

Interactive comment on “Determination of aerosol properties from MAX-DOAS observations of the Ring effect” by T. Wagner et al.

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Reply to referee 2

First of all we want to thank this reviewer for the positive assessment of our manuscript and the constructive and helpful comments. We followed most of the suggestions and give detailed explanations in (the few) cases where we disagree (see our detailed response below).

General comments

The MAXDOAS technique represents a relatively new and promising development in the area of atmospheric passive remote-sensing applications, with great potential for unattended monitoring of the vertical distribution of important tropospheric trace gases

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as well as aerosol optical properties. This paper explores the potential of Ring effect observations (a side product of scattered light DOAS observations), as an additional source of information to better constrain the determination of aerosol properties in MAXDOAS retrieval types, so-far based on intensity and O₂O₂ absorption measurements. A newly developed approach to quantitatively simulate the effect of inelastic rotational Raman scattering (Ring effect) in Monte-Carlo radiative transport calculations is used to investigate the dependence of the Raman scattering Probability (RSP) parameter for a large range of observational geometries and aerosol loads. Comparisons with a limited data set of MAXDOAS observations (2 days) show that main observed features of the RSP variations can be reproduced by the model and qualitatively understood. Although the main conclusions from the study, i.e. to what extent the Ring effect add pertinent and useful information to MAXDOAS aerosol determination, are still rather speculative and would certainly deserve more in depth analysis, the paper present enough of innovative and pertinent discussions and will certainly reach a large audience within the scientific community. Also the manuscript is clearly and concisely written. References to existing literature are comprehensive and figures of good quality. I recommend publication in AMT after careful attention to the specific comments and remarks given below.

Specific comments

Major point One important conclusion from the study (maybe the most important one) concerns the large Ring effect dependence in the zenith viewing mode. It is argued that this should allow aerosol retrievals to be obtained from traditional zenith-sky observations, with potential application to existing historical long time-series. Although this seems to be a quite exciting conclusion, I am a little bit skeptical about the actual applicability of the method since the RSP parameter not only depends on the AOD but also on the aerosol vertical distribution as visible from fig. 6a, which can in no way be inferred from zenith-sky measurements alone. The disturbing effect of thin cirrus clouds is also very strong, but this limitation is well identified in the paper.

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Reply: Maybe there is a misunderstanding at this point: for zenith sky observations the dependence of the Ring effect on profile is rather low (the effect of the change of the aerosol profile from 0-1km to 0-3km is only about 10% of the effect of changing the OD from 0 to 1. Of course, this influence is not negligible, but usually some general information on the (relative) aerosol profile might be available (e.g. boundary layer height) and the uncertainties can be largely reduced by using this information. But even without such information, the effect of the profile height is usually rather small; it becomes even smaller towards larger wavelengths (e.g. <5% at 500nm), because of the decreasing probability for scattering on molecules. We added this information at the end of section 5.3.

Minor points Abstract (and in several places throughout the paper): the authors insist on the argument that both Ring effect and O4 slant columns should have similar dependence due to the fact that both quantities depend on the light path length in the lower atmosphere. Although this is certainly true to some extent, only O4 directly depends on the light path length, while the Ring effect (which is directly linked to Raman scattering by air molecules) is also strongly modulated by the ratio of the intensities due to Rayleigh and Mie scattering. This explains the observed inverse relationship between RSP and intensities for near-sun measurements, when aerosol forward scattering becomes dominant (see e.g. figure 3). In brief, I think the similarity argument should not be stressed too strongly since it might give the wrong (or negative) impression that O4 and Ring effect show the same effect which is inexact.

Reply: We agree that our discussion was misleading here. We modified the text in the abstract and added a much more detailed discussion in the conclusions. This discussion includes not only the Ring effect and O4 absorption, but also the specific sensitivity of the observed radiance.

P. 730, L. 12: please note that the Hermans et al. O4 cross-section, although measured in the lab, have been obtained in atmospheric conditions (in a very long multi-pass absorption cell).

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Reply: Many thanks for this information. We changed the text accordingly.

P. 731, L. 15-20: the corrections for the O4 and RSP values in the reference spectrum are somewhat confusing. Some plots show negative RSP values (figs. 9 & 10), which suggests that these values have been sometimes subtracted from simulations instead of being added to measured quantities. It is also unclear how these values have been determined from radiative transfer simulations (under which assumptions ?)

Reply: We agree that the presentation of our results was confusing. We made all figures consistent now: the amounts of the Fraunhofer reference spectrum is always added. The amount was determined from RTM assuming a weak aerosol OD (the Fraunhofer reference spectrum was measured at 14.09.). This information is added to the text.

P. 734, L. 25: here, it is maybe worth to mention the reasons for the decreased radiance at low elevation angle (due to reduced light path length) and the increased radiance for zenith (I suppose due to multiple scattering effect)

Reply: We added the following information: 'The increased radiance for zenith view is caused by the additional the additional photons scattered by the aerosol particles. For low elevation angles, the situation is more complex: even in the absence of aerosols the optical depth along the line of sight is typically much larger than unity. Thus, additional aerosols do basically not enhance the overall probability for solar photons to be scattered into the instrument. The decrease of the observed radiance is instead mainly caused by increased probability for multiple scattering including multiple reflections at the low surface albedo.'

P. 738, L. 1: it is argued that a study of the wavelength dependence of the Ring effect might allow to discriminate the effect of clouds from those of aerosols. Without more discussion, this assertion seems rather speculative. What are the main arguments to support this statement?

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Reply: We added the following information: ‘While the wavelength dependence of scattering by cloud particles is weak (clouds are white), that of aerosol scattering shows a strong wavelength dependence. Thus the relation of the strength of the Ring effect at different wavelengths should be different for clouds and aerosols.’

Editorial comments P. 733, L. 24: typo – replace “telecsope” by “telescope”. Same line: add “s” to depth.

Corrected.

P. 734, L. 8: replace “potential effect” by “suspected presence”. Also refer to sect. 5.2 regarding the thin (cirrus) clouds issue.

Corrected.

P. 737, L. 1: replace “... influence of surface near aerosol...” by “... influence of near surface aerosol...”

Corrected.

The reference to Greenblatt et al. (1990) is missing

The reference is added.

The reference to Chance et al. (1991) listed at the end of the manuscript is not cited in the body text

The reference is deleted.

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