

***Interactive comment on* “Technical Note: Cloud and aerosol effects on rotational Raman scattering: Measurement comparisons and sensitivity studies” by A. Kylling et al.**

Anonymous Referee #2

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General Comments

The paper presents another formulation of the discrete ordinate radiative transfer equation (RTE) including first order rotational Raman scattering (RRS); the new model is used to make filling-effect comparisons against two measurement data sets, and to perform new cloud-related sensitivity studies of radiance and irradiance filling-in computed at various atmospheric levels, including inside clouds. The material is potentially suitable for publication in AMT, but there are a number of major issues that need to be addressed before the paper can be accepted. The main issues are listed next and particular line comments will be given separately.

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1. It seems that the main purpose of the paper is to present a new model for RT with RRS. If this is the case, then (1) there should be some justification (in the first paragraph of the Introduction) for the need to use RT models for simulating Ring spectra, and (2) there should be more detail presented on the model in Section 2. Radiative transfer with RRS sources is not easy, and there are essential details missing in this presentation.

2. I still have some problem with the organization. If the main aim is to present a new model, then Section 2 should be given over entirely to this task, and should be enhanced with parts of the Appendix and with additional details as noted below. Sections 2.1 and 2.2 should be moved to sections 3.1 and 3.2, and the very short section 2.3 can easily be incorporated at the beginning of section 4.

3. Although there are ~ 36 references quoted, the referencing in the text was uneven, not sufficient in some places and overabundant in others. For instance, the first paragraph of the introduction does not contain any references except Kattawar. There should be some reference to the importance of RRS and the usage of Ring spectra in ground-based and satellite applications, e.g. for ozone column and profile retrievals from space. Some references need to be added to cloud-property retrievals using Ring-based algorithms from OMI measurements (a reference to the 1995 concept paper by Joiner and Bhartia is not enough in this regard). On the other hand, it is not really necessary to give 4 or 5 references to issues relating to the instruments in the two measurement cases - these are minor considerations.

4. At no point in the paper can I see any reference to the number of discrete ordinates N used in any of the calculations. (The symbol N in the Appendix is never explained). Indeed one could be forgiven for thinking that the RRS sources are treated in the two-stream approximation, given the use of sums $j = \pm 1$ in Equation (A1). Please confirm that this is not the case. The choice of N is especially critical for studies with clouds and aerosols, and explicit choices of N need to be given for every simulation.

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5. In addition, there is no reference to the Delta-M scaling for dealing with sharply-peaked phase functions, or the need to perform very accurate single-scatter (SS) calculations with complete phase functions using the Nakajima-Tanaka ansatz. Again, these issues are critical for simulations with clouds and most especially if Mie-derived phase functions and associated expansion coefficients are used. Any such calculations using low values of N and not including "exact SS" and delta-M will involve large phase function truncation. The use of phrases such as "exact Mie calculations" is therefore misleading, and comparisons of "Henyey-Greenstein" with "exact Mie" results (Page 22525) cannot be convincing until these accuracy issues are stated and investigated.

6. The RTE with RRS can be thought of as a discrete ordinate system with a large number of additional particular integrals (PIs). These PI solutions are the most important new aspect in the RT modeling, and there is very little detail in the Appendix on their determination. We are simply referred to the "qdisort" solver. This is a major new application for this solver, and some detail would be useful - e.g. what type of PI is being solved here - exponential, linear, etc. There are also some issues of degeneracy and optical depth scaling that were treated in the paper by Spurr et al. (2008), but are not mentioned here. Specifically, all the inelastic source terms in Eq. A1 have exponential dependency; however, the exponential substitution method becomes degenerate when the source term exponential has the same form as the eigensolutions to the homogeneous discrete ordinate equations. This is the case with the diffuse-field Raman loss terms in particular. How did the authors deal with this in their model?

7. The choice of solar spectrum is important to RT with RRS, and its usage is critical. Eventually in paragraph 1 of Section 3.1 we learn indirectly that the ATLAS3 spectrum was used throughout the paper, but its resolution was not specified. In Section 3 (measurement comparisons), the calculation resolution is 0.05 nm. This is presumably different from that of the ATLAS3 spectrum, in which case was the ATLAS spectrum interpolated? It might have been better here to have used a high-resolution solar spectrum such as the Kurucz Kitt-Peak. Again, were the Raman-shifted cross-sections "binned"

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into 0.05 nm intervals, or was the calculation done "monochromatically"? (Reference is made here to the Spurr et al. (2008) distinction). In section 4, were the chosen H and K wavelengths actually taken from the ATLAS data set?

8. There are some issues with the model comparisons. The measurement data do not appear to be very accurate, and there are a number of model parameterizations (no ozone, variable albedo, choice of Angstrom coefficients, etc.) that are potential sources of error. A previous reviewer has noted the lack of an undersampling correction, and apparent differences in SZA values. In Figure 1b (b), the model/measurement ratios are 7-10% different at the start and end of the wavelength range yet much closer in the middle wavelengths - this requires an explanation. In addition, the filling effect in Figure 1b for the cloudless sky is hardly visible, but for the cloudy case there are marked differences between the solid and dotted lines - this is not what one would expect, and one wonders if the Figure is incorrectly labeled.

9. There are some serious issues with the sensitivity studies. Figures 4 and 5 are not consistent with results derived by other authors. Intuitively one would expect TOA upwelling RRS filling to decrease monotonically both with increasing cloud optical depth and higher cloud-top heights. This is certainly the case in de Beek et al. (Figure 5) and Spurr et al. (Figure 7). Yet in Figure 4 we find that the TOA radiance FI is a double-valued function of τ_{cloud} with a maximum at $\tau_{\text{cloud}} \sim 2$. And again, in Figure 5 for $\tau_{\text{cloud}} = 10$, we find a double-valued function of FI with cloud-top height; this non-uniqueness (especially Figure 5) has profound implications for cloud-property retrieval from space using RRS signatures, and contradicts results obtained from OMI data (Vasilkov et al, 2008).

10. Beyond a certain value of τ_{cloud} , clouds are essentially semi-infinite reflecting boundaries; the differences between $\tau_{\text{cloud}} = 100, 200$ and 500 in figure 5 are surprisingly large, as is the separation between the MLER value and the $\tau_{\text{cloud}} = 500$ value. Values between $\tau_{\text{cloud}} = 10$ and 50 would give some indication of the intermediate behavior, particular in the light of the above remarks on non-uniqueness.

11. There is no proper definition of the "irradiance" as obtained from the new model - is this the actinic or regular flux (hemispheric integrations of the radiance and cosine-averaged radiance fields)? Although the results for irradiance are new, the solar zenith angle behavior of RRS filling is well-known, and I think Figure 3 is not needed. The pseudo-spherical dependency has also been looked at before. I would recommend moving section 4.5 to section 4.1 (merging the text) and replacing Figure 3 with a modified form of Figure 9. [Figure 9 as it stands now contains mostly redundant information, and it is hard to pick out the pseudo-spherical effect - starting the x-axis at 70 degrees would be better].

Because of serious issues with the results in Figures 4 and 5, it is difficult to comment on subsequent results displayed in Figure 6-8, even though there is broad agreement in Figure 6 with previous results in the literature. Thus, conclusions drawn from the sensitivity studies cannot be properly assessed until these issues have been ironed out.

Particular Comments

Abstract

Please avoid the use of references in the Abstract.

Section 1

22516, L10. Please quantify this fraction inelastic scattering ($\sim 4\%$). "...dominantly oxygen and nitrogen," cut this. The acronym RRS should be used throughout for rotational Raman scattering.

22516, L18. gase \rightarrow gas. Add references here (see note 3 above).

22516, L21. Add references here (see note 3 above).

22516, L22. Numerous numerical \rightarrow Many numerical (alliteration is confusing).

22517, L4-L5. "Successive order of scattering codes are generally not well suited...".

This is a subjective statement, drop this sentence. Also, the Joiner et al. model was restricted to Rayleigh and Raman scattering of air molecules and contained no treatment of aerosols and/or clouds.

22517, L6, clouds explicitly → clouds and aerosols explicitly.

22517, L11-12. Both results...were shown → Results... were both shown.

22517, L20-21. Surely you mean "inclusion" and not "neglect". Suggest you rephrase this sentence. "Most applications" - are there any applications for which polarization may be important?

22517, L22. It is noted... this sentence has no added value, suggest you drop it.

22517, L26. What is subpercentage? Give a figure. You should mention in this section that the Vountas et al. model is also discrete-ordinate based, and that the Landgraf et al. model is based on Gauss-Seidel approximation of multiple scatter.

Section 2.

Section 2 should really only be about the RT model - this is the most important new thing in the paper and is the basis for everything else that follows. The "model description" should include some details currently relegated to the appendix. Although the Appendix is quite good, it is missing material on the RR spectroscopy, and there are some technical issues (for example associated with the use of the "qdisort" solver) which are missing or not adequately explained.

The RTE derivation used here and by most other groups, is that the solution with first-order RRS is viewed as a perturbation on the elastic calculation with Rayleigh scattering for air molecules. If you use the purely elastic Cabannes scattering instead as your starting point, then there is no "Raman loss term" in the RTE. This possibility was mentioned by Spurr et al. (2008) and van Deelen et al. (2007), and should be noted here.

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22518, L22 similar.. → similarly...write ĩĀćsca explicitly here.

22518, L23 specie → species.

22519, L11 wavelengths (sp)

22519, L12. The technical details... You should give a summary of the solution method here, using the material in the Appendix A as far as P22536, Line 15. At this point one can refer to the qdisort solution method in the Appendix (see note 6 above). Then you can insert material on delta-M, exact single scatter, number of streams (notes 4-5).

22520, L1-2. O3 is optically thin at 340 just as with NO2, not "negligible". Given the level of other uncertainties in this measurement modeling, it is likely that the ozone profile error is small and inclusion of this trace species would not affect results very much for the chosen window.

22520, L3. "little impact" on what? Please justify.

22520, L9. Eq. (4) is defined for irradiances, what about radiances? For the rest of this section, the symbol E is being used presumably just for irradiance. Does Eq. (5) apply also to irradiances only? More care is needed over the definitions, and the elastic/inelastic distinction should be preserved.

22521, L5. "The sky was relatively cloudless.." Cut this sentence, no added value.

22521, L17. Were these direct sun measurements taken by the same instrument?

22522, L6-9. Where did these assumptions about aerosols come from? How important are they?

22522, L23, at the center → near the center

22522, L24. "Furthermore". Cut this.

Section 3

"Heyney" should be "Henyey" (this section and elsewhere)

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Discussion Paper



22524, L17. "Data suggests..". Please rephrase this sentence. There are many sources of uncertainty here, and the best you can say is that these differences may in part be due to measurement uncertainties.

22524, L20. "exact Mie" → "Mie". see note 5 above.

22524. Why are the Mie tables "so-called"?

22524, L29. "Section 4" appears to be the wrong label here.

Section 4

See note 9 above. Remarks and discussions on results in Figures 6-8 are in general well-considered and probably relevant, but this reviewer is reluctant to comment further until the issues regarding Figures 4 and 5 have been resolved.

22527, L2 "after which it increases". Cut this (redundant)

22527, L20, and see note 9 above. This picture is not consistent with de Beek et al. results, and there is no explanation of the two-valued dependence on FI for TOA radiances.

22528, L11 "as a reflecting surface" → "as reflecting surfaces".

22528, L14. Need some more up-to-date references here that actually include OMI results.

22528, L17. "The LER and MLER.." This sentence has no added value (cut).

22529, L7-11. The sentence "Differences may be attributed..." is misleading. First, there could be any number of reasons for these differences, and unless there is good reason to mention one explanation over another, it is unwise to speculate. The reason suggested here is wrong anyway. It is clear from Siewert's (2000) and Spurr's (2002) papers that discrete ordinate particular integrals obtained either through exponential substitution or by Green's function methods are identical. Further, there is no

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such thing as a “standard discrete ordinate model”. Solving differential equations using Green’s functions is a “standard” procedure, and may be more general than the “qdisort” method mentioned in the Appendix.

22529, L26. “In Fig. 7 is shown the...” → Fig. 7 shows...”. It took me a little time to get oriented to these plots, it might be helpful to mention the solar and anti-solar principal planes. The caption in Figure 7 must be incorrect; the cloud should be between 2 and 3 km to make sense.

22530, L27. “Ground-based measurements” → “Ground-based retrievals”.

22531, L9-10. “Clearly, pseudo-spherical...” Cut this sentence (no added value).

22531, L13. “As such,” → “In this context,”

22531, L19-20. “nearly isotropic”. This is not a good description of these phase functions. Try “relatively smooth phase functions”. “..make variations in the radiance field small with..” → “ensure that the radiance fields show small variation with..”

22532, L12. “explains the spectral structures”. I think this is too strong and needs qualification, particularly in view of other sources of error as noted above and by other reviewers. Better to say “...helps to reproduce more accurately some of the spectral structures...”

22532, L16. Conclusions should refer to new work presented here. Bullets 1 and 3 (SZA and LER dependencies) are known already and should not appear here.

22532, L19, “levels” → “level”

Appendix A

22533, L22 - 22534, L2. Repetition of material in the introduction. Cut 3 sentences “Furthermoreincluded below”.

22535, L8. After noting the abundances, you should then add the sentence at the end

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of the Appendix ("A total of 233 Raman lines...."). In general, there is not enough detail on RR spectroscopy - at least, add an equation for the RRS cross-sections, and list the ingredients in this equation.

22535, L12-13. The symbols for phase function expansion coefficients are not explained. You could give their values for the Raman, Rayleigh and Henyey-Greenstein phase functions as useful examples.

22535, L15. "The transmission..." → "The solar beam transmission..."

Figures and Captions

It would be helpful where appropriate to indicate the viewing and solar angles used in the calculations, along with the surface albedo, number of discrete ordinates and use of delta-M.

22543. Fig 2 line 2. → "are shown in black".

22543. Fig 5 line 3. "Lambert" → "Lambertian". State the value used.

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