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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of a regional air quality model using satellite column NO_2: treatment of observation errors and model boundary conditions and emissions" *by* R. Pope et al.

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We thank the reviewer for his/her comments. These comments are repeated below surrounded by quotation marks. Our responses follow in normal text.

1-1: "Biogenic emission: Biogenic species (i.e., isoprene, mono-terpene, etc) play an important role even in NOx chemistry by controlling OH radical concentrations. For example, if isoprene emissions are overestimated, the estimated levels of NO2 can be higher than the actual levels of NO2 (because of slow NOx losses). The tropo-

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spheric chemistry can, sometimes significantly, influence the NOx analyses for summer episode. Thus, authors should clarify which biogenic inventory (e.g., MEGAN, GEIA, etc) was used in your study and explain how the biogenic inventory is uncertain (or reliable)."

As stated in Savage et al (2013) “Biogenic emissions of isoprene are from the monthly climatological data of Poupkou et al. (2010) at 0.125 degrees × 0.0625 degrees resolution. The use of climatological emissions for biogenic isoprene sources will diminish the ability of the model to respond to increased biogenic ozone precursor emissions during episodes, but this is not expected to be a major factor in the cases analysed in this paper. An interactive biogenic isoprene emission scheme is under development, but is not yet available for use in AQUM.”

The following text will be added at the end of Section 3.1: “Poupkou et al. (2010) provide the monthly climatology of biogenic emissions on a 0.125 degree × 0.0625 degree resolution. The use of climatological biogenic isoprene emissions will partially diminish AQUM’s representation of ozone from biogenic precursors. A new interactive biogenic isoprene scheme is under development by was not available for this study. However, this is a secondary issue in this paper as we focus on primary emissions of NOx, which affect ozone concentrations”.

1-2: "Biomass burning emission: I wonder whether biomass burning emission was considered in the CTM simulations. If it was considered, authors need to mention/describe it."

Again to quote from Savage et al “Biomass burning emissions of aerosols are taken from year 2000 values from the Global Fire Emissions Database (GFED) version 1 (Randerson et al., 2005). The choice of 2000 emissions is somewhat arbitrary, but these emissions have relatively little impact on our domain.”

We will add the following text at the end of Section 3.1 also: “Biomass burning emissions of aerosols come from the Global Fire Emissions Database (GFED) version 1

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(Randerson et al., 2005) for 2000. The use of biomass burning emissions from 2000 is somewhat arbitrary, but within AQUM's domain these emissions have relatively little impact."

1-3a: "Variations of NOx emissions: This issue can be an important factor in the analysis of seasonal trends of columnar NO2. Authors should provide the seasonal (or monthly) information of NOx emission. Also, this information can be useful to analyse the seasonal trends of columnar NO2 over the London and northern England regions in Fig.3."

We will add a new figure in line with the reviewer comments at the end of Section 3.1 in the discussion of AQUM NOx emissions. The figure shows the seasonal cycle (fraction) applied to the AQUM annual NOx (point and area sources) emissions total from the NAEI, ENTEC and EMEP. The seasonal cycle comes from Visschedijk et al., (2007).

1-3b:"Higher columnar NO2 over London and northern England from OMI observations are well captured by 3D-CTM simulation. However, usually, both NO2 columns from the CTM and satellite observations during winter are higher than those during summer (e.g., van Noije et al., 2006; Huijnen et al., 2010). Authors should explain the unusual trends of the NO2 columns (higher NO2 columns during summer) over the regions shown in Fig. 3."

This is true over continental Europe, however, as discussed by Pope et al., (2014) and van der A et al., (2008), the meteorological variability over the UK leads to different column NO2 signals over source regions. van der A et al., (2008) suggest that peak UK NOx emissions occur in July. Pope et al., (2014) suggest that the transport of column NO2 away from source regions due to strong winter dynamics outweighs the loss of UK source region column NO2 from enhanced summer photochemistry. Therefore, UK source region column NO2 is larger in summer than winter.

In Section 4.1 on page 21764, line 12, the following text will be added: “van der A et

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al., (2008) suggest that peak UK NOx emissions occur in July, while Pope et al., (2014) suggest that the transport of column NO2 away from source regions due to strong winter dynamics outweighs the loss of UK source region column NO2 from enhanced summer photochemistry.”

2-1: "Chemical LBCs: In the manuscript, authors mentioned that using chemical GEMS gives better result and it is consistent with the findings of Savage et al. (2013). However, authors do not mention what makes it better. What are the main differences between the GEMS and MACC LBCs? If the MACC is an improved version of the GEMS, what has been improved? Give some more detailed information on both the GEMS and MACC."

The MACC reanalysis used a more recent version of the ECMWF model (Integrated Forecast System), assimilated more satellite products and was run at a resolution of 80 km instead of 125 km. Savage et al found that the ozone bias from January to May when using GEMS to provide LBCs data had a larger negative bias than those made with the MACC LBCs. From May to the end of the year runs using the GEMS LBCs generally perform better, with a smaller positive bias.

The following sentence will be added in Section 3.2 at the end of line 10: “The MACC reanalysis uses a more recent version of the ECMWF model (Integrated Forecast System), assimilated more satellite products and was run at a resolution of 80 km instead of 125 km.”

2-2: "E2 (idealized point source tracer): In the E2 sensitivity test, authors tried to examine the spatial patterns of the tracer. The experiment remains unclear in the manuscript. i) How to idealize tracer from the point sources? ii) Which species are used as a tracer? It appears to be a “reactive species” having one-day lifetime. iii) What are the reasons to determine a tracer having one-day lifetime? The lifetime of NOx is less than 1 day (say, several hours) during summer (Schaub et al., 2007; Lamsal et al., 2010). iv) In Fig. 7-c, the columnar NO2 over other regions seems to be “zero”.

Did you consider only tracer's emission for this sensitivity simulation? If yes, what is the point of considering one trace species specially having one day lifetime? If no, was the tracer tagged for identification and how to tag the tracer in your model?"

We will improve the explanation of the implementation of this tracer in the manuscript to address these points. To answer the reviewer's specific comments:

i) The tracer is emitted at the location of UK NOx point source emissions with emissions which are the same as those of NOx from the power station sources in the model inventory.

ii) The idealised e-folding tracer is a separate transported tracer, using the same tracer transport scheme as the chemical tracers but not having any interactions with the chemistry scheme. It has a fixed lifetime, with a first-order loss applied to the tracer at every model time-step.

iii) The choice of a lifetime of 1 day is somewhat arbitrary and is indeed longer than the lifetime of NOx in summer at this latitude. We did not have the resources available to run tracers with a range of lifetimes for this case in order to estimate the most appropriate lifetime. However, we do not believe that this will have a significant impact on the results.

iv) Figure 7c only represents the idealised tracer field from UK NOx point source emission sources. Therefore, the concentrations over Europe and the sea are near zero. Yes, we only consider the tracer for Run E2 to look at the correlation of the point source emissions and mean biases in Figure 6a. This is not a “tagged” NOx tracer, as stated above – it is an idealised tracer with a simple first-order loss rate. The aim of this part of the study was to test whether there was a link between the emissions from the power stations and the observed areas of bias.

In line with comments i, ii, iii, the sentence on page 21761, line 20-22 “Run E2 uses an idealised passive tracer from the point sources with a lifetime of one day to examine if the tracer columns correlated with summer AQUM-OMI positive biases (see Sect. 4.3)." will be modified to "Run E2 introduces a new idealised passive tracer emitted from the UK point sources with the same emissions to that of the model NOx inventory. The idealised tracer can be transported like any chemical tracer, but cannot be lost through chemical reactions. Instead it is lost through its e-folding lifetime of one day. The point source tracer columns can then be examined to see if they are correlated with summer AQUM-OMI positive biases (see Sect. 4.3)." For comment iv, a new line has been added in Section 4.3 on page 2766, line 26: "The minimum tracer values of 0 x1015 molecules/cm2 are over the sea and continental Europe as there is no emission of the tracer there."

2-4: "Reaction probability of N2O5 (γN2O5): As mentioned in the manuscript, usually, the reaction probability of N2O5 is known to range from 0.02 to 0.001 (Riemer et al., 2003; Brown et al., 2006). It may exist somewhere between the two values. What are the reasons to choose only two extreme cases in the sensitivity simulations?"

We believe the range of γ between 0.001-0.02 is not extreme in the case of air quality modelling. Multiple air quality models use γ values between 0.0-1.0. For instance, Foley et al., (2010) used γ ranges between 0-0.06 in CMAQ and Menut et al., (2013) used γ values between 0.01-1.0 in CHIMERE. Macintyre and Evans (2010) explore the sensitivity of N2O5 uptake on aerosol for multiple γ values between 0.0-1.0. They state that intermediate γ values between 0.001-0.02 are the most sensitive values to this process. Therefore, we base the γ range in our study on Macintyre and Evans (2010). We did not explore other values of γ as this was not the main focus of this study.

3: "When the black polygonal regions are determined in Fig. 4, authors used the two values of MB and satellite error. Here, the satellite error is the criteria (i.e., the magnitude of the MB greater than the satellite error). In that case, how did you consider the satellite error (i.e., averaged value over domain or each pixel value)? For better understanding this analysis, authors should provide some ranges of satellite errors

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over specific regions (e.g. northern England + London, Benelux, Po valley, North Sea, etc)."

The satellite error is the average retrieval error with the random error component reduced, using the methodology in Section 2.2, for each pixel. A second table will be added to the revised manuscript to give information on some of the locations and example average retrieval errors. Please see the response to Reviewer 2 for more information.

4: "Where is Dartmoor located in England? For the sake of reader's convenience, you would better provide some geographical information (e.g., Dartmoor, Irish Sea, North Sea, Po Valley, and many regions mentioned in the manuscript), possibly in Fig. 2."

Please see the new table discussed in 3: and the response to Reviewer 2.

Specific Comment 1: "Merging two figures 3 and 4 in a 2 x 3 panel"

We did consider this before submission, but feel that if the figures were merged the images would become cramped and unclear. Therefore, we prefer to leave the figures as they are.

Specific Comment 2: "P. 21763, line 23 and P. 21764, line 4 (i.e., "around 0 – 3x1015" and "between 0 -6 x1015"). The minimum background columnar NO2 over background could not be "zero". Authors should provide approximate values."

These values will be changed to the minimum background values for the discussion of Figure 3 to "O(10ˆ13) -3 x10ˆ15 molecules/cm2, where O(10ˆ13) represents values in size of the order of 10ˆ13" and "O(10ˆ13) - 6 x10ˆ15" in Section 4.1 on page 21763, line 23, and on page 21764, line 4, respectively.

Specific Comment 3: "It is a quite interesting that by introducing N2O5 heterogeneous chemistry, these positive biases were "significantly" reduced even during the summer episode, indicating that columnar NO2 are significantly decreased. N2O5 is thermally unstable and decomposes to NO3 and NO2 at high temperatures. In other words,

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during summer, the decomposition of N2O5 is more active than the formation of N2O5. It would not have a significant impact on the columnar NO2 during summer. Thus, you would better provide other reasons in this part."

We agree with this comment over the rest of the domain in summer. However, over northern England, there is the largest loading of NOx and aerosols coming from the point sources. Therefore, even with decreased levels of N2O5, the model shows that this process is a significant sink of NOx. This process also occurs above the surface and in the free troposphere due to vertical transport and emissions entered in the model at 80, 180 and 320 metres (representing the emission from power stations with tall stacks). The temperatures are cooler here, so the thermal decomposition of N2O5 is reduced providing conditions suitable for heterogeneous chemistry to take place with the higher loading of NOx and aerosols. Therefore, we feel that the original conclusions stand and suggest no changes to the paper based on this comment.

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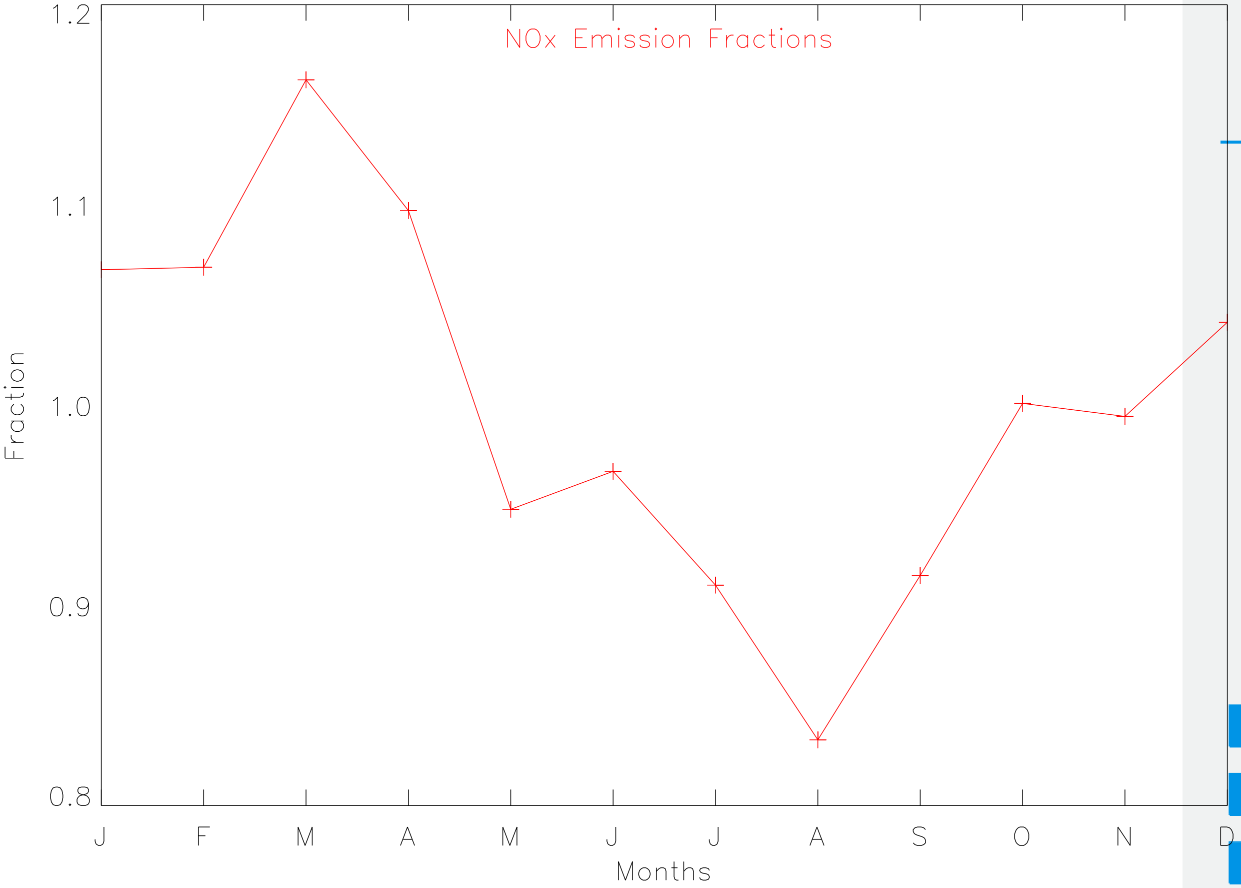

Fig. 1. NOx emissions seasonal cycle, based on Visschedijk et al., (2007), which is applied to AQUM's NOx emission annual totals.