Response to the Referee 2 report of the manuscript "Global satellite analysis of the relation between aerosols and short-lived trace gases", by J. P. Veefkind, K. F. Boersma, J. Wang, T. Kurosu, N. Krotkov, K. Chance and P. F. Levelt

In this document we give our responses to the referee report. We have copied the text of the referee report and address all of the comments and suggestions. The referee report text are in *italics*.

General:
I have mixed feelings about this manuscript. On the one hand I like the authors' approach to explore what can be learned from combining AOD and NO2/SO2/HCHO retrievals something which as far as I know, has not yet been attempted.

Response:
We thank the reviewer for the thorough review. This manuscript explores combined satellite retrievals of AOT and short lived trace gases (NO2, SO2 and formaldehyde). This is a new approach (or first attempt as the reviewer pointed out) that has strong potentials. We have chosen in this first paper to focus on the global and regional scales, rather than zooming into specific regions in detail. While this approach gives a top-down view and provides some new and interesting results, it is obvious that we cannot cover all details in this manuscript. Because of the top-down nature of this work, some aspects may be unanswered and we believe that this may have caused the mixed feelings of reviewer 2. However, we see this paper as starting point for future research that uses the combination of satellite data sets to focus on details of the processes in a limited region and we hope that we can inspire other researchers to do this as well.

On the other hand it is my feeling that the analysis does not reveal much more than what was already known. Claims that AOT to NO2 ratios can be used to estimate local efficiency of combustion are to my opinion not justified. Although combustion efficiency plays some role in determining such ratios, they are more dependent on the specific emission characteristics of the regions (traffic-power generation-industry-biofuel) and the specific abatement technologies installed for primary particles and SO2.
It further seems to me that many findings have been found by either individual sensors, surface obervations, and models, and the manuscript could be stronger pointing to what unique information is now derived by combining AOT and NO2 retrievals. In line 23 of the abstract a statement is made that the GEOS-CHEM model calculates grosly similar ratio ,'providing some confidence that we understand source, sinks etc.' This statement is of course somewhat peculiar, because there is already a host of evidence from different analysis methods for GEOS-CHEM as well as other models. If one turns it around one could also say that the consistency of model and satellite ratios provides some evidence that the satellite ratios are correct. I think that the paper would therefore need a better analysis of the potential erros involved with these ratios, before making strong claims (see below). This analysis is imperative to prove that the measured ratios are more realistic than the model results.

Response:
We think that our work shows some new and interesting results. First of all we show that spatial correlation between AOT and NO2 can be used to distinguish differences in the aerosol composition and underlying emission controls (comparison east vs. west Europe, comparisons industrial regions in Europe, US, China and biomass burning regions). To our knowledge, this is the first time that such a distinction can be made based on global satellite observations alone. Comparisons with a global model show that the large-scale features can be reproduced, however there are significant differences in several regions. In many of the industrial and biomass burning regions of the world, satellite instruments observe significant spatial correlation between the AOT and NO2, formaldehyde or both. We show that in China AOT is spatially correlated with NO2, SO2 and formaldehyde, although the spatial patterns differ and the strongest correlation is with formaldehyde. Finally we show how the combination of satellite data of AOT, NO2 and formaldehyde reveals the importance of biogenic emissions for AOT in the summer in the southeastern United States, which the GEOS-Chem model can not reproduce likely due to the too strong wet deposition in the model (as we discussed with the references in the manuscript).

We agree with the referee that the term combustion efficiency is not covering what we intended to say. Combustion efficiency refers to the combustion process itself, whereas the AOT to NO2 ratio is an indicator of the extent to which emission controls that are in place in a particular region. We therefore replaced the term "combustion efficiency" with "regional pollution control indicator" throughout the manuscript.

The statement in the abstract and in the conclusion have been changed to :
"Simulations with a global chemistry transport model (GEOS-Chem) capture most of these variations, although on regional scales significant differences are found."

Regarding the errors in the AOT to NO2 ratio we have followed the suggestion to add information on the estimated accuracy to section 2 (see below). We estimate the error in the ratio in regions dominated by anthropogenic sources to be 20-35%, which is much smaller than the observed global variations.

Finally, not explored in this study, but potentially more relevant is a stronger link to improving emission inventories. If one would 'zoom-in' into specific regions dominated by e.g. industrial emissions, an analysis of these ratios could perhaps lead to an increased knowledge of the effectiviness of e.g. particles abatement measures. I would recommend an stronger connection to emission inventories and to emission modellers.

Response:
We agree with the referee that this is a very interesting direction for research with combined satellite data sets. In the similar direction, we like to emphasize that the GEOS-chem results showed in this study is the result of a standard model run without tuning any emissions. Hence, the analysis presented in our paper, does show that current models are capable of not only capturing the large-scale pattern of aerosols or single aerosol species, but also capturing the large pattern of the AOT-NO2 ratios. Such agreement has important implications for emissions. For example, in biomass burning region, the agreement in ratios can be a good indicator that the emission factors for gases and particles are internally consistent. On the other hand, when the simulated ratios show good agreement with observations, but the AOT and gas amount in the model are both lower than in the observations; this indicates that our total fire emissions are too low, but the emission factors for the relevant species are appropriate. Hence, looking at the comparison of ratios can help us further identify the potential for improving emissions. In line with these ideas, we see this manuscript as a starting point, and will explore some of the possibilities on global and regional scales.

I have my doubts whether this should be published in it's present form, but I do suggest the authors to follow some of my suggestions to improve the significance of the manuscript. Possibly use of CO data could also improve the statements on combustion efficiency and biogenic emissions.

Response:
We agree with reviewer 2 that the term 'combustion efficiency' is inappropriate. We have replaced the term "combustion efficiency" with "regional pollution control indicator", as discussed above. We did not intend to claim that the AOT to NO2 ratio provides information on the local combustion efficiency. The local combustion efficiency depends strongly on the local ensemble of different emission categories and the extent of control for these sources. Nevertheless, we still believe that the ratio of AOT to NO2 provides an interesting characterization, at least on a regional scale, of large-scale combustion practices and can be used to discriminate between combustion practices in various regions of the world. We have not repeated our analysis with satellite measurements of CO, because most CO retrievals are especially sensitive to CO at higher altitudes, away from the source regions. In this work we focus mainly on pollution source regions, and sensitivity down to the ground is imperative. Even so, it is a good suggestion to include CO in future analysis of aerosols and trace gases, because it originates from the same combustion sources, has a longer lifetime than NO2, and therefore is a good tracer for long range transport of pollution plumes.

Detailed comments
p. 18919 Title: "global satellite analysis of the relation between aerosols and ..." would be more appropriate. While some use is made of SO2, and HCHO, the focus is on NO2.

Response:
We changed the title according to the suggestion.

p. 18920 l. 8 'this suggests'; I think nobody is doubting that aerosol and gases have common sources.

Response:
Aerosols and the short lived trace gases studied have partly common sources. For certain aerosol types, like sea salt and desert dust, this is not the case. We have changed the text to:
"This *shows* that *these* enhanced aerosol and trace gas concentrations originate ..."

C7382p. 18920 l. 13: I disagree that you can derive combustion efficiency from the ratio's. This would probably be true only if no abatement technology would applied. I would actually be nice to demonstrate how these ratios could change for road transport, power generation, etc, without and with various degrees of abatement tecnologies.

Response:
As mentioned above, we agree with the referee on this point and have replaced the term "combustion efficiency" with "regional pollution control index" throughout the manuscript.

p. 18926 l. 14 Describe here the uncertainty relate to using the ratios, what is the dependency on surface albedo?
We followed the suggestion of the reviewer and added the following text to this section:
"From the monthly averaged data the AOT to NO2 ratio is calculated. As discussed in section 3.3, this ratio can be used as regional indicator for pollution control measures. Given the above mentioned accuracies for the AOT and NO2 products and the fact that the errors in these products are not correlated, the accuracy of the AOT to NO2 ratio for regions where the concentrations are dominated by anthropogenic sources is estimated to be 20-35%."

p. 18928 l. 27 The choice of the 'Mediterranean' is rather strange since the focus of the paper is to say something about emission signatures and aerosol is mostly dust.

Response:
The Mediterranean is used as a kind of control region. As expected, no spatial correlation is found in this region between AOT and NO2 because the aerosol is mostly dust.

p. 18928 l.3 I guess these are annual avarages?

Response:
Correct. We changed the text accordingly.

p. 18927 l.23 For emission inventories it is difficult to give uncertainty ranges. To what extend can this product verify or falsify emissions from e.g. EDGAR; or put an 'measurement' based uncertainty range?

We think that this is possible but regional modeling has to be performed for this application. While it is beyond the scope of this work, we are planning to perform such studies.

p. 18929 l. 5 make clear what is different with the previous section (already include Europe). Seasonal and model comparison

Response:
We changed the text accordingly.

p. 18930 l. 25 this is not per se about combustion efficiency but more about abatement technology. This would be a spot to introduce analysis of these ratios.

Unfortunately we didn't understand this comment of the referee. The section that follows on p. 18930 l. 25 covers the analysis of the AOT to NO2 ratio.

Issues with conclusions=>see abstract

Response:
We covered this in our response on the abstract comments above.