Author comment on "Long-term changes of tropospheric NO2 over megacities derived from multiple satellite instruments" *by* A. Hilboll et al., doi:10.5194/acpd-12-31767-2012

Response to anonymous referee #1 (C12317)

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We thank the anonymous referee #1 for her/his valuable comments on our manuscript. We believe that they helped us improving the manuscript, making it considerably clearer. Our answers to the referee's questions and suggestions are as follows:

> The abstract promises "several ways", but at the end I found only two.

We view the levelshift method and the multi-instrument fit as two similar but different methods, so the manuscript includes three methdos in total. In the revised manuscript, we rephrased this sentence accordingly.

> "High" spatial resolution is often mentioned, but not defined.

We have addressed this issue by replacing "high" with "individual instrument's" in several places.

> The "physical principles", which sound grandiose, turned out to be just the effect of ground pixel size.

We thank the referee for pointing out that in general, our Section titles were not very accurate. In the revised manuscript, we have therefore re-named Sections 4, 5, and 6. Particularly, the section about the resolution correction factor Γ has been re-named to "Explicitly correcting for the difference in ground pixel size between GOME and SCIAMACHY measurements "

> I don't see how far the fitted trend function is a "statistic" model.

In the revised manuscript, we have rephrased several occurences to simply state 'trend model'.

> Why "levelshift"? Why not simply "offset"?

The referee is right in stating that in fact, the levelshift is an additive offset. However, we chose to follow the nomenclature used by Mieruch et al. (2008), which goes back to Weatherhead et al. (1998).

> 2. After reading the manuscript, it gets clear that the only instrumental difference that was "explicitly accounted for" is the spatial resolution of GOME1. This should be made clear in the abstract, and the ground pixel sizes should be mentioned already in the introduction, as they are crucial for this study.

We have rephrased the manuscript in several places to address the referee's concerns.

> 3. The authors claim that their method leads to lower uncertainties of the derived trends. I do not agree with this conclusion, as the authors just compare their uncertainties (for trends 1996-2011) to those of van der A for 1996-2006. The lower uncertainties may thus just be caused by the longer time period! This has to be revised appropriately (the trends+uncertainties have to be derived for 1996-2006 as well).

We thank the referee for this objection and agree that the reported lower uncertainties might be partly due to the extended time series covered in this study. Therefore, we repeated our levelshift trend calculations for the years 1996–2006. Our findings show that indeed, for the shorter time period, 'our' uncertainties are in the same range and thus comparable to those reported by van der A et al. (2008). Specifically, for 1996–2006, we derive trends of $+8.8 \pm 3.6$, $+0.48 \pm 0.23$, $+2.0 \pm 2.2$, and $+1.9 \pm 0.82$, each in $10^{14}\,\mathrm{molec\,cm^{-2}\,yr^{-1}}$, for Beijing, Mumbai, New York City, and Tehran, respectively. The trend results of all regions and megacities for this shorter study period are shown in a new supplement, accompanying the revised manuscript.

Consequently, we have removed the conclusion about 'our' uncertainties being lower than those of van der A et al. (2008) from the revised manuscript, stating that the two studies' uncertainties are

comparable, and adding a note about the importance of the time window used for the trend retrieval.

> 31768 5: and other instrumental offsets (GOME2A probably differs from GOME2B).
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> 31768 5-7: ”All these factors” have to ”be taken into account”, but in this study, only the spatial resolution is investigated!
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> 31768 9: ”Several”? ”explicitly account for the instrumental differences”? Please be more specific and honest to what has actually been done.
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> 31768 11: Which instruments are ”their”, and what is ”high”?
>
> 31768 12: I understand it now, but it’s very difficult to understand this before reading the paper. Please explain in an extra sentence.
>
> 31768 17: ”All”?
>
> 31768 28: ”+5-10” I suggest to write ”+5 to 10”.

We have rephrased these passages in the revised manuscript and believe that the abstract is now considerably clearer.

> 31769 3: show

As our manuscript does not quantitatively assess nonlinear changes, we have removed this sentence from the abstract.

> 31770: The key is the spatial resolution, so add it here.

We thank the referee to remind us that it is important to give the instruments’ spatial resolutions already in the introduction. In the revised manuscript, we followed her/his suggestion.

> 31770 16: New paragraph for DOAS.

We have changed this in the revised manuscript.

> 31771: Please add Hayn et al.: http://www.atmos-chem-phys.net/9/6459/2009/acp-9- 6459-2009.html

We thank the referee for pointing us towards the Hayn et al. paper, which we didn't include in the original manuscript. In the reivsed manuscript, we have added an according reference in the introductory section on previous work and also in the discussion of the impact of new emission sources.

> 31773 7-9: The OMI instrument deserves an own paragraph as well. Local time and reference(s) have to be added. OMI ground pixel size is 13x24 km2 in nadir, but, in constrast to GOME&SCIAMACHY, it is larger for higher viewing angles, and can be even larger than SCIAMACHY at the edges. This should also be indicated in table 1.

The referee is right in pointing out that the OMI instrument is different enough from the other three instruments to warrant a more detailed description. Therefore, we have added a note about local time and spatial resolution at high viewing angles to the description of OMI in the revised manuscript. Since now we explicitly state this issue in the main text, we believe it to be sufficient to say 'pixel size at nadir' in the column header of Table 1.

References for all four instruments are already given in the introduction, so we don't feel the need to repeat them here.

> 31774: The description of the stratospheric correction should be shortened as it is already given in literature.

To avoid unnecessary lenghts, we have shortened the description of the stratospheric correction algorithm to a mere reference to Hilboll et al. (2012) in the revised manuscript.

> 31776 3-5: I don't get this point - if there is some systematic effect, it should affect all the years from 2003 on, not only 2003!?

We thank the referee for pointing out that our argument was not very clear. Yes, any systematic effect will affect all years from 2003 onwards. However, in later years it will not be possible to directly see this effect, because the GOME value for that year is not available. Therefore, the 'naked eye' is only able to see this systematic effect in 2003, while for later years it is impossible to distinguish between temporal changes and this systematic effect. In the revised manuscript, we have rephrased this sentence to better explain this point.

> 31777 20 - 31778 5: A similar approach was introduced in Beirle et al., 2004, which should be referenced here.

In the revised manuscript, we have added an additional reference to Beirle et al. (2004) at this point.

> 31778 10: 5 SCIAMACHY pixels are still considerably smaller than 1 GOME pixel, which is mentioned later in the manuscript, but should be discussed here as well.

We believe that in the description of the method, it is sufficient to place the two pixel sizes so close to each other that the reader immediately notices their difference. Furthermore, this would lead to repetitions in the manuscript, since it is necessary to discuss this issue later in the "discussion" section.

> 31782: Concerning the problems over desert, I would suspect that the clouds may cause some problems. The determination of cloud fractions at the coastline of deserts is very challenging, as the ground pixels cover both bright sand and dark ocean. This can easily cause systematic differences between GOME and SCIAMACHY. The authors should compare the respective cloud statistics (pdf) for GOME, GOME corrected, and SCIAMACHY.

We agree with the referee that the determination of cloud cover in coastal desert areas is very challenging, and that this can lead to differences between GOME and SCIAMACHY NO_2 columns. However, as described on p. 31782, we observe that NO_2 columns from original SCIAMACHY measurements are

up to 10% higher than from the DOAS fits performed on spatially averaged SCIAMACHY spectra. This bias amounts for basically all the over-correction we observe between SCIAMACHY and resolution-corrected GOME measurements. However, this observation alone (SCIA_orig > SCIA_redres) is totally independent of GOME measurements and thus cannot be influenced by differences in cloud cover between GOME and SCIAMACHY. Furthermore, it is important to remember that we perform a cloud screening (with a 20% cloud fraction threshold) and do not explicitly correct measurements for the impact of clouds. Therefore, the worst that a problem in the cloud retrieval could cause is that we discard too many scenes as cloudy even though they are not. Also, a similar difference of up to 10% between original and reduced-resolution SCIAMACHY measurements can be observed over Riyadh, which is far enough away from any coastal areas to not be influenced by this coastline issue.

> 31783 13-14: Why not? I would think that the same method (adding spectra of several ground pixels to fill a GOME pixel) could be applied to OMI and GOME-2. For GOME-2, the situation might be even more comfortable, as its backscan could be used directly.

We thank the referee for pointing this out. We agree that in principle, one could also use the same method on OMI and GOME-2 data. However, this would be of limited use because of gaps in the resulting time series: In the end, one would have GOME data at each of the spatial resolutions of SCIAMACHY, OMI, and GOME2. So one could analyse the time periods 1996-2011 for GOME/SCIAMACHY, as we did in the present study, 1996-2003+2004-2011 for GOME/OMI, and 1996-2003+2007-2011 for GOME/GOME-2. We do not believe that this would be very helpful in the analysis of the long-time changes of tropospheric NO_2. However, in the revised manuscript we have added the thought that in principle, it would be possible to repeat the same approach with OMI and GOME-2 data.

> 31784 19: ”if”

In the revised manuscript, we have written “if and only if” instead of “if”.

> 31785 3: ”being the...”

We have changed this in the revised manuscript.

> 31785: In line 13, the authors state that the levelshift also accounts for differences beyond those caused by spatial resolution. In line 18, this statement seems to be forgotten.

We thank the referee for pointing us to this inconsistency in writing. In the revised manuscript, we have rephrased the sentence in line 18 accordingly.

> 31787: The discussion should include other trend studies for SCIAMACHY (Schneider & van der A) and OMI (Russell et al.).

We agree that the discussion should make more references to previous work. In the revised manuscript, we have therefore amended it with references to Vrekoussis et al. (2013), Russell et al. (2012), and Schneider and van der A (2012).

> 31788 12: See Russell et al.

In the revised manuscript, we have included a reference to Russell et al. (2012) at this point.

> 31791 3: How far is the levelshift really different from the multi-instrument fit? Couldn't it just be regarded as a multi-instrument fit with n=2?

It is true that on first sight, the levelshift seems to be a multi-instrument fit with n=2. However, the key difference between the two methods is that the multi-instrument fit allows for multiple observations at each timestep. Even though we have constructed the multi-instrument fit to be comparable (by assuring that each point in time has the same weight in the trend calculation, regardless of the number of instruments having observations at that time), this is statistically a fundamental difference. One of the consequences of this is that the estimation of trend uncertainties has to be calculated differenty (using bootstrap resampling).

> 31791 13: "... afternoon, the diurnal cycles of NO_2 and aerosols, as well as the spectral surface reflectance, can lead to ..."

In the revised manuscript, we have rephrased this sentence accordingly.

> 31793: Is this method just proposed or also applied to the time series? Please add the results to tables 4.

We also applied the method from Eq. 9 to the time series. For the revised manuscript, we have chosen to include the results in a supplement, which also contains the levelshift results for the 1996-2006 time period.

> 31794 10: ”instrumental differences”: only one, i.e. ground pixel size.

In the revised manuscript, we have rephrased this sentence accordingly.

> 31794 18: ”spatial dimension”: What should this mean?

In the revised manuscript, we have rephrased this sentence to be clearer.

> 31795 6: Has to be demonstrated.

We have investigated the uncertainty of the multi-instrument fit (Eq. 8) using the bootstrap method and included the results in the revised manuscript. These uncertainties are comparable to those of the levelshift method (Eq. 3) for the 1996–2011 time period. However, we found that the reduction in uncertainty compared to previous study seems to be mostly due to the longer time period covered in this study. We have therefore modified this paragraph of the conclusions in the revised manuscript to reflect this.

> Figure 1: It is helpful to have such a schematic figure, but it should be scaled correctly (320 vs. 60). Please add pixel widths of SCIAMACHY and GOME. I would skip ”pixel value” in the legend.

For the revised manuscript, we have followed the referee's suggestion.

> Figure 2 is discussed in section 4.3, so it should be moved after Fig. 4. Fig. 1 is sufficient for the introduction.

In the revised manuscript, we have followed the referee's suggestion.

> Fig. 6: The authors state that Gamma is noisy over ocean, but it is not pure noise, there are clear systematic patterns. How can they be caused?

We agree with the referee that Gamma is not just pure noise globally over the oceans. Mostly, the observed patterns are patches of varying variability. Apart from this, we suspect the 'non-noisy-looking' oceanic parts to result from sampling artefacts caused by the low coverage of the SCIAMACHY instrument, which in case of clouds (common over the oceans) can lead to very few measurements available per month.

> Fig. 12: The comparison is not that meaningful, as Gamma is a relative quantity, while levelshift is an absolute column density. Thus, for comparing the patterns, either multiply Gamma with the mean NO2 column, or divide the levelshift by it.

The referee raises a very good point. To make Γ and δ directly comparable, we have modified Fig. 12a in the revised manuscript to show the impact of the resolution correction factor Γ, defined as $\mathrm{VCD}^{\mathrm{GOME}}_{\mathrm{corr.}} - \mathrm{VCD}^{\mathrm{GOME}}_{\mathrm{orig.}}$ for the year 2002, instead of the raw Γ. With both subplots showing the same units at the same color scale, the excellent agreement between the two methods becomes immediately visible.

> Fig. 15: Please add the data from Fig. 14 for the same clipping and also without significance masking.

In the revised manuscript, we have followed the referee's suggestion and added a third subplot to Fig. 15, showing the results from Fig. 14 without significance masking, and in the same clipping as the other two subplots of Fig. 15.

References

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