

Interactive comment on “Improved satellite retrievals of NO₂ and SO₂ over the Canadian oil sands and comparisons with surface measurements” by C. A. McLinden et al.

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Response to Reviewer 2:

(Author responses in *italics*.)

The authors thank reviewer 2 for their thoughtful and thorough review. This paper certainly benefited from their efforts.

1 General comments

My main concern is the choice of journal for publication. In my opinion, the paper is of

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rather technical nature. While many improvements to operational retrieval algorithms are introduced, the paper does not provide new insight about the Earth's atmosphere and its underlying processes. I personally would be happier to see this article published in AMT, but maybe that's a matter of preference.

When the paper was submitted we included this text in our letter: “This paper fits nicely within the scope of ACP in that it focuses on remote sensing of the atmospheric trace gases. While some of the content of this paper is fairly technical, it nonetheless will be of a more general interest as it covers a highly visible topic (the Canadian oil sands) of also has general implications in that current satellite algorithms give results that are biased low. On this basis, we feel it is best suited for ACP as opposed to AMT.”

We defer to the judgment of the editor(s) on this point. If they deem it best suited for AMT then we respect their decision (although we stand by our statement above).

Either way, the following questions should be addressed by the authors before publication:

2 Specific comments

1.) On p. 21620 the authors describe their approach to deal with the temperature dependence of the absorption cross-sections. They state that for both NO₂ and SO₂ they use the exact same correction formula. However, it seems implausible to me that both cross-sections should exhibit exactly the same linear dependence on temperature. Some information on how this correction formula was derived, using which laboratory cross-sections, is needed to justify the chosen

This is simply a coincidence. The value of 0.003 for NO₂ was taken from Boersma et al. (2004) and it is also given in several others papers. The value of SO₂ was initially calculated by us, but later we also found it given in a recent paper, Bucsela et al., 2013. The Boersma reference was cited in the text (line 6, page 21620) and we have added the Bucsela reference here as well.

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2.) In the discussion of the validation of the VECTOR RTM on p. 21627, the authors state that the resulting AMFs differ by less than 3% on average and claim that "this agreement is acceptable given that not all input parameters were identical". Without further information, the reader starts wondering why not all inputs were chosen identically, and if the agreement could actually be worse if all inputs were identical. So either the authors should repeat the validation with identical inputs to both RTMs or, if not possible, they should justify why they think the differing inputs lead only to the observed 3% mismatch and are not causing more trouble.

We are a little confused by this comment, and perhaps the intent of this paragraph was not clear. We were trying to show that the VECTOR radiative transfer model used here can accurately compute air mass factors. To show this we tried to duplicate the AMFs supplied by the OMI data product using, to the fullest extent possible, identical input information. This has been clarified.

Completely identical inputs were not used as some were not available (e.g., individual temperature profiles and air density profiles). We have reworked this paragraph to (i) clarify that not all inputs were available, (ii) identify which ones were and which ones were not. The 3% difference is a result of a comparison of AMFs as calculated. A small difference is expected owing to the differences in the models (solution technique, numerical approximations, etc...) The fact that it is only 3%, and not larger, indicates that neither the model (nor the differences in the remaining inputs) lead to a significant difference. This was clarified also.

3.) In the EC-AMF sensitivity study (on p. 21631), the authors evaluate the influence of doubled PBL concentrations on the AMF and come up with a 6% decrease of the AMF. They claim that further increasing the NO₂ burden of the PBL would not significantly change this due to some saturation effect. However, the authors fail to substantiate this claim by further AMF calculations for, say, a 3- and 4-fold increased NO₂ burden of the PBL. Also, it would be interesting to know by how much emissions have actually increased since 2006.

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There was a brief explanation included in the original draft (page 21632, line 4-5): "AMFs were already sufficiently weighted towards the PBL that adding additional NO₂ there had only a modest impact". We have added an additional statement that we feel clarifies this sufficiently: "In the limiting case that all of the NO₂ resides in the PBL, further increases have no impact at all since only the profile shape is relevant". (The key here is that it is only the profile shape that impacts the AMF.)

4.) On p. 21635, the authors try to convert surface vmr to VCD, using CTM profiles. They assume that "the model can adequately capture the spatial and temporal behaviour of this ratio." However, the largest model uncertainties will be due to uncertainties in the emission inventories, impacting mainly the PBL and lower atmosphere. Therefore, I believe that the authors' assumption is not valid. It might be a better approach (and in line with the sensitivity study of the EC-AMF to PBL concentrations) to take the free troposphere profile from a CTM, only varying the PBL content according to the surface vmr, but maybe the authors can better justify their assumption or find a more elegant solution.

We agree that the uncertainties in emission inventories can be a key factor limiting the accuracy of this method. It is not clear to us exactly what the reviewer is suggesting: "It might be a better approach ... to take the free troposphere profile from a CTM, only varying the PBL content according to the surface vmr". We could use in-situ surface vmr, true, but then we could not be able to compare to it. But without bringing in additional information, the only way of mapping the column to a surface value is using the ratio predicted by the model.

The motivation for using the column-to-surface mapping in the first place was to calculate average surface vmrs in order to demonstrate that the new algorithm largely corrected a significant bias. The random component of this error would be eliminated by averaging over many years. The good overall agreement (in magnitude and spatial distribution) suggests that emissions uncertainties are not a large issue with the possible exception of the slow annual increase, which we attempted to estimate.

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5.) In Figure 3, the authors compare their albedo product at the SO₂ wavelength for 2005 and 2011. There is a large difference visible between the two plots. The authors should try to explain this trend in surface albedo in the discussion.

Figure 3 shows the NO₂ albedo for two years, 2005 and 2011, with the 2011 showing higher values over the surface mining area. A sentence was added explaining this: "Over the mining area the albedo is seen to increase with time which is consistent with the ongoing conversion of the darker forest into the brighter industrial land-use."

3 Technical comments

p. 21614/l. 11: The reference "Nowlan et al. 2011" is not included in the reference list at the end of the article.

Included.

p. 21617/fig. 2: As absorption happens on individual molecules, I would find it more instructive to see the vertical profiles as number density instead of vmr. Also, the shape factor could be calculated using number densities instead of vmr.

True. However, showing these a number density de-emphasizes the differences in the free troposphere which is one of the discussion topics this figure addresses. On this basis we feel it is better to leave them as VMR profiles.

p. 21617/l. 7: At the high latitudes of the study area, the LT of OMI overpass is shortly before 13:00. However, I don't expect this to significantly influence the results.

Agreed. At latitudes below 60 degrees we found this not to be an issue.

p. 21622/l. 12: For the GOME LER climatology, reference to Koelemeijer et al. 2003 should be given.

Yes, this has been added.

p. 21625/l. 29ff: As the authors state, the discussion of snow albedo seems misplaced

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in this study. I would prefer if the discussion would not be part of this manuscript. If the authors need treatment of snow albedo in a later study, they should include the discussion then.

We respect the reviewer's point, but we feel it is a judgment call. In the end it does not really derail the flow of the paper (one figure and one paragraph), and it does fit in with the larger idea in this paper of improving the treatment of surface albedo in UV/vis algorithms. In short, we view it as a valid and worthwhile contribution.

p. 21626/l. 11: Proper reference to the data product (publication, dataset name, data availability) of the used O₃ columns should be given.

Agreed. This oversight has been corrected and a sentence and reference identifying the algorithm that was used (Bhartia and Wellemeyer, 2002) added.

p. 21626/l. 19: A good reference for the aerosol influence on AMFs depending on their relative positioning is Leitão et al. 2010 (doi:10.5194/amt-3-475-2010).

Yes, it is. I (CMcL) appreciate you mentioning this as I was not aware of it.

p. 21626/l. 22: Please give a link/reference to the For McMurray sun photometer measurements.

A reference and a link to the Aeronet data web site was added here.

p. 21630/eq. 8: The numerator in the third summand under the root should be instead of

Corrected.

p. 21635/l. 26: The authors should provide information about the temporal representativeness of the WBEA measurements. Are daily averages taken, or only measurements of the LT of OMI overpass (which can be between 12LT and 14LT, depending on the viewing azimuth angle), or OMI overpass +/- X minutes, or ...

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Yes, this is an oversight. The WBEA measurements are averaged between local times of 1200 and 1400. This information has been added to the end of paragraph 2 at the beginning of section 5.

p. 21636/l. 17: The authors refer to Equation (4), but Eq. 4 is actually about albedo. Either I don't understand why albedo plays a role here, or the reference is wrong.

You are correct. This seemed to have creeping in during typesetting and was not caught. Anyway, it has been corrected.

p. 21649/Table 1: The authors speak of 30 altitude steps from 0 to 16km in 0.5km layers. However, going from 0 to 16 in 0.5 steps yields 33 layers?

The correct number is 32 (there are 32 layers, each 0.5 km thick, and collectively span 0-16km).

p. 21654/Figure 2: In the caption, the authors speak of "number density profiles", while the plot labels speak of "volume mixing ratio". Which is correct?

An artifact of previous version. This has been corrected to vmr.

p. 21655/Figure 3: A fifth panel, showing the data of (b) but integrated to the coarse spatial resolution of (a), would make it easier to compare the authors' albedo calculation to the original Kleipool et al. climatology, as the effect of spatial resolution and different albedo dataset would not be combined into one plot.

This is a good idea. The figure was reworked to include 6 panels, adding the panel suggested by the reviewer and the MODIS SO₂ albedo for 2011. The text was modified accordingly.

p. 21657/Figure 5: The OMI snow albedo in (a) should be shown at the product's original spatial resolution, without interpolation (as in Fig. 3a).

This was changed, as suggested.

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p. 21662/Figure 10: The WBEA stations are not really identifiable in the plots. The dots should be made bigger, and the caption should say that the plot actually shows the WBEA stations. (Or, alternatively, don't show the stations in this Figure, and fix the reference in the text to point to S1 instead).

Agreed. The station locations are hard to make out. They have been made clearer. We prefer to keep them here to highlight that the WBEA NO₂ stations miss the peak in the OMI NO₂ distribution.

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

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