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Interactive Comment

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.

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The paper describes a detailed investigation of the dependence of the Ring effect on the aerosol load, solar zenith angle and wavelength. The discussion and the conclusions are based on measurements and model results. In my opinion this paper is a very useful contribution and it should be published in ACP. Nevertheless, I am surprised that the authors seem to disregard that most of the basic findings and ideas of the paper were already introduced by Wagner et al. (2004). Wagner et al. also presented results of (MAX) DOAS observations and radiative transfer modelling of the Ring effect and discussed various dependencies of the Ring effect. The fact that the authors disregard the results of our paper is astonishing for me, especially because they cite it twice in the text indicating that they actually know it. (at both citations in the text, however, it seems to me that a reference to Greenblatt et al. (1990) would have been more appropriate)

Several basic ideas and results presented by Langford et al. were already introduced and discussed in Wagner et al., 2004; in the following I will give some examples:

a) the general dependence of the strength of the Ring effect (filling in, FI) on the relative contribution of Rayleigh and Mie scattering was discussed in detail in Wagner et al., 2004, e.g. in section 4.6: '....The magnitude of the Ring effect (e.g. expressed as the strength of the filling-in of a Fraunhofer line, see e.g. Grainger and Ring [1962]) is a measure for the relative contribution of light having undergone rotational Raman scattering to the measured total intensity. Rotational Raman scattering occurs only for scattering on molecules; the probability of a photon being Raman scattered is about a few percent [Bussemer, 1993; Chance and Spurr, 1997]. The magnitude of the Ring effect thus depends on the specific conditions of a given measurement. Two different cases have to be considered: a) Measurements, in which mainly single scattered photons are received by the telescope (e.g. zenith sky observations under clear sky at large wavelengths, see Table 3). In this case the relative contribution of Raman scattered photons decreases if additional (elastic) scattering on aerosols takes place. Consequently, the magnitude of the Ring effect decreases with increasing frequency of aerosol scattering. b) Measurements, in which mainly multiple scattered photons are received by the telescope (especially observations under cloudy sky). In this case additional aerosol scattering increases also the probability of a photon to be scattered again by a molecule and thus to undergo additional Raman scattering. Consequently, now the magnitude of the Ring effect increases with increasing frequency of aerosol scattering. This effect can be clearly seen for measurements under extended clouds (Fig. 6), when multiple Mie scattering takes place. As can be seen in Fig. 11, the amount of the Ring effect is indeed a very sensitive indicator of the specific contributions of molecular and aerosol scattering....'

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b) the decrease of the strength of the FI for increasing aerosol load was discussed in detail in section 4.6 (see point a) and presented in Fig. 11, Fig. 15.

c) the increase of the strength of the FI for multiple scattering inside clouds was discussed in detail in section 5.3.; measurements showing enhanced Ring effect (together with intensity measurements) were presented in Fig. 6.

d) Several measurements of the SZA dependence of the FI (and the intensity) were presented in Wagner et al. in Fig. 10 and 11. Nevertheless, Langford et al. claim on page 10159, line 23, that '...these measurements provide the first explicit demonstration of this phenomenon in radiance spectra that can be directly related to DOAS measurements....'

e) the relation of the FI to the measured intensity was discussed in Wagner et al. in section 4.6 and respective presented in Fig. 10, 11.

f) Langford et al. propose that the quantitative analysis and interpretation of the Ring effect as a new method for aerosol characterisation. (page 10171, line 1: ...suggests a new technique for accurate AOD measurements). This proposal was already made by Wagner et al., e.g. in the conclusions. There it is stated: '...e) Not only the O4 absorption can be analyzed but also the magnitude of the Ring effect and the (relative) intensity can be investigated. These quantities can also be used for the determination of aerosol properties...'

g) Wagner et al. investigated and discussed the influence of the scattering phase function in detail (see section 5.5., Fig. 15).

In addition to Langford et al., Wagner et al., 2004 also discusses and investigates in detail the dependence of the Ring effect (and the intensity) on the elevation angle and azimuth angle of the telescope. Also results of observations and radiative transfer modelling are presented. Wagner et al. also discusses the effects of absorbing aerosols (section 5.4)

Another basic idea of Langford et al. was already introduced by Wagner et al., 2002. There we describe the use of two Ring spectra to correct the wavelength dependence of the FI. In the analysis section of Wagner et al., 2002 it is stated: '...For the correction of the 'filling in' of the Fraunhofer lines in the spectra of scattered sunlight (the so called Ring effect [Grainger and Ring, 1962; Bussemer, 1991, Fish and Jones, 1995]) one or two Ring spectra are also included into the fitting routine. The first Ring spectrum was calculated assuming that Rayleigh-scattering was the dominant atmospheric scattering process, the second Ring spectrum assuming that Mie-scattering was the dominant atmospheric scattering process. Compared to the first Ring spectrum the amplitude of the second Ring spectrum increases towards smaller wavelengths. This reflects the strong difference of the wavelength dependence of Raman-scattering (and Rayleigh-scattering) compared to that of Mie-scattering [Wagner, 1999]. Using two Ring spectra in the DOAS analysis of scattered light spectra minimizes the errors of the fitting results, especially when large wavelength ranges are analyzed....'

Based on this facts, I recommend that the authors correct their manuscript at several places including proper references to both papers and changing the passages where they erroneously claim to have first presented measurements, model results or invented ideas.

Some additional minor comments

-On page 10158,line 23 it is stated that 'the agreement between both spectra is excellent'. The authors might be more precise here. The scale in which both spectra are shown, is not well suited for a detailed comparison. It would e.g. be more interesting to know, which fitting coefficient were obtained if (the logarithms of) both spectra are fitted to each other.

-on page 10158, line 26 the 'depth' of Fraunhofer lines is defined. I wonder why the authors do not apply the logarithm here (usually the (optical) depth of absorption lines is defined by the logarithm of intensity ratios.

-on page 10159,line 3 it is stated that 'This also minimizes any potential errors arising from a very weak O4 absorption...' In contrast to the rather broad band O4 absorption, at 344nm there is also a much narrower ozone absorption band, which might indeed interfere with the analysed Fraunhofer line. For typical atmospheric conditions, the optical depth is about twice of that of the O4 absorption. Did the authors correct for any potential interference?

-on page 10161 it is stated that 'the solar zenith angle dependence of the calculated FI arises from the ratio of the different phase functions for Raman and elastic Rayleigh scattering....'

While this dependence is certainly responsible for a large part of the observed SZA dependence, the authors might also say something about the additional effect of the increasing number of scatter events on the FI. On page 10163 it is e.g. stated 'that multiple scattering by molecules increases with SZA and the mean number of scattering events approaches 2 at SZA=80°.' An increasing number of scattering events will also increase the probability of a photon to be Raman scattered thus increasing the FI.

-On page 10164, line 5 it is stated that 'The number of Rayleigh scattering events approaches unity at long wavelengths in the absence of aerosols, but zero when even small amounts of aerosol are present.

I believe that aerosol scattering does in general not decrease the number of Rayleigh scattered photons, especially not to 'zero' (for the single scattering approximation, the Mie scattered photons just add to the Rayleigh scattered ones, increasing the total intensity). I guess the authors mean that the relative fraction of Rayleigh scattered photons decreases as the number of Mie scattered photons increases (see also point a above).

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