

Interactive comment on “Temporal characteristics of atmospheric ammonia and nitrogen dioxide over China based on emission data, satellite observations and atmospheric transport modeling since 1980” by Lei Liu et al.

Anonymous Referee #1

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This manuscript presents an overview of the temporal characteristics of various datasets relevant to NO_x and NH₃ abundance over China. The authors discuss trends in emission inventories (EDGAR and REAS), trends in satellite NO₂ and NH₃ columns (from OMI and IASI respectively), and trends in MOZART-4 model output for the region. Decreasing NO₂ since 2011 suggests that China's 12th Five Year Plan has resulted in successful emission reductions. On the other hand, the lack of a significant trend in NH₃ points to the growing importance of controlling and monitoring reduced nitrogen.

The authors are to be commended for compiling and exploring multiple datasets in

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determining patterns in reactive nitrogen over China. However, I have some general comments about the overall scientific significance and scientific quality. I look forward to hearing from the authors in this discussion phase.

(1) While the analysis of IASI NH₃ columns focusing on China might be somewhat new, I find the analysis of OMI NO₂ that is presented in this manuscript lacking in novelty or insight. In particular, I would refer the authors to de Foy et al. (2016) and to Liu et al. (2016). Both of these studies use OMI NO₂ observations from 2005-2015 to discuss long-term trends and the 2011 peak in NO₂ over China in detail. In my opinion, the observations made by the authors of this present manuscript have not added new insight into this discussion (and in fact treat the analysis with less rigor, as I will discuss below). In its current state, I am concerned that this manuscript does not represent a substantial enough contribution. I encourage the authors to refer to the above references and explicitly address what new insight is gained from their analysis.

(2) The inclusion of model results has added very little insight to the analysis. The MOZART model is driven by the EDGAR emissions to begin with (which are discussed in more detail separately). For both NO₂ and NH₃, I would expect the relationship between emissions and tropospheric columns to be pretty strong, so it's not clear what is expected to be learned by comparing trends in EDGAR emissions with trends in model output based on EDGAR emissions. Moreover, there is no analysis or discussion of the NO₂ model output at all, so why has this output been included in the figures? The authors must expand on or address why the model output has been included, and demonstrate clearly what insight is gained.

(3) The authors conclude the discussion section with implications for estimating long-term reactive nitrogen deposition. The discussion about uncertainty and challenges in estimating dry and wet deposition seems to be out of place in this manuscript. Of course, there is an obvious connection between emissions, atmospheric abundance, and deposition – but this manuscript does not bring up the question of deposition until this final section, so it appears as a digression. While I agree with the conclusion

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made by the authors (that more long-term data sets are needed), I feel their discussion has not presented any new concepts based specifically on the results presented in this manuscript. The connection between their analysis and insight into nitrogen deposition should be made stronger throughout the manuscript. Specifically, what has been gained from the analysis?

Technical Comment:

(1) In determining the trend in NO₂, the authors have calculated a linear fit to the monthly average data. I find this approach to be problematic, since the trend seems to be influenced strongly by an increasing seasonal amplitude. In my opinion, the authors need to remove (or account for) the seasonality before calculating a long-term trend. Specifically, the winter monthly means seem to be driving most of the increase in their linear fit – but these values have the highest uncertainty (borne out by the larger magnitude of the error bars compared to summer months). Accounting for seasonality in determining trends in NO₂ is common practice. This can be accomplished, for example, by fitting the seasonal amplitude separately (e.g. Lamsal et al. (2015)), or by calculating trends in seasonal averages (e.g. Russel et al. (2012)).

Specific comments:

line 88: The authors use of the term "widely" warrants more than two examples in the citation.

line 110: "is believed to have the highest spatial resolution". Surely this statement can be confirmed instead of believed.

line 117: I suggest the authors replace the expression "multivariate", since this term usually implies something different (i.e. modeling). May I suggest the authors use "multiple datasets" throughout the manuscript, instead of "multivariate".

lines 151-153: Repeating the thresholds for error consideration is redundant here.

line 202: Please also include the spatial resolution of the model simulation.

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line 223: "their thread values both positive". Please clarify this sentence.

line 232-233: I think the closer agreement with one other estimate does not necessarily mean the EDGAR estimate is "more reasonable". Please qualify.

line 255 (and elsewhere): The use of the expression "no big changes" does not have much scientific meaning. May I suggest "no significant changes" followed by the results of some statistical test?

line 256: The slope in NH₃ of 0.025×10^{15} is actually twice the slope of NO₂ (0.011×10^{15}), so can the authors clarify why the slope in NH₃ is not determined to be important or large? Should they clarify that they are speaking in relative terms to the atmospheric concentrations? What are the trends in %/year for NO₂ compared to NH₃?

line 305-306: Can the author confirm these numbers are coming from the reference in the preceding sentence (Wang et al. 2012)?

line 311: Can the authors explain why it would be better to calculate trends based on daily data? This would be unusual.

line 350, 351, and 353: Are the authors referring to the panels in Figure 6, not Figure 5?

line 359: "...this is the conclusion we really concerned." Please clarify this sentence.

line 360: "... the following discussion in this paragraph was all hypothetical". Are the authors referring to the next two sentences? This isn't much of a discussion.

line 373: "in high level". I suggest replacing this expression with something more clear.

line 401: "no big variations". Again, I suggest replacing this statement with something more scientifically/statistically clear.

References:

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de Foy et al. (2016), Scientific Reports, <http://dx.doi.org/10.1038/srep35912>

Liu et al. (2016), Environmental Research Letters, <http://dx.doi.org/10.1088/1748-9326/11/11/114002>

Lamsal et al. (2015), Atmospheric Environment, <http://dx.doi.org/10.1016/j.atmosenv.2015.03.055>

Russell et al. (2012), Atmospheric Chemistry and Physics, <http://dx.doi.org/10.5194/acp-12-12197-2012>

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