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Atmospheric Chemistry and Physics Discussions

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.

A. O. Langford et al.

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Reply to Comment by T. Wagner

We would like to thank Dr. Wagner for pointing out our omission or underrepresentation of the excellent work by him and his colleagues in our text and references. This will be corrected in our revised manuscript and the reference to Greenblatt et al. (1990) inserted where more appropriate. Dr. Wagner's comments are shown in italics and our responses in normal type. For brevity, we do not reproduce all of the excerpts from his papers quoted in his comments.

"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 conclu-

sions 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: . . .".

Advances in science do not occur in isolation and all "new" research is based in part on earlier work. This is certainly true for the Ring Effect, which has been studied extensively since the original observations by Shefov (1959) and Grainger and Ring (1962) more than 40 years ago. We have attempted to reference much of this early work in our introduction. Dr. Wagner is correct in pointing out that many of the basic ideas regarding the opposing effects of RRS and Mie scattering on the filling-in of Fraunhofer features were previously described in Wagner et al. (2002, 2004). However, we do not represent these ideas as having been "invented" by us as he suggests. Indeed, as we do point out, most of these ideas relating to the effects of Mie scattering by aerosols and cloud droplets and the resulting zenith angle dependence were in fact, first alluded to by Noxon et al (1979) and described explicitly by Kattawar et al. (1981) long before the publication of Wagner et al. (2002). To our knowledge, multiple Rayleigh scattering was first discussed by Fish and Jones (1995), and the effects of clouds (which we only mention in passing) by Dvorjashin (1995).

For example, Noxon et al. (1979) state: " With a clear unpolluted troposphere the effect appears to be due to rotational Raman scattering which generates a quasi-continuum of about 5% of the Rayleigh scattered intensity. With pollution the effects can be smaller

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or larger, depending upon whether the pollutants are more or less efficient at scattering . . .”.

And in Kattawar et al. (1981) we find “. . . aerosol scattering plays a part in the observed variations, partly in influencing the scattering angles and partly by diluting the molecular line-filling with unshifted sunlight.”, and “For small zenith angles, the strong forward lobe of the aerosol scattering will produce a strong unshifted component that will dilute the molecular scattering contributionĚthis effect should be less pronounced towards 90°, since the single scattering phase function for aerosols is much less intense here than in the forward direction.”

The latter comment qualitatively anticipates all of our results. What we have introduced in our paper is a set of measurements specifically designed to quantitatively probe this effect in order to simplify the modeling and quantitative interpretation of the data in order that the Ring Effect can be more effectively corrected in DOAS measurements. We use a direct-sun and zenith measurement, and our analysis is focused on the FI of individual Fraunhofer features to simplify the interpretation.

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. . . .’

As we admitted in our response to Reviewer 2, “First” claims are often problematic. This is particularly true in a field of research as storied as the Ring effect. We had hoped to circumscribe our “first” by including the admittedly ambiguous word “explicit”, and by confining the application to DOAS measurements. This was clearly insufficient. In any event, such claims rarely serve any useful purpose and we agree that it is best to remove it entirely. We would like to bring Dr. Wagner’s attention to the work by Karkoschka (1994) brought to our attention by Reviewer 2 identifying what the reviewer

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feels is the appropriate “first” reference here.

(From Reviewer 2) ” 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.

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: ‘. . .) 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. . . ’

I don’t quite understand this objection, we are certainly not proposing using the same methodology suggested in Wagner et al. (2004). I have attempted to clarify this by appending the text “based on the measurement of individual Fraunhofer line depths.” to the offending sentence. If the objection is more generic, I need only point out that it was Kattawar et al. (1981) who stated “It is entirely possible that this may be a new way of probing atmospheric aerosols.”

We have added the sentence “Wagner et al. (2004) also noted an inverse relationship between FI and the zenith sky brightness.” At the end of the paragraph discussing our Figure 2.

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 Ě one or two Ring spectra are also included into the fitting routine. The first Ring spec-

trum 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 Rayleighscattering) 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. . . .'

Dr. Wagner suggests that our suggestion of using a scaled Ring spectrum in DOAS retrievals is equivalent to the technique of using two different Ring spectra described by Wagner et al. (2002). As we have pointed out in the revised manuscript, our result is more general. Using only two Ring spectra in the analysis is essentially a first-order approximation of our technique since if the exponential pre-factor were expanded we would obtain an infinite series of Ring spectra.

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.

The linear correlation coefficient is R^2=0.996. Using the logarithms would be less instructive since the intensities over this wavelength range vary by only about a factor of two at our resolution.

-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.

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Our definition is equivalent to that used by Harrison and Kendall (1976) so that the results are more easily comparable to other FI studies.

-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?

The O_3 absorption is also negligible below 70° as is now stated in the text, i.e. "This also minimizes any potential errors arising from the absorption by O_3 (?1

-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.

We have changed the sentence on page 10161 to read "In the case of single scattering, the solar zenith angle dependence of the. . ." The increase in FI caused by second-order scattering is mentioned later in the same paragraph and elsewhere in the text.

-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

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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).

We were indeed referring to relative fraction as Dr. Wagner deduces. We have inserted the word “relative” into the sentence.

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