Reply to Reviewer Comment 2 (Anon. Ref. #1)

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We thank the anonymous reviewer #1 for the valuable suggestions how to improve our manuscript about NO_2 trends over India as observed from satellite and make its focus more clear to the reader.

> There is insufficient detail on the satellite products used. It is not clear whether the SCIAMACHY, GOME-2, and OMI data used here have been validated. Neither it is clear whether the data from the SCIAMACHY and GOME-2 (MetOp-A and MetOP-B) have been intercompared to check that they measure consistent columns over India. The paper should show for instance that GOME-2(A) and GOME-2(B) measure highly similar NO2 columns on the same day over India. Furthermore there is no information given on how the OMI NO2 product was generated, i.e. with a similar algorithm as GOME-2?

SCIAMACHY, OMI, and GOME-2/A have been shown to yield very consistent measurements over emission regions, e.g., by Hilboll et al. (2013).

NO_2 measurements from GOME-2 A and B have been extensively intercompared over several emission regions in Pinardi et al. (2015), showing excellent agreement between the sensors. While that study was performed using the operational EUMETSAT GOME-2 processor GDP4.8, the GDP retrieval algorithm has been modeled after the IUP-UB algorithm used in our study; consequently, the GOME-2 A/B retrievals for the IUP-UB retrieval algorithm can be expected to show the same level of agreement, which could be confirmed by internal tests (not published).

The OMI NO_2 product used in our study was generated with a DOAS retrieval based on a GOME-2 retrieval using a wide spectral window (425–497nm) presented in Richter et al. (2011), which was adapted for the special characteristics of the imaging instrument OMI (Richter et al., 2013).

The revised manuscript will contain a better description of the satellite datasets and references about all these points.

> The direct attribution of trends to socio-economic drivers is questionable. There are many factors influencing the relationship between economic activities, subsequent emissions, and the measured NO2 columns. To name the most important ones: (a) sampling – measurements taken during the monsoon period (cloudy) are not suitable to detect the influence of emissions on NO2 (why not reject the monsoon period from the analysis?), (b) atmospheric chemistry – it is well known that the relationship between NOx emissions and the NO2 column depends on chemical and meteorological circumstances, and there may be differences between years that influence the relationship, especially when NOx emissions are changing, (c) errors in the socio-economic and in the satellite data – if one or both data sets suffer from time-dependent errors, it becomes difficult to argue that similar trends in both data sets allow direct attribution. The authors seem to be aware of at least some of these issues, but do not address any of them other than making some remarks. I think they should make a much more convincing case for taking the satellite and socio-economic data at face value to make us believe there is a strong correlation between the two. In any case a more thorough analysis of sampling issues, intra-instrument consistencies and uncertainties is required, and the impact of variable meteorology and chemistry on the NO2 columns should be addressed with a model or otherwise.

We are well aware of the complexity of the relationship between economic activities, emissions, and measured NO_2 columns and agree with the reviewer's list of "most important" influencing factors.

That being said, this manuscript's main goal is to report on the changes in satellite-observed NO_2 over India, most notably the surprising slow-down of observed NO_2 columns in spite of a growing economy and no sufficiently noteworthy changes in technology. This focus is reflected in the manuscript's title, which does not refer to socio-economic data at all. As part of this report, we find it important to point out the strong *correlation* between this NO_2 increase and various socio-economic factors, which does not necessarily imply *causation*. A robust, quantitative analysis of the potential *causal* relationship between socio-economic factors and observed NO_2 columns is, while admittedly both interesting and important, however far beyond the scope of the present study.

In the revised manuscript, we will make the focus of the study more clear, give care to avoid implying clear causal, quantitative relationships between economy and NO_2, and provide a better description of the potential caveats of direct attribution.

> The claim that the economy may grow without increased NO2 pollution on page 12 is very difficult to follow. The figure 6 shows very similar NO2 levels between 2003 and 2015 over Tamil Nadu, but also that energy production from fossil fuel combustion has increased strongly between 2011 and 2015. I can understand that if fossil electricity generation is driving NO2 pollution, we expect SCIAMACHY and GOME-2 to follow the yellow line from 2003 to 2012. But, elsewhere in the paper, we are led to believe that NO2 increases when coal-burning starts, so why would this then not be the case over Tamil Nadu after 2012? It would help if the NO2 column values were given, and also the NOx emission contributions from the various sources.

We agree with the reviewer that in view of the strong increase on coal-fired electricity production in recent, the lack of NO_2 increases after 2011 is surprising. However, NO_2 pollution by coal-fired power plants is mostly a local phenomenon of which, given the location of the newly constructed power plants close to the coast or state border, only part (i.e., the part being over land and inside state territory) can influence the NO_2 levels which our study attributes to the state of Tamil Nadu. As we lay out in the manuscript, we believe that these changes are not high enough (yet) to significantly influence the state-wide averages. In the revised manuscript, we will make this reasoning more clear to the reader and formulate this fact as the open research question that it is. Also, Fig. 6 will be given a second y-axis giving the NO_2 columns from the GOME-2 instrument.

> P2, line 12: the Burrows et al. 2011-reference is not included in the reference list.

The updated manuscript will contain Burrows et al. (2011) in the reference list.

> P2, L14: the vertical integration limits used in the retrieval should be given, i.e. what defines the tropopause?

The DOAS method measures the *vertically integrated* NO_2 amount in the atmosphere, which is only subsequently corrected for the influence of stratospheric NO_2 (which comes from independent measurements or from an external data source, i.e., model data). The definition of the tropopause becomes important only in this post-processing step. It should be noted that the actual tropopause height is not critical in NO_2 retrievals because of the very low NO_2 concentrations in the altitude region between 8km and 20km.

That being said, methods to define the tropopause vary between data products, from a simple constant or latitude-dependent assumption to accurate calculations from meteorological model data. In our case, we use ECMWF ERA-Interim reanalysis data to calculate the tropopause from the potential vorticity fields; details are given in the referenced Hilboll et al. (2013b) publication.

Since the mentioned sentence (p. 2, l. 14) belongs to the introduction of our manuscript and does not apply to any actual data set in particular, we believe that the integration limits should not be mentioned at that place.

The revised manuscript will however mention our method to define the tropopause in the *Methods* section.

> Page 3, Lines 33-34: please explain why anthropogenic emissions are lowest in August.

In the manuscript, we state that the *measured tropospheric column densities* VCD_{trop} are lowest in August. The seasonality of NO_2 columns over India is mostly driven by meteorology, i.e., the minimum in August is mostly caused by the monsoon (see ul Haq et al., 2015, and Ghude et al. 2013) and by the dependence of NO_2 life-time on photochemistry (again, leading to lower NO_2 concentrations in summer, when the sun is high). This is especially pronounced in regions of strong (anthropogenic) emissions, as only there a significant amount of NO_2 is released to the atmosphere which can actually be washed out.

> Page 4, an indication on the accuracy and reliability of Indian socio-

> economic data would be welcome.

All Indian socio-economic indicators used in this study have been collected from official government sources. The revised manuscript will contain a note about their reliability.

> P5, section 2.7: there is no discussion on how uncertainty in the monthly mean is taken into account in the trend analysis. This should be done especially in view of the sometimes sparse sampling of SCIAMACHY data (between 0-5 measurements per month).

The uncertainty in the monthly mean is not taken into account in the trend analysis. However, the sparse sampling of SCIAMACHY data can be assumed to not pose any limitation on our trend estimates, as measurements by GOME-2 and OMI, which have different, independent sampling patterns, yield similar results.

Regarding sampling due to cloud cover, Wonsick et al. (2009) could show that the peak amplitude of the diurnal cycle of cloud cover over India is rather low (below 30%), and that between the instruments' measurement times there is no large variation. While this does not rule out the possibility of our trend estimates being influenced by sampling issues caused by the cloud cover, it seems unlikely that this issue would cause any systematic effect on our results.

In the revised manuscript, we will give a short account of the sampling issue.

Also, the revised manuscript will include trend estimates for NO_2 columns retrieved from the OMI instrument as an additional dataset, showing the robustness of the trend estimate results.

> P6, Figure 2: it is not clear if the trends in the NO2 columns in Figure 2 have been obtained for retrievals without clouds. If so, do the bars represent proper 'annual means'? Or rather monsoon-filtered annual means?

Figure 2 shows annual mean NO_2 columns, which are calculated as average of the 12 monthly means of cloud-filtered NO_2 VCD_{trop} over the respective regions. This means that each month contributes equally to the displayed annual mean, i.e., monsoon months are not filtered out.

> P7, Figure 3: please include estimates of the uncertainties of the monthly means in the Figure.

The uncertainties of the monthly means are hard to quantify, as very different factors (spatial sampling, temporal sampling, sampling for meteorological condition due to cloud filter, intra-monthly variability of NO_2 VCDs, ...) contribute and a thorough analysis of their individual importance and interdependence can only be estimated with complex model sensitivity runs. Given the highly uncertain uncertainty estimates, we therefore choose to not give any quantitative estimates.

While the quantification of the uncertainties is a very interesting (and we agree, also important) study, it lies clearly outside the scope of the present article, which wants to report on NO_2 increases over India as observed from space.

That being said, the trend method has proven to be robust against outliers caused by measurement noise (Hilboll 2014).

> P7, L12-13: Figure 3 a really strong seasonal cycle over India with a factor of 2-4 differences between winter and summer NO2 columns. It seems implausible that these differences can be explained from the difference in NOx-lifetime alone. Have the authors checked other reasons for this seasonal variability, e.g. emission variability or the influence of air mass factors on the variability? Are slant column densities normalized with a geometric AMF also varying this strongly between Summer and Winter?

NO_2 columns over India have been reported to show a strong seasonal cycle in previous studies (see, e.g., Ghude et al. 2013). The especially pronounced seasonal cycle is a known feature of the IUP-UB NO_2 product, which is partly caused by the used AMFs which are derived from a monthly climatology of NO_2 vertical profiles derived from the MOZART-2 model. However, it has been shown (see Hilboll 2014) that the seasonal cycle is only being enhanced by these AMFs, as SCDs normalized with a geometric AMF also show a pronounced seasonal cycle.

That being said, one should note that the strong seasonal cycle does not impact significantly on the estimated annual change rates. The amplitude of the seasonality is one of the fit parameters in our trend model and has been shown (Hilboll 2014) to not significantly impact on the resulting NO_2 trends.

> Page 8, Line 3-4: it is unclear why a “reduced growth rate” (of traffic-related NOx emissions) would contribute to NO2 decreases. If emissions are still growing, I’d only expect a decrease in NO2 concentrations if the emissions increase pushes the photochemical regime into the titration phase.

We agree with the reviewer’s remark, and will re-phrase this paragraph in the revised manuscript.

> P8, L11-18: this paragraph on the delayed monsoon and its possible influence is merely speculating. My suggestion would be to analyse whether the decrease in 2014/2015 is due to the later monsoon in a more quantitative way via model simulations or other supporting data.

We thank the reviewer for the honest criticism of our, admittedly speculative, argument. While we believe that this is a very interesting aspect, performing dedicated model simulations for the investigation of this point is however outside the scope of the present article, which wants to mainly report on the increase of NO_2 over India as observed from satellite. In the revised manuscript, we will therefore give less emphasis on this and suggest future studies be performed to investigate this aspect.

> Page 8, line 12: pai?

This refers to the Monsoon reports for 2014 and 2015 by Pai and Bhan; we will fix the citation in the revised manuscript.

> Page 8, Line 24: it is unclear how the relative annual change rate in Figure 4 was calculated.

The trend analysis is already briefly discussed in Section 2.7. In the revised manuscript, we will add a reference to that section to the Figure caption.

> P8, L26-29: please indicate the cities of Ballari etc. on the large map of India. Not all readers will be familiar with the names of cities and

> regions in India.

In the revised manuscript, the locations mentioned in the text will be indicated in a map, where feasible.

> P9, L2: with a sudden increase in 2010, how can you trust the linear regression trend analysis? This should be better explained.

A linear regression trend analysis can only give an average growth rate of the study period. In case of newly constructed emission sources, e.g., steel furnaces or power plants, the resulting slope of the regression line depends just as much on the length of the study period as on the actual increase in NO_2 concentrations.

That being said, since our study uses the same time period for all linear regression trends, the results do allow comparing *average* NO_2 increases between different locations.

> P13, L5-7: this part is rather vague. Please clarify why this needs to be in the paper.

We have removed this passage from the revised manuscript.

> P13, section 3.4 seems like stating the obvious, and rather belongs in an introduction section.

We agree; the revised manuscript will contain the contents of Section 3.4 in the introduction.

References

Burrows, J. P., Platt, U., & Borrell, P. (2011). The Remote Sensing of Tropospheric Composition from Space (1st ed.). Heidelberg: Springer.

Ghude, S. D., Kulkarni, S. H., Jena, C., Pfister, G. G., Beig, G., Fadnavis, S., & van der A, R. J. (2013). Application of satellite observations for identifying regions of dominant sources of nitrogen oxides over the Indian Subcontinent. Journal of Geophysical Research: Atmospheres, 118(2), 1075–1089.

https://doi.org/10.1029/2012JD017811

Hilboll, A., Richter, A., & Burrows, J. P. (2013). Long-term changes of tropospheric NO2 over megacities derived from multiple satellite instruments. Atmospheric Chemistry and Physics, 13(8), 4145–4169. https://doi.org/10.5194/acp-13-4145-2013

Hilboll, A., Richter, A., Rozanov, A., Hodnebrog, Ø., Heckel, A., Solberg, S., ... Burrows, J. P. (2013b). Improvements to the retrieval of tropospheric NO2 from satellite – stratospheric correction using SCIAMACHY limb/nadir matching and comparison to Oslo CTM2 simulations. Atmospheric Measurement Techniques, 6, 565–584. https://doi.org/10.5194/amt-6-565-2013

Hilboll, A. (2014). Tropospheric nitrogen dioxide from satellite measurements: SCIAMACHY limb/nadir matching and multi-instrument trend analysis (PhD thesis). Universität Bremen, Bremen. Retrieved from http://nbn-resolving.de/urn:nbn:de:gbv:46-00103664-15

Pinardi, G., Lambert, J.-C., Yu, H., De Smedt, I., Granville, J., Van Roozendael, M., & Valks, P. (2015). O3M SAF Validation Report (O3M SAF Validation Report No. SAF/O3M/IASB/VR/NO2). Retrieved from http://acsaf.org/docs/vr/Validation_Report_NTO_OTO_DR_NO2_GDP48_Nov_2015.pdf

Richter, A., Begoin, M., Hilboll, A., & Burrows, J. P. (2011). An improved NO2 retrieval for the GOME-2 satellite instrument. Atmospheric Measurement Techniques, 4, 1147–1159. https://doi.org/10.5194/amt-4-1147-2011

ul-Haq, Z., Tariq, S., & Ali, M. (2015). Tropospheric NO2 trends over South Asia during the last decade (2004-2014) using OMI data. Advances in Meteorology. Retrieved from http://www.hindawi.com/journals/amete/aip/959284/

Wonsick, M. M., Pinker, R. T., & Govaerts, Y. (2009). Cloud Variability over the Indian Monsoon Region as Observed from Satellites. Journal of Applied Meteorology and Climatology, 48(9), 1803–1821. https://doi.org/10.1175/2009JAMC2027.1