Responses to Anonymous Referee #1

For clarity we repeat the reviewer comments in normal font, followed by our responses in blue italic font.

In their paper "Recent satellite-based trends of tropospheric nitrogen dioxide over large urban agglomerations worldwide", Schneider et al. report on NO2 trends derived from SCIAMACHY satellite observations over megacities and large agglomerations. They give results for 66 regions, provide a brief statistical analysis of the distribution of trends, investigate the average behaviour of NO2 in all agglomerations combined and also separated by geographical region, briefly investigate the impact of spatial resolution on the derived trends and finally analyse the link between population growth and NO2 change.

NO2 is an interesting proxy for air pollution and one of the atmospheric composition parameters which can be observed well from space. Previous publications have highlighted large changes in tropospheric NO2 amounts over the last 15 years but have not covered as many individual regions as this paper. As I'm sure that this overview (mainly Table 3 of the paper) will be of interest to many readers, and because the study is well written and sound, I can recommend it for publication in ACP after consideration of the points made below.

We thank the reviewer *for the overall positive assessment.*

General Comments:

The amount of new material in this manuscript and the progress beyond the state of art (and beyond the last paper of the same authors) is rather limited, and one would hope that a follow-up publication on a topic so thoroughly investigated in literature as NO2 trends from satellite would contain more original data and conclusions.

The manuscript is intended as a follow-up to the Schneider et al. 2012 JGR paper allowing us to go into more detail about specific trends over a very large number of urban agglomerations worldwide. To our knowledge, our study provides what is currently the most comprehensive analysis of NO2 trends over urban agglomerations worldwide, significantly expanding the number of study sites from that of previous studies (e.g. Schneider et al. 2012 or Hilboll et al., 2013). As such, one of the main outputs of the study is the overview of trends for a large number of urban sites computed from a reliable and homogeneous data source (as mentioned by the reviewer above). In addition to the, at least to our knowledge, most comprehensive global overview of NO2 trends over urban areas to date, the paper further provides the first-ever comparison of NO2 trends with changes in population density.

Nonetheless, following the reviewer's advice we have now significantly expanded several sections of the manuscript to go into more detail on several relevant issues. We have substantially improved the background section by providing a much broader overview of the relevant literature. We also added an additional figure showing an example of the trend fitting results and discuss this additional figure

in detail, as well as the implications of errors in the model fit on the trend assessment. Furthermore, we significantly expanded the section on the relationship between population growth and NO2 trends and in the revised manuscript now provide additional material discussing potential reasons for the observed clustering effects.

The link to the MACC-II project and thus the Special Issue is limited to the source of funding. The paper does not discuss in any way the connection to the COPERNICUS system, presumably because there is none, and I think it should not be part of this Special Issue.

The study was carried out through partial funding provided by the MACC-II project. More specifically, the OBS subproject, as part of which we performed this work, has the goal of evaluating satellite data related to Copernicus. Future Copernicus satellites such as Sentinel-5p, Sentinel-5, and Sentinel-4 will continue the NO_2 record provided by SCIAMACHY and other satellite instruments. Thus, the study of NO2 trends from predecessor satellite instruments is very relevant for users of NO2 data from the future Copernicus satellites, both for extending the NO2 trends and for assessing the quality of NO2 data from these future satellites. Furthermore, there is a strong link between trends in satellite-based NO2 columns and changes in NOx emissions, which in turn have an important effect on the quality of the results provided by the Copernicus Atmospheric Monitoring Service (CAMS). For these reasons, we think it is appropriate for this paper to be included in the MACC-II special issue.

In order to clarify these points and to highlight the relevance of our study for Copernicus we have revised this section and further added additional material in the introduction.

Throughout the paper and the figure captions, the authors use the term "concentrations" where they really mean "tropospheric columns". This has to be corrected.

We note that many other studies have used the term "concentrations" in lieu of "columns" when talking about satellite-derived tropospheric NO2 data; examples include van der A et al. (2008), Castellanos and Boersma (2012), and Curier et al. (2014). However, we do agree with the referee that "tropospheric column" is the more technically correct term and we modified the manuscript accordingly.

Figure 1 is nice but has two problems that the authors should fix:

1. There is a clear stripy pattern visible which is not present in other SCIAMACHY NO2 figures published in the literature.

2. There is clear indication for a shipping signal between Central America and Europe. To my knowledge, this is an artefact which should not be present or at least should be mentioned in the figure caption.

1) The origin of the stripy pattern is not entirely clear, but likely related to an east-west difference across the swath in combination with the repeating orbit cycle. Earlier studies (e.g., van der A et al. 2008) have noticed this artefact; however, this effect was not as obvious in, e.g., van der A et al., due to a different color scale used in their figure. Since the stripy pattern occurs only over the ocean and since

*the tropospheric columns there are extremely low (< 0.3 * 10^{15} molec cm^2), they occur in regions where NO2 measurements are mostly below the detection limit of the SCIAMACHY instrument. Because of this, we think it is justified to modify the color scale slightly to mask out the stripy pattern. Please note that because none of our urban agglomeration study sites exhibit such low long-term average values or are located anywhere near the stripes which are visible only over the oceans, the analysis carried out in this paper is unaffected by the striping issue.*

*2) This linear feature is a known artefact and not a real shipping signal. It originates from the TM4 Chemical Transport Model used in the TEMIS retrieval process. While it is an artefact in the dataset, it should be noted that the signal is extremely small (approximately 0.32 * 10^{15} molec cm^2 versus 0.24 * 10^{15} molec cm^2 in the surrounding background over the Atlantic Ocean, i.e., an enhancement of roughly 0.06 * 10^{15} molec cm^2). While this issue is known, it has hitherto not been visible in similar figures because of the different color scales used. Again, please note that none of the urban agglomerations studied here are affected by this artefact as they are not located anywhere in their vicinity.*

We very slightly modified the lower part of the color scale of Figure 1 to bring it more in line with what previous studies have shown and mask the striping artefacts which are irrelevant for this study. We further included a comment in the Figure caption noting that the faint linear feature in the North Atlantic is not a real signal but an artefact originating from the particular model-based retrieval methodology used by TEMIS. The updated Figure 1 is shown below.

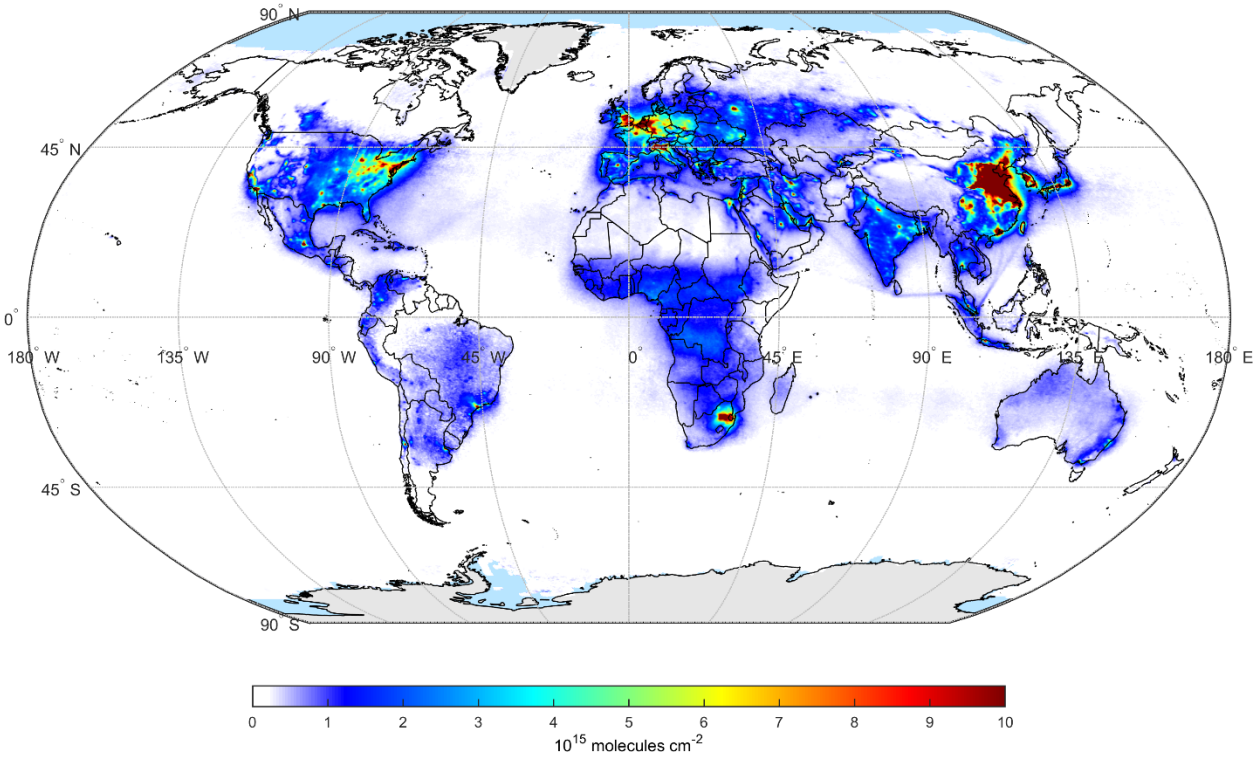


Figure 1 updated based on the reviewer comments. The striping pattern, which was only visible due to the very detailed color scale that we used (in particular for the lower part of the scale), is now gone. The faint linear feature in the North Atlantic is now mentioned in the updated Figure caption.

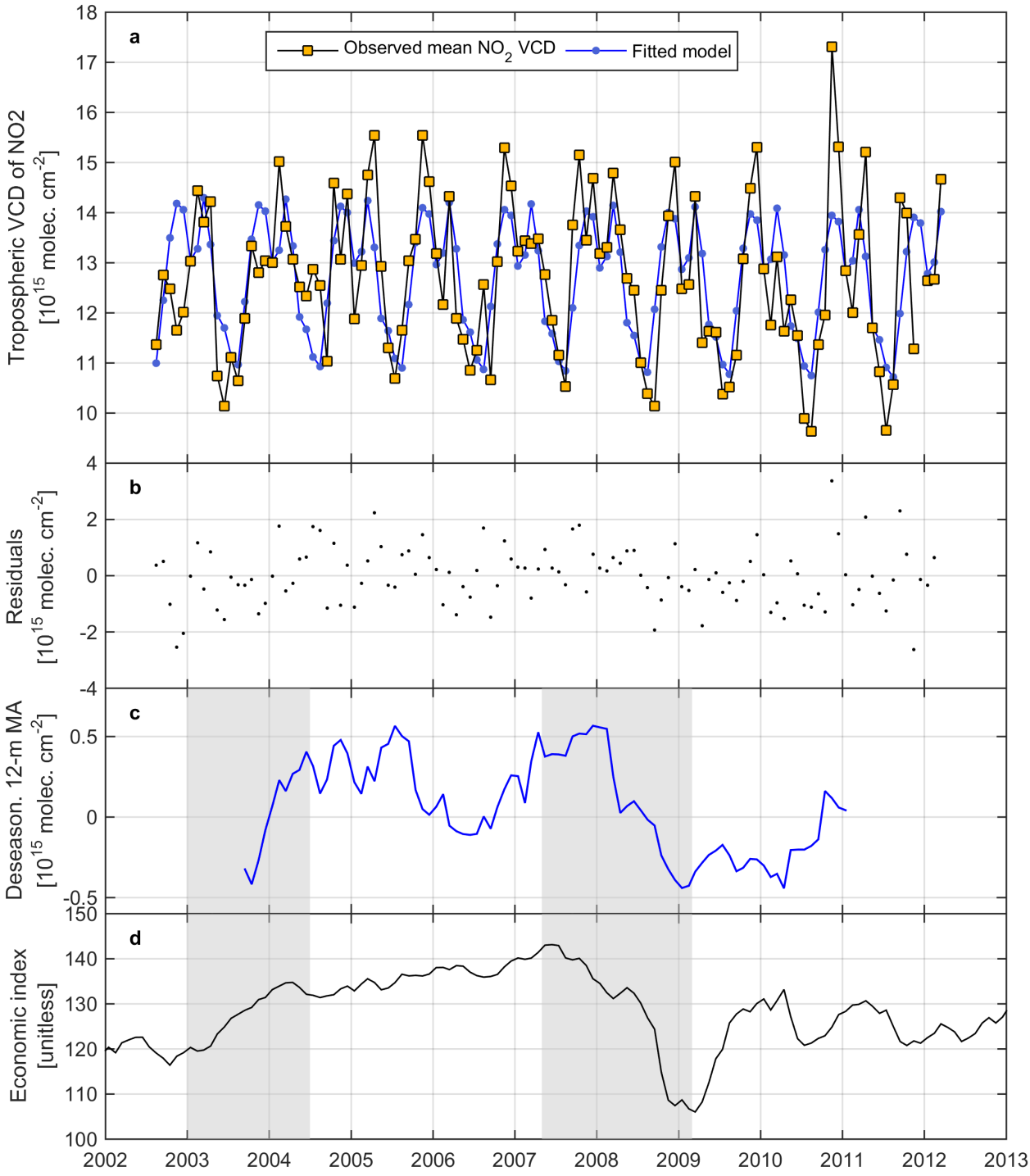
The 2008 drop in Figure 4 is obvious and tempting to link to the economic downturn. However, if we believe this, what is then the explanation for the rapid increase in 2004?

We were at first slightly puzzled about what could be the cause for this increase in 2004. It turns out this effect might also be related to changes in economic activity, which we were able to show by comparing the data to an index of economic health (see updated Figure 5). This leading economic index shows an increase throughout 2003 that precedes the NO2 concentration increase. Similarly, this economic index shows a decrease in mid-2007 preceding the big drop in NOx emissions associated with the global economic crisis.

We took this opportunity to revisit the underlying analysis and improve the Figure (former Figure 4, Figure 5 in the revised manuscript) in several aspects. Firstly, we included a time series of the fitted model together with the global average time series of NO2 VCDs. Secondly, we included an additional panel showing the residuals of the model fit. Thirdly, we included an additional panel showing the temporal behavior of an economic index (the ECRI Weekly leading index). We further added gray boxes identifying to the reader the two periods of interest (2003-2004 and 2007-2008).

We also included an additional description of the Figure and a discussion and interpretation of the results:

"Figure 4c also indicates an increase from late 2003 through early 2004. The reason for this behavior is not as obvious. To better understand this feature, Figure 4d shows a monthly-averaged time series of the Economic Cycle Research Institute (ECRI) leading weekly economic index (https://www.businesscycle.com/ecri-reports-indexes/all-indexes). While this index is primarily intended as an indicator of the economic outlook for the United States (higher values indicate a more positive outlook), we use it here as a proxy for the global economic situation. Both the increase in 2003/2004 and the rapid drop in 2008 are clearly seen in both datasets, suggesting a link between the NO2 levels over large urban agglomerations and the economic situation. It should be noted that other features such as the small temporary decrease in NO2 levels in 2006 (Figure 4c), or the moderate rise of the NO2 levels in 2010, are not reflected as clearly by the economic index. However, the latter could be interpreted as a slightly delayed response, keeping in mind that the NO2 levels shown in Figure 4c are global averages whereas the economic index shown in Figure 4d is for the United States. Overall, these results indicate that the global average NO2 levels over major urban agglomerations reflect large-scale changes in the global economic situation that happen over a relatively short timeframe."



IMPROVED AND EXPANDED FIGURE 4. Average time series of SCIAMACHY-derived tropospheric NO2 column computed over all study sites including a fitted trend model (a), the model residuals (b), the 12-month moving average of the deseasonalized time series (c), and the ECRI Weekly Economic Index. Gray boxes in the two lowermost panels mark periods where the overall NO2 levels in the studied urban agglomerations are indicative of overall economic activity.

As this study uses values from individual grid cells, it would be good to add information how exactly the SCIAMACHY observations were mapped into the grid.

We included a brief description of the gridding process in the revised manuscript (Section 3.1).

Specific Comments:

p24313,l2: "strongly increased emissions" – relative to what?

Revised to "very high emissions"

p24314,l17: ERS-1 => ERS-2

Many thanks for this correction. This has been fixed.

p24315,l2: using method => using a method

Fixed.

p24316,l14: in spite of its name, SCIAMACHY does not have imaging spectrometers so I'd suggest to drop "imaging" here

Thanks for this advice. We dropped the term "imaging" in the revised manuscript.

24318,l11: statistic model => statistical model

Fixed.

p24318,l21: it might be worthwhile to mention here that the seasonality is assumed to be constant over time

Agreed. We added a clarifying statement on this.

p24319,l17: Adding 1015 here doesn't make sense as the units just have to be the same as for the mean columns.

Agreed. This has been taken out and replaced by "given in units of molecules cm^{-2} $year^{-1}$"

p24319,l20: Is VCDtrop deseasonalised?

No, it is the long-term average of the original column values. We added a statement on this in the revised manuscript.

p24321,l12: value => values

This has been corrected.

p24321,l17: "most rapidly increasing trend" – I guess not the trend is increasing but the columns

Correct. We replaced this phrase with "is the site with the largest relative trend" in order to make this less ambiguous.

p24325,l24 "politically motivated emission reductions" => suggest to rephrase this to something like "emission reductions linked to changes in legislation"

We changed this phrase based on the reviewer's suggestion.

p24326,l20: Something is wrong with this list – 45 sites have statistically significant trends, of those 34 increasing, 24 of which are statistically significant?

This statement was unclear – thank you for bringing this to our attention. We changed this to

"Overall, 44 of the 66 study sites showed statistically significant trends (either positive or negative) at the 95% level. In total, 34 sites exhibited increasing levels of tropospheric NO2 throughout the study period, 24 of which were found to be statistically significant. In addition, 32 sites showed decreasing levels of tropospheric NO2 during the study period, of which 20 sites did so at statistically significant magnitudes."

p24327,l6: characteristics patterns => characteristic patterns

Fixed.

Table 2, what is N?

We understand this refers to Table 3, not Table 2 as the latter does not contain an N. As for Table 3, N was not defined in the caption. We therefore added a statement about N in the caption of Table 3.

Figure 2: studies urban => studied urban

Corrected.

Figure 3: trend over => trends over

Corrected.