

***Interactive comment on* “Influence of the North Atlantic Oscillation on air pollution transport” by T. Christoudias et al.**

T. Christoudias et al.

christoudias@cyi.ac.cy

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Dear Editor,

We would like to thank the referees for their careful reading of our manuscript, acp-2011-633, "Influence of the North Atlantic Oscillation on air pollution transport".

We are most grateful for the comments, constructive criticism and very useful suggestions received on how to improve the paper.

Please find attached our detailed answers to the questions and a new version of the paper, which we hope satisfactorily address the points raised during the discussion.

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1 Anonymous Referee #1

The study by Christoudias et al. analyzes the influence of the NAO on air pollution transport, notably modifications of transport from North America and from Europe. There have been only very few studies dedicated to this topic so far and I therefore consider the present study as a valuable contribution. There had been a few studies on modulations of African dust transport by NAO which should probably be acknowledged in the present manuscript as well (e.g. Moulin et al. published in Nature 1997). The study is based on a 50-year model simulation to include several positive and negative NAO periods in order to deduce robust patterns. The model simulates the transport and loss of two idealized tracers representing i) a non-soluble CO-like gas and ii) a water soluble aerosol-tracer to assess not only changes in transport pathways but also the influence of changes in precipitation. This setup is suitable to address the scientific questions asked. The manuscript is concise and well-structured and overall rather well written.

Added citation to Moulin et al. (Nature 1997)

The main problem with the study, however, is that it is very weak and sometimes lacking regarding data interpretation and conclusions. The reader is left with a strong feeling that more data analysis could have been performed and more firm and relevant conclusions could have been drawn. In fact, I do not agree with some of the interpretations of the simulated patterns and some relevant conclusions that should have been drawn appear to have been overlooked (see below).

I therefore would like to propose the following modifications before the manuscript is acceptable:

- In Sect. 2.2 it should be clearly stated that the model produces its own meteorology/climate variability, only forced by the SSTs, which is not identical to the true observed variability during the period 1960-2010, but that the characteristics

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of NAO variations (amplitude, frequency, duration of phases) are similar to the observations or describe the relevant differences if there are any. It is important for the reader to understand that the model produces its own but (presumably) realistic NAO. The title of Sect. 2.2 should be changed accordingly to “North Atlantic Oscillation as represented by EMAC”.

Added paragraph to highlight the fact that the NAO variability in our study is model-generated and changed the title of Sec. 2.2 to reflect this.

- Section 3.2, which is the key section of the manuscript, should be strongly revised and the discussion should be enhanced. Abstract and conclusions should be adapted accordingly.
 1. Section 3.2 (p 25975, l 18) suggests that correlation patterns in Fig. 7 are generally reversed between CO and aerosols, which is not the case (or I don't understand what the author is trying to say). For both CO and aerosols the surface concentration anomalies are positively correlated with the first EOF over the northern part of the North Atlantic and negatively over the southern part. I don't see a reversal in this pattern.

Indeed, for both CO and aerosols, the anomalies are correlated over the North and anti-correlated over the South Atlantic. Removed all instances from the text.
 2. Section 3.2 (p 25975, lines 22-24): Figure 7 shows an anticorrelation (blue colors) for aerosols for the equatorial region and extending to Africa. The interpretation presented, namely that this is due to increased moisture and precipitation over the tropics for seasons with low NAO, is incorrect. An anticorrelation with NAO means that aerosol concentrations during low/negative NAO phases are elevated, not reduced, thus opposite to what would be expected from enhanced precipitation. Under negative NAO conditions transport is directed more strongly to the east and southeast as opposed to a more northeastward flow under high NAO. I think this is nicely seen in the

CO tracer at least over the western parts of Africa where the blue colors suggest elevated CO during negative NAO conditions. However, the correlation for CO turns into a positive one over the eastern parts of Africa. This is interesting but not commented. To me this looks like enhanced transport of North American CO during high NAO along an anticyclonic path first leading towards northern Europe and then southwards to eastern Europe and finally to Africa. I think this is understandable since a stronger Azores High during high NAO leads to enhanced north to south transport over Eastern Europe. This pathway has a different effect on the water-soluble tracer probably because of efficient wash-out along this path even leading even to reduced aerosols during high NAO.

Added discussion to the text.

3. Section 3.2. The discussion of the patterns of the European tracers in Fig. 8 is very lacking. CO and aerosols are reduced over most of central Europe (blue colors) during high NAO conditions. The paper does not discuss this feature, nor does it explain the positive anomalies over northern Africa and the Arctic, nor does it explain the strongly negative correlations over the eastern North Atlantic. I think the story is actually quite simple: During a positive NAO both the Icelandic Low and the Azores High are more pronounced than normally. This leads to a stronger separation of the flow over Europe, with enhanced northward transport over the northern parts of Europe and enhanced southward transport over the southern parts. The European tracer is thus depleted over central Europe, as it is either moved rapidly towards southern Europe and northern Africa (creating positive anomalies there) under the influence of the Azores High, or rapidly to the Arctic under the influence of the Icelandic low. During low NAO conditions, conversely, the air is more stagnant over Central Europe allowing European pollutants to accumulate. The strong negative anomaly over the western Atlantic is simply an effect of reduced direct outflow of European air pollution to this

region due to the enhanced north-south pressure gradient and associated westerlies. These points should be discussed.

Added discussion to the text.

Minor points:

- P 25968, line 3: suggest to change to “transport and removal of idealized insoluble gaseous and ..”

Added “idealized”.

- P 25969, line 20: Please explain the advantage of simulating such a long time period.

Added sentence to indicate advantage of long time period simulation.

- P 25971, line 8: Add a comma in “.. dry and wet deposition processes, respectively.”

Added comma.

- P 25971, line 19: “extend” → “extent”

Corrected.

- P 25972, line 3: What are biogenic emissions used for? To my knowledge CO has no biogenic sources, at least not directly.

Although Carbon Monoxide is not directly emitted from plants (as the referee correctly mentioned), it is indeed emitted by processes that can be considered biogenic. Schade and Crutzen (1999) estimate global sources of 30-90 Tg CO/yr for photochemical production from dead biomass, and 40 Tg CO/yr for thermal decomposition of dead organic matter. Additionally, ~ 13 (6-25) Tg/yr of CO are emitted by the ocean (see Bates et al (1995), Bergamaschi et al (1999)).

1. Schade, G. W., Hofmann, R.-M. and Crutzen, P. J. (1999), CO emissions

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from degrading plant matter. *Tellus B*, 51: 889-908. doi: 10.1034/j.1600-0889.1999.t01-4-00003.x

2. Bates, T. S., K. C. Kelly, J. E. Johnson, and R. H. Gammon (1995), *Regional and seasonal variations in the flux of oceanic carbon monoxide to the atmosphere*, *J. Geophys. Res.*, 100(D11), 23,093-23,101, doi:10.1029/95JD02737.
3. Bergamaschi, P., R. Hein, M. Heimann, and P. J. Crutzen (2000), *Inverse modeling of the global CO cycle 1. Inversion of CO mixing ratios*, *J. Geophys. Res.*, 105(D2), 1909-1927, doi:10.1029/1999JD900818.

- P 25972, lines 15-22. It seems to me that the only chemical reaction actually needed is the loss reaction $\text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H}$. We are not interested in the further fate of H as it is not modeled in this version. The reactions as they are stated now are simply wrong as there is no stoichiometry between the left and right-hand sides.

The CO reaction with OH and the cycle to reproduce OH occur on a much shorter time scale than the model time-step. The chemical integration between timesteps would thus produce a decrease (consumption) of the OH reactants, unless they are artificially kept constant. The reaction presented in the paper (even though stoichiometrically invalid) does not remove OH during the chemical integration time. This is not a problem as we are only interested in the decomposition of CO and not in any other component. The global atmospheric distribution of OH, is re-initialised at every time-step and nudged towards prescribed values.

- P 25974, line 10-11: I think in this equation the variable q actually refers to “mass mixing ratio in units of kg/kg”, certainly not to “concentration” which would have units of mass per volume. This can easily be seen when dp is replaced by $-\rho * g * dz$ assuming hydrostatic equilibrium, where rho is the density of air. Changed “concentration” to “mole ratio”.

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- P 25974: Why is high NAO associated with an EOF component (> 1) and low NAO with (< 0)? Why not symmetric (> 0.5) and (< -0.5)? Figure 4 suggests that high and low NAO phases are quite symmetric about the zero-line.

The NAO index is symmetric about zero by definition. We use asymmetric very high (> 1) and normal or low (< 0) phases following the method used by Hurrell (1995) to obtain results that are directly comparable to the published observations.

2 Anonymous Referee #2

This is a very cleanly designed and well written manuscript that diagnoses the impact of North American (NA) and European (EU) pollution sources on the surface in Europe, dividing the patterns of transport into positive and negative phases of the NAO. It is a valuable contribution to the community and fits the mandate of ACP. There are some weaknesses that can and should be fixed before publication.

1. The HTAP assessment (see below) did extensive multi-model evaluations of NA and EU sources of pollution (actually, they did the full O₃ calculation, which is only minimally approximated here as tagged CO tracer). They also discussed the NAO's impact (see quote below). This paper must reference that document and the suitable references in it (even though it is 'grey literature').

Added reference to the HTAP assessment and suitable references in the Introduction.

2. The use of tagged tracers is always suspect when dealing with active tropospheric chemistry as they can/should influence the chemistry of themselves and other pollutants. The use of monthly mean OH values, as used here, would be suspect except that the problem of equating CO (with lifetime of 2-4 months) with

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O3 (2-4 weeks) is a bigger approximation. Given the CO lifetime, the fixed OH is probably OK, unless we are looking at some numerical derivatives of the chemistry. In fact, the use of CO is interesting as can be seen in the Figs 7&8, the NA and EU lat-alt patterns are almost the same because the CO has gone around the world several times, and what we are seeing is the shift on the north-south latitudinal gradient with the NAO, not really the short-term pollution transport.

We prescribe OH to prevent it from being depleted by the reaction with OH as we are not pursuing a comprehensive chemistry study.

3. (page 3) I cannot understand the "simplification $\text{OH} \Leftrightarrow \text{HO}_2$ " – or the nudging since OH is fixed? Explain better.

We agree that mentioning the simplification $\text{OH} \Leftrightarrow \text{HO}_2$ may have been confusing and have reformulated the text.

4. (page 4) On the vertically integrated transport it is important to know the time scale of the integration. If the authors use monthly means for these transports, then they then I do not believe the fluxes can be correct since they would miss the big events. I presume that these are integrated hourly and this should be stated.

We produce 10-hourly output, which we average and aggregate over the entire simulation period, which we expect to provide adequate statistical analyses.

5. Fig 6 – I am having trouble with the units here. Mole fraction is the abundance of CO, but the fluxes should have units of mass per interval (kg/m/s?)

The units are derived from the eqn: $\vec{Q} = \frac{1}{g} \int_0^{ps} q\vec{u} dp$ as: $\frac{\vec{u}[\text{m/s}] \times dp[\text{Pa:kg/m.s}^2]}{g[\text{m/s}^2]}$ which simplifies to kg/m.s.

6. For the aerosols, the correlation maps are misleading if the absolute abundance of aerosols is so small as to be trivial. The free-trop abundances are trivial (see HTAP results), especially at 200 hPa, so the plots in Fig 7&8 give a mistaken impression.

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