

Response to Reviewer 1

Reviewer's comments are in italics, followed by our responses in non-italics.

This paper compares NO₂ and its variability in the UKCA model over South and East Asia to OMI observations. The impact of sampling at the time of day corresponding to the satellite overpass and incorporating the OMI averaging kernel is investigated. The evaluation reveals areas and seasons of bias in the simulated NO₂ and suggests several possible factors that could contribute to the bias. The study provides a robust evaluation of the UKCA simulation of NO₂ over South and East Asia. The manuscript would benefit from additional discussion of whether the results can be generalized to other models or regions to broaden its scientific significance.

This comment about generalizing is responded to below.

General comments:

1. In the discussion, can you compare your results to other modeling studies or comment on whether the same biases are likely to affect other models? This could broaden the applicability of the study.

We appreciate that being able to broaden the applicability of this study would be very useful. However, direct comparison to other modelling studies is difficult outside of something like a dedicated MIP (model intercomparison project), and, as far as the authors are aware, a large study like that has not been conducted since the early ACCENT model intercomparison (van Noije et al., 2006). That study mainly discussed differences between modelled and measured NO₂ in terms of discrepancies between NO₂ retrieval methods and the emissions used as inputs to the models rather than atmospheric processes within the individual models. Our study mainly describes the mechanics of how to meaningfully compare modelled and satellite measured NO₂ over S/E Asia, analyses recent trends, and discusses some potential reasons for model-measurement differences. A deeper understanding of these differences, firstly for the UKCA model, would be very useful. Extending the analysis to other models would also be very useful. However, our study is already fairly lengthy, and we feel that expanding it further is mainly beyond the scope of this particular paper. Nevertheless, we do extend the discussion to highlight that similar biases may well be present in other models, and further research, such as within a MIP dedicated to analysing NO_x budgets within models and evaluating column NO₂, would be of great benefit to the wider improvement and assessment of atmospheric chemistry models.

2. While the study focuses on South and East Asia, it would be helpful to include a figure or some discussion of how the model compares to OMI observations globally. Are the biases found over South and East Asia present in other regions as well? This might provide additional information on what the most likely cause is.

We again agree that extending the analysis to look globally would be beneficial, however, we decided to focus our study on S/E Asia. Archibald et al. (2020; their Figures 18 & 19) shows a global comparison, for a similar model set-up. The reasons for focussing on S/E Asia are laid out in the manuscript: e.g., it is the region of the world with the largest NO₂ columns, the greatest pollution problems, and also has shown large and divergent trends in NO₂ over the period studied. All these reasons make S/E Asia an obvious initial target region. It would definitely be interesting to know if the biases we see are present elsewhere, and we agree this

may help pin down the causes. But we feel it is beyond our scope within this paper to extend the analysis globally; we hope to do this in future studies.

3. Uncertainties in NO_x emissions and emission trends are another potential cause of model biases. More discussion is needed on the NO_x emissions used in the model and their uncertainties.

We agree that emissions may cause some of the model biases and now include some discussion of this. Whilst it is possible that emissions in industrial regions are overestimated and remote emissions are underestimated, we think that a process bias (such as related to insufficient heterogeneous removal and/or PAN formation) is probably a more likely contributor to the model-measurement discrepancies that we see, but further investigation is needed to make stronger statements.

Specific comments:

Page 5: Do you sample the model every day or only on days when there is a retrieval for a specific grid box? Is the model-obs comparison affected by clear-sky sampling?

The UKCA model is sampled daily at the satellite overpass time ($13:45 \pm 15$ minutes local time). For model-observation comparisons, only days and grid boxes with valid satellite retrievals are included. This ensures consistency in spatial and temporal sampling, reducing the potential for errors associated with inconsistent sampling. Although satellite data is filtered for clear-sky (i.e., cloud fraction < 0.2) conditions, the model is sampled under all conditions, including clouds, which may introduce bias. However, the UKCA model is nudged to ERA-Interim data, so when the real world has clear skies, the model should too, reducing transport and sampling inconsistencies and enhancing the reliability of the comparisons.

Line 145: How were these regions chosen?

Apart from whole country (India and China) analysis, we also selected NO_x hotspot and cleaner regions to capture contrasting pollution profiles across South and East Asia. For instance, West China has surface NO_x emission intensity that is approximately 30 times lower than East China, while South India's emissions intensity is about half that of North India (see new Figure S1, below). This approach allows for a comprehensive analysis across both high-emission and cleaner regions, providing broader insights into NO_x distribution. We have added this clarification in the manuscript.

Fig. S3: The caption of Fig. S3 says "trends" but the figure appears to show a timeseries.

We have modified the caption of Figure S3 (Figure S4 of the revised manuscript).

Figs. S5 and S6: Are these discussed in the text?

Figures S5 and S6 (Figures S6 and S7 of the revised manuscript), which provide diurnal and seasonal insights into boundary layer height variations, have now been explicitly discussed in the manuscript. These figures are referred to in Sections 3.1 and 3.3 to explain how BLH dynamics influence NO₂ distribution and model performance.

Line 195: Can you explain why the averaging kernel makes a larger difference in winter?

In winter, the boundary layer height is lower, confining NO₂ closer to the surface within the planetary boundary layer (PBL). This trapping effect reduces the sensitivity of OMI, as the satellite struggles to detect NO₂ concentrated at lower altitudes. Consequently, the averaging kernels (AKs) have lower values near the surface, which potentially influence the comparisons between model and satellite data. When the dot product is taken between the AK and the model sub-column profile, the lowest sub-column contributions are substantially reduced, affecting the derived tropospheric column amount. Additionally, increased aerosol loads and shallow boundary layers prevalent in winter further limit the satellite's ability to observe NO₂ near the surface. These factors collectively enhance the influence of the AK in winter, where corrections for vertical sensitivity are most pronounced. Studies by Boersma et al. (2008, 2016) emphasize the importance of AKs in mitigating discrepancies between model and satellite data under such conditions, while Martin (2008) highlights their role in adjusting for seasonal and vertical variability in NO₂ profiles.

Line 197: The statement “clearly demonstrates the importance of...” may need a caveat as the difference looks quite small in summer.

We agree that the statement could benefit from clarification. While applying the averaging kernels (AKs) significantly reduces the winter bias (from ~80% to ~50%), the difference is indeed smaller in summer, with biases close to zero. We have modified the statement to reflect that the importance of the AKs varies seasonally, with a more pronounced effect in winter than in summer.

Section 3.5: It would be helpful to plot a timeseries or trends for the NO_x emissions for each of the regions.

We have now included time series of NO_x emissions intensity (emissions per unit area) for each region, as suggested (Figure S1).

Section 3.5: How do the trend magnitudes compare in terms of % trends?

We now include plots of % trends (Figures S8 and S9).

Discussion: Could errors in the assumed emission trends also be a cause of model-obs mismatch? This should be discussed.

Errors in the assumed emission trends may well also be part of the mismatch between model results and observations, and we now include this in our discussion. However, the overall upward then downward trend in emissions over China, and upward trend over India are also seen in the satellite data, suggesting the signs of trends in regional emissions are well represented. Modelled trends are larger than seen in the measurements, particularly in China in winter. Sensitivity experiments with reduced emissions trends would be required to quantitatively explore this as a cause of the mismatch. Such experiments would be useful but have not been performed in this study; we consider detailed sensitivity experimental modelling is beyond the scope of this study but would be very interesting for future research.

References

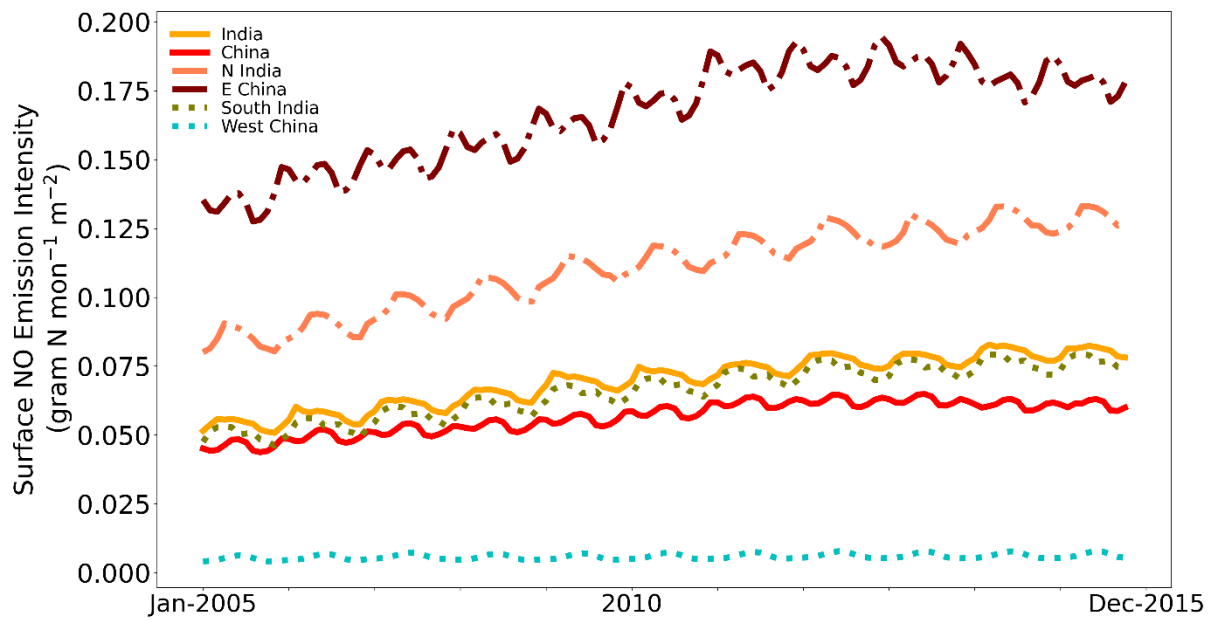
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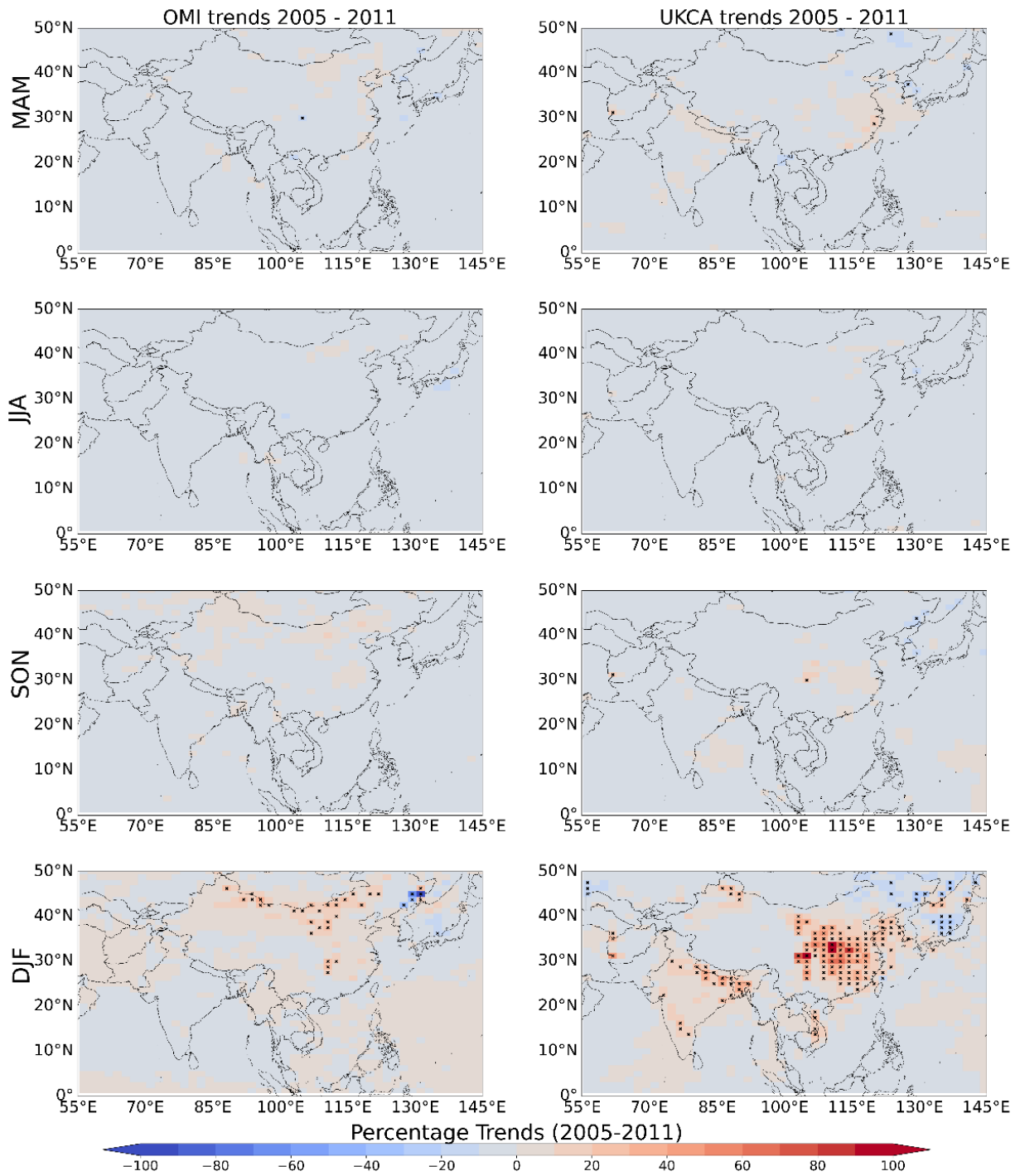
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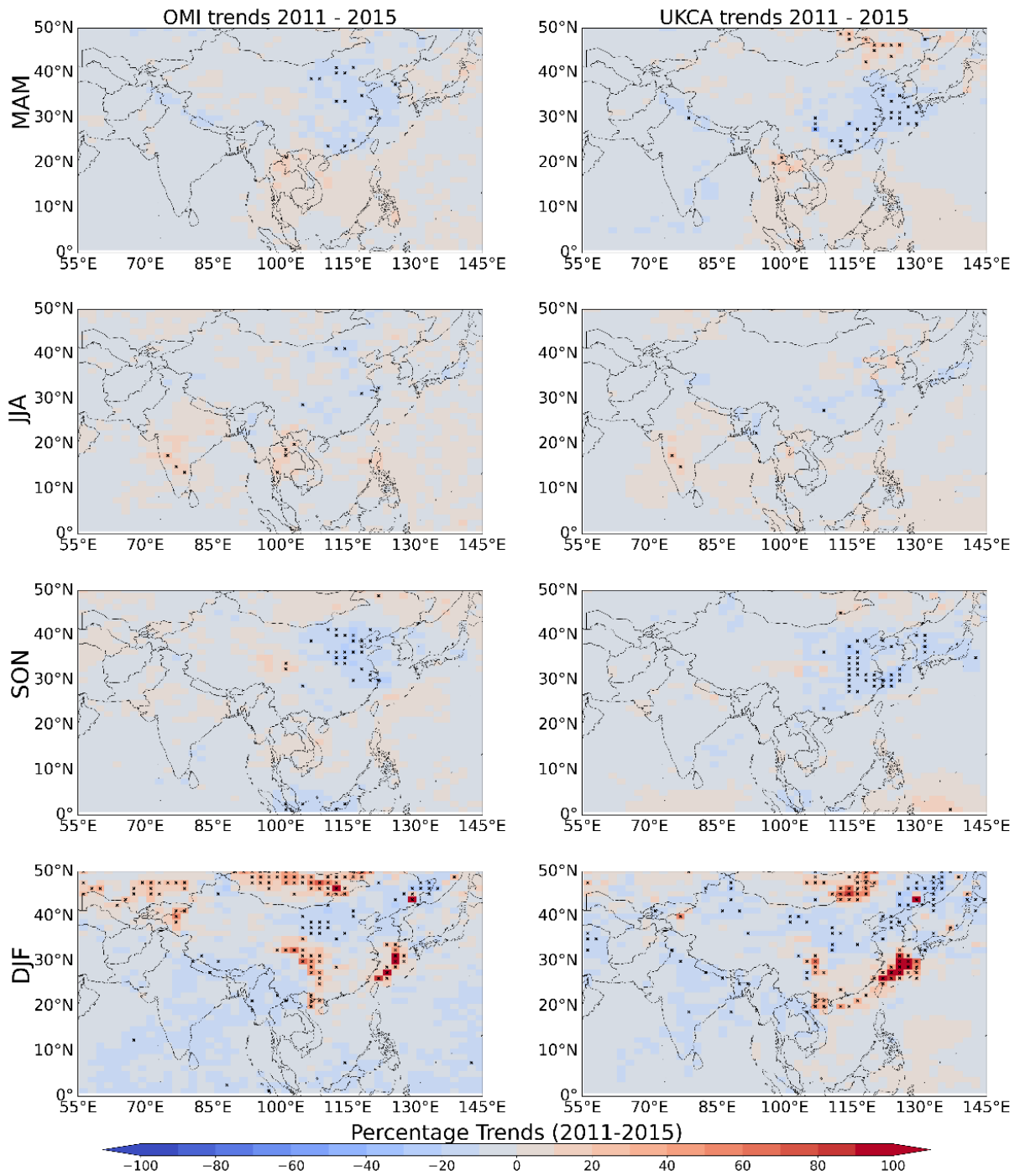
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New Figure S1 Monthly (2005-2015) NO surface emission intensity ($\text{g N month}^{-1} \text{m}^{-2}$) over India and China, along with the polluted (North India and East China) and relatively clean (South India and West China) regions analyzed (see Figure 1c).



New Figure S8 Percent trends of tropospheric column NO₂ from 2005 to 2011 from OMI (left) and UKCA (right) for the four seasons. Absolute trends are shown in Figure 9. Grid boxes with significant trends are indicated by crosses.



New Figure S9 Percent trends of tropospheric column NO₂ from 2011 to 2015 from OMI (left) and UKCA (right) for the four seasons. Absolute trends are shown in Figure 10. Grid boxes with significant trends are indicated by crosses.