

***Interactive comment on* “On the variability of the Ring effect in the near ultraviolet: understanding the role of aerosols and multiple scattering” by A. O. Langford et al.**

Anonymous Referee #2

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General comments

This discussion paper contributes significantly to the literature on the Ring effect. The paper begins with a well written and researched introduction. The paper presents a novel, simple and effective approach to model multiple Raman scattering for atmospheric applications. The paper addresses relevant scientific questions within the scope of ACP. The paper presents novel, good-quality data as well. There is a well-presented demonstration of the wavelength dependence of the Ring effect in the remote sensing of NO₂ columns from near ultraviolet spectra. Substantial conclusions are reached, for example, on the importance of higher order scattering. Assumptions

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are valid and clearly outlined. Some improvement is required to more clearly outline the scientific method. The description of experiments and calculations is not sufficiently complete and precise to allow reproduction by fellow scientists (traceability of results). The results are sufficient to support the interpretations and conclusions. The authors have not credited related work in a few instances. Some additional references are necessary (see below). The title clearly reflects the contents of the paper. The abstract provides a concise and complete summary, although the authors should mention that they have developed a parameterization for filling in. The overall presentation is adequate. The language is fluent and precise. Mathematical formulae, symbols, abbreviations, and units are correctly defined and used.

Specific comments (to the authors)

2. Measurement techniques

It is not easy to visualize the “daisy” pattern. Is this important to one’s understanding of the setup? Perhaps this detail is not necessary. Is there any internal scattering from the Hg/Ar calibration lamp leading? This could lead to a filling in of Fraunhofer lines. Perhaps the authors could quantify this by measuring the intensity of the lamp at wavelengths far from any lamp emission lines (with no second input from sky or sun). A reference for the lack of fluorescence of PTFE at 344 nm should be given since only the direct solar measurements used the plate. Heitz et al. studied the weak visible fluorescence of PTFE with 514 nm excitation [Appl. Phys. A 69 [Suppl.], S467-S470 (1999)].

The sentence “Forward scattered light . . . was shown to be negligible by measuring the change in intensity when the telescope tracking was switched off” leaves it up to the reader to figure out how the intensity changed as the sun moved out of the field of view (FOV). I assume the measured intensity dropped off significantly when the sun left the FOV. This should be stated explicitly.

The term “global component of solar irradiance” is not one with which I was familiar

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prior to reviewing this paper. I suggest “total irradiance”, the term used by Michalsky et al. (2001). 3. Measurement results

The SZA range was restricted to <70 deg. Are the measured intensities at SZA=79 deg so small that uncertainties go from being negligible at SZA=70 deg to being non-negligible? Including the SZA range of 70-80 deg would provide an interesting test of the authors’ ability to simulate the effects of aerosols.

HOW WELL DOES A SCALED SINGLE SCATTERING RING SPECTRUM FIT THE OBSERVATIONS? You are in the position to answer this question and it would be valuable to contrast the single and multiple scattering Ring spectra.

“These measurements provide the first explicit demonstration of this phenomenon in radiance spectra that can be directly related to DOAS measurements.” The authors have overlooked the work of Karkoschka [Icarus 111: 174, 1994] who clearly showed the filling in of Neptune and Uranus to be greater than Jupiter and Saturn and related it to the stronger contribution by aerosol scattering for the latter planets. That study relied on optical absorption spectroscopy to investigate, for example, the possible existence of water vapour in Jupiter’s atmosphere. The authors should remove their claim of a “first” here.

The authors should add the word “simply” in the blank to “calculated ___ from the Rayleigh phase function normalized ” so that the reader is clear that there are no RTMs used up to this point in the paper.

4. Model descriptions

“. . . from a forward Monte Carlo model developed by one. . .” - Refer to the model description paragraph which follows later in the section.

“it does not appreciably change the shapes of the Fraunhofer lines. . .” -The shape of the lines are changing, it’s the shape of the filling-in which is relatively constant between first and second order scattering.

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“Molecular parameters are taken from Sioris and Evans (1999).” -The appropriate reference for molecular parameters is:

Sioris, C. E., The filling in of absorption lines in sky spectra due to rotational Raman scattering, Ph. D. thesis, 135 pp., York Univ., Toronto, May 2001.

and/or

C. E. Sioris and W. F. J. Evans, Impact of rotational Raman scattering in the O₂ A band, *Geophys. Res. Lett.* 27, 4085-4088 (2000).

What simplifications have been made in your single scattering model relative to the one using the approach of Sioris et al.? Is it mostly in the way the Raman shifting is handled?

The description of the RRS model is slightly lacking, forcing most readers to try to understand why binning is done when the spectrum has already been interpolated to the pixel center wavelengths. After the sentence on binning, you could simply add a sentence such as: “The binning is required for the computation of inelastic scattering component to determine the pixel in which the Raman shifted light falls.”

“These phase functions are essentially independent of wavelength” -The phase functions listed in Eqns 1-2 are completely independent of wavelength, as they are only a function of scattering angle. Phase functions for Rayleigh scattering that include part or all of the rotational Raman band have a weak wavelength dependence (far from resonant frequencies).

“Since only one of the scattering angles is constrained” -Change “constrained” to “unconstrained” so that similarity of SZA dependence between single and double scattering intensity is explained.

Citing “Health” is insufficient. The web address should be inserted into the text, according to EGU citation guidelines.

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5. Model results and comparison to measurements

“This expression assumes that scattering orders greater than two are unimportant”.
-change to e.g. “. . . that double molecular scattering is sufficient”.

“. . . RRS model of Sioris and Evans” The appropriate reference for this model is: C. E. Sioris, W. F. J. Evans, R. L. Gattinger, I. C. McDade, D. Degenstein, E. J., Llewellyn, Ground-based Ring effect measurements with the OSIRIS DM, *Can. J. Phys.*, 80, 483-491, 2002.

“The scattering angle dependence of FI2 . . .” A reference to: C. E. Sioris and W. F. J. Evans, Modeling higher order radiation fields using iterated integrals of phase functions, *J. Quant. Spectrosc. Radiat. Transfer*, 72, 227-236 (2002).

and/or a polar plot of the angular dependence of FI2 would be useful here.

6. Implications for DOAS retrievals

It is somewhat surprising that including NO₂ absorption does not improve the fit between 305 and 365 nm.

Near Eqn. 5, the following work should be cited:

Liu, X., K. Chance, C. E. Sioris, R. J. D. Spurr, T. P. Kurosu, R. V. Martin, and M. J. Newchurch (2005), Ozone profile and tropospheric ozone retrievals from the Global Ozone Monitoring Experiment: Algorithm description and validation, *J. Geophys. Res.*, 110, D20307, doi:10.1029/2005JD006240.

There are at least two previous studies on the impact of clouds on Ring effect. In the final paragraph, you could acknowledge this with a reference to, for example:

R. de Beek, M. Vountas, V. V. Rozanov, A. Richter, and J. P. Burrows, “The Ring Effect in the cloudy atmosphere,” *Geophys. Res. Lett.*, vol. 28, pp. 721-724, 2001.

Technical corrections

Rotational -> rotational (although others have capitalized as you have)

3. Measurement results “Rayleigh scattering contribution calculated from the Rayleigh phase function” -change “from” to “using”

“The calculated FI are. . .” -> “The FI observations are. . .”. “calculated” sounds like a model calculation but you are referring to your measured FI here.

4. Model descriptions

“. . .and the average weighted by the level density.” -Insert “is” between “average” and “weighted”.

5. Model results and comparison to measurements

“. . .scattering results from. . .” -> “model results from. . .”

“. . .when the aerosol scattering phase function is large.” -> “. . .when aerosol scattering is most intense.”

6. Implications for DOAS retrievals

“NE” -> “northeast”

Figure captions

Figure 1. Include SZA and indicate am or pm.

Figures

Figure 2. The colour of the crosses should follow the same day-based colour scheme as is used for FI, so the reader isn't left guessing.

Figure 3a. Exponents on the y-axis are not legible.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 10153, 2006.

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