

Dear Editor,

The authors would like to thank the referees for their very thorough and constructive reviews. We believe that the paper is much improved as a result of addressing these referees' comments. In particular, we have clarified the text to address both referees' comments on the background conditions. We also request during typesetting the figures are printed as large as possible, as asked by the reviewers.

Our responses follow each specific comment of the referees with any changes to the text shown in italics.

Referee #1

The study of Knowland et al. examine the impact of extra-tropical storms in the North Pacific and North Atlantic on springtime O₃ and CO distribution. The authors considered the most intense (95th percentile) storms (60) in each basin and focused on the period 2003-2012, using MACC reanalysis. By composing the storms, the study quantifies the changes in CO and O₃ horizontally and vertically throughout the storm. The analysis is thorough and the manuscript is in general well written. The results are interesting; however, I think the authors should present more analyses related to the implication of their results (see major comment below), before being published in ACP.

We address this comment below.

Major comment

Referee's comment: "Hess and Lamarque (2007) have shown how ozone at the surface is modulated by the Arctic oscillation during the spring months and Pausata et al., (2012) have shown that the NAO is driving ozone variability in large parts of Europe, both at low levels and in the middle-upper troposphere, suggesting that increased baseline ozone over western and northern Europe may be due to the prolonged NAO phase during the 90s. It would be interesting whether the authors would expand the part related to the implications of their results: For example, better quantifying – together with what has been suggested by Referee #2 – the effects of stratospheric ozone transport at the surface and how this could impact the trend seen in the mid-high latitudes of the Northern Hemisphere.

It would be interesting to investigate in which phase of the NAO these stronger storms develops and then analyze those that impact western and northern Europe (e.g. Mace Head) and a similar analysis could be done for the pacific storms. The authors show that at the end of the DI there can be O₃ anomalies at the surface of 5 ppb. May an increased number of such strong storms (during NAO + ?) have an impact on baseline ozone and influence trends over western and northern Europe."

Author's comment and changes in manuscript: We thank the referee for these suggestions. We address Referee 1's comment on the effects of stratospheric ozone transport in our response to Referee 2 who asks about this in detail.

We are aware of the studies above on the NAO and this is a good point. However we do not find a correlation between the frequency of intense storms and the NAO (Pearson correlation of -0.02 when comparing the monthly frequency of MAM 95th percentile NA storms to the monthly Hurrell pc-based NAO index). We do find a weak correlation between the frequency of the strong storms and indices over the Pacific (Pearson correlation of 0.33, 0.19, -0.10 when comparing the monthly frequency of MAM 95th percentile NP storms to monthly PNA, PDO, SOI indices, respectively). Hence, we do not believe that the phase of the NAO is impacting the cyclogenesis of these very intense springtime storms i.e. that intense storms preferentially form under a given NAO phase. The

phase of the NAO would likely influence the direction of storm propagation and eventually where in Europe the storm would impact. However, this study focuses on the distribution around the storm centers at the time of maximum vorticity which occurs mainly over the middle of the oceans. We are currently preparing a manuscript where we investigate the relationship between the NAO, storm direction and composition, storm tracks in all four seasons over the North Atlantic sector. We have added a second paragraph to Sect. 3.1 discussing this result:

Eckhardt et al. (2004) found that WCBs during strong positive NAO winter months are generally more frequent over the North Atlantic (about 12%) and are more frequent over northern Europe compared to strong negative NAO winter months where WCBs are more frequent over the western North Atlantic. The frequency of the intense springtime storm tracks in each region was tested against different indices to investigate if stronger storms develop during particular phases. There was no correlation between the frequency of the MAM 95th percentile NA storm to the monthly PC-based NAO index of Hurrell et al., 2003. There are weak/no correlations between the frequency of the MAM 95th percentile NP storms to the monthly Pacific North American (PNA: downloaded from the NOAA Climate Prediction Center) and to the monthly Pacific Decadal Oscillation (PDO: downloaded from <http://jisao.washington.edu/pdo/PDO.latest>) respectively. These most intense storms do not show a robust relationship to these teleconnection indices, however the sample size (~ 60 storms over each region) is small and the criteria to select the storms was not limited to the months of strong NAO phases. Further study is required to investigate the spatial relationship between storm tracks and composition (Knowland et al., in prep.).

Minor comments

Referee's comment: "I feel the introduction needs some further discussion. For example, the authors state that meteorological conditions play an important role in the intercontinental transport of pollutants citing a viewpoint (Akimoto, 2003) rather than the studies that have shown that. Here below I suggest some studies that would be relevant to discuss (in the last section of the manuscript as well) in relation to the intercontinental transport of pollutants and also in light of my previous comment on NAO.

Long range air pollutant transport in general: Duncan and Bey, 2004; Hess and Mahowald, 2009; Christoudias et al., 2012; Pausata et al., 2013.

Long range Ozone transport specifically: Lelieveld and Dentener, 2000; Creilson et al., 2003; Lamarque and Hess, 2004; Pausata et al., 2012."

Author's comments and changes in manuscript: Akimoto (2003) was used primarily as a reference to the fact that O₃ and CO have long enough lifetimes to be transported on hemispheric scale. We do reference other studies that report on the long-range transport of pollutants (Pg 27096 Ln 25-29, Pg 27097 Ln 11) but had focused on those studies that reported a link with storm processes explicitly: such as movement of pollutants in the warm conveyor belt. As suggested, we have now added further introductory text on pollution transport pathways and these references to the Introduction (Pg 27097 Ln 11):

Furthermore, the different transport pathways of constituents can be linked to atmospheric circulation changes associated with different phases of teleconnection patterns such as the North Atlantic Oscillation (NAO), El Nino/Southern Oscillation (ENSO), and the Pacific-North American (PNA) pattern (e.g. Li et al., 2002; Creilson et al., 2003; Eckhardt et al., 2004; Lamarque and Hess, 2004; Christoudias et al., 2012; Pausata et al., 2012, Lin et al., 2014).

Referee's comment: "It is not completely clear to me the definition and the choice of the background averaged conditions. The authors should better clarify it."

Author's comments:

Referee #2 also asked for more clarification on the selection of the background conditions and raises issues of interannual variability. We hope to satisfy both referees with the additional text as outlined in our responses to Referee #2's comments.

Referee's comment: "Some figures are too cluttered to be well understood. I would suggest:

- a) Reducing the number of panels specifically in figures 4, 5 and 6.
- b) I would place the O₃ in NP close to the O₃ in NA and CO in NP close to CO in NA for a better comparison of the two basins.
- c) Please fix the colorscale in some figures since one cannot distinguish the contours (e.g., Figs. 8 and 9).
- d) In figures 2 and 3, are all 12 levels really necessary? For example, having the O₃ concentration at 950-925 and 900 hPa does it add anything to the key message you want to convey?"

Author's comments and changes in manuscript:

a) There are currently six levels in the stacked distribution plots. We chose each level specifically to capture the different locations within the troposphere and lower stratosphere. We request the figures are presented as large as possible in the final published version.

b) In preparation of this manuscript we had explored the alternative presentation of the figures as the referee suggested but we found it is better to display the figures comparing the composition within air streams of the same composite storm.

c) We have revised the color scales in Figs 8-13 for clarity as suggested. Changes to the text have been made to reflect the changes in the color scale where necessary.

d) Figures 2 and 3 have been adjusted to show every 100 hPa from 1000 to 200 hPa; figure captions have been revised accordingly.

Referee #2

"Knowland et al. present an analysis of trace gas composition within intense North Pacific and North Atlantic mid-latitude cyclones. The authors apply two compositing methods to the MACC reanalysis CO and O₃ fields, along with meteorological parameters. The airstreams within a mid-latitude cyclone are clearly identified, despite being hidden behind some background gradients in CO/O₃, and changes in CO/O₃ are quantified. The analysis is straightforward and of interest to the community, even if the results are not terribly novel.

The manuscript is very well written and I suggest that it be accepted for publication in ACP after the authors address the minor issues listed below."

General Comments

Referee's comment: "The storm composites are adequately described, but not straightforward for the reader unaccustomed to their use. I recommend a figure showing the use of each composite method on a specific storm, which would much more clearly indicate the procedure used."

Author's comments and changes in manuscript: We have amended the text to point the reader to the schematic in Catto et al., 2010. Additionally, we have added a reference to Appendix A in Bengtsson et al., 2007 which is the paper that first described this method.

Pg 27102 Ln11: a spherical “cap” based on a given radius is used to define the area of coverage by the storm (*See Appendix A of Bengtsson et al. (2007) for full details on track compositing*).

Pg 27102 Ln27: *A schematic of the compositing method can be found in Figure 3 of Catto et al., 2010.*

Referee's comment: “More explanation and defense of the definition of the background “average conditions” is necessary. There is the potential for significant year-to-year variability in background values of both CO and O₃ in these regions and apparent signals could simply arise from a long-term trend in precursors, distant exceptional sources (biomass burning/lightning), or atypical transport pathways. An alternative would be to have a threshold relative vorticity value signaling the absence of a storm.

The latter figures (4-13) clearly show that the background definition is likely acceptable and I am ready to accept that these concerns regarding the background conditions are misguided, but evidence to this end is needed.”

Author's comments and changes in manuscript: We believe that the interannual variability of the emissions over the 10 year period and any possible trend in the precursors and transport pathways are included in the background composites as all ten years are still sampled and we have added such a statement to the text:

Pg 27104 Ln13-16 (sentence split in two): *The interannual variability of the emissions over the 10 year period is included in the “average condition” composites. The sensitivity to different offset time-periods requires further study.*

We argue that an alternative method based on a threshold relative vorticity value would in practice be more subjective and would not likely add additional information to the background. The method to calculate background composites has been previously used by Grandey et al., 2011.

As the referee has noted, the figures show the background definition is “likely acceptable”. We rely on the meteorology fields to simulate a background environment without storms. We have amended the text to forward reference Fig. 5 which clearly shows the background environment is without the influence of storm dynamics:

Pg 27104 Ln10: *These meteorological conditions can be clearly seen in Fig 5.*

Referee comment: “While the difference between the storm and the background state is clear in Figures 4-13, this is not the case in Figures 2-3. In addition, the definition of the background state is essential to properly quantify anomalous CO and O₃.”

Author's comments and changes in manuscript: The referee is correct that there are smaller differences in the area-averaged O₃ and CO between the storm-centered composites (Fig. 2) and the background composites (Fig. 3) because we are taking an average over a large area. As we state on Pg27105 Ln19-21, the differences in the area-averages indicate the substantial influence of the storm dynamics on the composition. To strengthen the discussion between the storm-centered

composite life cycle (Fig. 2) and the background composite life cycle (Fig. 3), we have added text focusing on the gradients in the aavg-O₃ and aavg-CO:

Pg27106 Ln 13, we have changed “greater increase” to “steeper gradient” that it now reads: *In the storm-centered life cycle plots, there is a steeper gradient compared to the background composites in aavg-O₃ at the upper levels (200 and 300 hPa) and mid-tropospheric levels (400 and 500 hPa; Fig. 2a and c, Fig. 3a and c) from about 2 days prior to maximum ζ₈₅₀ until about a day after.*

Pg27106 Ln 17, we have modified the sentence to now read: *The fact that the storm-centered composite aavg-O₃ is above or just below 150 ppb at 400 hPa in both regions around the time of maximum ζ₈₅₀ (Fig. 2a and c) while the background composite aavg-O₃ is well below 150 ppb at 400 hPa (Fig. 3a and c) indicates that stratospheric air can reach to below 400 hPa as part of the DI in these intense storms.*

In order to differentiate better between the storm-centered and background composite lifecycle plots, we have removed the ζ₈₅₀ black line from Fig. 3. Therefore we have added “Fig. 2” to the (black line) on Pg 27105 Ln 11.

Referee’s comment: “The figures are well constructed when viewed on the computer screen, but much harder to interpret when printed. Please make sure that the printed final versions of these figures are large enough to do them justice.”

Author comments: We request that the journal prints the figures as large as possible in ACP format.

Referee’s comment: “Section 3.2 describes the change in ozone and CO within during an intense storm and in the background composite. Many of the features are the same: an increase in ozone and decrease in CO at altitude with minimal changes near the surface. Perhaps the discussion of the upper atmosphere (pressures lower than 400hPa) should be dropped? The effect of the DI is clear on this data, it makes the upper changes in CO/ozone stronger, but it also hides near-surface changes. In my opinion, it is the 1000-500hPa airflows that are of greatest interest. The latter figures (4-13) are much more convincing.”

Author comments: The main conclusion in Sec. 3.2 is the greater increase in O₃ and greater decrease in CO at upper levels in the troposphere seen by the composite storm’s interaction with the stratosphere. The lifecycle plots are able to give some indication as to the change in composition while the storm grows and decays. Hence we decided to leave this text which has been amended for clarity as described in the comment above.

Referee’s comment: “The airstream analysis is very interesting. I wonder if it can be expanded to provide an estimate of a strat-trop flux? This would be of great interest to the community and add a novel result to this manuscript.”

Author comments: We thank the reviewer for this comment. We have carefully considered what additional work we could do as we agree this would be interesting, but it could in itself be another paper due to the complexity of STE dynamics (see Reutter et al., 2015, ACPD). In the MACC dataset there are limited meteorological variables available. However, the MACC and ERA-Interim reanalyses are produced from very similar IFS versions. We find that the MACC and ERA-interim data for the same period pick up the same intense storms. Hence we have sampled the potential vorticity (PV) on pressure levels from the ERA-Interim dataset within the 20° spherical cap of the most

intense springtime storms. We diagnose the storm-centered composite dynamical tropopause and the background composite dynamical tropopause by the widely accepted 2 PVU level (e.g., Holton et al., 1995) in Figs. 4,5,8-10 and 12. (1 PVU = $10^{-6} \text{ m}^2 \text{ kg}^{-1} \text{ s}^{-1}$)

We have added the following text to Sects. 2.1, 2.3, 3.3, and 3.4 and revised the appropriate figure captions. The inclusion of a dynamical tropopause in addition to the chemical tropopause allows us to make firmer statements about the location of stratospheric air and likely STE.

Section 2.1

Pg 27100 Ln 26: Add a last sentence. *The MACC reanalyses did not archive potential vorticity (PV) data. TRACK detects the same intense storms in the ERA-Interim reanalyses for the 2003-2012 period as in the MACC reanalysis (not shown), therefore we used PV data from ERA-Interim to determine the dynamical tropopause at PV=2 PVU (e.g., Holton et al., 1995).*

Section 2.3

Pg 27103 Ln 16: Add a last sentence. *To diagnose the dynamical tropopause, the ERA-Interim reanalysis PV on pressure levels was composited.*

Section 3.3

Pg 27108 Ln 18: After second sentence add *“The dynamical tropopause outlined in white shows that the high O₃ and low CO values clearly depict air in the stratosphere.”*

Pg 27108 Ln 27: amend to *“At upper levels (200 and 300hPa), the storm centred composites (Fig. 4) and background composites (Fig. 5) depict air of mainly stratospheric origin with high O₃ and low CO (no emission sources) values, to the north of the dynamical tropopause (white line, Figs. 4 and 5).*

Section 3.4.1

Pg27111 Ln 19: *“Outside of the WCB, high values of O₃ are found at upper levels and at the storm centre and whose spatial patterns closely aligned with the tropopause delineation including the dip of the tropopause at the storm centre which may induce stratosphere-troposphere exchange. O₃ - poor...”*

Section 3.4.2

Pg 27113 Ln 3, after Fig. 12.: *“O₃-rich air from the stratosphere descends in the DI to ~400 hPa...”*

Pg 27113 Ln 12: *“Above 500 hPa, the stratospheric air has descended in alignment with the tropopause pattern but does not follow the isentropes (Fig. 10a and c);”*

Referee’s Specific Comments

Referee’s comment: “Page 27094, Line 26 – When is the area-averaged ozone higher? During the passing of the storm? After it passes? This sentence is unclear.”

Author’s comments and changes in manuscript: All percentages given in the Abstract are stated for the area-averaged O₃ or CO at the time of maximum intensity (initially stated in line 19). We have changed the Abstract text Pg 27094 Ln 19 to clarify this:

“The influence of the storm dynamics compared to the background environment on the composition within an area around the storm center at the time of maximum intensity is as follows. Area-averaged O₃ at 300 hPa is enhanced by 50 and 36% and by 11 and 7.6% at 500 hPa, for the NP and NA regions, respectively.”

Referee’s comment: Page 27100, Line 13 – “due too low NO_x” should be “due to low NO_x”

Author's changes in manuscript: Text has been changed.

Referee's comment: "Figure 7 – The negative values should have a different dashed pattern than that used for the axis; the contours are easily lost."

Author's changes in manuscript: The negative values have been changed from dotted to long dashed contours. The Figure caption text on Pg 27135 Ln3 has been changed accordingly.

Referee's comment: "A relevant publication to include: Polvani and Esler (2007), Transport and mixing of chemical air masses in idealized baroclinic life cycles, *J. Geophys. Res.*, 112, D23102, doi: 10.1029/2007JD008555."

Author's changes in manuscript: We added the following text regarding the different life cycle types to the Discussion Sec. 4 on Pg 27116 Ln12:

Polvani and Esler (2007) used idealized tracer transport in different baroclinic life cycles to distinguish the transport and mixing of air masses within the troposphere and lower stratosphere in two types of cyclone development (LC1 and LC2). The WCB or DI are characterised by either anticyclonic (LC1) or cyclonic (LC2) isentropic advection. In reality, systems likely have both types of advection or may transition from one type to another (Thorncroft et al., 1993; Schultz et al 1998; Polvani and Esler, 2007). Our composites are likely a mix of both types, as we find anticyclonic advection at the end of the WCB, cyclonic motion around the storm center in both the WCB and DI in the mid-troposphere, and anticyclonic advection behind the cold front at lower levels. It may be of interest to select storms from the MACC reanalysis according to their development type to see if similar storm structure as Polvani and Esler is obtained. However, this would likely be difficult, due to the fact cyclones may change type during their development and they may have a hybrid structure making them difficult to associate with either of the two conceptual models.

New References:

Bengtsson, L., Hodges, K.I., Esch, M., Keenlyside, N., Kornblueh, L., Luo, J.J. and Yamagata, T. (2007). How may tropical cyclones change in a warmer climate? *Tellus A*, 59, 539-561.

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Eckhardt, S., Stohl, A., Beirle, S., Spichtinger, N., James, P., Forster, C., Junker, C., Wagner, T., Platt, U. and Jennings, S.G. (2003). The North Atlantic Oscillation controls air pollution transport to the Arctic. *Atmos. Chem. Phys.*, 3, 1769-1778.

Eckhardt, S., Stohl, A., Wernli, H., James, P., Forster, C. and Spichtinger, N. (2004). A 15-year climatology of warm conveyor belts. *J. Clim.*, 17, 218-237.

Grandey, B.S., Stier, P., Wagner, T.M., Grainger, R.G. and Hodges, K.I. (2011). The effect of extratropical cyclones on satellite-retrieved aerosol properties over ocean. *Geophys. Res. Lett.*, 38.

Holton, J.R., Haynes, P.H., McIntyre, M.E., Douglass, A.R., Rood, R.B. and Pfister, L. (1995). Stratosphere-troposphere exchange. *Rev. Geophys.*, 33, 403-439.

Hurrell, J., Kushnir, Y., Ottersen, G. and Visbeck, M. (2003). The North Atlantic Oscillation: Climate Significance and Environmental Impact, vol. 134. Eds. Geophysical Monograph Series.

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Thorncroft, C.D., Hoskins, B.J. and McIntyre, M.E. (1993). Two paradigms of baroclinic-wave life-cycle behaviour. *Q. J. R. Meteorol. Soc.*, 119, 17-55.