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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Improved satellite retrievals of NO_2 and SO_2 over the Canadian oil sands and comparisons with surface measurements" *by* C. A. McLinden et al.

Anonymous Referee #1

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This paper builds upon previous work (McLinden et al. 2012) that demonstrated enhanced NO2 and SO2 over Canadian oil sands mining areas using satellite-derived tropospheric vertical column densities (VCDs) from the available NASA and Dutch retrievals. Here, the authors develop new AMF calculations based on high spatial resolution inputs from a regional air quality model (GEM-MACH), and MODIS observations of surface reflectivity. Using the new "EC-AMFs", tropospheric NO2 and SO2 VCDs differ significantly from either the DOMINO or Standard Product retrievals, with up to a doubling of observed NO2 and SO2 directly above the polluted mining area. This product was validated against ground-based observations by converting the calculated

VCDs to a surface concentration, followed by corrections for high biases in the molybdenum converter-based instruments and smoothing and clear-sky biasing in the OMI observations.

The work focuses on the improvements in retrievals based on higher resolution geophysical and chemical inputs. While in this respect it is similar to some previous work (and the results are therefore not surprising) the novelty of the present study is in the application of these methods to the Canadian oil sands, an area of great global interest where land cover and emissions are said to be changing rapidly, and furthermore in the derivation of new AMFs for SO2 in addition to NO2. This work represents a constructive contribution in the science of air quality over oil sands operations. For these reasons, the work will be of great interest to other investigators and should be published. The data are well presented and support the authors' conclusions, and for the most part the methods are explained satisfactorily. I therefore recommend publication in ACP, however I would like the authors to address the following comments.

General comment:

In the work of McLinden et al. 2012, the authors report an increasing trend in NO2 of 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 NO2 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? 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? Could the significance of the trend be much larger than the contribution of this error? 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 (at-

tenuated, 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.

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?

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. 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.

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. 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?)

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

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? 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.

Minor/technical comments:

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

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

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

-Table 1: Shouldn't the NO2 profile also be considered in this table, where the "node" values are months?

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

-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

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

averaged in the."

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 Vt = St/M. If not exactly inconsistent, it is at least confusing and this confusion continues throughout because "M" appears in the presence of both "V" and "Vt" 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 "Vt" as it has been defined?

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