

The paper “Use of rotational Raman measurements in multiwavelength aerosol lidar for evaluation of particle backscattering and extinction” submitted by I. Veselovskii et al. describes one practical solution for isolating lines of pure rotational Raman spectra (PRRS) of atmospheric nitrogen and oxygen molecules and suggested by the authors for implementation in the lidars aimed at the remote sensing of aerosol scattering properties. The approach presented is shown as an alternative one to the vibrational Raman lidar method. The advantages of the solution proposed are given in the paper in a form of theoretical analysis and also proved by comparing the measurement results achieved with both techniques.

However, some more editing work could be still suggested. Thus, when comparing pure rotational and vibrational Raman lidar techniques the authors demonstrate some but not all pro and contra arguments of one approach and the other. For instance, the fact that blocking elastic scattering component from penetrating into molecular channel would be technologically more demanding in case of PRRS (due to a smaller spectral shift from the laser line), this is not mentioned in the paper explicitly. With this aspect missing one does not really draw a complete picture on the topic when reading the paper.

Also when giving a reference to other groups deriving information on atmospheric temperature from PRRS the authors did not mention those who made the first and fundamental steps in developing the technique rather talking about quite recent efforts that do not overwrite the previous work.

Discussing the specific temperature dependence of pure rotational Raman signal appearing in case when only few lines of spectra are accepted by the bandwidth of molecular channel, the authors have presented only two alternative solutions: either combining in one signal all the lines of the spectra or taking only those of them that when summed together manifest only a small temperature sensitivity. Thus the approach of many other groups (even cited in the paper) of isolating in separate channels the lines with opposite temperature sensitivity for atmospheric temperature profiling and combining those separate channels in one sum for aerosol measurements is completely ignored, even if those groups demonstrated some good results in deriving aerosol optical properties from PRRS.

The following list gives some more comments to these and few other points that could be recommended for revision.

1. The sentence given in the Introduction saying that “*First of all, cross section of vibrational Raman scattering is almost three orders lower comparing to Rayleigh one...*” is a bit misleading while the intensity ratio of these two lines is close to 1700 in case of excitation at 532 nm.
2. Few lines later in the Introduction it is mentioned that “*As compared with Raman nitrogen measurements at 387 nm, daytime skylight values are higher at 608 nm and the quantum efficiency of detectors is lower*”, in which the second statement regarding the quantum efficiency is at least questionable. In contrast to the detectors mentioned in the paper the module H7422-40 from Hamamatsu, for instance, demonstrate exactly the opposite properties.
3. Another statement to be clarified says “*Furthermore, the backscatter cross section of anti-Stokes component of pure RR nitrogen scattering exceeds that of vibrational scattering by approximately a factor of 15-20, when all lines for nitrogen and oxygen are integrated*” (page 2). It would be good to have a short extension to the statement explaining the reason for this span 15-20 to exist?
4. The key element of the paper is the solution proposed to solve the issue of the residual temperature dependence of pure rotational Raman signal that comes from the fact that not all the lines of the rotational spectra are accepted by the

spectral bandwidth of the channel. Due to its central position to the paper this element appears to be the most deficient point in the article as the list of the alternative solutions mentioned is not complete with only two possibilities discussed. The first option, to sum all the lines of PRRS, is rejected with the reason that *“measuring all Stokes and anti-Stokes lines with equal efficiency is technologically very difficult”*. This argument is a bit difficult to accept as the filtering technique required to implement this option could be, for instance, a “double chamber diffraction grating polychromator” similar to what is presented by Kim and Cha 2007 (the reference given in the manuscript itself). This option could be rejected as the least optimum due to some other reasons (like the spectral bandwidth of the channel would be too large), but technologically it is similar to what is developed by other groups. The second option, the *“... alternate approach considered in this paper is to select a portion of the RR spectrum characterized by a low temperature sensitivity inside one of the spectral branches”* is proposed as the only feasible solution. However, the third option of isolating in different spectral channels the lines of PRRS with opposite temperature sensitivity and summing these signals in one while applying appropriate factors for each channel is completely omitted. It could be shown, for example, that with four spectral channels built as it is described in “Recent Advances in Atmospheric Science” Ed.: L.Fiorani, V.Mitev, INOE, Bucharest, 2010 ISBN: 978-973-88109-6-9, (two channels in Stokes and two channels in Anti-Stokes branches of PRRS), the residual temperature sensitivity of the balanced sum of these four signals will be much smaller than it is with the solution proposed in the paper submitted (see the figure discussed below in the comment 23).

5. The following sentence given in the first paragraph of the section 2 *“... it was shown (Whiteman, 2003a, 2003b) that the temperature dependence of Raman scattering can be significant when considering measurements of Rayleigh-Mie and Raman lidar”* is not self-explaining. Which “Raman scattering” is meant here?
6. The description to the equation (2) claiming that the sigma *“... is molecular differential backscattering cross section, which includes Cabannes line together with vibrational and rotational Raman contributions”* would assume an unfeasible large spectral bandwidth of the molecular channel.
7. In page 6 *“In the simulations performed here the particle bimodal size distribution was represented as ...”*, it is not mentioned what is going to be simulated and what is the model taken.
8. There is no any comment given to explain the choice of the parameters selected for the particle size distribution. Why it is 0.1 μ and 1.2 μ for the two modes considered, and why the standard deviation is chosen that way?
9. Typo in page 6: *“find mode”* instead of *“fine mode”*
10. Page 7, the sentence *“As mentioned, the spectral dependence of A has the most influence for PSD with a predominant fine mode ...”* is not really clear. Should it be understood as *“...the spectral dependence of A is the most strong for...”*?
11. Page 7, *“The backscattering and extinction coefficients for the chosen size distribution were used to generate noise-free synthetic lidar signals.”* Were the molecular signals also simulated? This is not mentioned and the model used is not described.
12. In page 7, when reading that *“...the values of α and β were calculated using Ångström exponents...”* one can only guess the coefficients are calculated from the synthetic lidar signals. Also the inversion technique used is not specified.
13. The results presented in Table 1 for the Angstroem exponent are given with two significant digits behind the decimal dot, it would be much easier to understand

the relation between data in Table 1 and Table 2 if in Table 2 the data for the absolute deviation of the Angstroem exponent are given with the same format (two digits behind the dot).

14. Page 8, "...the uncertainty of extinction... doesn't exceed 2% for both wavelengths, when A355_532 is used", this is not obvious from the Table 2, some more explanation is required.
15. Page 8, "In contrast to extinction the uncertainties Eb depend on range since the differential transmission between the two Raman wavelengths is involved..." Should it be the laser and the Raman wavelengths instead of "the two Raman wavelengths"?
16. Page 8, "The uncertainty is negligible near the reference height ($z_0=4000$ m) and it rises with increasing $z-z_0$ ". Correctly speaking only the absolute difference of the $z-z_0$ should increase.
17. Page 8, when discussing the biases introduced to the backscatter coefficient due to improper choice of the Angstroem exponent, "For 532 nm these uncertainties are 4% and 6%, which is significantly lower than at 355 nm", it would be more informative and concrete if replacing the definition "significantly" by "approximately by factor of two".
18. Page 8, the sentence "an increase of ΔA to $\Delta A = 1$ doubles the uncertainty" to be complete should be extended probably as "an increase of ΔA from 0.5 to $\Delta A = 1$ doubles the uncertainty".
19. Page 9, when providing a reference to the equation (12) as "Penney et al., 1974; Behrendt and Nakamura, 2002; Adam, 2009" it would be more appropriate to refer to the originators of the theory rather than pointing to the quite recent papers of Behrendt and Nakamura, and Adam.
20. Page 9, when introducing the equation (13) as "The factors $b(J)_{s,as}$ for Stokes and anti-Stokes lines are approximately:" what is meant by the "approximately"?
21. Page 10, there are few points to comment in the sentence "The RR line with least temperature sensitivity is in the vicinity 530.4 nm ($J=9$) for O₂ and in vicinity of 530.2 nm ($J=7$) for N₂...":
 - a. It is the line number 9 with the least temperature sensitivity in Anti-Stokes branch of PRRS of O₂, i.e. the quantum number representing the rotational state J is equal to 11.
 - b. And it is the line number 7 (but the rotational quantum number $J=9$) in case of the Anti-Stokes branch of PRRS of N₂.
 - c. When indicating the wavelength of the least temperature sensitive lines it is good to remind that they are given for the case when the Raman spectra are stimulated at 532.12 nm.
22. Page 10, it is specified that "Computations were performed with T varying in the range 230-300K..." A bit wider interval, 200-310K, would be more appropriate to consider if aiming to upgrade the tropospheric lidars.
23. This is to extend the comment 4 involving the "X-function" category introduced in page 10 of the paper submitted. The measurement distortion presented in Fig.4 is recomputed for the PRRS channel configuration proposed in the paper and also for another configuration that would represent the third possible solution described above in the comment 4, see the Figure (A) below. The green line in Fig.(A) is the exact copy of the green line from the Fig.4 of the paper submitted (calculated assuming 100% filter transmission for the interval of 529.2-531.2 nm and zero transmission otherwise). The red line in Fig.(A) is calculated for the PRRS channel configured as it is described in L. Fiorani and

V. Mitev “Recent Advances in Atmospheric Science”. Fig.(B) adopted from this paper illustrates the configuration.

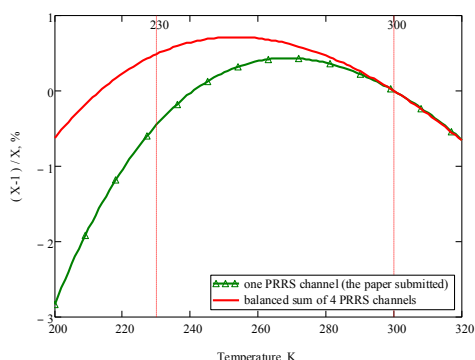


Fig.(A)

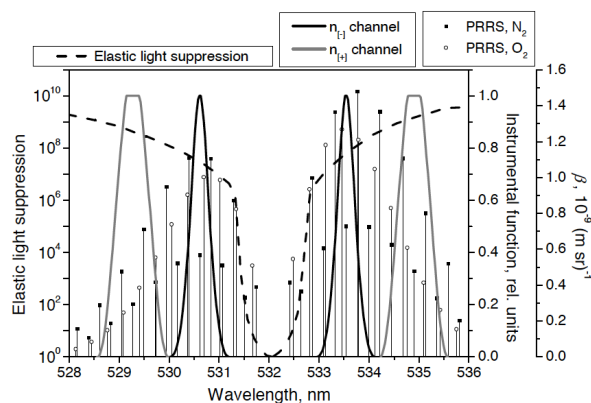


Fig.(B)

The smaller temperature sensitivity of the “X-function” achieved with the balanced sum of 4 PRRS channels is due to the absolute freedom in selecting the balancing factors. Similar performance should be also expected with the two-channel configuration developed, for instance, by Jens Reichardt for RAMSES lidar, or presented by Radlach et al. 2008 (this paper is listed in the references of the manuscript submitted).

24. Page 11, the approximation given in the sentence “...we estimate that the total RR cross section, including both Stokes and anti-Stokes branches of N₂ and O₂, exceeds that of vibrational nitrogen by approximately a factor of 40” slightly overestimates the effect, the more accurate number is expected to be about 38.
25. Page 12, in the sentence “We therefore estimate that the scattered power in the RR channel is a factor of 15 higher than that of vibrational nitrogen Raman channel at 608 nm (using a filter transmission of 70% for the vibrational channel)” the factor of 15 seems to be underestimated. It should be about 18 if assuming the “factor of 40” above.
26. Page 12, reading the sentence “The spectral components at 608 nm and 532 nm were separated by a dichroic mirror DM with approximately 95% being reflected to 608 nm channel” one can definitely guess that it is the 95% of the light intensity at the 608 nm, which is reflected to the 608 nm channel. Still it would be good to express this fact more explicitly.
27. Page 13, “This figure clearly demonstrates the improvement of the extinction calculation when the RR signal at 530 nm is used instead of that of vibrational scattering at 608 nm.” Error bars plotted in the figure would help to see this improvement clearly.
28. Page 13, it is not clearly communicated what is meant by “This” in the sentence “This has been one of the advantages of the technologically more complex approach of High Spectral Resolution Lidar (HSRL) to the task of measuring aerosols with a sufficient number of wavelengths to support microphysical inversions.”
29. Page 14, a typo in the either-or construction in the sentence “either anti-Stokes of Stokes components”.
30. Fig.3, a dedicated axis should be plotted for the PRRS lines.

In general the paper produces a good impression and with the appropriate editing work invested to address the comments given could be recommended for publishing.