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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Intercomparison exercise between different radiative transfer models used for the interpretation of ground-based zenith-sky and multi-axis DOAS observations" *by* F. Hendrick et al.

F. Hendrick et al.

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At first, we would like to thank Dr U. Friess for his helpful comments and suggestions.

Specific Comments

Referee comment: It is stated in the introduction (P. 7932, L. 4) that the NDSC network consists of about 20 stations and 5 primary sites. To my knowledge, there are many more primary and complementary sites, even if only the stations with DOAS instruments are considered. Please check this.

Reply: In the revised version of the manuscript, we have used the following sentence for describing the NDSC network: 'The NDSC consists of about 75 globally distributed stations combining various observation techniques.'. This concise description will be clearer for the reader.

Referee comment: P. 7932, L 17: You state that off-axis measurements '...enable the measurement of tropospheric species'. This statement is too strong since zenith-sky measurements also have a certain sensitivity to tropospheric trace gases. Please consider rewording (e.g., 'enhanced sensitivity compared to zenith-sky measurements').

Reply: We agree with the rewording suggested by Referee U. Friess. The text has been changed accordingly.

Referee comment: Maybe it would be worth mentioning in the introduction that a potential application of radiative transfer models is the usage as forward models for the inverse modelling of tropospheric trace gas profiles using MAX-DOAS measurements (see, e.g., the ACP paper of Bruns et al. [2004] on retrieval of profile information from airborne MAXS2830 DOAS measurements). An accurate radiative transfer modelling is essential for inverse modelling.

Reply: We agree and have added a comment on this in the revised version of the manuscript. It should be noted that the Bruns et al. (2004) paper has been published in Applied Optics, not in ACP.

Referee comment: P. 7937, L 13: 'A significant part of the OClO layer is likely to be in the Earth's shadow region and therefore can only be probed with multiply scattered light' - this is not exactly true: in the Earth's shadow region (above the instrument), single scattered light also traverses the trace gas layer vertically before it reaches the instrument. I would suggest to state that multiply scattered light has a stronger relative impact in the Earth's shadow region.

Reply: We have followed this suggestion in the revised version of the manuscript.

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Referee comment: P. 7937, L 20: You explain the persistence of relatively large SCDs for NO2 and OClO at high SZA compared to BrO with photochemistry. However, OClO and NO2 SCDs also decrease at very high SZAs, and it would be worth mentioning that this decrease is owing to geometrical reasons (it happens when the average scattering altitude moves above the trace gas layer). The SZA where the maximum SCD is reached is thus determined by a combination of photochemistry and altitude of the trace gas layer. In that respect, it might have been useful to perform model simulations also without chemical enhancement to separate geometrical from photochemical effects (please treat this as a comment only, I am aware that further calculations by all participants cannot be easily done).

Reply: In the revised version of the manuscript, we will mention that (1) OClO and NO2 SCDs also decrease at very high SZAs and (2), the SZA where the maximum SCD is reached is determined by a combination of photochemistry and altitude range of the trace gas layer.

Referee comment: P 7938, L 20: The difference between the models is explained by 'the step between the SZA values corresponding to the concentration tables is increasing at large SZA'. I do not understand this. Is it because the trace gas profiles are given as a function of time and need to be interpolated to SZAs? Again, model runs without chemical enhancement would have been useful to better understand the discrepancies between the model results.

Reply: The trace gas profiles are given as a function of the altitude and SZA. Since our photochemical box-model PSCBOX provides output with a constant time step of 6 minutes, the corresponding step in SZA increases with increasing SZA: e.g., for OClO and BrO profile tables, the SZA step is about 0.05° at 78° SZA and 0.70° at 90° SZA. These tables can be interpolated linearly or exponentially, depending on the model. Since (1) the step in SZA in the tables increases with SZA and (2), the change in concentration due to photochemistry are the largest at high SZA, the interpolation effect should be the largest above 90°. The difference between the model results can be also

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partly explained by a geometrical effect. In the ray-tracing, the trace gas concentration is calculated in each atmospheric layer along a given sun ray path. Depending on the model, it is done by interpolating the trace gas concentration table at the altitudes and local SZAs along the ray path corresponding either to the centre of the layers or to the bottom and top of the layers. In this last case, the concentration in the layers is given by the mean of the bottom and top values. This geometrical effect can lead to different concentration values for a given layer and ray path and it will be stronger at large SZA where the light path in the layers is the longest (according to Sarkissian et al. (1995), its impact on SCDs can reach up to 4% above 90° SZA).

Referee comment: Top of P. 7939: I do not understand the explanation why the Monte Carlo model has problems to deal with the OClO simulations, and I feel that this point needs clarification. Is the term 'scattering point altitude' referring to the altitude of the last scattering event in the zenith? If so, then a photon scattered below the trace gas layer should already have had a long (slanted) path through the trace gas layer, but you argue that its path is short. On the other hand, you state that a scattering point within the trace gas layer 'causes the photon to continue the flight on a slant path within the profile, leading to a sharp increase of the SCD'. If you talk about the last scattering event in the zenith, this statement makes no sense since the photon will not (or very unlikely) reach the detector. If it is not the last scattering event, then the photon could also be scattered at an angle that causes it to leave the trace gas layer after a short distance.

Reply: Indeed the term "scattering point altitude" is misleading. In our paper, it refers to the first scattering event the photon encounters after its atmosphere entry. If this event occurs above the OClO layer, then the path through this layer is short. If the event occurs within the OClO layer, it has had a long path through this layer. If the event occurs below, then the path through the OClO layer also has been long, though shorter then in the second case. If the profile is very confined vertically, as in the case of the OClO, and opposed to e.g. stratospheric NO2 or ozone, small variations in the

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altitude of the first scattering event, arising from statistical fluctuations, lead to large variations in path length and hence to large fluctuations in the simulated SCD. Only very high numbers of photons can reduce this noise.

Referee comment: Section 4.1: The NO2 profile used for the MAX-DOAS simulations is possibly not the best choice. The NO2 surface mixing ratio is only about 0.2 ppb (Fig. 5). This is representative for relatively unpolluted conditions under which MAX-DOAS measurements do not have a very high sensitivity for tropospheric NO2. On the other hand, the NO2 concentrations in the upper troposphere appear to be unrealistically high. In my opinion, a more 'typical' scenario for MAX-DOAS applications would have been a profile with an NO2 enhancement in the boundary layer, similar in shape to the tropospheric part of the HCHO profile.

Reply: In the NO2 profile used for the MAX-DOAS simulations, the volume mixing ratio in the troposphere has been fixed to 0.21 ppbv, which is about a factor of 10 larger than the value given for the troposphere in AFGL 1976. We agree with Referee U. Friess concerning the fact that a more realistic scenario for NO2 SCD simulations in MAX geometry would have been a profile shape in the troposphere similar to the tropospheric part of the HCHO profile. However, we think that the large concentration values in the free troposphere in the NO2 profile gives us the opportunity to test the models for two different profile shapes in the troposphere: a first one where the trace gas species is mostly present in the boundary layer (HCHO) and a second one where the trace gas species is situated in both the boundary layer and free troposphere (NO2).

Referee comment: Section 4.2: The impact of the azimuth angle on the modelled SCDs is only discussed for an aerosol-free atmosphere. It would be worth mentioning that the presence of aerosols should cause a much stronger azimuth effect owing to the strong preference of forward scattering by particles (whereas the Rayleigh phase function is symmetric in forward and backward direction). It is no surprise that the azimuth effect is smaller for NO2 than for O4 if you define it as the relative change in SCD as a function of azimuth angle: a significant fraction of NO2 is located in the stratosphere, where the

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azimuth effect should be very small. Furthermore, you refer to the study of Wittrock et al. [2004], who discusses the azimuth effect for O4 around 370nm, whereas your NO2 simulations were performed at 422nm. From my own studies on O4 AMFs, the azimuth effect in an aerosol-free atmosphere should be stronger at shorter wavelengths (here I am a bit confused, because you report that the azimuth effect for HCHO is smaller than for NO2 - perhaps this is because HCHO is only present in a thin layer above the surface). Moreover, the calculations from Wittrock et al. have aerosols included, which makes a large difference in the azimuth effect. Therefore I feel that the statement that Wittrock's calculations were performed under 'similar conditions' (P. 7941, L. 12) is not correct.

Reply: We agree with Referee U. Friess that the comparison of our results with the Wittrock et al.'s ones is not relevant, due to the differences between both studies in the model initialization settings (wavelength, trace gas species profile, aerosol conditions). So in the revised version of the manuscript, we don't refer anymore to Wittrock et al. (2004) for the discussion on the relative azimuth effect. As suggested by Referee U. friess, we have added a comment on the fact that the presence of aerosols should cause a much stronger azimuth effect.

Referee comment: The UHEI Monte Carlo off-axis simulations show a quite strong scatter (Figures 6 and 7). Can you comment on this? Is this caused by statistical fluctuations of the modeled photon paths?

Reply: Yes, it is statistical scattering. Higher photons numbers can reduce it, though take more time for large series of values.

Referee comment: Section 5: You use an aerosol extinction profile corresponding to a very clear atmosphere (k Ÿ 0.04/km at the surface, corresponding to a visibility of about 100 km) to investigate the impact of aerosol scattering. I could imagine that higher aerosol loads would yield larger differences between the model results. Can you comment on this? Have you performed any tests with higher aerosol loads?

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Reply: In the present study, we did not perform any tests with higher aerosol loads. However, such tests are currently performed in a new intercomparison exercise currently led by the University of Heidelberg in the framework of the EU Network of Excellence ACCENT. Preliminary results show that the differences between the altitude-dependent AMF (the so-called box-AMF) calculated by the different models are larger when the aerosol load is high, especially for box-AMF corresponding to altitude below 1000 m. More information on this new intercomparison exercise as well as preliminary results are available at the following web page: http://satellite.iup.uni-heidelberg.de/index.php/RTM_Workshop/149/0/.

Referee comment: Can you give further information about the aerosol settings (absorption coefficients and asymmetry factor)? Or, more importantly: what kind of aerosol composition corresponds to these settings?

Reply: The absorption coefficient profile and the asymmetry factor will appear in the revised version of the manuscript (in Fig. 8 and Table 6, respectively). All the aerosol settings have been derived from the aerosol model of Shettle (1989) included in the IASB and NILU RT models using a surface visibility of 100 km. The settings in the boundary layer and troposphere correspond to mixture of water soluble and dust-like aerosols representative of a rural environment. In the stratosphere, sulphuric acid aerosol settings corresponding to summer background conditions have been used.

Referee comment: P. 7943, last paragraph: I don't understand why the concept of Monte Carlo modeling should lead to a different sensitivity to surface albedo compared to the analytical models. Compared to Rayleigh and Mie scattering in the atmosphere, Lambertian reflection (and I suppose that all models treat the Earth's surface as a Lambertian reflector) is a very simple process that should also be reproduced realistically by Monte Carlo modelling.

Reply: For low (0.0) and medium (0.2) surface albedo conditions, the UHEI model results agree well with the results from other models. For high (0.9) surface albedo

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conditions, the UHEI model gives significantly larger SCD values, especially above 70° SZA. The modelling of the absolute radiance still poses a challenge to backward Monte Carlo approaches, which is also under investigation by other groups. At high albedos, light reflected off the ground is unattenuated and increases the signal of absorbers near the ground, so the absolute radiance begins to play a significant role in the SCD calculations. The present RTM intercomparison exercise was aimed at detecting those subtle effects, and the results will help to optimize the modelling. So will comparison against measurements, e.g., as performed in Weidner et al. (2005).

Referee comment: Table 1: It is stated that Raman scattering is included in the SCIATRAN model. Is this feature really used for the calculations presented in the paper? If not, please indicate.

Reply: Raman scattering is not included in the calculations presented in our paper. We have modified Table 1 accordingly.

Technical Corrections

Referee comment: The term 'observed' is frequently used for the description of model results. I find this a bit inappropriate since 'to observe' usually refers to observations (measurements) and not to the output of numerical models.

Reply: We have corrected this by replacing the term 'observed' by 'obtained' or 'is found'.

All the technical corrections listed in pages S2834-S2840 have been performed except the following one:

P. 7938, L. 26: replace 'as' with 'than'

The use of 'as' is here appropriate because in MS mode, as in SS mode, larger discrepancies are obtained above 90° SZA compared to lower SZAs.

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