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."

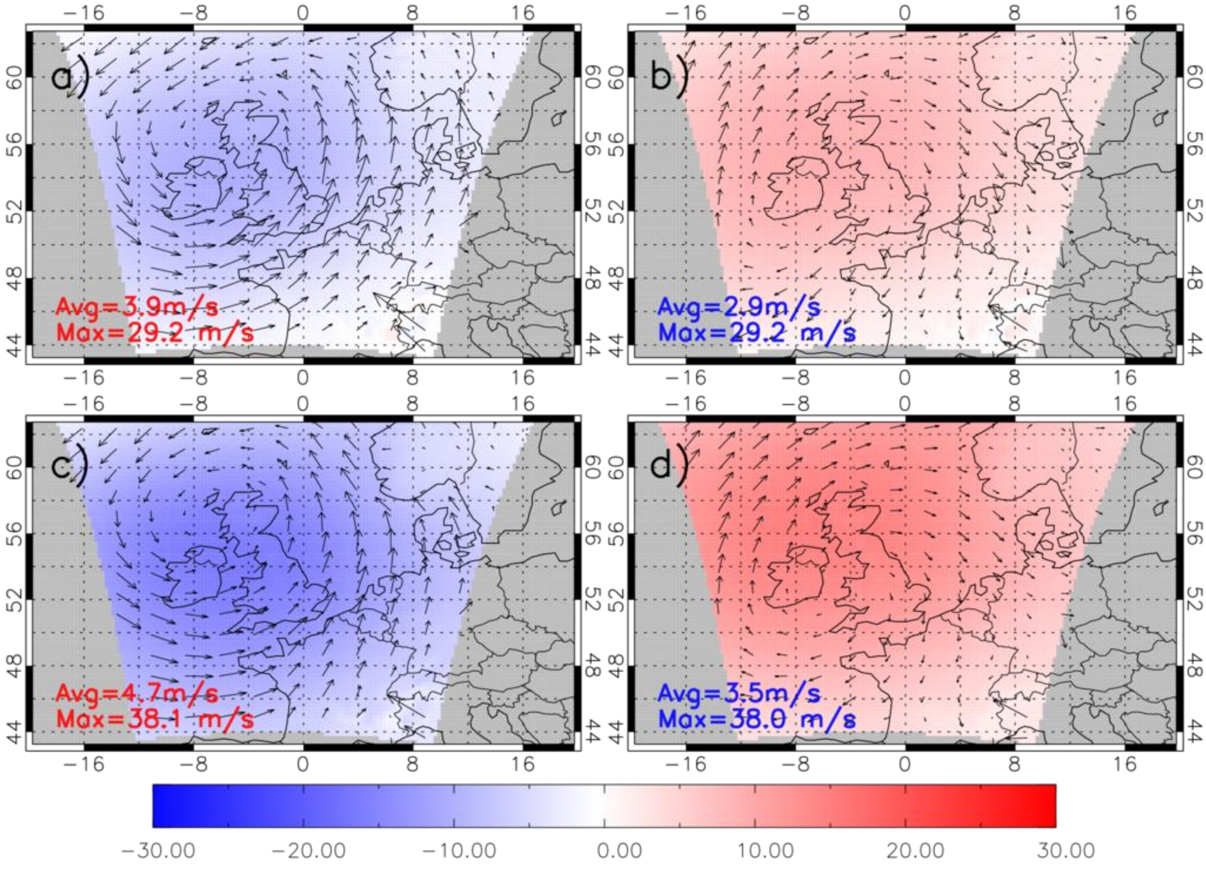

Figure 1: Pressure anomalies (hPa) relative to the seasonal average (2007-2010) with the wind circulation over plotted. a) summer cyclonic, b) summer anticyclonic, c) winter cyclonic and d) winter anticyclonic, all derived from the Lamb Weather Types.

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.

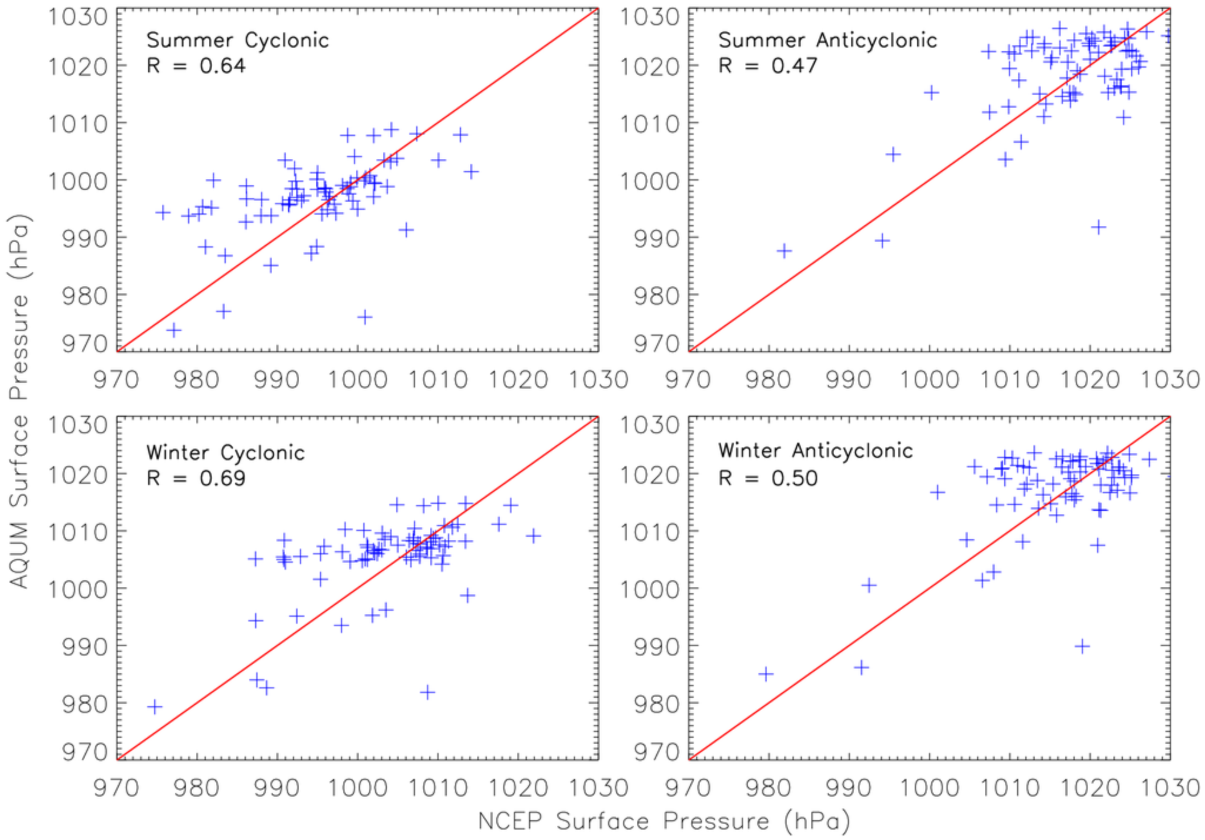

Figure 2: AQUM surface pressure vs. NCEP surface pressure (2006-2010), both composited under summer and winter cyclonic and anticyclonic conditions. The correlations are based on Spearman's Rank with a significance level of $p < 0.001$.

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.

Reviewer 2 Responses

We thank the reviewer for his/her comments. Our responses are below (in red text - edits to the manuscript in blue italics) following the reviewer's comments in black text:

1. A weakness of the study is that it doesn't fully achieve its stated aim to "determine the controlling factors" in the relationship between column NO2 and synoptic meteorology. This isn't a major failing, but the paper is less useful than it would otherwise be. The idealised tracer approach hasn't been fully exploited to quantify the influence of transport or to provide a more critical test of lifetimes from the observations. This would require a small amount of additional analysis, but I believe it would add substantial value to the study. My other comments are relatively minor and are outlined below.

We agree with the reviewer's comment and will soften the statement. We will change "determine the controlling factors" to "explore the relative importance of various factors" in the abstract.

2. What is the lifetime of NO2 in the model, and how does this compare with the idealised tracer analysis? If the relevant model fluxes have not been diagnosed it should still be possible to estimate the mean lifetime from the regional tropospheric abundance and emissions.

We are trying to use the idealised tracers to estimate the lifetime of NO_2. The fact that we are using a limited area model makes it impossible to calculate the lifetime because fluxes through the boundaries are likely to be a strong sink in some conditions and a source under others. The model does not at present have the capability to output the relevant chemical fluxes directly.

We will add "*Also, the direct lifetime of NO_2 cannot be determined as fluxes through the model boundaries are likely a strong sink or source under different conditions.*" on Page 18590 Line 13.

3. Does the tracer analysis provide any new insight into how modeled and observed NO2 lifetimes differ?

The tracers here were used to try and estimate the most representative tracer lifetime of AQUM NO_2 under the different spatial regimes. By doing so, the tracer anomaly field which most accurately matches the NO_2 field were correlated (R^2) to find how much of the spatial variability in the AQUM NO_2 is explained by transport as the lifetime (chemistry) is fixed. They are primarily used to diagnose the importance of transport on column NO_2.

On Page 18590 Line 9, we will add the following text "and an approximation for the model lifetime of NO_2".

4. What are the implications of this for removal processes, for the magnitude of emissions, or for the balance between transport and chemistry processes? This would allow a stronger and more quantitative statement than the current one "showing that transport is an important factor..." (which is true, but not very informative).

The literature does not provide a lengthy discussion on the controlling processes for NO_2 over Europe. Savage et al., (2008) suggest that meteorology is the key processes controlling the spatial variability of NO_2. In this study, we investigate which overarching processes are important. We use these tracers as a first-order estimate of NO_2 control processes as they were quicker and simpler to implement. We did not have the time or resources in this study to undertake this type of analysis. To undertake a complete investigation of the AQUM production and loss processes (both meteorological, chemical and emission) was beyond the scope of this study. This is future work would be interesting to carry out. Therefore, we will add the following text to, Page 18593 Line 25 "*As follow on work from this study, we intend to perform a sensitivity analysis of different AQUM processes to determine the governing factors on the distribution of column* NO_2.", to the conclusions about future work.

5. The "best fit" lifetimes of 6h in summer and 12h in winter are identified based on Fig 9. However, marking off the fraction of pixels for modeled NO2 in Figure 8 under each season/condition would provide verification of this and might allow assessment of equivalent tracer lifetimes intermediate between those modeled.

The AQUM column NO_2 significant anomaly domain fraction was calculated at 0.02, 0.04, 0.07 and 0.09 for summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. Reading across to the respective tracer profiles in Figure 8, the approximate NO_2 life times are 6.0, 4.5, 11.0 and 7.0 hours for summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. This matches the tracer results in that summer NO_2 lifetimes are shorter than that of winter. It should be noted though that this approach does not take into account the magnitude of the anomalies.

We will over plot these values onto Figure 8 and add the following text "*To verify this result, the AQUM column* NO_2 *significant anomaly domain fraction was calculated at 0.02, 0.04, 0.07 and 0.09 for summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. Reading across to the respective tracer profiles in Figure 8, the approximate* NO_2 *lifetimes are 6.0, 4.5, 11.0 and 7.0 hours, respectively. This supports the tracer results in that summer* NO_2 *lifetimes are shorter than that of winter. It should be noted though that this approach does not take into account the magnitude of the anomalies*." on Page 18592 Lines 4.

6. Section 4.2: How do the NO2 emissions in the model vary by season, and how much does this contribute to the observed seasonal column differences? Greater wintertime emissions will contribute to greater absolute anomalies even without differences in NO2 lifetime.

The AQUM NO_x emissions are annual totals from different sources (point, area) and different datasets (NAEI, EMEP). A seasonal cycle (scaling factor) is applied to these annual emissions totals per grid square from Visschedijk et al., (2007). The model emissions and seasonal scaling factors are discussed in Pope et al., (2015) and Savage et al., (2013). We will add a new sentence about the emissions in section 3.2, Page 18583 Line 15 "The emissions are initially annual totals, however, the seasonal scaling factor from Visschedijk et al., (2007) is applied. See Pope et al., (2015) for more information.", and the differences between seasons. The Visschedijk et al., (2007) reference will be added to the reference list.

7. It would be helpful to provide a brief assessment of the likely meteorological biases in the analysis given that both cloud cover and tropopause height are strongly influenced by the synoptic system.

Yes this is true that cloud cover and tropopause height will be influenced by different synoptic regimes. Under anticyclonic conditions, there will be less cloud cover when compared with cyclonic conditions and the NO_2 composites will be made from a larger sample. However, as shown in this study and in Pope et al., (2014), there is still sufficient satellite data to make sensible composites of column NO_2. As for the tropopause height, this is included in the satellite data. The tropopause information used in the OMI product is based on TM4. For the AQUM, the NO_2 profiles, which have been co-located in time and space with OMI retrievals, have been interpolated onto the satellite pressure grid, the subcolumns calculated and totalled up to the pressure level in the satellite data where the TM4 tropopause occurs. Hence, only tropospheric data is included in the model and satellite NO_2 columns.

The following information "*Initially, the AQUM NO_2 profile is interpolated to the satellite pressure grid. The AKs are then applied to the NO_2 sub columns using Eq. 2. The AQUM sub-columns are then summed up to the satellite tropopause level.*", Page 18584 Line 16, will be added in section 4.1. Then "*For more information on OMI tropospheric column NO_2, we refer the reader to Boersma et al., (2008).*".

8. How is the stratospheric contribution to the NO2 column removed, and how might this influence the comparison between cyclonic and anticyclonic conditions?

In the response above, the pressure fields in the satellite data will be representative of anticyclonic/cyclonic conditions. Therefore, the satellite tropopause level will vary in height according to the weather systems in which the column NO_2 is being retrieved. Therefore, no stratospheric information will be included in the model column NO_2. As for the DOAS technique used for the satellite data, the total column is retrieval, the stratospheric component simulated using TM4 (and assimilated information), which is then subtracted from the total column to give the tropospheric column.

We refer the reviewer to our response in comment 7.

9. In addition, the chemical lifetime of NO2 will differ under the different synoptic conditions, and this is likely to exaggerate the contrast between cyclonic and anticyclonic conditions that is currently attributed to transport. How much effect is this likely to have?

The fact that most of the variability in NO_2 column is explained by a tracer with a fixed lifetime shows that the differences between regimes has only a small contribution from this process. This is outlined on Pages 18589-90 Lines 28-18.

10. Standard statistical metrics are discounted in section 4.2 as providing only a partial evaluation, but in combination these approaches remain powerful. Supplementing the new approaches with these conventional metrics (demonstrating their weaknesses if necessary) would comfort any readers suspicious about why the normal statistics are not used.

Rank	Metrics			
	Correlation	**Regression**	**RMSE**	**New Method**
1	Summer Anticyclonic	Summer Anticyclonic	Summer Anticyclonic	Summer Cyclonic
2	Summer Cyclonic	Summer Cyclonic	Summer Cyclonic	Winter Anticyclonic
3	Winter Anticyclonic	Winter Cyclonic	Winter Anticyclonic	Winter Cyclonic
4	Winter Cyclonic	Winter Anticyclonic	Winter Cyclonic	Summer Anticyclonic

Table 1: Highlights the skill rank of the seasonal synoptic regimes for which AQUM can simulate column NO_2 when compared with OMI column NO_2 using correlation, slope-of-regression, RMSE and the method proposed here. 1: Best AQUM-OMI agreement, 4: Worst AQUM-OMI agreement.

We will add this table to show that even though the correlation metrics, regression and RMSE produce similar results, the new method proposed here shows something different. Like the correlation and RMSE, our method has summer cyclonic, winter anticyclonic and winter cyclonic in the same order. However, summer anticyclonic has the worst comparisons using our method. This is because in the anomaly fields (Figure 4b), our method shows AQUM does not simulate significant negative biases whereas the other metrics show the best agreement. This justifies our new method as it takes into account the significance of the anomalies unlike the other metrics.

We will add the following paragraph on Page 18589 Line 5:

"In Table 2 we justify using our approach of using the anomaly clusters and FGE when compared with other statistical metrics. The table highlights the order in which AQUM most successfully reproduces the OMI column NO_2 anomalies when sampled under the seasonal synoptic regimes. Like the correlation and RMSE, our method has summer cyclonic, winter anticyclonic and winter cyclonic in the same order. However, summer anticyclonic has the worst comparisons using our method. This is because in the anomaly fields (Figure 4b), our method shows AQUM does not simulate significant negative biases whereas the other metrics show the best apparent agreement. This justifies our new method as it takes into account the significance of the anomalies, unlike the other metrics."

Here Table 2 in the manuscript is Table 1 in the response document.

11. The description of the clustering approach (p.18587) isn't clear. The term "cluster" suggests
distinct groupings, but the text suggests that this is just done for all positive and all negative anomalies to give two values of phi. Please clarify this description. How sensitive is the approach taken here to different choices of the significance criterion?

Indeed, the term "cluster" refers to significant positive and negative anomalies. However, these are distinct groupings of anomalies, so we feel that the current definition is ok. We will add the following text, Page 18587 Line 8 "Here, we use the term "cluster" to represent a grouping of positive or negative significant anomalies.", to clarify the use of the word "cluster" in our analyses.

As for the sensitivity of the WRT, where we use a 95% significance level, many of the significant anomalies will be significant to the 99% level and higher. If this significance level is lowered (e.g.to 90%) more pixels would become significant but the likelihood of them occurring by chance increases. Therefore we will stick with the level of 95% significance.

Minor Comments:

Brackets or slashes are used to denote alternatives in a number of places, e.g., "cyclonic (anticyclonic) conditions..." in the abstract (lines 1-2). This shorthand is difficult for the reader to follow and should be replaced by the full text to provide a slightly more wordy (but much clearer) description. The main occurrences are: p.18578,l.1-2, p.18579,l.20-21, p.18586,l.6-7 and l.19-20.

In line with Reviewer 1's comments, we will change this aspect of the text.

p.18583,l.17: "....will dominate" - some justification needed here.

We will include the reference "Zhang e al., (2003)". They show that in the mid-latitudes (USA) that NO_x soil emissions are an order of magnitude smaller than anthropogenic NO_x emissions. Therefore, the new text on Page 18583 Line 17 will be *"emissions from transport and industry in this region will dominate (Zhang e al., 2003)".*

p.18580, l.8: remove "manage to"

Will be removed.

Figure 9: Please choose a different color scale, as the most interesting contrasts are between the 6h and 12h tracers which are both colored green.

We will change the colour scale.

References:

Pope, R., Savage, N., Chipperfield, M., Arnold, S., and Osborn, T.: The influence of synoptic weather regimes on UK air quality: analysis of satellite column NO2, Atmos. Sci. Lett., 15, 211–217, doi:10.1002/asl2.492, 2014.

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The influence of synoptic weather regimes on UK air quality: regional model studies of tropospheric column NO_2

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Abstract

Synoptic meteorology can have a significant influence on UK air quality. Cyclonic ~~(anticyclonic)~~ conditions lead to the dispersion ~~(accumulation)~~ of air pollutants away from ~~(over)~~ source regions, while anticyclonic conditions lead to their accumulation over source regions. Meteorology also modifies atmospheric chemistry processes such as photolysis and wet deposition. Previous studies have shown a relationship between observed satellite tropospheric column NO_2 and synoptic meteorology in different seasons. Here, we test whether the UK Met Office Air Quality in the Unified Model (AQUM) can reproduce these observations and then use the model to ~~determine the controlling~~ explore the relative importance of various factors. We show that AQUM successfully captures the observed relationships, when sampled under the Lamb Weather Types, an objective classification of midday UK circulation patterns. By using a range of idealised NO_x-like tracers with different e-folding lifetimes, we show that under different synoptic regimes the NO_2 lifetime in AQUM is approximately 6 h in summer and 12 h in winter. The longer lifetime can explain why synoptic spatial column NO_2 variations are more significant in winter compared to summer, due to less NO_2 photochemical loss. We also show that cyclonic conditions have more seasonality in column NO_2 than anticyclonic conditions as they result in more extreme spatial departures from the wintertime seasonal average. Within a season (summer or winter) under different synoptic regimes, a large proportion of the spatial pattern in the UK column NO_2 field can be explained by the idealised model tracers, showing that transport is an important factor in governing the variability of UK air quality on seasonal synoptic timescales.

1 Introduction

Local air quality (AQ) can be influenced significantly by regional weather systems through the accumulation and dispersion of atmospheric pollutants over and away from source regions and populated areas. Local air quality can also be influenced by changes in atmospheric chemistry processes. For example, increased cloudiness will reduce photolysis rates below cloud and increased precipitation can lead to enhanced removal of pollutants by wet deposition.

Many studies have used synoptic weather classifications to investigate the influence on AQ. These include objective classifications such as the Lamb Weather Type (LWT) and the North Atlantic Oscillation (NAO) Index. The LWTs are an objective description of the daily midday atmospheric circulation over the UK based on mean sea level pressure reanalysis data (Jones et al., 2013). The NAO Index is based on the pressure gradient between the Icelandic low and the Azores/Gibraltaran high pressure systems (Jones et al., 1997). In winter this pressure gradient has a significant influence on UK weather, where the positive phase can result in mild wet winters and the negative phase can lead to cold stable conditions (Osborn, 2006).

Previous studies including Demuzere et al. (2009); Tang et al. (2011); Lesniok et al. (2010) and McGregor and Bamzelis (1995) have used surface observations of air pollution to look at these AQ – regional weather relationships. Pope et al. (2014) and Thomas and Devasthale (2014) were two of the first studies to use earth observation (EO) of atmospheric pollutants, in combination with measures of synoptic weather, to investigate the influence of regional weather on AQ. Pope et al. (2014) used the LWTs and Ozone Monitoring Instrument (OMI) tropospheric column NO_2 between 2005 and 2011 (note that in the following we often refer to "tropospheric column NO_2" as "column NO_2"). They found that anticyclonic ~~(cyclonic)~~ and cyclonic conditions lead to the accumulation ~~(transport)~~ and transport of air pollutants over ~~(away from) source regions~~and away from source regions, respectively. They also successfully detected the leeward transport of column NO_2 away from source regions under certain wind directions, similar to Beirle et al. (2011) and Hayn et al. (2009). These two studies used OMI column NO_2 and wind information to analyse NO_2 transport from the isolated megacity Riyadh, Saudi Arabia and Johannesburg, South Africa, respectively. Zhou et al. (2012) found significant impacts of wind speed and precipitation on OMI column NO_2 over western Europe. Savage et al. (2008) investigated the interannual variability (IAV) of satellite NO_2 columns over Europe, finding that meteorology influences NO_2 IAV more than emissions. Thomas and Devasthale (2014) found that Atmospheric Infrared

Sounder (AIRS) CO at 500 hPa from 2002 to 2013 over the Nordic countries increased by 8, 4, 2.5 and 1 % under southeasterly winds, northwesterly winds, the positive phase of the NAO and anticyclonic conditions, respectively. The clearest conditions were under northeasterly winds and the negative phase of the NAO when cleaner Arctic air was transported into the Nordic region. When looking at the Global Ozone Monitoring Experiment (GOME) column NO_2 and the NAO, Eckhardt et al. (2003) found that significant positive phases lead to the reduction in column NO_2 over western Europe. However, Pope et al. (2014) did not ~~manage to~~ find any clear evidence for this relationship.

This paper uses both satellite observations and the UK Met Office's operational Air Quality in the Unified Model (AQUM) to extend on the work of Pope et al. (2014). We investigate the differences in the air quality-synoptic weather relationships found by Pope et al. (2014) by attempting to quantify the dominant processes involved, for example, is atmospheric chemistry or weather more important in governing the links between synoptic meteorology and air quality in different seasons? First, we assess the ability of AQUM to simulate UK air quality under different synoptic regimes found in the OMI data. This is defined as "dynamical" model evaluation, i.e. assessing a model's ability to simulate changes in air quality stemming from changes in emissions and/or meteorology (Dennis et al., 2010). This follows the work by Pope et al. (2015) who used "operational" model evaluation, i.e. statistical analyses aimed at determining the agreement between the model and observations (Dennis et al., 2010), to perform the first evaluation of AQUM against satellite observations. Then, we use AQUM e-folding tracers with specified lifetimes designed to assess the impact of meteorology, emissions and chemistry on UK AQ.

The paper is structured as follows: Sect. 2 discusses the LWTs and OMI column NO_2 data. The model setup and application of OMI averaging kernels (AK) is discussed in Sect. 3. Section 4 shows our OMI/AQUM–LWT results for 2006–2010 and our conclusions are presented in Sect. 5.

2 Data

2.1 Lamb weather types

Lamb (1972) originally had a manual methodology of classifying the UK weather patterns but that has been superseded by automated methods. The objective (automated) LWTs, developed by Jones et al. (2013) based on the algorithm of Jenkinson and Collison (1977) and using the NCEP reanalyses midday mean sea level pressure data described by Kalnay et al. (1996), classify the atmospheric circulation patterns over the UK according to the wind direction and circulation type. The LWTs (Table 1) are grouped into three vorticity types (neutral vorticity, cyclonic and anticyclonic) and eight wind flow directions unless solely classified as cyclonic or anticyclonic. The left column and top row of Table 1 show the grouped classifications used by Pope et al. (2014) to composite OMI column NO_2 data between 2005 and 2011. In this study we focus on ~~2006–2010~~ 2006-2010 to match the AQUM simulation period~~.~~, but only focus on seasonal cyclonic and anticyclonic conditions. Therefore, from here on in, references to “OMI-LWT” or “AQUM-LWT” comparisons relate to the analysis of OMI or AQUM tropospheric column NO_2 fields under seasonal cyclonic or anticyclonic conditions. For more information on the application of the LWTs to composite OMI and AQUM column NO_2 see Pope et al. (2014).

2.2 Satellite data

OMI is aboard NASA’s EOS-Aura satellite and has an approximate UK daytime overpass of 13:00 local time (LT). It is a nadir-viewing instrument with pixel sizes between 16–23 and 24–135 km along and across track, respectively, depending on the viewing zenith angle (Boersma et al., 2008). The tropospheric column NO_2 data used here is the DOMINO product version 2.0, which comes from the Tropospheric Emissions Monitoring Internet Service (TEMIS) (Boersma et al., 2011a, b) and is available from http://www.temis.nl/airpollution/no2.html. We have binned NO_2 swath data from 01 January 2006 to 31 December 2010 onto a daily 13:00 LT $0.25^\circ \times 0.25^\circ$ grid between 43–63° N and 20° W–20° E. All satellite retrievals were quality controlled, and retrievals/pixels with geometric cloud cover greater than 20 % and poor quality data flags (flag $= -1$ including retrievals affected by row anomalies and flagged by the Braak, 2010 algorithm) were removed. Several studies including Irie et al. (2008) and Boersma et al. (2008) have validated OMI column NO_2 against surface and aircraft measurements of tropospheric column NO_2 with good agreement within the OMI uncertainty ranges. Therefore, we have confidence in the OMI column NO_2 used in this study.

3 Air Quality in the Unified Model (AQUM)

3.1 Model setup

The AQUM domain covers approximately 45–60° N and 12° W–12° E, on a rotated grid, including the British Isles and part of continental Europe. The grid resolution is $0.11^\circ \times 0.11^\circ$ in the horizontal and the model extends from the surface to 39 km on 38 levels. It has a coupled online tropospheric chemistry scheme, which uses the UK Chemistry and Aerosols (UKCA) subroutines. A complete description of this chemistry scheme, known as Regional Air Quality (RAQ), is available from the online Supplement of Savage et al. (2013). It includes 40 tracers, 18 non-advected species, 23 photolysis reactions and 115 gas-phase reactions. It also includes the heterogeneous reaction of N_2O_5 on aerosol as discussed by Pope et al. (2015).

For aerosols, AQUM uses the Coupled Large-scale Aerosol Simulator for Studies In Climate (CLASSIC) aerosol scheme. Aerosols are treated as an external mixture simulated in the bulk aerosol scheme. It contains six prognostic tropospheric aerosol types: ammonium sulphate, mineral dust, fossil fuel black carbon (FFBC), fossil fuel organic carbon (FFOC), biomass burning aerosols and ammonium nitrate. It also includes a fixed climatology for biogenic secondary organic aerosols (BSOA) and a diagnostic scheme for sea salt. For more details of the aerosol scheme see Bellouin et al. (2011).

Meteorological initial conditions and lateral boundary conditions (LBCs) come from the Met Office's operational global Unified Model (25×25 km) data. The chemical initial conditions come from AQUM's forecast for the previous day and the chemical LBCs are provided by the global Monitoring Atmospheric Composition and Climate (MACC) reanalyses (Inness et al., 2013). Pope et al. (2015) showed that for 2006, using the ECMWF GEMS (Global and regional Earth-system Monitoring using Satellite and in-situ data) reanalysis (Hollingsworth et al., 2008) LBCs provided more accurate forecasts than using the MACC LBCs. However, the GEMS LBCs are only available for 2006–2008. Therefore, we have used the MACC LBCs, which are available for the full period analysed here.

The model emissions were generated by merging three datasets: the National Atmospheric Emissions Inventory (NAEI) (1×1 km) for the UK, ENTEC (5×5 km) for the shipping lanes and European Monitoring and Evaluation Programme (EMEP) (50×50 km) for the rest of the model domain. NAEI NO_x emissions consist of point and area sources. Area sources include light industry, urban emissions and traffic, while elevated point sources are landfill, power stations, incinerators and refineries. Typically, the point source emissions are $100\,g\,s^{-1}$ in magnitude, while the area sources tend to be $10\,g\,s^{-1}$. The emissions are initially annual totals, however, the seasonal scaling factor from Visschedijk et al. (2007) is applied. See Pope et al. (2015) for more information. NO_x lightning emissions are parameterised based on model convection (O'Connor et al., 2014). AQUM does not include any soil NO_x sources, but large emissions from transport and industry in this region will dominate (Zhang et al., 2003) .

AQUM was run for 5 years from 01 January 2006 to 31 December 2010. Five years provide a sufficient model data record to test the OMI column NO_2–LWT relationships. There are a few missing days for the 5 year simulation as the MACC LBCs do not exist over the full period (i.e. 4–6 June 2007 are missing).

As AQUM is a limited area NWP model, with meteorological boundary conditions from an operational NWP analysis and short (24 h) forecasts, the representation of large-scale weather systems via the LBCs is likely to be highly consistent with the NCEP reanalyses, used to calculate the LWTs. Jones et al. (2014) also show high correlations between LWTs derived with NCEP reanalyses and those from another independent reanalysis (20CR). ~~Any inconsistency between the NCEP reanalyses and AQUM flow fields will tend to worsen the comparisons between observations and AQUM~~

We have sampled the AQUM surface pressure and winds under summer and winter anticyclonic and cyclonic conditions (Figure 1). In this study, summer ranges from April to September and winter is October-March. This is between 2007-2010 as u and v winds were 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). 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. Therefore, we choose to sample AQUM column NO_2 fields using the LWT classifications derived from the NCEP reanalysis in Table 1.

3.2 OMI averaging kernels

Since OMI retrievals of column NO_2 range in sensitivity with altitude, the OMI AKs must be applied to the model for representative comparisons. The OMI retrievals use the Differential Optical Absorption Spectroscopy (DOAS) technique and the AK is a column vector. Following Huijnen et al. (2010) and the OMI documentation (Boersma et al., 2011b), the AKs are applied to the model as:

$$y = \boldsymbol{A} \cdot \boldsymbol{x} \tag{1}$$

where y is the total column, $\boldsymbol{A}$ is the AK and $\boldsymbol{x}$ is the vertical model profile. However, here the tropospheric column is needed:

$$y_{\text{trop}} = \boldsymbol{A}_{\text{trop}} \cdot \boldsymbol{x}_{\text{trop}} \tag{2}$$

where $\boldsymbol{A}_{\text{trop}}$ is:

$$\boldsymbol{A}_{\text{trop}} = \boldsymbol{A} \cdot \frac{\text{AMF}}{\text{AMF}_{\text{trop}}} \tag{3}$$

AMF is the atmospheric air mass factor and AMF_{trop} is the tropospheric air mass factor. Initially, the AQUM NO_2 profile is interpolated to the satellite pressure grid. The AKs are then applied to the NO_2 sub columns using Equation 2. The AQUM sub-columns are then summed up to the satellite tropopause level. For more information on ~~the~~ OMI tropospheric column NO_2, we refer the reader to (Boersma et al., 2008) and for more information on the effect of OMI AKs on AQUM column NO_2 see Pope et al. (2015).

4 Results

4.1 OMI tropospheric column NO_2–LWT relationships: 2006–2010

As AQUM was run for 2006–2010, the OMI column NO_2–LWT analyses performed by Pope et al. (2014) are repeated for this time period to assess whether the synoptic weather – AQ relationships are consistent between the 7 year period presented in that study and the 5 years analysed here. Figures 3 and 4 show the influences of cyclonic and anticyclonic conditions in winter and summer on column NO_2 from OMI. ~~Here~~Again, summer ranges from April to September and winter is October–March. These extended seasons give more temporal sampling of OMI column NO_2 and better composites under the weather regimes. Under cyclonic conditions, column NO_2 is transported away from the source regions, while anticyclonic conditions aid its accumulation. Figure 4 highlights significant (95 % confidence level – based on the Wilcoxon Rank Test, Pirovano et al., 2012) anomalies of up to $\pm 5 \times 10^{15}$ molecules cm^{-2} over the North Sea ~~/~~and UK under cyclonic conditions. The reverse is found under anticyclonic conditions. The spatial extent of the anomalies is greatest in winter for both vorticity regimes. Therefore, there are no significant differences between the synoptic weather – air quality relationships based on the 5- and 7 year comparisons. Hence, the LWT–OMI 5 year comparisons act as the baseline for comparisons between AQUM column NO_2 and the LWTs.

4.2 AQUM tropospheric column NO_2–LWT relationships

AQUM column NO_2, composited under the LWTs, displays similar patterns to OMI (Fig. 5). For this comparison, AQUM output has been co-located spatially and temporally with each OMI retrieval and the averaging kernel applied. In winter, under cyclonic conditions AQUM column NO_2 ranges between $10–13 \times 10^{15}$ molecules cm^{-2} over the UK and Benelux source regions (Fig. 5c). Over the western and eastern model domain, column NO_2 ranges between $0–4 \times 10^{15}$ molecules cm^{-2} and $5–8 \times 10^{15}$ molecules cm^{-2}, respectively. Under winter anticyclonic conditions column NO_2 over UK and Benelux source regions is $16–20 \times 10^{15}$ molecules cm^{-2} and the background column NO_2 ranges between $0–8 \times 10^{15}$ molecules cm^{-2} (Fig. 5d). Larger background column NO_2 over the North Sea in Fig. 5c is indicative of cyclonic westerly transport off the UK mainland and the Benelux region, while larger source region column NO_2 in Fig. 5d highlights anticyclonic accumulation of NO_2.

When compared with OMI (Fig. 3c), AQUM sampled under the winter cyclonic conditions (Fig. 5c) shows transport of more column NO_2 over the North Sea ranging between $5–8 \times 10^{15}$ molecules cm^{-2} and covering a larger spatial extent. Under anticyclonic conditions (Fig. 5d), AQUM column NO_2 is lower ~~/higher~~ than OMI over the London and Benelux region ~~/~~by 2–3 $\times 10^{15}$ molecules cm^{-2}. However, AQUM column NO_2 is higher than OMI over northern England by 2–3 $\times 10^{15}$ molecules cm^{-2}. The AQUM-OMI winter anticyclonic background column NO_2 is similar, ranging between 0–5 and 5–10 $\times 10^{15}$ molecules cm^{-2} over the sea and continental Europe, respectively.

Both OMI and AQUM show similar patterns in summer for both vorticity types, but with lower spatial extents than winter. Interestingly, the OMI cyclonic UK source region column NO_2 is larger in summer (8–10 $\times 10^{15}$ molecules cm^{-2} – Fig. 3a) than winter (6–8 $\times 10^{15}$ molecules cm^{-2} – Fig. 3c), but AQUM does not simulate this (Fig. 5a and c). AQUM summer cyclonic UK source region NO_2 ranges between 6–8 $\times 10^{15}$ molecules cm^{-2}, while in winter it is 10–12 $\times 10^{15}$ molecules cm^{-2}.

The AQUM and OMI transport and accumulation similarities and differences can be seen in Figs. 4 and 6, which show anomalies of the composite averages calculated as differences with respect to the 5 year seasonal means. Under winter cyclonic conditions, both AQUM (Fig. 6c) and OMI (Fig. 4c) show significant negative ~~/~~and positive anomalies of similar magnitude over the UK ~~/North Sea~~and North Sea, respectively. Winter anticyclonic conditions lead to an accumulation of AQUM (Fig. 6d) and OMI (Fig. 4d) column NO_2 over UK and English Channel, causing significant positive anomalies of 1–3 $\times 10^{15}$ molecules cm^{-2}. The summer AQUM (Figs. 6a and b) and OMI (Figs. 4a and b) synoptic-column NO_2 spatial patterns are similar in extent and magnitude. They are similar to the winter equivalents, but cover a smaller spatial extent. Therefore, on the regional scale, we can say that AQUM captures the OMI column NO_2–LWT relationships with similar significant anomalies from the period average.

For a more complete dynamical model evaluation the differences between AQUM and OMI column NO_2 have been quantified. To compare the spatial extent of the anomaly fields from AQUM and OMI under the different seasonal weather regimes metrics such as correlation, slope of the linear regression and RMSE could be used, but these have limitations. Correlation only accounts for the spatial patterns of the anomalies and not the magnitude. Also, it does not account for the significance of the anomalies. Linear regression should indicate the best AQUM-OMI agreement when tending towards a 1 : 1 fit. However, this metric does not account for anomaly significance either. RMSE does not always give a good indication of the error in the anomaly field magnitudes or in the spatial extent of the significant anomaly clusters. Here, we use the term "cluster" to represent a grouping of positive or negative significant anomalies. For instance, if an anomaly cluster for AQUM has a smaller spatial extent than OMI, the error magnitudes will be larger where the two are different, degrading the comparisons. Comparisons can also be degraded if the anomalies in AQUM and OMI are similar but offset slightly (e.g. should the model anomaly cluster be offset to the east by 0.5°).

A more appropriate method to compare AQUM and OMI column NO_2 under the four regimes, which we do here, is to analyse both the spatial extent of the significant anomalies and their magnitude. For each of the seasonal synoptic regimes the number of significant positive and negative column NO_2 anomalies (pixels) were calculated. This represents the spatial extent of significance. The anomalies were grouped into separate counts of the positive and negative anomaly clusters as they show independent features across the model domain. To ascertain the magnitude of the anomaly clusters, the average positive and negative anomaly was calculated. This means that the spatial extent and size of the anomalies are both accounted for. We then define the cluster density to be the product of the respective cluster size (i.e. number of pixels) and its average anomaly magnitude giving:

$$\phi_{\pm} = \alpha_{\pm} \times \eta_{\pm} \qquad (4)$$

where ϕ is the anomaly cluster density, α represents the size of the anomaly cluster, η is the average magnitude of the anomaly cluster and $\pm$ indicates if it is the positive or negative anomaly cluster density. The AQUM and OMI anomaly cluster densities were then compared using the fractional gross error (FGE). FGE is a normalised metric of the model's deviation from the observations which performs symmetrically with respect to under- and over-prediction, and is bounded by the values 0–2 (for more information see Savage et al., 2013; Pope et al., 2015). In this study's context, the FGE is represented by:

$$\mathrm{FGE}_{\pm} = 2\left|\frac{\phi_{\mathrm{AQUM}_{\pm}} - \phi_{\mathrm{OMI}_{\pm}}}{\phi_{\mathrm{AQUM}_{\pm}} + \phi_{\mathrm{OMI}_{\pm}}}\right| \qquad (5)$$

In Fig. 7, the AQUM-OMI positive and negative FGEs for the four seasonal/synoptic cases are plotted against each other in red. The smaller the FGE, the closer the AQUM-OMI column NO_2 comparisons are under the seasonal synoptic regimes. A goal zone of $x = 0, y = 0$ would show that AQUM can accurately simulate the column NO_2–LWT relationships seen by OMI. However, this method only works if the anomaly clusters are in similar locations in the AQUM and OMI fields. From observation of Figs. 4 and 6, the anomaly dipole clusters cover the same regions in both datasets and spatial variances (R^2), discussed in more detail at the end of the section, show high associations between the two (i.e. the anomaly clusters are in similar locations). Therefore, we suggest that we can use this methodology to assess the skill of AQUM in simulating seasonal synoptic relationships seen in the OMI data, by looking at the size and magnitude of the anomaly clusters. In Fig. 7 we have added 4 arbitrary zones which indicate the closeness to the goal of $x = 0, y = 0$.

Summer cyclonic conditions give the best comparisons with positive and negative FGEs of approximately 0.4 and 0.45, respectively. This falls in Zone 1, closest to the (0,0) goal zone. Winter anticyclonic conditions have the next best agreement as the negative FGE shows small differences of under 0.1. Therefore, AQUM under these conditions can accurately represent the OMI negative anomaly pattern. However, the positive FGE is approximately 0.75 resulting in a comparison skill in Zone 2. The winter cyclonic conditions present FGE values of approximately 0.7 for both anomaly clusters falling into Zone 2 as well. Summer anticyclonic conditions show the poorest comparisons falling in Zone 4 with reasonable agreement in the positive FGE of 0.4–0.5, but 1.5 in the negative FGE. This appears mostly to be a result of the smaller magnitude and extent of the negative anomalies in the proximity of the North Sea within the model, where they are significant for much fewer pixels (Fig. 6b) than in the observations (Fig. 4b).

In Table 2 we justify using our approach of using the anomaly clusters and FGE when compared with other statistical metrics. The table highlights the order in which AQUM most successfully reproduces the OMI column NO_2 anomalies when sampled under the seasonal synoptic regimes. Like the correlation and RMSE, our method has summer cyclonic, winter anticyclonic and winter cyclonic in the same order. However, summer anticyclonic has the worst comparisons using our method. This is because in the anomaly fields (Figure 6), our method shows AQUM does not simulate significant negative biases whereas the other metrics show the best apparent agreement. This justifies our new method as it takes into account the significance of the anomalies, unlike the other metrics.

The spatial variance (R^2) between AQUM and OMI column NO_2 anomalies (both significant and non-significant) is 0.70, 0.61, 0.68 and 0.59 for summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. This represents the proportion of spatial variability in OMI column NO_2 anomalies captured by the AQUM column NO_2 anomalies for each seasonal synoptic regime. For all the seasonal regimes, the association between the AQUM and OMI anomaly fields is significant large, with peak associations in the anticyclonic comparisons. ~~This gives us~~ As the associations are strong, the anomaly spatial patterns are located in similar locations, as can be seen in Figures 4 and 6. Therefore, this provides us with further confidence to use the methodology discussed in ~~Eq. (5)~~ Equation 5 to analyse the size and spread of the significant anomalies for each seasonal synoptic regime. Interestingly, even though AQUM does not simulate the significant negative anomalies over the North Sea (worst comparisons in Fig. 7) under summer anticyclonic conditions (Fig. 6b), it does capture the spatial variability in the OMI anomalies (Fig. 4b) better than under the other regimes.

4.3 AQUM tropospheric column tracer–LWT relationships

Section 4.2 has shown that AQUM successfully reproduces the relationships seen by OMI column NO_2 when sampled under the LWTs. Therefore, AQUM can be used as a tool to diagnose the influence of meteorology and chemistry on the distribution of NO_2 under the seasonal weather regimes. Here idealised tracers are introduced into AQUM with e-folding lifetimes of 1, 3, 6, 12, 24 and 48 h. They are emitted with the same loading and over the same locations as the model NO_x. This method of using e-folding tracers has been applied in inverse modelling of NO_x emissions from satellite data. For example, Richter et al. (2004) used SCIAMACHY column NO_2 measurements and simple approximations of NO_x loss (i.e. a fixed lifetime of NO_x) to estimate shipping emissions over the Red Sea. These idealised tracers will indicate the importance of transport and atmospheric chemistry governing the relationships between column NO_2 and seasonal synoptic weather. If transport is the main factor governing the air quality distribution under the different synoptic regimes, then a fixed lifetime tracer would have similar anomaly fields to NO_2. On the other hand, if changes in chemistry are driving or significantly contributing to the different regime anomalies, then a certain fixed lifetime tracer would be unable to capture the observed differences. Therefore, depending on which of the tracers with different lifetimes results in anomaly fields most similar to the AQUM column NO_2 anomalies, for winter and summer cyclonic and anticyclonic regimes, the relative importance of the processes can be determined and an approximation for the model lifetime of NO_2.

As the chemistry of NO_x is complex, with non-linear relations via ozone, diurnal cycles and varying emissions, a simple e-folding tracer will never truly match the NO_2 distribution. However, this approach is less complex than investigating chemical budgets and wind fields, which are not available from the AQUM for this study. Also, the direct lifetime of NO_2 cannot be determined as fluxes through the model boundaries are likely a strong sink or source under different conditions. Therefore, the tracers will indicate transport and chemical representation to a first-order approximation, and can be used to answer questions such as “Does the use of tracers support the well-known fact that the chemical lifetime of NO_2 is shorter in summer than in winter?; if so does synoptic meteorology have a smaller effect on NO_2 columns in summer than in winter?”.

The same method of compositing AQUM column NO_2 has been applied to the e-folding tracer columns. The tracer anomalies under the seasonal synoptic conditions are shown in Figs. 8 (summer) and 9 (winter) with OMI AKs applied. The tracers successfully reproduce the spatial patterns seen in the AQUM and OMI column NO_2 sampled under the different seasonal synoptic regimes. However, the area size of the tracer anomalies (both the negative and positive clusters) are a function of the tracer lifetime. In the case of the tracers with 1 and 3 h lifetimes (tracer_1 and tracer_3), the anomaly cluster areas are small. The short lifetime means that there is less column tracer to be accumulated or transported under anticyclonic or cyclonic regimes. With the longer lifetimes, $\text{tracer}_{24\text{ and }48}$, these anomaly cluster areas cover a larger proportion of the domain. This pattern can be seen in Fig. 10, where as the lifetime increases from 1 to 48 h, the cluster size of significant pixels (positive and negative totals combined) increases from a fraction of 0.0 to 0.3–0.5 (depending on seasonal synoptic regime). This clearly shows that the lifetime of the tracer is important and has an impact on the spatial pattern (area size) of the tracer column anomalies.

The summer and winter anticyclonic curves in Fig. 10 are very similar reaching approximately 0.35 for tracer_{48}. This suggests that under anticyclonic conditions differences in meteorology between the two seasons have relatively little impact on the area of significant tracer columns. Thus the chemistry is playing an important role in the summer to winter differences in the spatial distributions. However, under cyclonic conditions, the winter anomalies are somewhat larger than the summer ones, reaching approximately 0.51 and 0.47, respectively, for tracer_{48}. Here differences in meteorology between summer and winter are playing a more active role suggesting that winter cyclonic systems are more intense than summer ones. ~~Wind data were not output in the AQUM model runs, so 2006–2010 winter~~ 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 ~~summer average wind flows over the UK from ECMWF ERA-Interim (available at – **?**) were investigated (not shown here). Over the northern and western parts of the AQUM domain , the wind speeds in winter are around 5–12and tend to be larger than in summer (3–9)~~2.9 m/s. Therefore, the synoptic type wind speeds are stronger in winter. Thus, the stronger transport in winter probably explains the difference in the cyclonic curves in ~~Fig.~~ Figure 10.

The analysis performed previously for the FGEs of the AQUM and OMI column NO_2 anomaly cluster densities (Fig ~~.~~7) was repeated for the FGEs of the AQUM column NO_2 and tracer column anomaly cluster densities in Fig ~~. 11.~~ 11. Therefore, in Equation 5, $\phi_{AQUM_\pm}$ has been replaced with $\phi_{tracer_\pm}$ and $\phi_{OMI_\pm}$ has been replaced with $\phi_{AQUM_\pm}$. The aim is to find which tracer lifetimes most accurately represent the NO_2 lifetime under the seasonal synoptic regimes. Overall, $\text{tracers}_{1,\,3 \text{ and } 48}$ have the least accurate lifetimes with skill comparisons in Zone 4, because the domain coverage of the tracer anomalies is either too small or too large (the winter tracer$_{48}$ regimes fall into Zone 3). The most accurate tracer lifetime for summer cyclonic and anticyclonic regimes is tracer$_{6}$, with FGE values between 0.3 (Zone 1) and 0.6–0.7 (Zone 2), respectively. The winter cyclonic and anticyclonic regimes are most accurately represented by tracer$_{12}$; both of them fall into Zone 1 with FGE values lower than 0.4. This is more consistent with chemical processes in summer than winter acting as a loss of $\mathrm{NO_2}$. To verify this result, the AQUM column $\mathrm{NO_2}$ significant anomaly domain fraction was calculated at 0.02, 0.04, 0.07 and 0.09 for summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. Reading across to the respective tracer profiles in Figure 10, the approximate $\mathrm{NO_2}$ lifetimes are 6.0, 4.5, 11.0 and 7.0 hours, respectively. This supports the tracer results in that summer $\mathrm{NO_2}$ lifetimes are shorter than that of winter. It should be noted though that this approach does not take into account the magnitude of the anomalies.

Having found the best representations of the seasonal synoptic regimes' lifetimes, the respective tracer anomaly fields were correlated against the AQUM column $\mathrm{NO_2}$ anomalies. Since the tracer lifetime was fixed, the variance between the tracer fields and the column $\mathrm{NO_2}$ represents the proportion of meteorological variability in the spatial pattern of the anomalies within the season (the emissions for each seasonal synoptic regime $\mathrm{NO_2}$ – tracer comparison are equal). The variances (R^2) are 0.92, 0.87, 0.80 and 0.75 for the summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. Therefore, a large proportion of the seasonal variability in the spatial patterns, under the seasonal synoptic regimes, is explained by the meteorology (e.g. transport) and the remaining variability is due to the chemistry and emissions.

5 Conclusions

The ~~LWTs–OMI~~ LWTs (cyclonic and anticyclonic) - OMI tropospheric column $\mathrm{NO_2}$ relationships discussed by Pope et al. (2014) for a ~~7~~7-year period have been analysed for the ~~2006–2010~~ 2006-2010 period simulated by AQUM in order to investigate the model's ability to capture the impact of synoptic weather on tropospheric column $\mathrm{NO_2}$.

AQUM column NO_2, composited in the same way as OMI data by using the LWTs directly, successfully captured the OMI column NO_2 ~~–LWT relationships~~anomalies for cyclonic and anticyclonic LWT conditions. Under anticyclonic conditions, AQUM column NO_2 accumulates over the source regions, while it is transported away under cyclonic conditions. This also shows that the representation of weather systems through the model LBCs is sufficiently consistent with the NCEP reanalyses that the LWTs derived from NCEP can be used to investigate the influence of synoptic weather regimes on air quality.

To determine which processes are important in driving these relationships, idealised tracers were introduced into the model using the NO_x emission sources and selected lifetimes ranging from 1 to 48 h. The tracers reproduce the AQUM column NO_2 anomaly fields under the different seasonal synoptic regimes, but the relationships found depend heavily on the lifetime. A 1 h lifetime was clearly too short and a 48 h lifetime clearly too long, resulting in smaller/larger anomaly patterns when compared with the model column NO_2. The most representative tracer lifetimes are 6 h in summer and 12 h in winter, which is consistent with enhanced photochemistry in summer. The variance (R^2) between the most representative tracer lifetimes for the seasonal synoptic regimes and the corresponding AQUM column NO_2 spatial anomaly fields were calculated. This resulted in R^2 values ranging between 0.75 and 0.92. Therefore, within seasons (i.e. summer and winter), under the synoptic regimes, a large proportion of the spatial pattern in the UK column NO_2 fields can be explained by these tracers, suggesting that transport is a significant factor in governing the variability of UK air quality on seasonal synoptic timescales. We also show that cyclonic conditions have more seasonality than anticyclonic conditions as winter cyclonic conditions result in more extreme spatial column NO_2 distributions from the seasonal average.

This study shows that to a first-order approximation atmospheric chemistry is, as expected, more influential in summer than in winter. During summer the NO_2 lifetime decreases due to enhanced NO_2 photolysis and OH chemistry, which explains the less spatially significant synoptic weather-air pollution relationships detected for that season in OMI column NO_2 (Pope et al., 2014). This work also shows that the Met Office AQUM can reproduce the large-scale accumulation of ~~air pollution~~ tropospheric column NO_2 over the UK under anticyclonic conditions.

As follow on work from this study, we intend to perform a sensitivity analysis of different AQUM production and loss processes of NO_2 to determine the governing factors on the distribution of column NO_2.

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Table 1. The non-bold elements show the 27 basic Lamb Weather Types with their number coding. LWTs also include ~~—1~~ -1 (unclassified) and ~~—9~~ -9 (non-existent day). ~~In this work these LWTs are~~ Pope et al. (2014) grouped the LWTs into 3 circulation types and 8 wind directions, as indicated in bold characters by the outer row and column. However, in this study we focus on the cyclonic and anticyclonic conditions.

This Work	**Anticyclonic**	**Neutral Vorticity**	**Cyclonic**
	0 A		20 C
North-easterly	1 ANE	11 NE	21 CNE
Easterly	2 AE	12 E	22 CE
South-easterly	3 ASE	13 SE	23 CSE
Southerly	4 AS	14 S	24 CS
South-westerly	5 ASW	15 SW	25 CSW
Westerly	6 AW	16 W	26 CW
North-westerly	7 ANW	17 NW	27 CNW
Northerly	8 AN	18 N	28 CN

Table 2. Highlights the skill rank of the seasonal synoptic regimes for which AQUM can simulate column NO_2 when compared with OMI column NO_2 using correlation, slope-of-regression, RMSE and the method proposed here. 1: Best AQUM-OMI agreement, 4: Worst AQUM-OMI agreement.

Rank	**Correlation**	**Regression**	**RMSE**	**New Method**
1	Summer Anticyclonic	Summer Anticyclonic	Summer Anticyclonic	Summer Cyclonic
2	Summer Cyclonic	Summer Cyclonic	Summer Cyclonic	Winter Anticyclonic
3	Winter Anticyclonic	Winter Cyclonic	Winter Anticyclonic	Winter Cyclonic
4	Winter Cyclonic	Winter Anticyclonic	Winter Cyclonic	Summer Anticyclonic

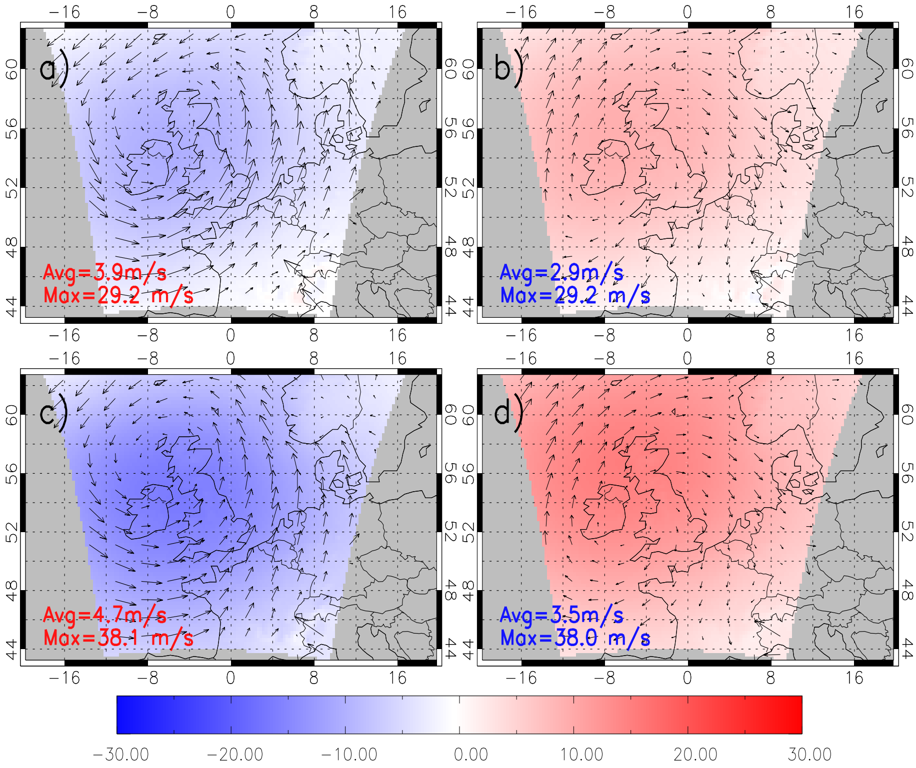

Figure 1. Pressure anomalies (hPa) relative to the seasonal average (2007-2010) with the wind circulation over plotted. a) summer cyclonic, b) summer anticyclonic, c) winter cyclonic and d) winter anticyclonic, all derived from the Lamb Weather Types.

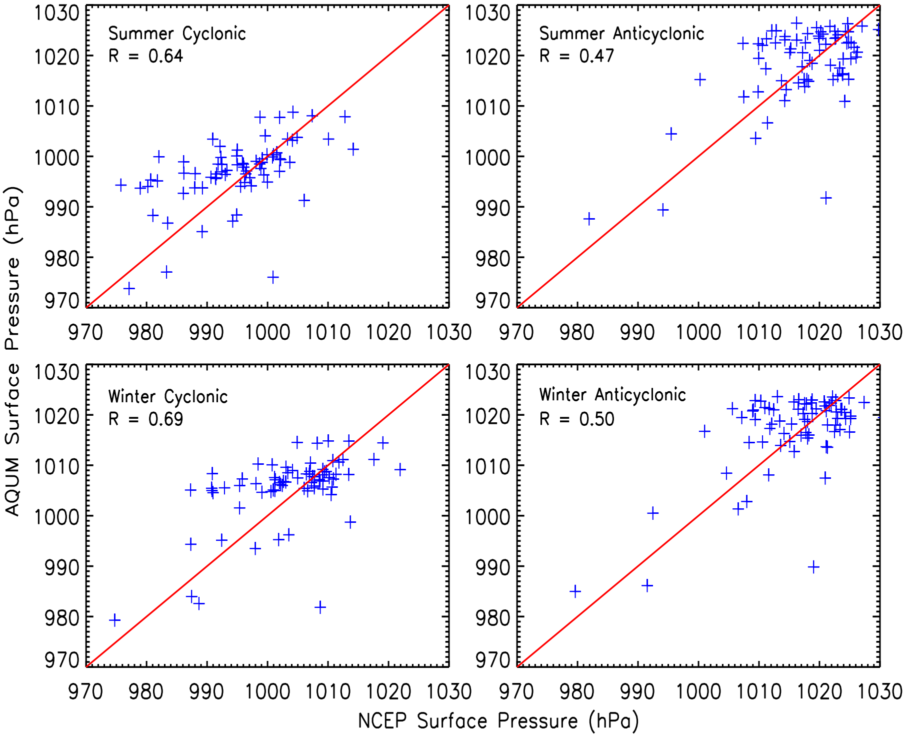

Figure 2. AQUM surface pressure vs. NCEP surface pressure (2006-2010), both composited under summer and winter cyclonic and anticyclonic conditions. The correlations are based on Spearman's Rank with a significance level of $p < 0.001$.

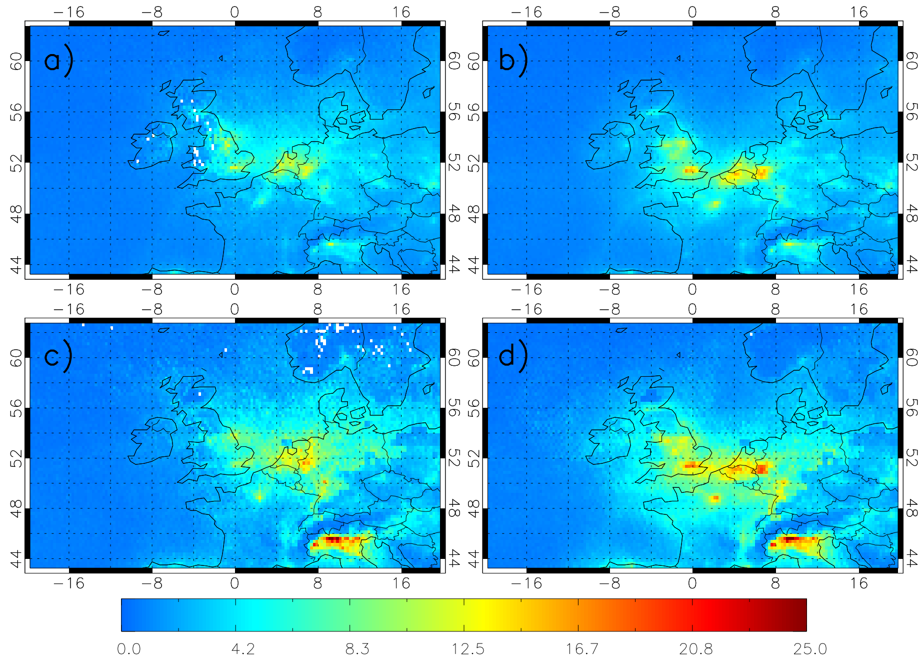

Figure 3. Composites of OMI tropospheric column NO_2 (10^{15} molecules cm^{-2}) for **(a)** summer cyclonic, **(b)** summer anticyclonic, **(c)** winter cyclonic and **(d)** winter anticyclonic conditions during 2006–2010.

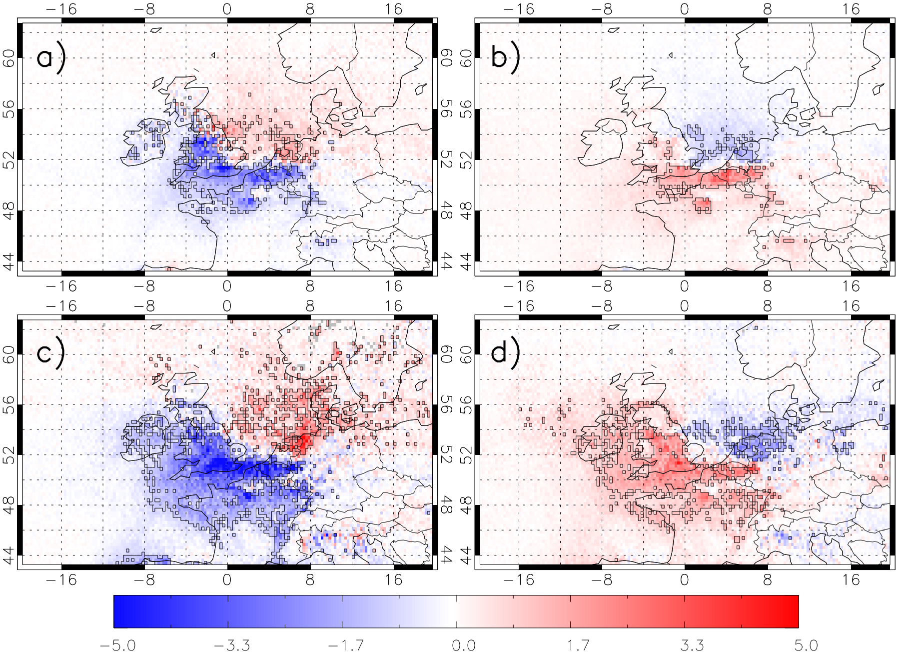

Figure 4. Anomalies of OMI tropospheric column NO_2 composites (calculated as the deviations with respect to the seasonal 5 year averages, 10^{15} molecules cm^{-2}) for **(a)** summer cyclonic, **(b)** summer anticyclonic, **(c)** winter cyclonic and **(d)** winter anticyclonic conditions. Black boxes indicate where the anomalies are statistically significant at the 95 % level.

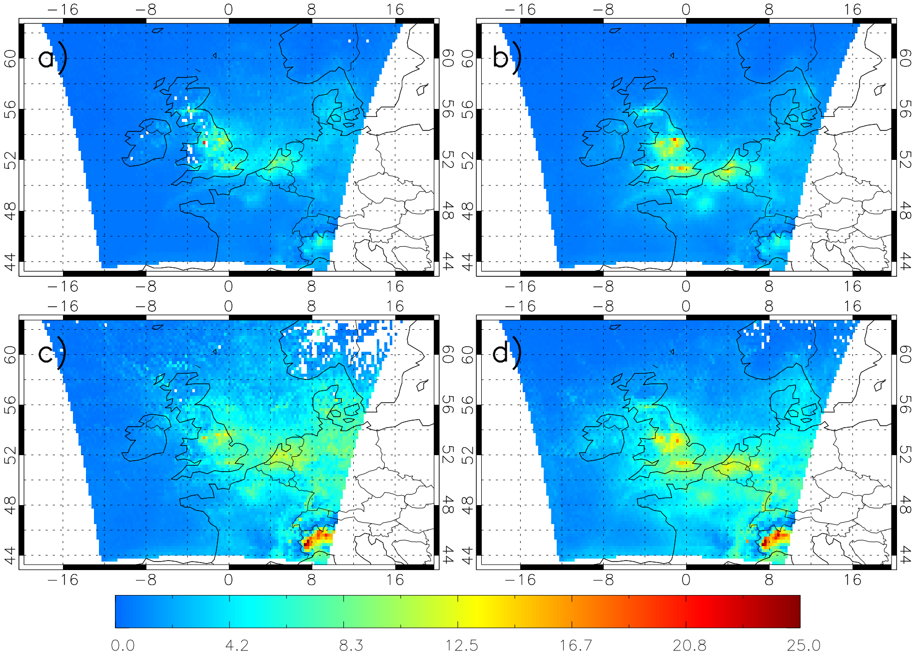

Figure 5. Composites of AQUM tropospheric column NO_2 (10^{15} molecules cm^{-2}) for **(a)** summer cyclonic, **(b)** summer anticyclonic, **(c)** winter cyclonic and **(d)** winter anticyclonic conditions (OMI AKs applied) during 2006–2010.

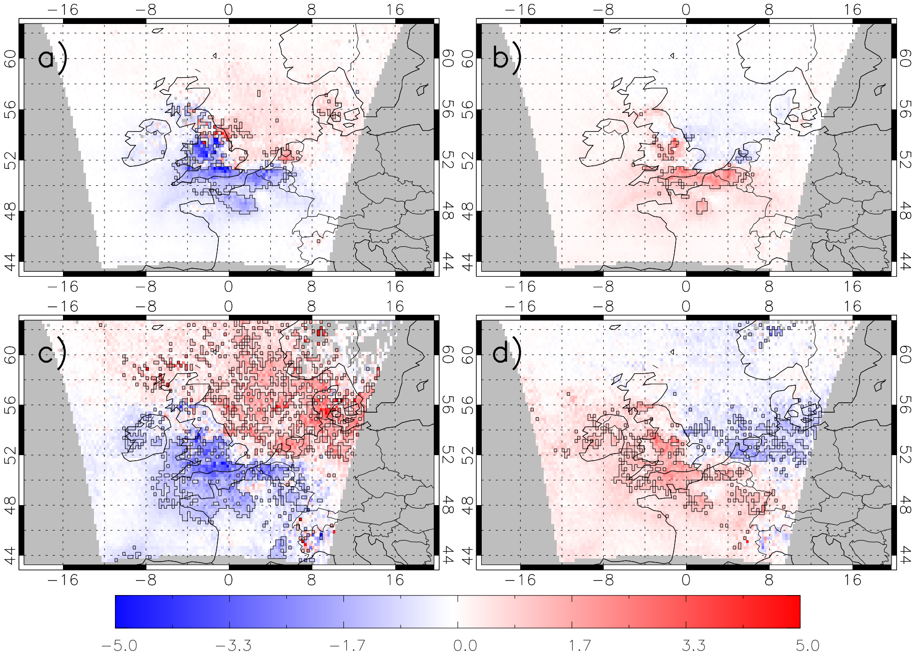

Figure 6. Anomalies of AQUM tropospheric column NO_2 composites (calculated as the deviations with respect to the seasonal 5 year averages, 10^{15} molecules cm^{-2}) for **(a)** summer cyclonic, **(b)** summer anticyclonic, **(c)** winter cyclonic and **(d)** winter anticyclonic conditions (OMI AKs applied).

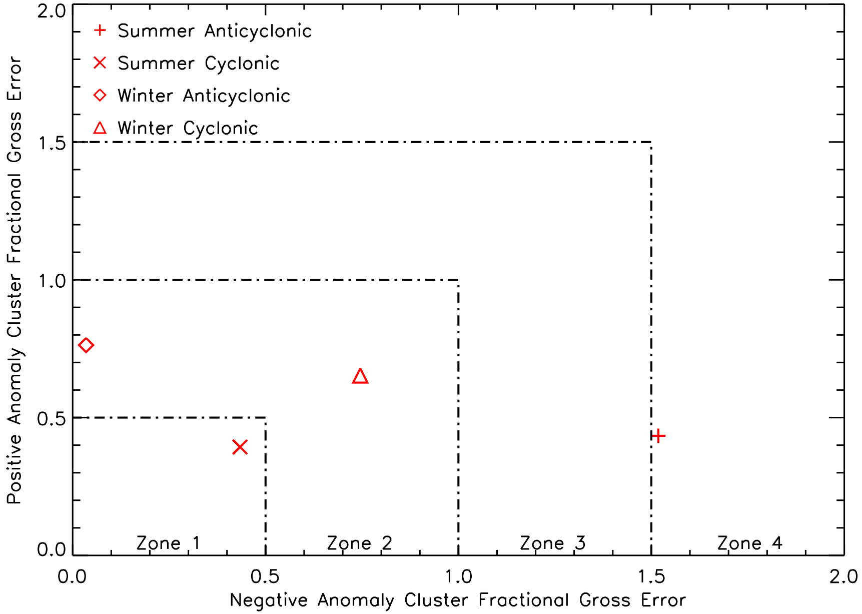

Figure 7. The fractional gross error of the AQUM-OMI positive and negative anomaly cluster densities are plotted against each other for different seasonal synoptic regimes. The best agreement between AQUM-OMI column NO_2 is at the goal zone $(x = 0, y = 0)$ showing no error. Zones 1–4 represent areas of skill between 0.0–0.5, 0.5–1.0, 1.0–1.5 and 1.5–2.0. The lower the zone, the better the comparison is.

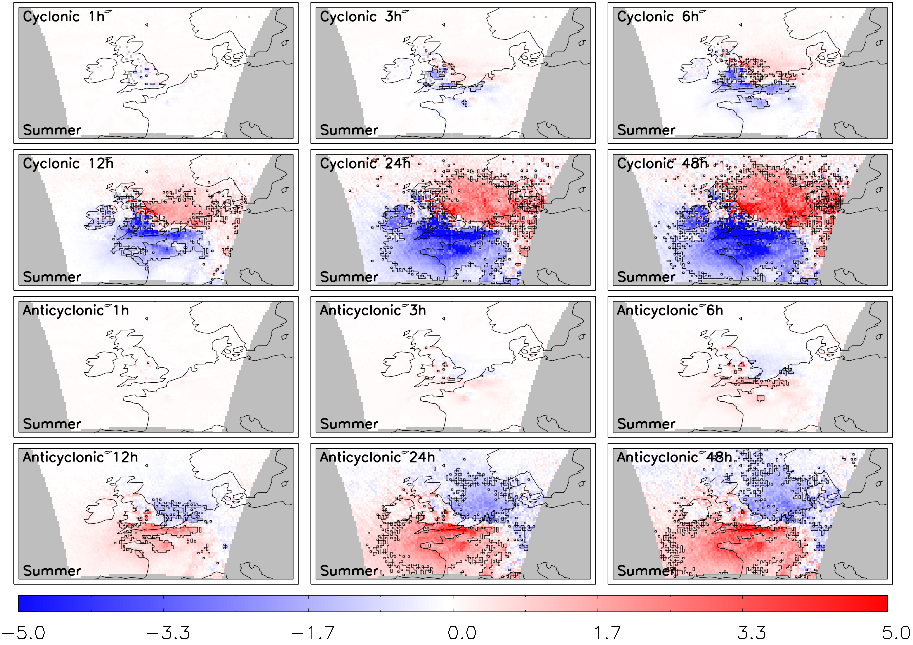

Figure 8. Summer AQUM column tracer anomalies (10^{15} molecules cm^{-2}) with different lifetimes for cyclonic and anticyclonic conditions (OMI AKs applied).

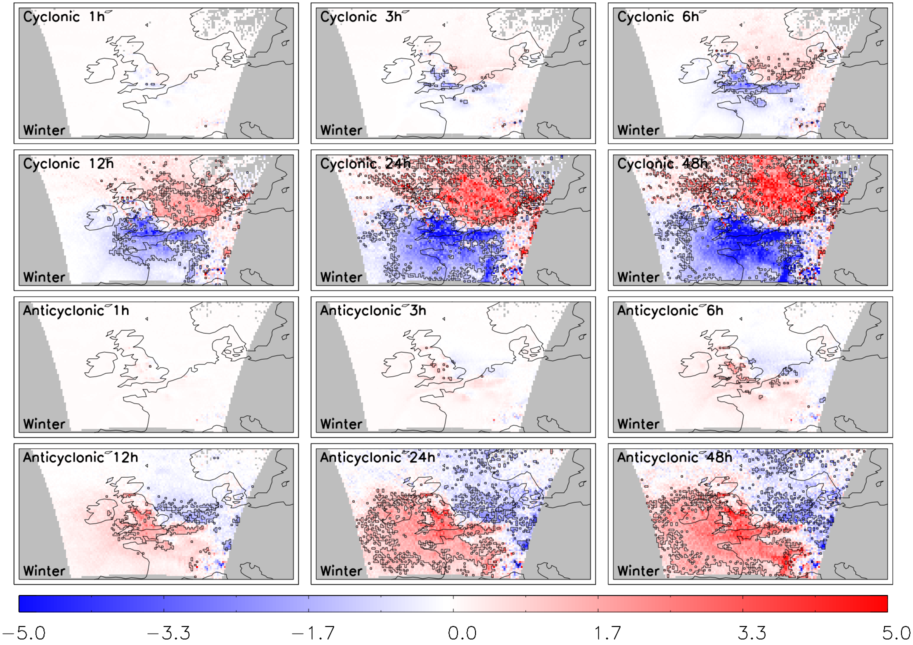

Figure 9. Winter AQUM column tracer anomalies (10^{15} molecules cm^{-2}) with different lifetimes for cyclonic and anticyclonic conditions (OMI AKs applied).

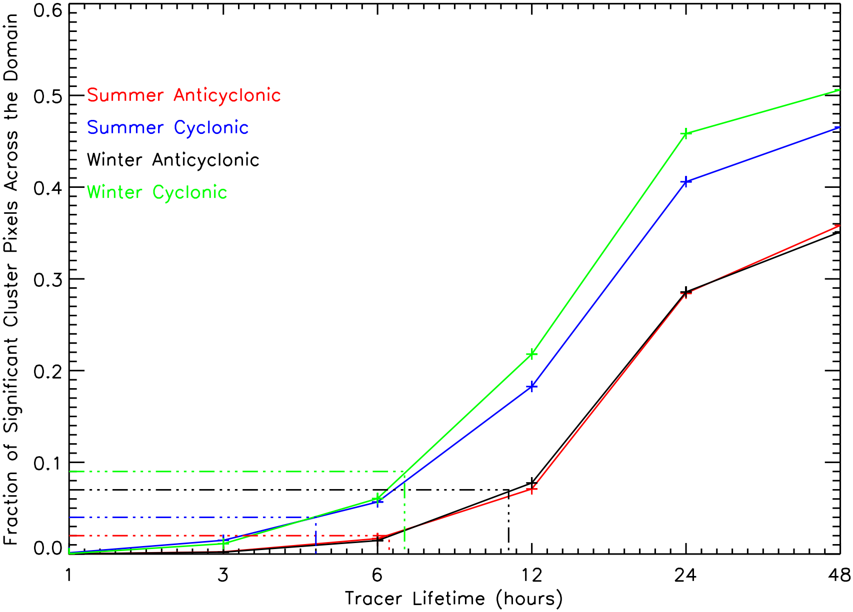

Figure 10. Proportion of the AQUM domain covered by significant anomaly pixels as a function of tracer lifetime for the different seasonal synoptic regimes. Red, blue, black and green represents the summer anticyclonic, summer cyclonic, winter anticyclonic and winter cyclonic conditions, respectively. Dashed lines represent the approximate life time of AQUM column NO_2 under the seasonal synoptic regimes based on the domain proportion of significant anomalies (pixels) in Figure 6.

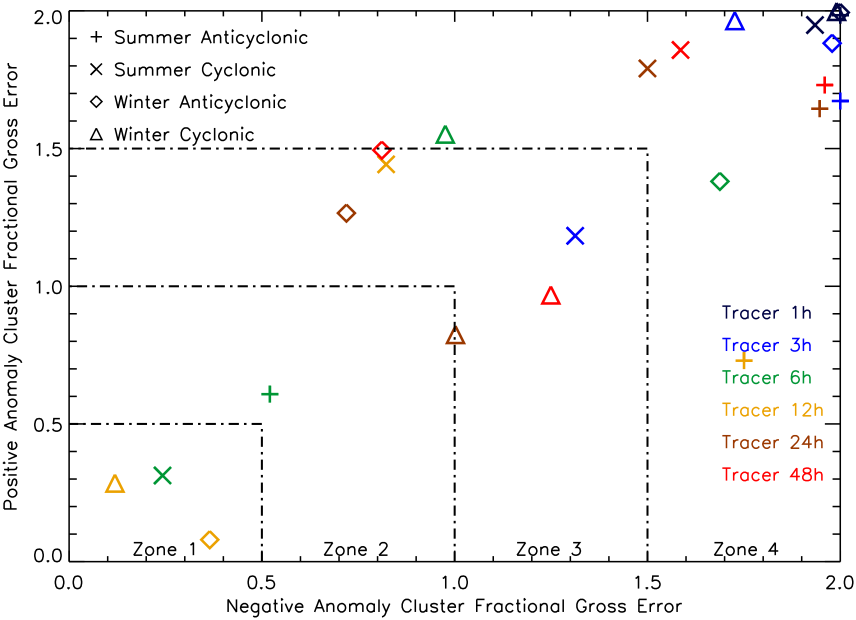

Figure 11. The same as Fig. 7, but for the anomaly cluster densities of AQUM column NO_2 – AQUM tracer columns. The different colours refer to the AQUM tracer experiments with e-folding lifetimes of 1, 3, 6, 12, 24 and 48 h.