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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.

U. Friess (Referee)

uf5@le.ac.uk

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General comments

This paper describes the results of an intercomparison exercise between several radiative transfer models for the interpretation of zenith-sky and off-axis DOAS measurements. A realistic modelling of the radiative transfer through the atmosphere is of paramount importance for a quantitative interpretation of trace gas observations from

scattered sunlight measurements, such as the conversion of the slant column density to the vertical column density and the retrieval of vertical profile information from multi-axis DOAS measurements. In particular, the intercomparison of radiative transfer simulations in MAX-DOAS geometry is, to my knowledge, performed here for the first time, and provides an assessment of the quality of radiative transfer models regarding the interpretation of tropospheric trace gas measurements using this relatively new measurement technique. For these reasons, the topic of the paper fits well within the scope of ACP. The comparison of the radiative transfer models is performed under consideration of the most important quantities influencing the radiative transfer (e.g., vertical trace gas profile, surface albedo, aerosols, viewing geometry) and the conclusions drawn provide a useful assessment of the comparability of the different radiative transfer models. Apart from some weaknesses in the usage of the English language, the paper is well written and clearly structured and I recommend it for publication in ACP after some minor corrections as detailed below.

Specific comments

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.

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').

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 MAX-

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DOAS measurements). An accurate radiative transfer modelling is essential for inverse modelling.

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.

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).

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.

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

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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.

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.

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 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

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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.

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 modelled photon paths?

Section 5: You use an aerosol extinction profile corresponding to a very clear atmosphere (k ˜ 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?

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?

P. 7943, last paragraph: I don't understand why the concept of Monte Carlo modelling 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.

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.

Technical corrections

The term 'observed' is frequently used for the description of model results. I find this a

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bit inappropriate since 'to observe' usually refers to observations (measurements) and not to the output of numerical models.

P. 7931, L. 9: '... taking into account the photochemical enhancement': delete 'the'

P. 7931, L. 13: replace 'angles' with 'angle'

P. 7931, L. 17: 'the aerosol scattering': delete 'the'

P. 7931, L. 20: replace 'difference values between' with 'differences ranging between'

P. 7931, L. 21: 'All the initialization data': replace with, e.g., 'The complete set of initialization data'

P. 7932, L. 4: replace 'observations' with 'observation'

P. 7932, L. 6: replace 'Network operations' with 'The network operation'

P. 7932, L. 7: replace 'have provided' with 'has been providing' (still ongoing activity)

P. 7932, L. 20: replace 'Due to' with 'Owing to'

P. 7932, L. 22: replace 'heights' with 'altitudes'

P. 7932, L. 29: 'In the case of': delete 'the'

P. 7933, L. 4: 'The so-called photochemical enhancement effect': replace 'The' with 'This'

P. 7933, L. 6: replace 'heights' with 'altitudes'

P. 7933, L. 7: replace 'concentration' with 'concentrations'

P. 7933, L. 12: replace 'models interface' with 'model interface'

P. 7934, L. 8: 'This task has been achieved': replace 'task' with 'aim' or 'objective'

P. 7934, L. 10: replace 'ground' with 'ground-based'

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P. 7934, L. 12: 'the photochemical enhancement': replace 'the' with 'with'

P. 7934, L. 25: 'the aerosols': delete 'the'

P. 7935, first sentence: I think you need to put a 'the' in front of the name of each institute

P. 7935, L. 8: replace 'name' with 'acronym'

P. 7935, L. 8: 'All the models': delete 'the'

P. 7935, L. 9: '... taking into account the photochemical enhancement': delete 'the'

P. 7936, L. 1: replace 'heights' with 'altitudes'

P. 7936, L. 8: replace 'ISAC' with 'ISAC-CNR'

P. 7936, L. 16: '... and when possible, ...': insert a comma after 'and'

P. 7936, L. 18, and captions of Figures 1 - 3: replace 'color plot' with 'contour plot'

P. 7936, L. 19: 'the altitude': delete 'the'

P. 7936, L. 25: '... and in case of ...': insert a comma after 'and'

P. 7937, L. 7: '... BrO and NO2 ...': insert 'SCDs' after 'NO2'

P. 7937, L. 8: 'In the case': delete 'the'

P. 7937, L. 9: replace 'SZA' with 'SZAs'

P. 7937, L. 13: 'the largest': delete 'the'

P. 7937, L. 13: replace 'Given that, ...' with 'Therefore, ...' or 'Thus, ...'

P. 7937, L. 14: put 'Earth' in upper case

P. 7937, L. 17: replace 'display' with 'show'

P. 7937, L. 20: '... increase with the SZA ...': delete 'the'

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P. 7937, L. 22: insert 'the' before 'MS'

P. 7938, L. 1: replace 'In the case of the ISAC-CNR mdoel, simulated SCDs are' with 'However, the SCDs simulated by the ISAC-CNR model are'

P. 7938, L. 6: delete 'the one'

P. 7938, L. 7: 'the photochemical enhancement': delete 'the'

P. 7938, L. 10: delete 'the ones observed'

P. 7938, L. 12: replace 'difference values' with 'differences'

P. 7938, L. 18: 'the photochemical enhancement': delete 'the'

P. 7938, L. 20: delete 'observed'

P. 7938, L. 21: replace 'increase of' with 'increase in'; insert 'the' before 'models'; replace 'step' with 'steps'

P. 7938, L. 22: replace 'is' with 'are'

P. 7938, L. 23: insert 'the' before 'agreement'

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

P. 7939, L. 3: '... the stratospheric OCLO ...': delete 'the'

P. 7939, L. 7: replace 'does react very sensitively' with 'is very sensitive'

P. 7939, L. 8: replace 'variation' with 'variations'

P. 7939, L. 10: replace 'increase of the' with 'increase in'

P. 7939, L. 12: replace 'induced' with 'induces'

P. 7939, L. 15: replace 'a near future' with 'the near future'

P. 7939, L. 18: 'the photochemical enhancement': delete 'the'

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P. 7939, L. 20, 'limited our comparison in': replace 'in' with 'to'

P. 7939, L. 24: replace 'and in the case of' with 'and, in case of'

P. 7939, L. 25: 'of elevation': delete 'of'

P. 7939, L. 25: start a new sentence after 'above the horizon'

P. 7940, L. 3: replace 'appear' with, e.g., 'are shown'

P. 7940, L. 4: 'the altitude': delete 'the'

P. 7940, L. 5: 'in the case of': delete 'the'

P. 7940, L. 11: insert 'the' before 'NO2'

P. 7940, L. 12: 'maximum difference values': replace with 'maximum differences'

P. 7940, L. 17: replace 'Concerning the UBRE model, it underestimates ...' with 'However, the UBRE model underestimates ...'

P. 7940, L. 18: it's a bit inappropriate to say that the UBRE model 'underestimates' the IASB model. Replace with something like 'the modelled SCDs are smaller than'

P. 7940, L. 18: replace 'maximum' by 'at most'

P. 7940, L. 19: replace 'The relative differences are also ...' with 'Furthermore, the relative differences are ...'

P. 7940, L. 20: replace 'in contrast with' with 'in contrast to'

P. 7940, L. 21: replace 'a SZA' with 'an SZA'; delete 'of' before 'elevation'

P. 7940, L. 22: consider rephrasing in order to avoid saying that one model 'underestimates' or 'overestimates' the other.

P. 7940, L. 23: replace 'observed maximum' with 'a maximum'

P. 7941, L. 1: replace 'through the calculation of' with 'by calculating'

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P. 7941, L. 8: 'the SZA': delete 'the'; 'of elevation': delete 'of'

P. 7941, L. 9: replace 'is observed' with 'occurs'

P. 7941, L. 15: replace 'than the one observed for NO2' with 'compared to NO2'

P. 7941, L. 18: replace 'The aerosols' with 'Aerosols'

P. 7941, L. 22: replace the sentence 'Given this effect ...' with 'It has been shown by Wagner et al. (2004) that the impact of aerosols on O4 SCDs can provide a new method to derive information on atmospheric aerosols.'

P. 7942, L. 1: 'the aerosols': delete 'the'

P. 7942, L. 5: delete 'the ones'

P. 7942, L. 8: 'the aerosol': delete 'the'

P. 7942, L. 11: 'In the case': delete 'the'; 'the aerosol': delete 'the'

P. 7942, L. 13: replace 'when the elevation angle increases' with 'at higher elevation angles'

P. 7942, L. 14: 'impact of the aerosol scattering': delete 'the'

P. 7942, L. 15: replace 'than NO2 SCDs' with 'than on NO2 SCDs'

P. 7942, L. 15: 'comprised': do you mean 'confined'?

P. 7942, L. 24: 'the aerosol': delete 'the'

P. 7943, L. 7: replace 'to higher absolute AMFs' with 'in higher absolute AMFs'

P. 7943, L. 8: replace 'horizon' with 'off-axis'

P. 7943, L. 9: replace 'due to' with 'owing to'

P. 7943, L. 10: 'the largest': delete 'the'

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P. 7943, L. 18: replace 'increase of the ground albedo value' with 'increase in ground albedo' (perhaps 'surface albedo' is better)

P. 7943, L. 18: replace 'increase of the HCHO SCD' with 'increase in HCHO SCDs'

P. 7943, L. 21: 'in the same conditions': replace 'in' with 'under'

P. 7943, L. 24: replace 'increase of the SCD values are systematically higher using the UHEI model' with, e.g., 'increase in SCDs calculated by the UHEI model is systematically higher'

P. 7944, L. 11: Please state that the comparisons in MAX-geometry were performed in the absence of aerosols.

P. 7944, L. 24: 'the aerosol scattering': delete 'the'

P. 7945, L. 1: replace 'observed' with, e.g., 'achieved'

P. 7945, L. 12: avoid using 'observed'

P. 7945, L. 12: insert 'the' before 'agreement'

P. 7945, L. 12: 'spread value': delete 'value'

P. 7945, L. 16: replace 'due to' with 'owing to'

P. 7945, L. 22: delete 'observed'

P. 7945, L. 25: 'All the initialization data': replace with, e.g., 'The complete set of initialization data'

Table 5: 'of elevation': delete 'of'

Captions of Figures 1, 2 and 3: Replace 'Color plot' with 'Contour plot' and delete 'the' before 'SZA'

Caption of Figure 7: Replace 'Here are plotted the relative differences ...' with 'The relative differences ... are plotted.'

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Caption of Figure 9: 'Impact of the aerosol scattering': delete 'the'

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