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 1590 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, 12.5 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, 12.5 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 18594 Line 16, will be added in section 4.1. Then on Page 18582 Line 4 "*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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Savage, N. H., Pyle, J. A., Braesicke, P., Wittrock, F., Richter, A., Nüß, H., Burrows, J. P., Schultz, M. G., Pulles, T., and van Het Bolscher, M.: The sensitivity of Western European NO2 columns to interannual variability of meteorology and emissions: a model-GOME study, Atmos. Sci. Lett., 9, 182–188, 2008.

Savage, N. H., Agnew, P., Davis, L. S., Ordóñez, C., Thorpe, R., Johnson, C. E., O'Connor, F. M.,and Dalvi, M.: Air quality modelling using the Met Office Unified Model (AQUM OS24-26):model description and initial evaluation, Geosci. Model Dev., 6, 353–372, doi:10.5194/gmd-6-353-2013, 2013.

Visschedijk, A., Zanveld, P. and van der Gon, H.: A high resolution gridded European Emission Database for the EU integrated project GEMS, TNO report 2007-A-R0233/B, 2007.

Zhang, R., Tie, X. and Bond, D. W.: Impacts of anthropogenic and natural NO_x sources over the US on tropospheric chemistry, PNAS, 100,1505-1509, 2003.