Reviewer 1 Responses

We thank the reviewer for his/her comments. We have responded to the comments below. The original comments are in black text, followed by our responses in red (edits to the manuscript are in blue italics).

1. "Even though the general ideas and the conclusions drawn from this study are clear, it is hard to judge the methodology followed in this analysis. For instance, the selection methodology of LWTs is unclear, i.e. which locations are used for this, and what are effectively the predominant wind patterns during winter/summer anticyclonic and cyclonic weather conditions. I find it unfortunate that meteorological data is not available from the AQUM model, considering its pivotal role in this study. Now such information needs to be inferred from figures like those showing the idealized tracers. Even a sketch with an overview of prevailing winds during cyclonic / anticyclonic conditions could be helpful for understanding the anomalies seen in NO2."

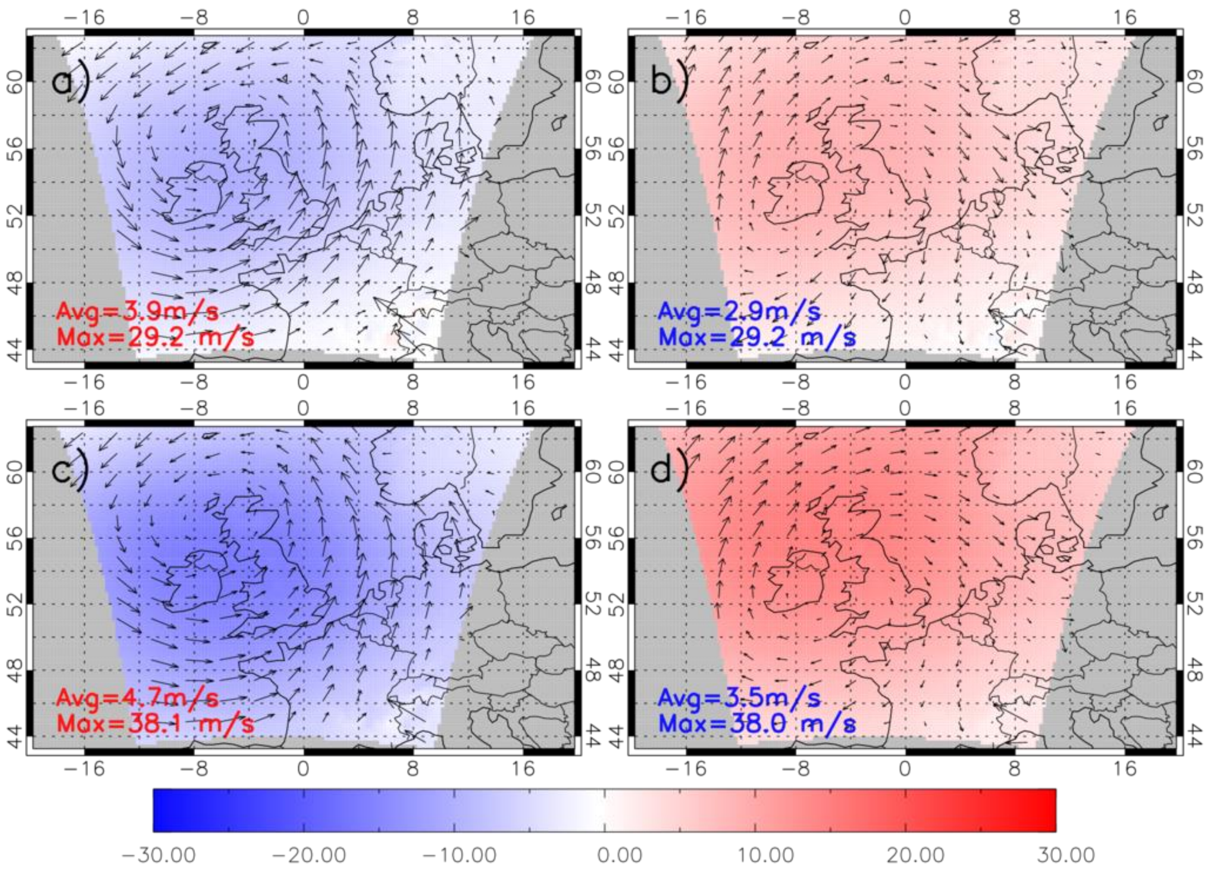

Figure 1: Pressure anomalies (hPa) relative to the seasonal average (2007-2010) with the wind circulation over plotted. a) summer cyclonic, b) summer anticyclonic, c) winter cyclonic and d) winter anticyclonic, all derived from the Lamb Weather Types.

To address this comment we will add a new four-panel figure. It shows the midday AQUM surface pressure anomalies, relative to the seasonal average, for a) summer cyclonic, b) summer anticyclonic, c) winter cyclonic and d) winter anticyclonic conditions. The wind circulation has been over plotted. Please note the wind barbs are scaled relative to the maximum wind speed in each plot. Therefore, we put the average and maximum wind speeds on the separate panels. The maps represent data between 2007 and 2010 as we used wind output available from a previous model run.

In summer cyclonic conditions, the anomalies are negative (-10 to 0 hPa) with anticlockwise circulation. This is similar to c), winter cyclonic, but the average domain winter wind speed (4.7 m/s) is greater than summer (3.7 m/s). In both cases, the flow over the UK is south-westerly, transporting pollution out over the North Sea. In the cases of summer (b) and winter (d) anticyclonic conditions, the pressure anomalies are positive (10 hPa in summer and up 20 hPa in winter) and the circulation is clockwise. Again, the domain flow is faster (3.5 m/s) in winter than summer (2.9 m/s).

2. Specifically the authors mention that '...any inconsistency between the NCEP reanalyses and AQUM flow fields will tend to worsen the comparisons...' and therefore tend to use the NCEP meteo data. This is only allowed when NCEP and AQUM flow fields are sufficiently identical. Even though this is likely the case, it is not explicitly shown nor mentioned.

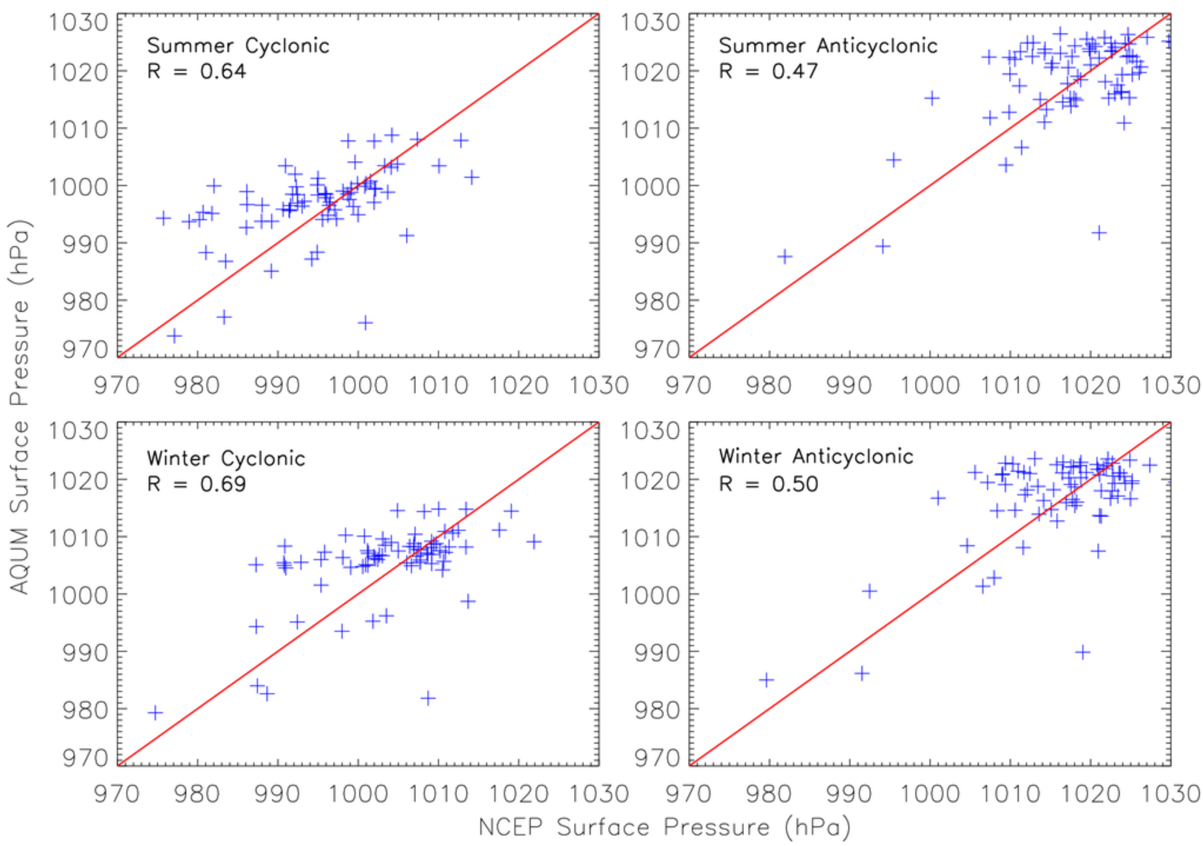

Figure 2: AQUM surface pressure vs. NCEP surface pressure (2006-2010), both composited under summer and winter cyclonic and anticyclonic conditions. The correlations are based on Spearman's Rank with a significance level of $p < 0.001$.

We investigated both NCEP and AQUM surface pressure composited under the seasonal synoptic conditions between 2006-2010. Surface pressure was used to test the consistency between both datasets as it is the primary variable used to generate the LWTs. The spatial pattern in both datasets, under the seasonal synoptic conditions, yielded correlations of between 0.47-0.69 at the 99.9% significance level using Spearman's Rank. These are relatively strong and significant correlations showing a consistency between the data products (the correlation values will have been degraded after the AQUM pressure data was interpolated to the coarser NCEP resolution).

These are our inclusions in the manuscript to address the reviewer comments 1 and 2:

We will replace "*Any inconsistency between the NCEP reanalyses and AQUM flow fields will tend to worsen the comparisons between observations and AQUM.*" on Page 18583 Lines 27-28 with "*We have sampled the AQUM surface pressure and winds under summer and winter anticyclonic and cyclonic conditions (Figure 1). This is between 2007-2010 as wind speed output was unfortunately not saved for 2006. Under cyclonic conditions, the pressure anomalies from the seasonal average range between -10 to 0 hPa and -20 to 0 hPa in summer and winter, respectively. Under anticyclonic conditions, the summer and winter anomalies range between 0-10 and 0-20 hPa. These pressure anomalies are consistent with cyclonic and anticyclonic conditions. Under anticyclonic conditions, the circulation is clockwise and is stronger in winter (domain average of 3.5 m/s) than summer (domain average of 2.9 m/s). Both the cyclonic regimes have anticlockwise circulation with stronger flow in winter (average of 4.7 m/s) than summer (3.9 m/s).*

We have also correlated the surface pressure spatial pattern from AQUM and NCEP, sampled under the seasonal synoptic regimes, using the Spearman's Rank Test (Figure 2). This yielded correlations of between 0.47-0.69 at the 99.9% significance level. These are significant correlations showing consistency between the AQUM and NCEP surface pressure data (the primary variable used to generate the LWTs). Overall, the AQUM and NCEP data are consistent and AQUM produces sensible meteorological fields when sampled under the LWTs. Therefore, we choose to sample AQUM column NO_2 fields using the LWT classifications derived from the NCEP reanalysis in Table 1.".

Now since we no longer use the ECMWF wind data, we will remove the discussion on the tracers (Page 18591 Lines 13-18) and in the acknowledgements. The text on Page 18591 will be replaced with "*From Figure 1 the average AQUM domain winter wind speed is 4.7 and 3.5 m/s under cyclonic and anticyclonic conditions. In summer, the equivalent average AQUM domain summer wind speeds are 3.9 and 2.9 m/s. Therefore, the synoptic type wind speeds are stronger in winter.*"

3. In fact, from my understanding the use of AQUM meteorological fields rather than those from NCEP would make more sense, as these fields are more consistent with the model tracer fields.

The LWTs are an independent dataset to the AQUM. Jones et al., (2013) used the NCEP reanalyses and an objective algorithm to generate the LWTs. Therefore, if we were to create LWTs using the AQUM winds and pressure fields, we would have to apply this algorithm here. However, since we have shown in the comment above that the AQUM and NCEP data (used to calculate the LWTs) are consistent, we argue that we can directly composite the AQUM tracer fields under the current LWTs.

After our response to comments 1 &2, we will add the text "*The most reliable method to examine the influence of meteorology on AQUM column NO_2 would be to apply the LWT algorithm used by Jones et al., (2013) on the AQUM pressure fields directly. However, as we have shown AQUM and NCEP to have consistent meteorological fields, it is simpler to directly sample the AQUM under the existing LWTs*.".

4. Secondly, despite the exhaustive introduction of the FGE score based on the 'anomaly cluster density' it is still difficult to appreciate this metric, for instance, in relationship with the correlation. The correlation is also reported, but shows a different message on the relative performance of the model under various LWTs. It would be helpful to expand on this relationship (or on its absence).

The correlation and FGE show different things in the analysis. The correlation/variability was used to look at the overall spatial relationships between the OMI and AQUM NO_2 anomalies under the four seasonal synoptic conditions. By finding a good R^2 value it proves that the anomalies fields are similar. Therefore, the anomaly features will be in similar spatial locations. We also know his from inspection of the anomaly Figures 2 & 4. This provides quantitative evidence and we are confident to use the FGE metric which investigates magnitude and significance, which R^2 does not. The FGE has been used to specifically look at the column NO_2 which is influenced by these weather regimes and to determine how well the model can capture it. Therefore, there is no direct link between the two metrics and they are used to diagnose different scientific questions.

Page 18589 Line 11 "*As the associations are strong, the anomaly spatial patterns are located in similar locations, as can be seen in Figures 2 and 4. Therefore, this gives us confidence to use the methodology discussed in Eq. (5) to analyse the size and spread of the significant anomalies for each seasonal synoptic regime*".

5. In this respect it is also interesting to note that the correlation as well as the FGE score with respect to OMI observations are better for idealized tracer fields than for the actual model NO2 field. This is contra intuitive considering the missing chemistry treatment. It could be worth to expand on this.

The idealised tracer fields are not compared to the OMI fields using the FGE. The standard AQUM NO_2 field is compared to the OMI NO_2 field. The idealised tracer fields are then compared with the AQUM NO_2 fields using the FGEs to find the most representative idealised tracer lifetime under the four seasonal synoptic regimes.

Too make this clearer in the text, we have replaced *"The analysis performed previously for the FGEs of the AQUM and OMI column NO_2 anomaly cluster densities (Fig 5) was repeated for the FGEs of the AQUM column NO_2 and tracer column anomaly cluster densities in Fig 9.*" on Page 18591 Lines 21-23 with *"The analysis performed previously for the FGEs of the AQUM and OMI column NO_2 anomaly cluster densities (Fig 5) was repeated for the FGEs of the AQUM column NO_2 and tracer column anomaly cluster densities in Fig 9. Therefore, in Eqn. 5, $\phi_{AQUM\_+/-}$ has been replaced with $\phi_{AQUM\_tracer\_+/-}$ and $\phi_{OMI\_+/-}$ has been replaced with $\phi_{AQUM\_+/-}$.*".

6. It would have been interesting to study specific chemical or physical loss processes which are influential to the NO2 lifetime, and could improve the scores presented here.

It would indeed have been interesting to try and diagnose the importance of different chemical/physical processes, but this was beyond the scope of this work. This is future work that we would like to carry out, which we will state in the conclusions.

7. Furthermore, the introduction of the four zones appears arbitrary to me and not really helpful for the discussion and may be omitted.

The four zones are indeed arbitrary (which we state in the text), but we feel that it makes the figure and discussion somewhat clearer in terms of showing how the AQUM compares with OMI under the different seasonal synoptic regimes.

8. Finally, the authors introduce all 27 LWTs, and suggest from the abstract, conclusion and section headers of 4.1-4.3 that they have validated all LWT relationships. However, in their analyses they only discriminate between two families of cyclonic and anticyclonic weather types. In my opinion the authors should change the manuscript throughout in this respect, more clearly stating the chosen selection of LWTs for cyclonic and anticyclonic conditions only.

We agree with the reviewer that we only talk about a subsection of the LWTs. In the revised paper we will introduce all the classifications as done in section 2.1, but clarify that we focus on cyclonic and anticyclonic conditions only in this study.

9. Sec. 2.1: The methodology for selection of LWTs is described rather cryptic. Please expand on this (see suggestions above).

The comments the reviewer refers to have been addressed in responses 1-3.

10. Pp 18585, l62: "Large background columns NO2 over the North Sea is indicative of cyclonic westerly transport off the UK mainland..." : Isn't there are contribution from NO2 originating from the continent?

We show that in Figure 1 the AQUM wind fields under cyclonic conditions transport UK NO_2 out over the North Sea. This can be seen in the old Figures 2 and 4 (cyclonic anomalies) for OMI and AQUM. There is also transport of continental Europe column NO_2 out over the North Sea, as seen in the same figures.

We will alter the text above to "Large background column NO_2 over the North Sea is indicative of cyclonic westerly transport off the UK mainland and the Benelux region".

11. Pp18586: "Under anticyclonic conditions ..." : please brake sentence to two to improve readability.

We will replace this method throughout the manuscript to the style suggested by both reviewers (i.e. two separate sentences).

12. Pp 18586, l10 -l15: This is indeed an interesting observation. Do you have any suggestion why AQUM does show a different anomaly than OMI?

There are many potential reasons for this. However, without a detailed sensitivity study, it is difficult to identify potential causes or speculate on any possible reasons. Such sensitivity experiments are beyond the scope of this study. This is something we would like to research in the future and will state so in the conclusions.

13. Pp 18589, l 7: The correlation appears highest for summer anticyclonic, while for this case the FGE score is lowest. Do you have an explanation for this?

Overall, the AQUM captures the spatial variability of the OMI column NO_2 under summer anticyclonic conditions. However, AQUM does not capture the positive anomaly field seen in the observations. Therefore, the correlation (R^2) is good but the FGE comparisons are poorer. Please see the response to comment 4.

14. Pp 18591, l 13-l16: This is a clear weakness of this paper, as mentioned above. The manuscript would benefit from a closer inspection of the prevailing weather systems.

This will be covered by the analysis of Figure 1.

Pp18592, l21: "NO2-LWT relationships": please change to something like: "captured the OMI column NO2 anomalies for cyclonic and anticyclonic LWT conditions".

We have changed this in line with the reviewer's comment.

15. Pp 18593, l22-l24: "This work..." I don't think this can be concluded from the current study, considering that the authors do not evaluate the absolute NO2 values during anticyclonic conditions. Also 'accumulation of air pollution' is obviously much wider than NO2 anomalies, as it should also include evaluations of other pollutants such as ozone. Finally it is unfortunate that for summer time anticyclonic conditions the FGE score performs worst (even though the summertime positive anomaly indeed appears in line with those from other weather conditions.

We disagree with the reviewer's comment here. The discussion of Figures 1 and 3 in Sections 4.1 and 4.2 shows that AQUM and OMI have similar spatial column NO_2 patterns. The anomalies in Figures 2 & 4 are of similar spatial location and magnitude in general, which are deviations from the seasonal average conditions. Therefore, the absolute values are similar and the deviations from the seasonal average are similar, so the seasonal averages will be similar. We quantitatively analyse the anomalies under the seasonal synoptic regimes, with the purpose to see if the AQUM could capture the changes in column NO_2 associated with changes in meteorology.

However, the reviewer is correct about "air pollution" on Page 18593 Line 23 and this will changed to "tropospheric column NO_2".

For the reviewer's comment "Finally it is unfortunate that for summer time anticyclonic conditions the FGE score performs worst (even though the summertime positive anomaly indeed appears in line with those from other weather conditions." we refer him/her to our response to comment 4.

Table 1: It is misleading to present all weather types, when only anticyclonic and cyclonic conditions are studied.

We will change Table 1 to show bold text of just cyclonic and anticyclonic conditions. We also state that that we only focus on these two weather types.

Figures 5, 8 and 9: Please improve readability of legends on axes and within the figure (font thickness).

We will increase the font and line thickness on these figures.

References:

Jones, P. D., Harpham, C., and Briffa, K. R.: Lamb weather types derived from reanalysis products, Int. J. Climatol., 33, 1129–1139, doi:10.1002/joc.3498, 2013.