Three-dimensional radiative transfer effects on airborne, satellite and ground-based trace gas remote sensing

Marc Schwaerzel et al.

Response to the Reviewer's Comments

We thank the two reviewers for their positive comments, critical assessment and useful points to improve the quality of our paper. In the following we address their concerns point by point. Changes in the paper are shown in blue. We hope we clarified all concerns and that the revised manuscript has improved.

Reviewer 2

Reviewer Point P 2.1 — Pg3, Ln43: The first review pointed out that it would be difficult to obtain the relevant profile information required to use the 3D scattering weight information. Turning this problem on its head, I think it would be worthwhile commenting on the potential to constrain the horizontal gas distribution if you could scan the instrument azimuthally i.e. is there enough information present to invert concentrations radially in the same manner that different viewing zeniths can be used to partially infer the vertical profile. I think this was alluded to in the conclusions but could be expanded on.

Reply: This is an important point. We added a sentence on this approach in the conclusions:

> On the other hand, measuring different azimuth angles with a MAX-DOAS instrument could be used to constrain the 3D fields of trace gases (e.g. Dimitropoulou et al., 2019).

Reviewer Point P 2.2 — P3, Ln 190:"This hypothesis could be tested by including more streams "I agree with the reasoning, but wouldn't it be easy to rerun the SCIATRAN simulation with a higher stream number just to confirm that it converges towards the monte carlo method so you can make a more definite statement?

Reply: Since we used 1D-layer AMFs from a previous study (Wagner et al., 2007), we have neither the SCIATRAN executable used in that study nor the exact input files to replicate the simulations. We therefore think that the effort required to set up SCIATRAN just for this one test is too large, also because the difference only affects altitudes that are less relevant for our study.

Reviewer Point P 2.3 — Pg 13, L239:The discussion on the line-of-sight sensitivity in this paragraph is useful for gaining some physical intuition for the observations. It would be useful to more systematically explore this as a function of AOD/view geometry, to provide guidelines for situations that permit the interpretation of when the majority of photons are coming to the instrument by single scatter into the path of the detector. It is possibly more relevant to only consider the line-of-sight within the assumed boundary layer, where the largest horizontal variation of NO2 is expected to be.

Reply: This is a valid point and we agree that such information could be useful for further studies. In this paper, we want to show the general importance of 3D in radiative transfer modelling for trace gas retrieval. We feel that quantifying effects of particular conditions (i.e. input parameters) and particular measurement setting on an instrument sensitivity would be complex and out of scope of this study.

In section 4 (Figure 6) we already considered the line-of-sight only within the boundary layer.

Reviewer Point P 2.4 — I think somewhere it would be helpful to mention how long the monte carlo calculation stake perhaps in a until of time/photons to get an idea of how long the scene calculation stake

Reply: We addressed this question in the response to the first reviewer. We added a paragraph in section 5.1.

> The computational cost of calculating 3D-box AMFs is considerably larger than for 1D-layer AMFs. The computational time for calculating 3D-box AMFs for the scenarios here (see Table 1 with SZA=20°, SAA=90°, VAA=90° and VZA=2°) is around 218 seconds with 1 million photons using a single core of our local machine (Intel Xeon W-2175 CPU @ 2.5 GHz). The computational time for the corresponding 1D-layer AMFs is only about 4 seconds with 1 million photons. Note, however, that even less photons would be sufficient to obtain a similar noise level as for the 3D-box AMFs.

Reviewer Point P 2.5 — Pg 3, Ln 69: The following equation for SCD more accurately captures the way you have described it

$$SCD = \frac{1}{n}\sum_{i=1}^{n}\int_{path_i} c(l)dl \tag{1}$$

Reply: Our methods section starts from general equations to specific equations. The suggested equation already implies a solution calculated with a Monte-Carlo solvers. Therefore we did not apply the suggested change.

Reviewer Point P 2.6 — Pg 5, Ln 98:I think MCARaTs is another Monte Carlo RTM with the capability ofcomputing AMFs (https://sites.google.com/site/mcarats/home)

Reply: The MCARaTs RTM computes 1D-layer air mass factors and was part of the Wagner et al. (2007) RTM intercomparison study. However, we didn't find any indication that it is also able of computing/outputting 3D-box AMFs. We added two citations of the model in the introduction.

> In the past decades, numerous RTMs have been developed with the possibility to calculate one-dimensional layer AMFs (e.g. Berk et al., 1999; Postylyakov, 2004; Rozanov et al., 2005; Wagner et al., 2007; Spurr et al., 2001; Iwabuchi, 2006; Iwabuchi and Okamura, 2017). The computation of layer AMFs is implemented in most trace gas retrieval algorithms for satellite and ground-based observations applied today (Boersma et al., 2011; Irie et al., 2011; Wenig et al., 2008; Wu et al., 2013).

Reviewer Point P 2.7 — Pg 15, Ln 293: The SCCs are larger than the VCDs (panel a) because the AMFs(panel c) are generally larger than 1”SCC -> SCD. Also, is this a tautology?

Reply: We corrected the typo and do not think the sentence is a tautology.

Reviewer Point P 2.8 — Pg 17, Fig. 8x=19 km and y=13 km -> x=1.9 km and y=1.3 km

Reply: We corrected the typo.

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