“Development and characterisation of a state-of-the-art GOME-2 formaldehyde air-mass factor algorithm” by Hewson et al. [2014]

Response to Reviewer 2

General Comments:

The subject of the manuscript is suited for AMT. The manuscript is well written, and the work and results are explained clearly. The most interesting results are the model resolution effect illustrated over South America and the full RTM error calculation. The other improvements have minor impacts on the final AMFs. However, some aspects of the baseline UoL AMF algorithm, namely the aerosol correction and to a lesser extent, the full radiative transfer calculation for each observation, are not described in this study, while they are specific to the Leicester algorithm. I think that the authors missed the opportunity to extend their previous studies over South America to the global scale. As I can understand the difficulty to run more sensitivity tests at the global scale, I recommend publication after minor revisions.

We thank the reviewer for their positive and useful comments that have helped to improve the manuscript. We have addressed all their specific issues and have adjusted the manuscript accordingly where necessary.

Specific Comments:

Introduction: Lines 14-22: “There is, therefore, a pressing need to improve AMF calculations and reduce uncertainties wherever possible. Accordingly, this paper details a new algorithm, which attempts to improve the accuracy of HCHO AMFs by performing scene-specific full-radiative transfer calculations and through more advanced treatment of the input a priori information.” I agree with the first sentence. However, the improvement of the AMF accuracy by performing full RTM calculations is not addressed in this paper. In Barkley et al. 2012 and 2013, a LUT was used (as reported in Table 1). So this is an improvement that deserves to be described (it is mentioned in the abstract, the introduction and the conclusion).

We have now added an extra section and supplementary figure discussing the AMF differences due to the use of a LUT versus the full radiative transfer calculation (baseline) approach.

Section 2: I suggest merging section 2 with section 3 and shortening the theoretical explanations, especially as they have already been published before (for example equations in Section 2 can be found in Gonzalez Abad et al., 2015).

We prefer to retain both these sections within the manuscript so a reader can understand the theoretical basis of AMFs and put into context our AMF algorithm against current implementations. Another reviewer has asked for further clarification on the AMF computations, hence we cannot shorten this section. Apologies.

Section 3: Table 1: please add a column with the baseline AMF calculation from this study.

We ask the reviewer to note that our baseline AMF algorithm is presented in Table 3, facilitating an easy comparison to the other AMF algorithms shown in Tables 1 and 2.

Line 19: please note that a GOME-2 albedo dataset is under development.
See http://www.knmi.nl/_tilstra/Reports/GOME2_surface_LER_ATBD_v1.6_20141113.pdf)

Thank you for bringing this to our attention! We have now implemented the new GOME-2 albedo data into our final AMF algorithm.

Lines 26-33: AMF and error calculated using look up tables are also scene-specific, as the surface altitude, albedo, cloud properties, profiles, geometry, etc: : : are taken into account for each individual observation. However, LUTs entail interpolation errors. Please reformulate.

Yes, good point. We have adjusted the text from “Scene specific AMFs are expected to be…” to “AMFs derived from full RTM calculations are expected to be…”, to avoid this misinterpretation.

Section 4 I acknowledge the authors for the details that have been added about the aerosols. It improves the manuscript.

No problem, we appreciate suggestions to improve the paper.

Section 5 Overview: I suggest to follow the same order as in the following sections, and therefore to swap points (3) and (4).

Swapped around.

Section 5.2: Please add a figure showing the HCHO and AOD profiles at the model resolutions of 4°x5° and 0.5°x0.667°, for a particular location and month. The resolution of the CTM has one of the largest impacts on the total AMFs. Therefore, I think it is important to extend this section, showing plots of vertical profiles at different spatial resolutions, and above emission areas.

A figure and supporting text has been added to the manuscript, which show an example profile over the Amazon from 4°x5° and 0.5°x0.667° model simulations. This highlights HCHO and AOD profile differences, both in terms of magnitude and shape.

Figure 3: What is the reason for the AMF enhancement in March, North of the Amazon at the Guyana border? If this is altitude, I wonder why this effect disappears in August.

This is not an altitude effect but rather due to differences in the simulated HCHO distributions (i.e. profiles) owing to the different GEOS-Chem model resolutions (0.5x0.667 vs 4x5). In the Amazon nest-grid simulation, high HCHO columns are simulated in March over a region of predominately high biomass burning emissions (and to a lesser effect enhanced isoprene emissions) near the Guyana border, whereas in the coarse 4x5 simulation this high HCHO feature is 'washed' out over the larger grid cells. The biomass burning over the region is not present in August so the effect vanishes in that month.

Section 5.4: At the global scale, a model resolution of 0.5x0.667 is not realistic. Therefore, I would like to see the improvement brought by the area weighting and surface pressure correction of the profiles, when working with a model at a lower resolution. For example, is it possible to add a line on the histogram of figure 3, corresponding to 2x2.5, pressure corrected?

In Figure 3 (red line) of the manuscript we show the histogram of AMF differences when using the GEOS-Chem 2x2.5 model data with the IJ selection method (i.e. not area-weighted) compared to the default baseline AMF algorithm (which uses data from the 4x5 simulation). As stated in section 5.3 and 5.4 effect of area weighting (AWM) and pressure correcting the model inputs is small when compared to the use of different model resolutions. This is shown for Figure 3 histogram differences when the AMF algorithm uses GEOS-Chem 0.5x0.667 data via the IJ selection method (blue line), area weighting selection method (green line), and area weighted and pressure corrected method (aqua line); essentially the histograms are indistinguishable.

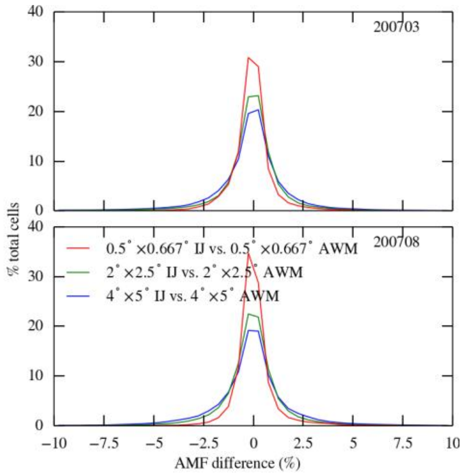


Figure R1: Attached is a plot showing the AMF differences between the IJ and area-weighting (AWM) profile selection methods for each of the different GEOS-CHem model simulations (4x5, 2x2.5 and 0.5x0.667).
That is,
Green Line =
100% x (AWM 2x2.5 – IJ 2x2.5) / IJ 2x2.5

Blue Line =
100% x (AWM 4x5 – IJ 4x5) / IJ 4x5

Red Line =
100% x (AWM 0.5x0.67 – IJ 0.5x0.67) / IJ 0.5x0.67.

The effect is similar when using the different methods on the 2x.2.5 data (as illustrated above by Figure R1), hence over plotting the extra histograms on Figure 3 just makes it lose clarity and potentially more confusing to the reader, hence we prefer to keep Figure 3 as it is now. However, to make clearer to the reader of the overall difference of the IJ versus AWM methods for a given simulation, we now included

Figure R1 in the supplementary material.

Equations of section 5.4 are from Zhou et al. and could be skipped.

We prefer to retain this information within the manuscript so a reader can understand our approach without having to read further material.

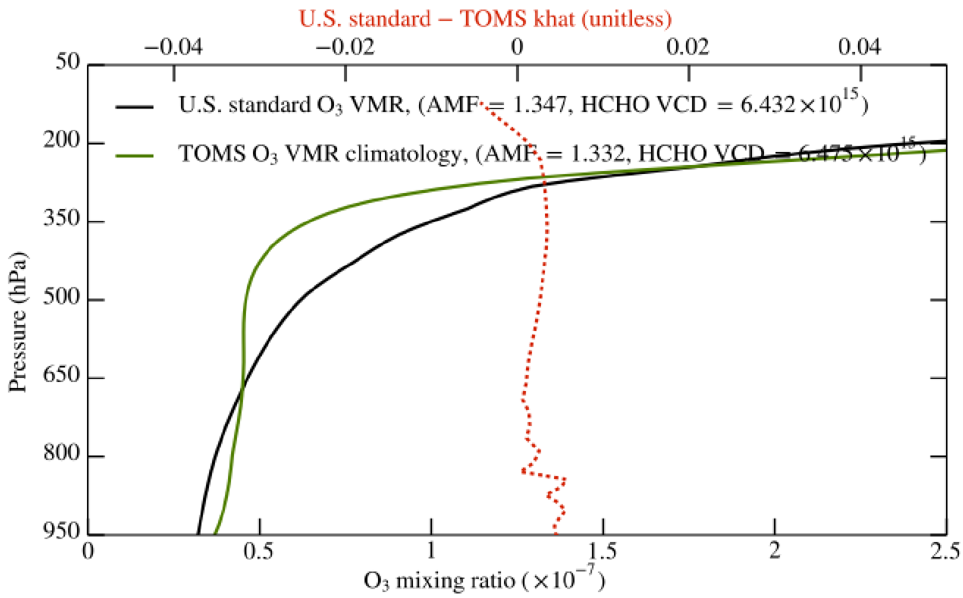
Figure 4: please specify in the caption the model resolution that has been used. If this is 0.5x0.667_, then please add a third line with 4_x5_.

Model resolution added.

Section 5.5, Line 8: Figure S1 Section 5.7, Line 11: Figure S2

Now corrected.

Section 5.7: The use of O3 profiles climatology is rather new for HCHO AMF calculations, and deserves to be published, even if the impact is negligible. Please explain how differences in ozone profile compared to US atmosphere lead to a diminution of the AMFs over regions of moderate to high surface elevation.



The above plot shows O_3 VMRs from the TOMS climatology (green) and from the default U.S. standard atmosphere (black), for a location over the southern Himalayas near the Indian border (76.815, 31.434), 20070321. Surface pressure is taken from the GEOS-Chem 4x5 degree simulation. The red dashed shows the difference in scattering weights from LIDORT (clear-sky); that is baseline algorithm (using US ozone profile) minus the AMF algorithm using the TOMS ozone climatology. Use of the TOMS ozone profiles generally results in slightly lower scattering weights than the US standard atmosphere, especially over regions of higher elevation where the divergence between the profiles is more pronounced (as stated in manuscript). The lower scattering weights result in lower AMFs and hence the observed widespread AMF decrease shown in Figure S2 of the supplementary material.

Section 5.8, lines 7-15: Aerosols and model spatial resolution effect. This is interesting and could be further developed, maybe with a figure showing the AOD profiles at 2 different model resolutions, as suggested for section 5.2.

As mentioned above, a figure that shows the HCHO and AOD profiles (black and organic carbon) has now been included in the manuscript with reference to Section 5.2; this extra figure clearly shows how important model resolution is in simulation of aerosol AOD. Since the impact of model resolution on dust AOD and its effect on the AMFs, occur mainly over the oceans where VOC emissions are small, it is not so critical to the paper that this aspect is explored here.

Section 6 In equation 6.1: all the input parameters used in the AMF calculation should be included in this equation. Where is the term for aerosols?

The default AMF calculation does not include a term for aerosols. Adjustment of the HCHO profile accounts partly for the change in HCHO vertical distribution relative to aerosols. However, we have now added another equation that explains how the uncertainties due to aerosols were determined in section 6.1.

Line 21: I would rather say that the greatest source of AMF uncertainty is associated with the HCHO profile shape and its relative vertical distribution compared to cloud altitude and AOD profile (same comment for the conclusions).

We have refined the main take message of the paper to reflect this distinction (in abstract/conclusions).

Section 6.1, line 26: distributed.

Corrected.