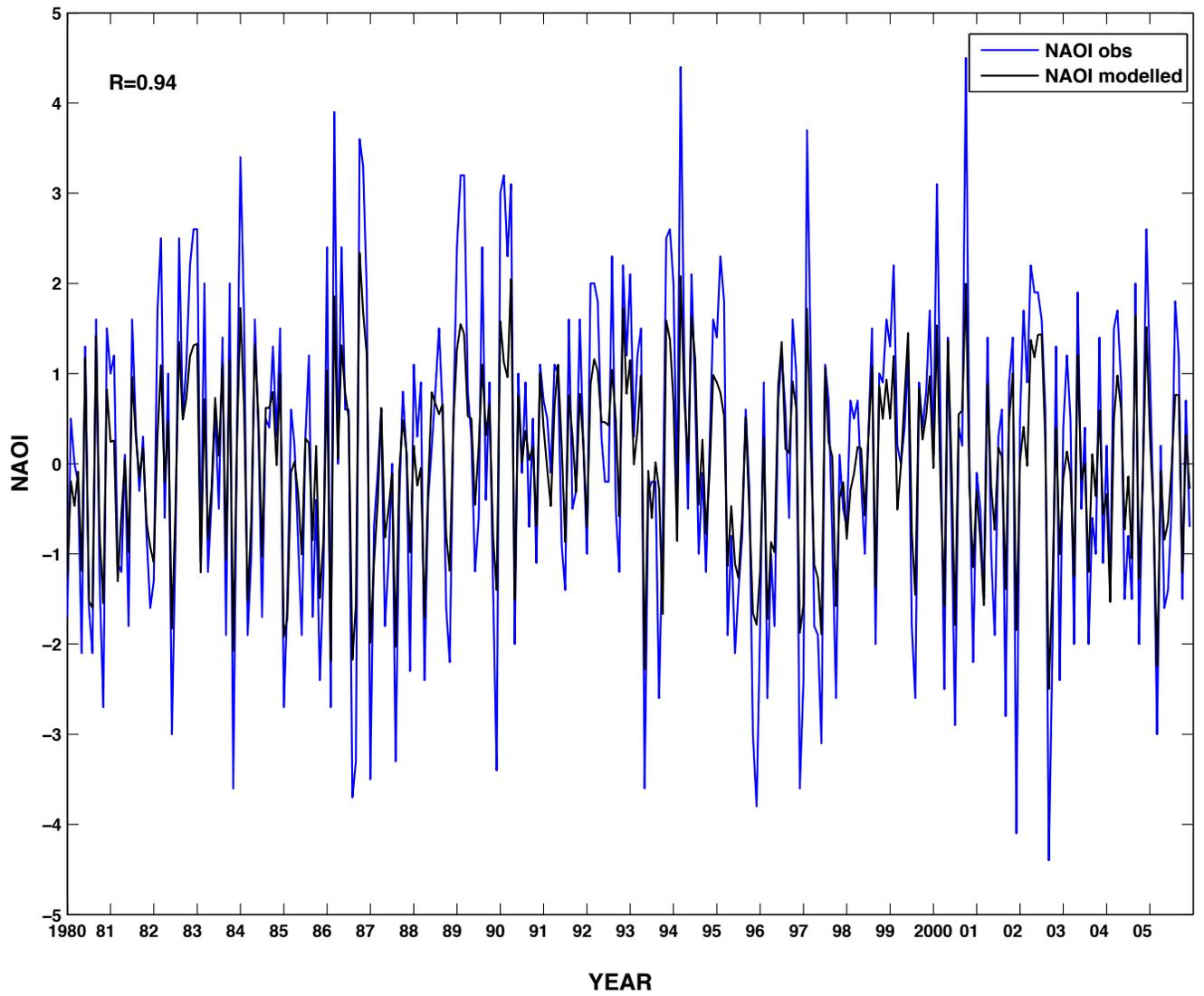


Fig. R1: Monthly station based (blue) and modelled (black) NAOI.

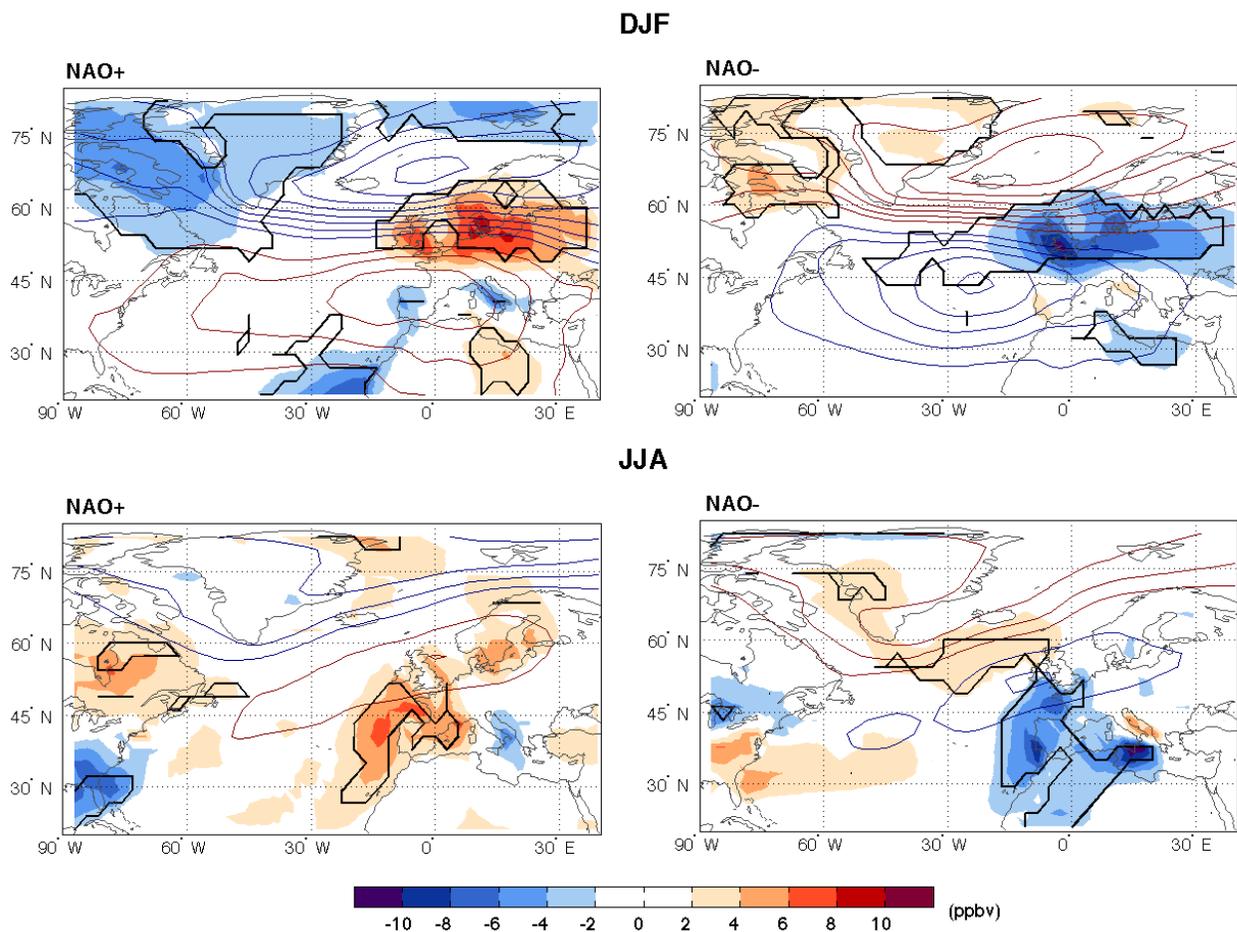


Fig. R2: Positive (red contours) and negative (blue contours) SLP and surface O₃ concentration (color shading) anomalies associated with the ensemble average of positive (right) and negative (left) NAO phase winter (top) and summer (bottom) months. Contours have 1.5 hPa interval. The bold black contours indicates O₃ anomalies significant with 90% confidence.

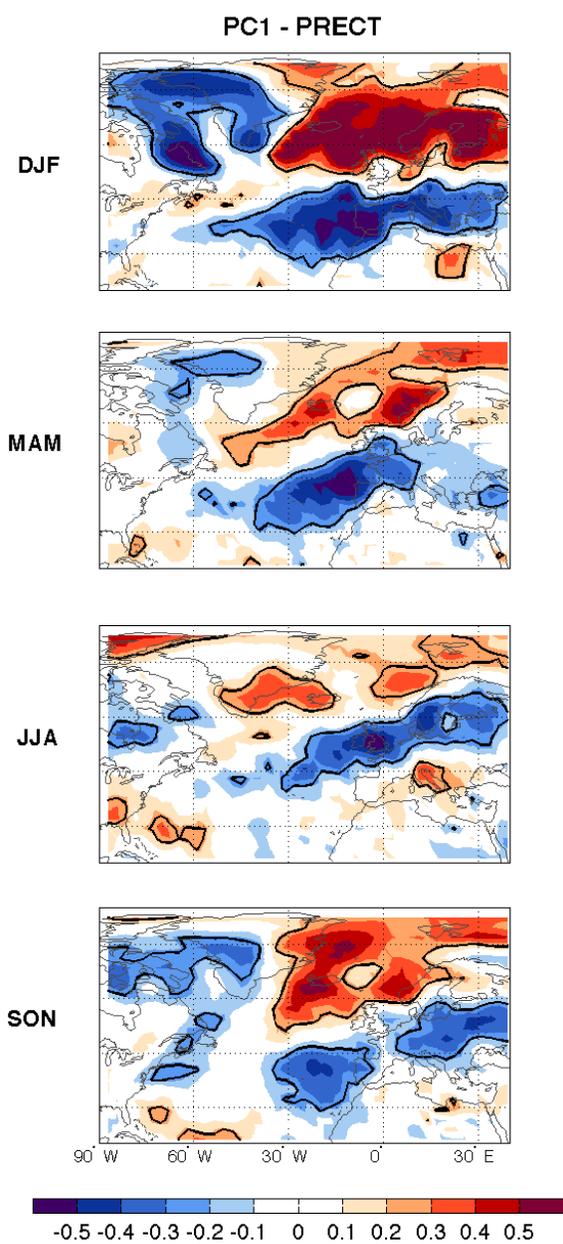


Fig. R3: Correlation between PC1, and surface O₃ concentrations for each season. The contours indicate where the correlation is significant with 90% confidence.

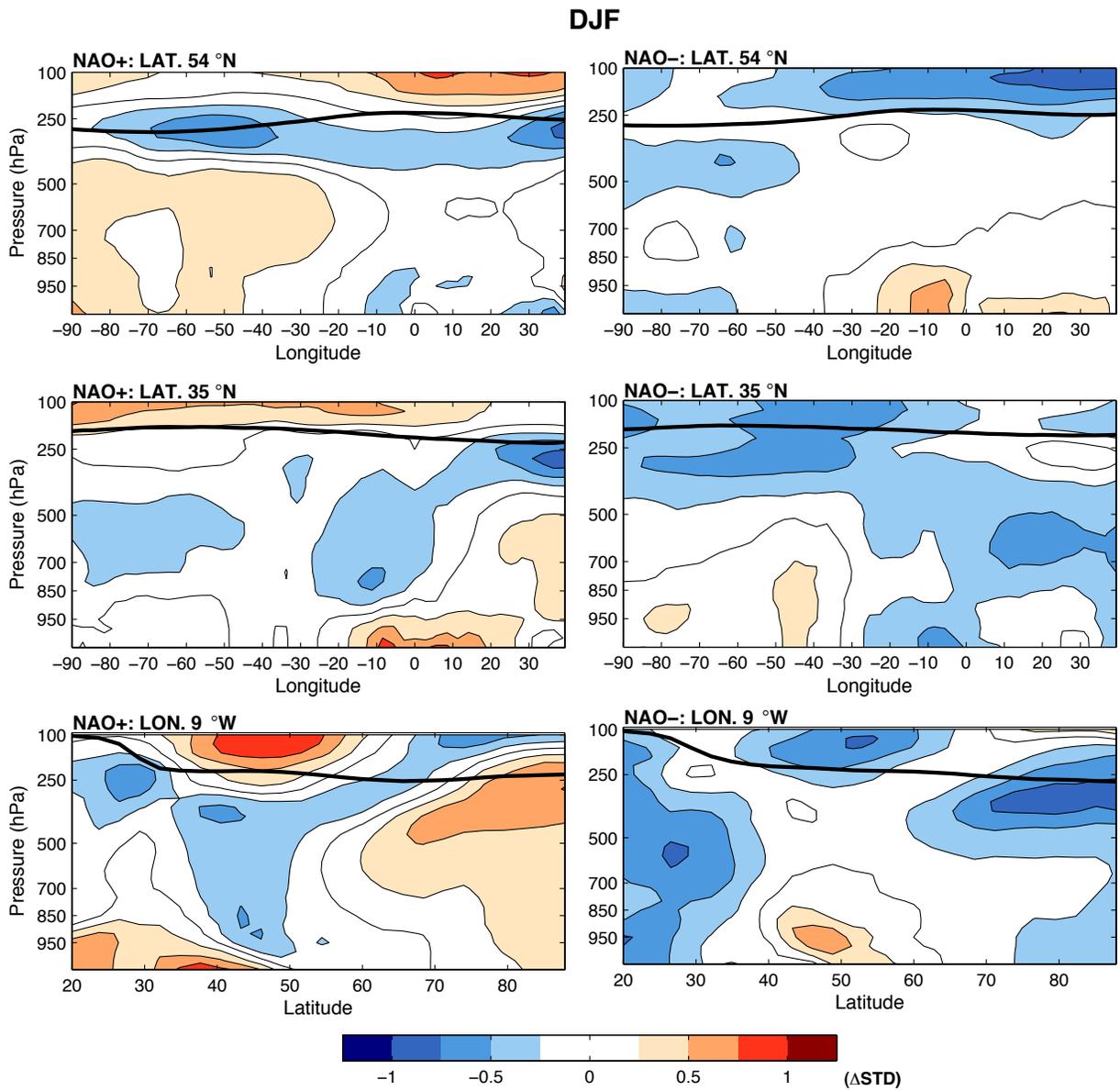


Fig. R4: Transect of normalized CO concentration anomalies for a given latitude (upper and middle panels) and longitude (lower panels) as a function of pressure in winter. CO concentration anomalies are normalized by CO standard deviation. Thick solid line indicates the tropopause height.

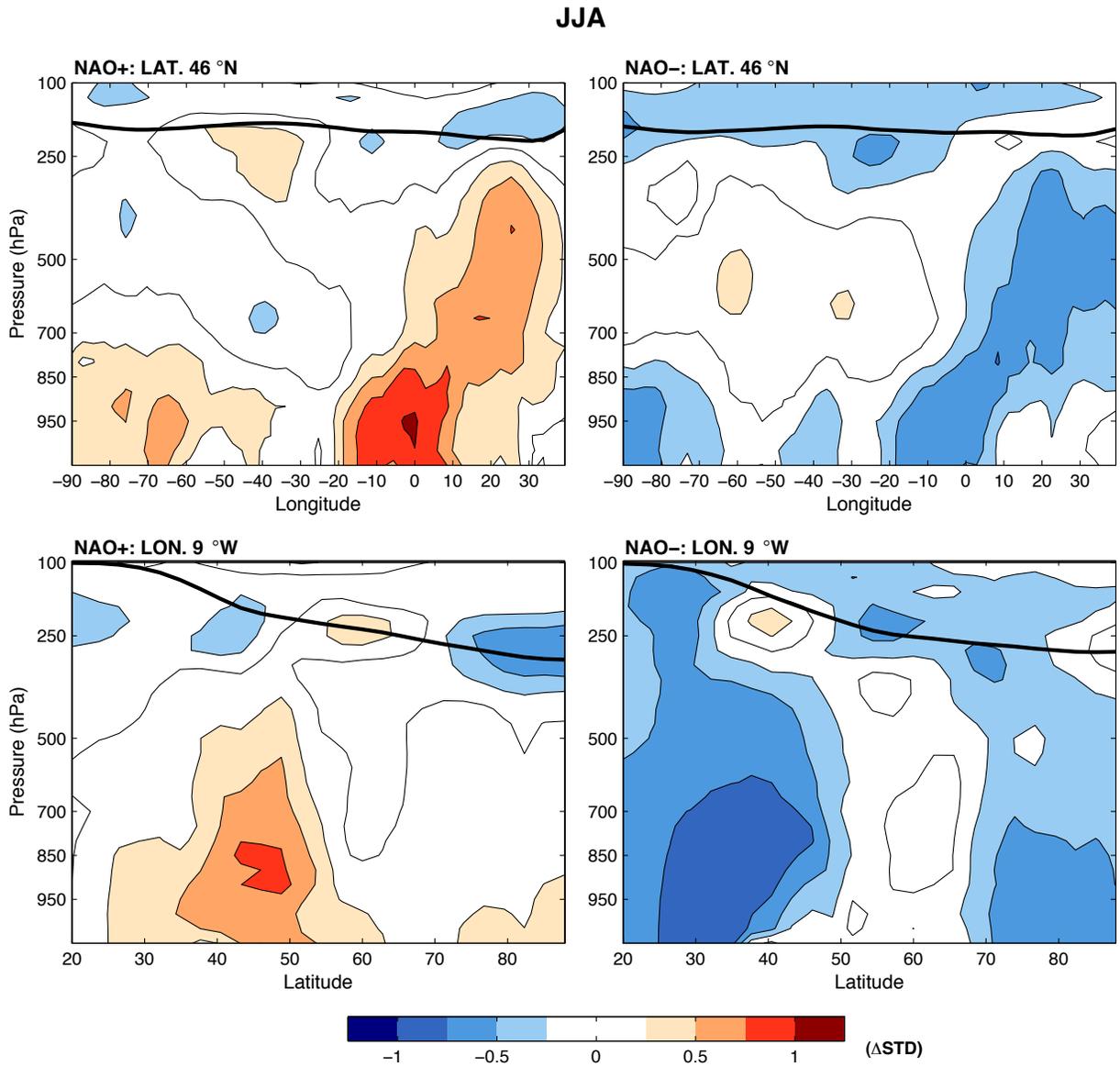


Fig. R5: Transect of normalized CO concentration anomalies for a given latitude (upper panels) and longitude (lower panels) as a function of pressure in summer. Δ concentration anomalies are normalized by CO standard deviation. Thick solid line indicates the tropopause height.

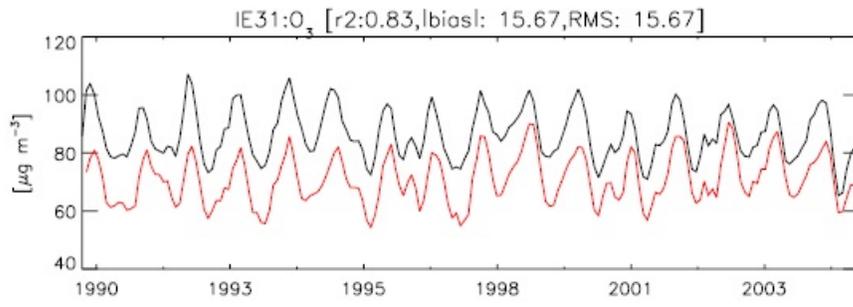


Fig. R6: O₃ seasonal cycle at Mace Head for observation (red) and model (black).

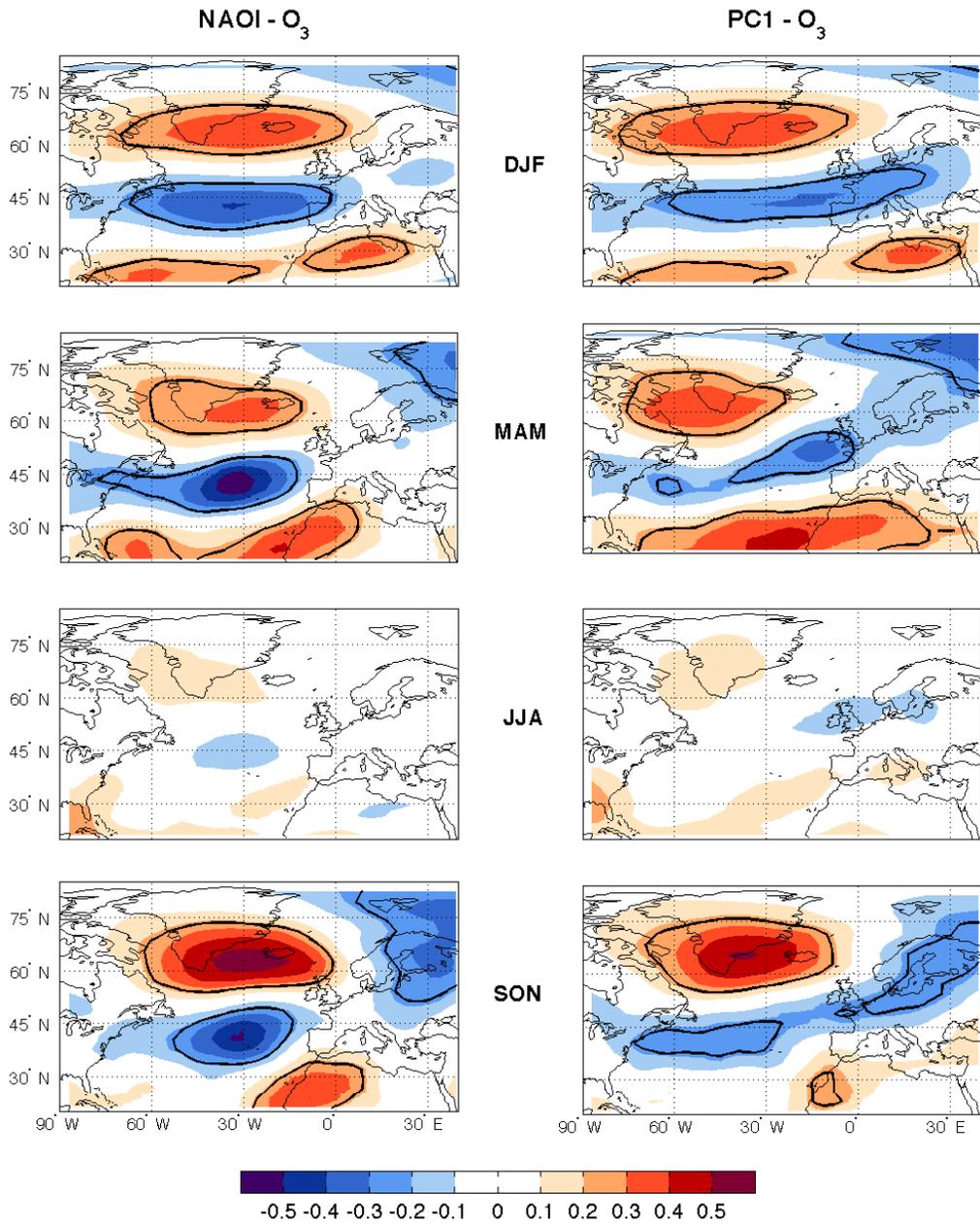


Fig. R7: Correlation between PC1, and total O₃ concentrations for each season. The contours indicate where the correlation is significant with 90% confidence.

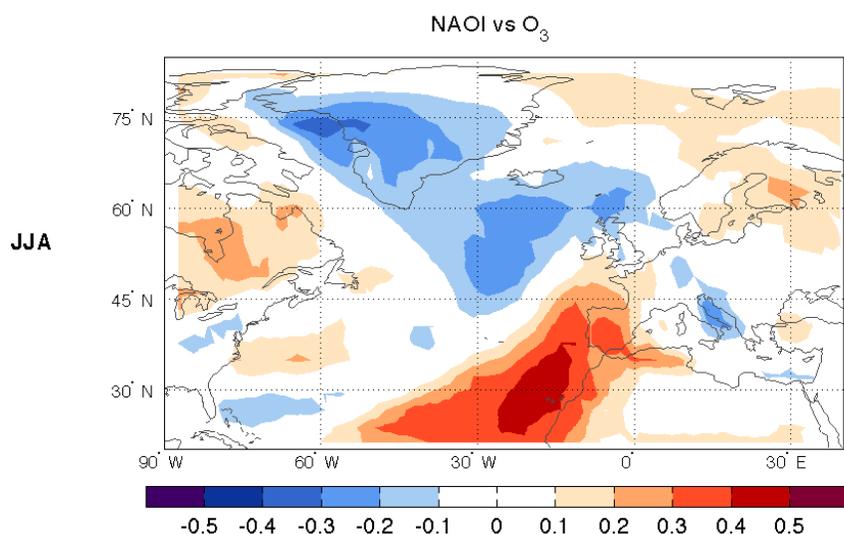


Fig. R8: Correlation between NAOI calculated using the new locations and surface O₃ concentrations in summer (JJA).

References

- Appenzeller, C. Weiss, A. K., and Staehelin, J.: North Atlantic Oscillation modulates Total Ozone Winter Trends, *Geophys. Res. Lett.*, 27, 1131–1134, 2000.
- Barnston, A. G. and Livezey, R. E.: Classification, seasonality and persistence of low-frequency atmospheric circulation patterns, *Mon. Weather Rev.*, 115, 1083–1126, 1987.
- Guerova, G., Bey, I., Attié, J. L., Martin, R. V., Cui, J. and Sprenger, M.: Impact of transatlantic transport episodes on summertime ozone in Europe, *Atmos. Chem. Phys.*, 6, 2057–2072, 2006.
- Hurrell, J. W.: Decadal trends in the North Atlantic Oscillation: regional temperatures and precipitation, *Science*, 269, 676–679, 1995.
- Hurrell, J. W., Kushnir, Y., Ottersen, G., and Visbeck, M.: An overview of the North Atlantic Oscillation. In *The North Atlantic Oscillation: Climatic Significance and Environmental Impact*, AGU monograph, 13, 1–35, 2003.
- Pausata, F. S. R., Li, C., Wettstein, J. J., Nisancioglu, K. H., and Battisti, D. S.: Changes in atmospheric variability in a glacial climate and the impacts on proxy data: a model intercomparison, *Clim. Past*, 5, 489–502, 2009.
- Pausata, F. S. R., Li, C., Wettstein, J. J., Kageyama, M., and Nisancioglu, K. H.: The key role of topography in altering North Atlantic atmospheric circulation during the last glacial period, *Clim. Past*, 7, 1089–1101, 2011.
- Pozzoli, L., Janssens-Maenhout, G., Diehl, T., Bey, I., Schultz, M. G., Feichter, J., Vignati, E., and Dentener, F.: Re-analysis of tropospheric sulfate aerosol and ozone for the period 1980–2005

- using the aerosol-chemistry-climate model ECHAM5-HAMMOZ, *Atmos. Chem. Phys.*, 11, 9563-9594, 2011.
- Rast, J., Schultz, M., Aghedo, A., Bey, I., Brasseur, G., Diehl, T., Esch, M., Ganzeveld, L., Kirchner, I., Kornbluh, L., Rhodin, A., Roeckner, E., Schmidt, H., Schroede, S., Schulzweida, U., Stier, P., and van Noije, T.: Evaluation of the tropospheric chemistry general circulation model ECHAM5-MOZ and its application to the analysis of the inter-annual variability in tropospheric ozone from 1960–2000 chemical composition of the troposphere for the period 1960–2000 (RETRO), MPI-Report (Reports on Earth System Science), in preparation.
- Stohl, A., Bonasoni, P., Cristofanelli, P., Collins, W., Feichter, J., Frank, A., Forster, C., Gerasopoulos, E., Gaggeler, H., James, P., Kentarchos, T., Kromp-Kolb, H., Kruger, B., Land, C., Meloen, J., Papayannis, A., Priller, A., Seibert, P., Sprenger, M., Roelofs, G. J., Scheel, H. E., Schnabel, C., Siegmund, P., Tobler, L., Trickl, T., Wernli, H., Wirth, V., Zanis, P., and Zerefos, C.: Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO, *J. Geophys. Res.-Atmos.*, 108(D12), 8516, 2003.