

We thank both referees for their constructive comments and suggestions and have addressed them in a revised version of the manuscript. Our responses to the reviewer's comments and detailed changes made to the manuscript follow. Additional minor editorial changes have been included in our revision.

### **Response to Anonymous Referee #1**

Main comments:

This is a very interesting study showing how OMI observations of tropospheric NO<sub>2</sub> columns provide clear indications for trends in NO<sub>x</sub> air pollution over the U.S. between 2005 and 2011. The strong and statistically significant negative trends over the urban parts of the U.S. stand in clear contrast with smaller negative trends over more remote parts of the U.S. The paper also presents some nice ideas on how to use spatial and temporal patterns in the OMI data to distinguish between different sources and their trends. Although these ideas are innovative and worth pursuing, I find the paper weak in the more detailed messages it attempts to deliver.

For example, in section 4.1 the authors try to tell us that OMI also detects accurate reductions over locations with power plants, because “trends in CEMS emissions show generally good agreement with OMI observations.” (P15428). But they never show a substantial quantitative CEMS result to solidify their claim. I think the authors should show a Figure with both CEMS and OMI NO<sub>2</sub> trends for the Seminole plant, to make the case that both methods indeed see the same stepwise reduction following measures.

*CEMS data from the Seminole power plant has been added to Fig. 2g.*

Also, Table A1 should be extended to also include the CEMS reductions for the power plants. The reader can then judge the quality of the agreement between OMI and CEMS trends. Actually, the paper now only refers to CEMS findings in prose, which is a bit meagre after telling us that CEMS will be used in the paper ‘to evaluate the consistency of trends in emissions’ (P15425).

*In addition to including the trend at the CEMS power plant in Fig. 2g, we have added a figure (Fig. 3) showing the relationship between summertime CEMS emissions and OMI NO<sub>2</sub> observations for 2005-2011 so that the reader may judge the quality of the agreement between the datasets. We believe that this representation is easier to interpret and more informative for our purposes than including the trends in Table A1. We have also added the following text to discuss the relationship between CEMS measurements and OMI observations: “Figure 3 shows the average summertime CEMS emissions versus OMI NO<sub>2</sub> column densities for each power plant analyzed in this study. There is a strong linear relationship between emissions and OMI observations at the more isolated plants in the western US. While there is a similar relationship for the eastern US power plants, OMI measurements show that both a higher and more variable background concentration and closer proximity to urban sources complicates the direct comparison of trends in emissions from eastern power plants as measured by CEMS with columns as measured by OMI.”*

In section 4.2 the focus is on the economic impact on the trends. Here the authors define the recession period as 2007-2009, but the economic recession in the U.S. has been reported to start only in December 2007, in other words, most of 2007 should be characterized as pre-recession.

Unless the authors have something to show for it, the recession period should be chosen as 2008-2009 instead of 2007-2009.

*We chose 2007 for exactly the reason the reviewer describes, it is an appropriate year against which to measure the changes induced by the recession. The year 2007 is included in the trend for the recession years as a reference for comparing years 2008 and 2009 when NO<sub>2</sub> concentrations were impacted by changes in the economy. We are not suggesting that 2007 was significantly impacted by the recession; but are including it to examine the change between 2007 and the recession years following. We added the following to clarify this point: “The years 2007 and 2009 are included in the recession and post-recession time periods, respectively, as references against which to measure changes in subsequent years.”*

In section 4.3 the authors try to distinguish different signals in the OMI observations by using trends in weekdays and weekends over cities for the periods preceding, during, and following the economic recession. Although this is a clever idea, I think the presented results are not convincing. I have three reasons for this:

1. Neither of the weekday or weekend trends before, during, or after the economic recession, is significantly different from another, so it is very difficult to draw any conclusions based on them. *While the differences in trends are small, the trends reported are summaries for all cities in the US and describe the average change among cities and the standard deviation among regions (as described in Section 3). We believe that the consistent relationship between weekday and weekend observations among cities for each of the three time periods studied is very interesting and that agreement with results from mobile source emission studies referenced in this work is an important result.*

2. The argument of the authors is very prozaic and it is not clear how they attribute the stronger weekday reductions in later years to reduced truck activity. A Figure illustrating the line of reasoning is clearly missing. For instance a Figure with time on the x-axis and NO<sub>2</sub> and truck activity for urban areas on the y-axis for the weekday and weekend case would already help. *We’ve added a reference to a recent paper by McDonald et al. (JRG, 2012) that shows the reduction in heavy-duty fuel consumption (and resulting NO<sub>x</sub> emissions) since 2007. We have also added the following text to address this issue: “McDonald et al. (2012) confirmed that heavy duty diesel fuel consumption increased steadily from 1990–2007 but has since decreased, yielding a significant decrease in NO<sub>x</sub> emissions in the United States.”*

3. No reference for the 10-14% reduction in truck activity is given – this is a critical piece of information.

*The reference for the 10-14% reduction in truck activity was added to P15431 L9.*

In the Conclusions different trend numbers are cited than in the main text (-8% instead of -9% per year during recession, and -3% instead of -4% after recession).

*The -9% and -4% refer to trends on weekdays only while the -8% and -3% refer to trends on weekdays and weekends combined.*

Also the claim that ‘we show that : : : a reduction in diesel truck activity has had a larger impact on emission reductions since the start of the recession’ is currently not justified by the results as discussed above. This should be toned down.

*The added reference to the recent work by McDonald et al. (discussed in comment 2 above) clarifies the impact of heavy-duty diesel emissions on the recessionary trends.*

Specific comments:

P15425, L15-18: please provide some more detail and justification about your choice for the optimal radius that captures plumes while at the same time ‘dampens’ the mixing in of background signal.

*Modified Section 3 to read: “Circular regions surrounding each location were chosen with radii large enough to capture NO<sub>2</sub> plumes from each source during summer 2005 (concentrations exceeding  $\sim 4 \times 10^{15}$  molecules cm<sup>-2</sup>), while minimizing a dampening of the signal from averaging in the low background concentrations (black circles in Fig. 1a).”*

P15426, L22-25: it would be appropriate to cite Beirle et al., ACP, 2003 here since they first observed this effect from space.

*The following was added to/modified in Section 4.1: “Additionally, we observe the well-documented weekend effect, characterized by larger NO<sub>x</sub> concentrations on weekdays than weekends. The weekend effect was first observed using space-based observations by Beirle et al. (2003) and in the US, is caused by a larger proportion of emissions from mobile sources that vary according to the weekly cycle in human activity.”*

P15429, L3-7: it is unclear whether the 10-14% reductions in freight transport in the 2007-2009 timeframe hold for California (as the reference seems to suggest) or should be interpreted as a national average number. Also the relation of these sentences immediately to preceding parts on city-trends in the southeastern and southwestern U.S. are not clear to me.

*Clarified that the 10-14% is a nationwide number in the text: “Large reductions observed for 2007–2009 are consistent with heavy duty diesel activity reports showing a reduction of 10–14% in nationwide freight transport”.*

P15429, L11-14: please explain if the 13% reduction in coal-powered energy generation in the US is associated with the economic recession, with the transition to renewables, or with reduced demand as a consequence of relatively mild summers. Section 4.4: if I understand correctly, the authors assume -34% for C<sub>m</sub>, determine C<sub>BEHR</sub> from the local (?) OMI trends, and assume F<sub>m</sub> and F<sub>n</sub> from the NEI database, in order to infer C<sub>n</sub>, the trend in non-mobile NO<sub>x</sub> sources. Please clarify.

*We are unable to determine whether the reduction in coal-powered energy generation in the US results from a transition to renewables or reduced demand and are not aware of any studies that address this. The cause of this reduction is beyond the scope of this work.*

P15432, L22: please clarify what parameter has been correlated with what.

*Modified the text for clarification: Both increasing and decreasing trends observed by OMI are linear for 2005–2011 (not shown), with fits to the trends in summertime observations in regions where absolute changes are greater than  $\pm 5\% \text{ yr}^{-1}$  yielding R<sup>2</sup> values ranging from 0.33 to 0.86.*

Section 4.6: here the work by Lamsal et al., GRL, 2011. should be cited. That study investigated the OH feedback specifically with GEOS-Chem for the world.

*The citation was added to section 4.6 and to the references.*

Figure 3, colour bar: the light blue indicates changes between 0 to -25%. I suggest to split this up in two parts for consistency with the other colours.

*The color bar is divided to show locations where reductions are within a standard deviation of the mean reduction and regions where the change was larger or smaller, as indicated in the caption. We believe that this color bar provides a useful distinction between trends in different locations-so that one can easily identify regions where changes were particularly large or small for 2005-2011.*

## **Response to Anonymous Referee #2**

The paper "Trends in OMI NO<sub>2</sub> observations over the US" by Russell et al. analyses a 7-year dataset of satellite observations of NO<sub>2</sub> with respect to trends and weekly patterns. The authors find different trends before, during, and after the economic recession, and deduce the reduction of mobile and non-mobile sources separately from the weekday vs. weekend trends. The study is appropriate for publication in ACP. The analysis is sound, and the paper is well written. I recommend publication after dealing with the comments below.

- The conclusions drawn are sometimes too optimistic - e.g. concluding that "the economic recession had a significant impact on NO<sub>x</sub> emissions" is a too strong statement given the actual annual changes of -6\_5%yr<sup>-1</sup> before the recession and -8\_5%yr<sup>-1</sup> during the recession. The change of both slopes is far lower than the respective uncertainties.

*The +/- 5% is the standard deviation, describing the variance among locations—not the uncertainty which we believe is minimized by averaging the observations over an entire season (as discussed at the end of Section 3). Reductions during the recession are consistently larger than those before the recession, leading to our conclusion that the economic recession had a significant impact on NO<sub>x</sub> emissions.*

- Given the big advantage of satellite measurements, I don't see why the trends are only shown for selected locations. The authors should provide maps of the relative and absolute trends. These maps might replace Fig. 1, as "NO<sub>2</sub> in the Central US was anomalously high in 2011", i.e. the informative value of Fig. 1 is limited. Also the area affected by the NO<sub>2</sub> increase, and its absolute values, would be more informative than the current Fig. 7.

*We believe that the rural backgrounds measured with satellites are subject to different systematic errors than urban locations. In general, we prefer to avoid conflating the two issues. We have produced the maps suggested by the reviewer, however, trends in rural regions are complex and as our brief discussion highlights, we do not fully understand them. We have chosen to retain Fig. 1 as it highlights the significant changes in NO<sub>2</sub> from 2005 to 2011 in major source regions of the country.*

- The increase in remote regions is interesting, but could not be explained satisfactorily in the paper. Concerning this aspect:

a) Is there any evidence for increasing NO<sub>x</sub> from ground-based monitoring stations?

*Unfortunately, there are not any ground-based monitors in the remote regions for comparison. The following was added to Section 4.5: "Additionally, there are no ground-based monitoring sites in the regions to verify the trends."*

b) The authors skipped the cross-track rows affected by the row anomaly (RA) completely from their analysis (i.e. also for the years before it appeared), to avoid artefacts in the derived trends. However, as far as I am aware, this was not done for the NASA stratospheric estimation algorithm; while for 2005, the complete swath was used to derive the stratospheric pattern, the last years of the considered period are either affected by the RA itself (if included), or by the exclusion of the affected pixels, which are distributed quite asymmetric along the swath. Thus, the RA could definitely result in a trend of the stratospheric correction - probably too small to affect the signal over urban hotspots, but perhaps the explanation for the observed positive trend in background levels. This hypothesis could be tested by e.g. comparing monthly mean total vertical columns in 2005-2006 for the complete swath vs. the reduced swath used for the current analysis.

*We have completed the analysis suggested by the reviewer, analyzing monthly averages for 2005 in remote regions, by both including all cross-track scenes and by excluding cross-track rows affected by the row anomaly. We find no consistent bias between the two methods. On average, we find that excluding the rows affected by the anomaly results in a decrease in the average column over remote regions of 1.33%. Additionally, we find that excluding the data affected by the row-anomaly results in a decrease in the observed column in 57% of the observations but an increase in the observed columns in the remaining 43% of the observations.*

c) The positive trend should be presented and discussed in absolute numbers rather than in relative changes, given the very low tropospheric column densities over the remote areas.

*Absolute changes in tropospheric column density have been added to the text.*

- Some figures have to be revised. In Fig. 1, the unit is missing. Figures 2 and 4 are very small, labels are hardly readable.

*Units were added to the Fig.1 caption. Labels on Figs. 2&4 were made larger.*