

## ***Interactive comment on “Improved satellite retrievals of NO<sub>2</sub> and SO<sub>2</sub> over the Canadian oil sands and comparisons with surface measurements” by C. A. McLinden et al.***

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Response to Reviewer 1:

(Author response in *italics*.)

*The authors thank reviewer 1 for their thoughtful and thorough review. This paper certainly benefited from their efforts.*

General comment:

In the work of McLinden et al. 2012, the authors report an increasing trend in NO<sub>2</sub> of

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10.4 %/year resulting from local increases and enhancements in spatial extent. The present work focuses on calculating accurate AMFs in order to satisfactorily derive realistic NO<sub>2</sub> VCDs for the same area, but then does not address how this improvement impacts the trend that was observed using the currently available products. This is an obvious extension of the work that would not require a large amount of space. Could the authors report on this? Or at least address why they have decided not to report annual trends from this new product?

*As the reviewer points out, an obvious extension of this work is to use this new data product to re-evaluate the trends in NO<sub>2</sub> and update what was reported in McLinden et al. (2012). However, wanting to focus on correcting algorithmic issues and also to demonstrate that the corrections were appropriate, we feel adding in the issue of trends and properly addressing the uncertainties that go along with trends would be too much for this paper. (As is currently stands the paper is nearly 15,000 words, 4 tables and 12 figures!)*

While the constant 2006 emissions that are assumed in the model for this new product may pose a problem (and the authors estimate it could result in AMF errors up to 6%), how does this uncertainty compare with the newly calculated trend?

*The 6% is a sensitivity study and would only be appropriate if the concentrations in the PBL did double over the length of the OMI time series, or between 2006 and 2011, depending on how one viewed it. So the 6% decrease in AMF could also be estimated at about a 1%/year decrease in AMF, which amounts to a 1%/year in VCD. Neglecting this effect (assuming it is appropriate) would mean the trend is underestimated by 1%/year. We have added the following sentence here: " In the evaluation of the trend, an unaccounted 6% decrease in AMF over the OMI mission, say 2005 to 2011 or 6 years, would be equivalent to about a 1%/year decrease in AMF. This would amount to an underestimate of the trend in VCD by about 1%/year."*

Could the significance of the trend be much larger than the contribution of this error?

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The readers have no sense of whether indeed it is important or not in this context. It would be nice to know whether the trend was reproduced (attenuated, accentuated, or not at all?) in the new satellite-derived tropospheric VCDs. My sense is that this manuscript is not too long to exclude such an addition.

*While we have not as yet finalized the updated trend, it will not vary much from the original number and the 1%/year would be a relatively minor correction (or a 1%/year addition to the uncertainty budget, depending on how best to utilize this value).*

Comments on the error budget:

In Section 4.1, the authors state that the “primary goal” of this work was to address systematic errors in the current data products. While the sum total of these systematic errors seems to have been addressed (we see for example the ratio of the new “EC AMF” to DOMINO AMF in Figure 6), the authors do not specifically break down how replacing particular inputs of the current data products contributed to reducing the systematic error. It would have been nice, and in my opinion most useful to future investigators, to report how much of the improvement in the new product could be explained by, for example, a higher resolution absorber profile alone. How much did the other improvements (surface reflectivity, more correct treatment of snow, topography) contribute individually? The results of such an analysis might be different from previous work addressing these issues at other locations, given the unique latitude, land cover changes, snow cover, and emission patterns of this particular region. Considering their stated “primary goal”, can the authors speak to this at all?

*Yes, this is a good point, although only relevant for NO<sub>2</sub> as the original SO<sub>2</sub> AMF was a constant. We performed some additional sensitivity studies to address this issue. The key changes were (i) using profiles from a high resolution model based on recent emissions instead of a coarse resolution model using outdated emissions and (ii) use of a higher resolution, annually varying surface albedo instead of an annually invariant, low resolution albedo. Disentangling the higher model resolution aspect from the updated*

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*emissions was not straightforward and so those two aspects were assessed together.*

*In the interest of space, we felt it reasonable to describe the more important aspects of this study without a detailed analysis or introducing new figures or tables. We added an additional paragraph describing this analysis and a summary of the important numbers.*

Also along the lines of the error budget, I would ask the authors to defend explicitly their choice of ranges in calculating error in the EC AMFs. In Table 3, column 3 the authors state the “parameter uncertainty” in cloud fraction, cloud pressure, albedo, surface pressure, and column ozone, but it is not obvious why these particular ranges were chosen. I would prefer to see the reasoning behind these choices in the manuscript, particularly if there should be references to other work (for example, is there a reference that states the surface albedo derived from MODIS is 0.02?). Without this reasoning, the error budget has little meaning.

*Of the random error terms, the cloud fraction and cloud pressure are taken from the OMI cloud retrieval algorithm paper - the reference is now provided in the table. The column ozone value of 20 DU (about 5%) is from a validation study for the OMI-TOMS data product. Profiles shape was discussed above. Surface pressure uncertainty was derived from the variability from the forecast model over the course of one month, and albedo is based on results from a validation study. Footnotes were added to Table 3 with this addition information and, where necessary, additional references were added. The systematic terms are from sensitivity studies and hence more speculative. The reasoning for these is given in the text and, in any event, are not used incorporated into the total uncertainty as they are more speculative.*

Touching on my question in the paragraph above, is it possible that these errors could represent the difference between the the inputs used in the currently available retrieval products and the inputs used for this new EC AMF? If not, this doesn't seem to address the “primary goal” of the work unless I have misunderstood their meaning.

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*We interpreted this question as - are the differences in inputs between what are used now and what the standard algorithm uses within their uncertainties? In short: perhaps, but the answer is not that straightforward. In general, error in the input information will be some combination of random or systematic. The key improvement in the input information used here - spatial resolution that is better than the OMI measurements - means a sizable component of the systematic error will be reduced. Likewise, accounting for the temporal evolution will help as well. In this sense this does address our primary goal.*

The only choice of "parameter uncertainty" that is explained clearly is the profile shape, where the authors evaluate the uncertainty by recalculating the AMFs using profiles from GEOS-Chem. In this case, my sense is that the error is not properly addressed whatsoever. Profile shape in the GEM-MACH model may or may not be represented realistically in the same way that profile shape in GEOS-Chem may or may not be—so the error could be largely underestimated depending on how similar the modeled processes are.

*This is a fair statement, and both sources of profiles could be completely wrong at times. In that sense this is more of a sensitivity study. However, we have used the GEOS-CHEM profiles for this purpose as they have been used extensively for the calculation of AMFs for several data products and have been compared extensively to aircraft-derived profiles. Thus, we feel, they are reasonable overall, and reference(s) has been added to studies which demonstrate this. While in some ways this is not that satisfying an answer, given all the variables involved, the bottom line is that this is best we can do is to estimate this uncertainty.*

Could the authors take time in the manuscript to elaborate on the model processes that will affect vertical profile in each model. (Would a comparison between models represent a reasonable range that encompasses true profile shapes in order to calculate this uncertainty?)

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*We believe that a discussion on the model processes is beyond the scope of this paper, although we have highlighted references in the paper for the interested reader. Again, we suggest that the GEOS-CHEM model is as good as any in simulated the profile shape, and so differences with it, we believe, represent a reasonable method for quantify the uncertainty.*

Comments on the comparison with surface observations:

The wind-sector averaging prior to calculating a GB observation average is an interesting attempt to get around the problem of pixel-vs-point measurements. Some previous work has calculated a wider temporal average in ground-based observations hoping to represent spatial variability in the temporal. Is the pre- wind-sector averaging approach used here an original approach to this particular problem?

*We are not sure. To our knowledge this is first time it has been used (at least for this application).*

Or are there precedent examples (if so, it would be nice to see references).

*We are not aware of any references.*

Unless I am misunderstanding the authors, the 2-d Gaussian distribution estimate is another interesting approach to what is essentially the same problem (pixel-vs-point). Why have the authors chosen to use both approaches at once instead of comparing the two? There needs to be some further clarification as to how the two approaches address different problems, in order to justify using both.

*The wind-sector averaging was used to better capture the average vmr in the vicinity of the ground station (instead of simply the upwind direction), and in that respect it does not necessarily even involve the satellite resolution issue. The Gaussian was meant to help estimate the effect of the OMI pixel size / relate the area measurement to a point. In other words, each is aimed at correcting a different measurement in order to make their intercomparison "fairer". There are specific circumstances (for example, when the*

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station is right at the edge of an OMI pixel), where it might be better not to perform the wind sector averaging. Likewise, high winds will distort the Gaussian and make it less appropriate to use. The bottom line is that on average we believe this approach is justified.

The error estimate for the molybdenum-converter based ground instruments ("CF") is based on half the difference between the means of the CF calculated by both GEM-MACH and GEOS-Chem (supplementary material). I would ask that the standard deviation of the CF from each model at overpass time also be included in the supplementary material, to give the interested readers an estimate of the possible range this factor takes on at each location (i.e. by how much are the individual observations are being transformed). The difference of the average CF between the two models gives us no indication of the range of CF values applied at each site.

*This has been added, as requested, to the supplemental material.*

Finally, the authors chose to address the potential "clear-sky bias" in the OMI measurements by cloud-screening data from GEM-MACH. Could the authors state more clearly how cloud-screening was applied to model data? How are clouds diagnosed in the meteorology of the model (all we are told is that GEM is the Canadian weather forecast model)? Is this then related directly to a cloud-fraction as would be seen by OMI?

*Clouds in GEM-MACH are obtained from 4 different condensation schemes representing different phenomena. Belair et al. (Mon Weather Rev., v133, p1938, 2005) provides a description and evaluation of them. Essentially the 4 schemes produce the clouds associated with (1) the low-level stratocumulus clouds found ahead of synoptic systems (2) the shallow/intermediate cumulus clouds found in their rear portion; (3) the deep convection clouds; and (4) the non-convective clouds. The last two results from very well-known algorithms: Kain Fritsch and Sundqvist respectively. The first two are from less-known schemes: MoisTKE, and Kuo Transient; and those two are described*

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*in the paper above. We added this reference to the description of the GEM-MACH model (section 2.3)*

*The model quantity used is the total column cloud fraction over all cloud types (this statement was added to the paper). This should be comparable with the effective (geometric) cloud fraction.*

If a model-data screening approach is to be used (instead of sampling the ground data), it would also be good to know how likely it is to reproduce the same "bias" when the model is randomly sampled. Given this discussion, I ask that the authors also state what fraction of data-days in this region was actually removed using radiative cloud fraction of 0.2.

*This is a good question, but unfortunately the WBEA ground stations do not measure clouds or cloud cover (they measure solar radiation, but it is not straightforward to convert this). However, along these same lines, we investigated the distribution of cloud cover in the model and compared it with OMI, which we believe is more informative.*

*The GEM-MACH cloud fraction distribution is reasonably comparable to the OMI (peak at cloud fraction of zero and general decrease thereafter), with the exception that the GEM-MACH has a secondary peak for cloud fraction of 1, which OMI does not. In terms of fraction of days with clear skies, OMI has about 45%, and GEM-MACH about 35%. Increasing the GEM-MACH cloud threshold so that 45% of measurements are considered cloud-free, for consistency with OMI, leads to no appreciable differences in the results.*

The above paragraph has been added to the paper.

Minor/technical comments:

-Abstract: May I suggest the authors state in the abstract the time period for which this study is performed (2005-2011)

*Yes, this was added.*

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-p.21614, line 11: Citation of Nowlan et al. (2011) is missing in the bibliography

*This reference was added.*

-p.21616, line 12: Citation of Kelly et al. (2012) is missing in the bibliography

*This reference was added.*

-Table 1: Shouldn't the NO<sub>2</sub> profile also be considered in this table, where the "node" values are months?

*No, this table refers to the independent variables used to directly compute the air mass factors. So the profiles change monthly (and spatially), but this is accounted already as in the 'altitude' dependence.*

-Figure 1 Caption: Remove "the" in "Map of the Canada showing..."

*Corrected.*

-Figures 3 and/or 5: Just a suggestion, but a representation of the typical OMI pixel size on these figures could make a nice addition to further justify the higher resolution data

*This was done as suggested.*

-Figure 7 Caption: The second last sentence was cut off: "These data have been averaged in the."

*The caption was corrected. This fragment was deleted.*

General comment on the equations and symbols:

I find the authors inconsistent or at least unclear in their representations of "M", "S", and "V". On page 21619, lines 4-5, the symbol "M" is defined generally as the ratio of the SCD (S) to the VCD (V) so that  $M = S/V$  (as repeated in Equation 2). However, for Equation 1, the same symbol "M" is strictly redefined as the 'tropospheric' air mass factor, so that  $V_t = S_t/M$ . If not exactly inconsistent, it is at least confusing and this

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confusion continues throughout because "M" appears in the presence of both "V" and "V<sub>t</sub>" in subsequent equations. While the symbols and their interpretation are mostly obvious to those who work closely with these retrievals, I worry that it can be confusing for readers who are less accustomed to it. I would suggest carefully going through all equations and their descriptions in the text to be more clear when it is possible. For example, in Equation 7 and 8 the symbol "V" is used, but don't the authors really mean "V<sub>t</sub>" as it has been defined?

*Yes, equation 7 has been corrected. We have also gone through the symbols and made sure the use of subscripts was made more consistent. For example,  $\epsilon_b$  was changed to  $\epsilon_{S_b}$  in equation (8).*

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 21609, 2013.

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