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How Well Do We Know Rayleigh Cross Sections?

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Abstract

Rayleigh scattering cross section in air are needed for the transmission and light scattering corrections to air fluorescence data. The purpose of this note is to provide a bibliography of references on Rayleigh scattering and to summarize the current knowledge of the Rayleigh cross section in air.

1 Introduction

Air fluorescence detectors are sensitive to light in the wavelength range 300nm $\leq \lambda \leq$ 420nm. In this range Rayleigh scattering typically dominates the scattering corrections to the fluorescence data. The point of this note is to provide a list of reference to the Rayleigh cross sections, and based on these papers to present the best value, with uncertainties, for the Rayleigh cross section in air.

In papers published in 1871 [1] and 1899 [2] Lord Rayleigh developed a theory to describe light scattering by ideal gases. A good modern treatment is given by Chandrasekhar [3]. For air an important modification is the allowance for anisotropic scattering (by the air molecules). As a consequence the Rayleigh cross section is given (Eqn 256 of [3]) by:

$$\sigma = \frac{8\pi^3}{3} \cdot \frac{(n^2-1)^2}{N^2\lambda^4} \cdot \frac{3(2+\rho_m)}{6-7\rho_m} \tag{1}$$

where $n(\lambda)$ is the index of refraction of air, λ is the wavelength of light, N is the number of scatterers per unit volume, and ρ_m is the *depolarization factor*. $\rho_m = 0$ for isotropic scattering. Throughout the second half of the last century, the depolarization factor, ρ_m, has been included in equations dealing with Rayleigh scattering. An important issue is the correct value for ρ_m. Suggestions for this factor cover the range $0.028 \leq \rho_m \leq 0.05$. Historically Bunner [4] (Eqn 2.65 of [4]) [1] referenced Baum and Dunkelman [5] (Eqn 2a of [5]) which evaluated the Rayleigh cross section using $\rho_m = 0.04$. In contrast the recent study of Bucholtz [6] favors the value $\rho_m \approx 0.03$. Although ρ_m may vary slowly with wavelength [6] this variation is less than the $\sim \pm 0.01$ current uncertainty in ρ_m.

The normalized Rayleigh differential scattering cross section, or Rayleigh phase function, is given (Eqn 255 of [3]) by:

$$\frac{1}{\sigma}\left(\frac{d\sigma}{d\Omega}\right) = \frac{3}{16\pi(1+2\gamma)} \cdot \Big((1+3\gamma) + (1-\gamma)cos^2(\theta) \Big) \tag{2}$$

where $\gamma = \frac{\rho_m}{2-\rho_m}$. For isotropic scattering $\gamma = 0$ and for small values of ρ_m then $\gamma \approx \rho_m/2$. For isotropic scatterers, the components of the electric field (in, normal) to the scattering plane scatter with relative amplitudes ($cos^2(\theta)$, 1) respectively. Thus Rayleigh scattered light at $\theta = \pi/2$ is fully polarized. This is no longer true for anisotropic scatterers with relative intensities (Eqn 252 of [3]): $(2\gamma + (1-\gamma)cos^2(\theta), 1+\gamma))$ respectively. At $\theta = \pi/2$ the ratio of terms is $2\gamma/1+\gamma \equiv \rho_m$ explaining the name *depolarization factor* for ρ_m.

In summary values of $\rho_m \neq 0$ result in a change in both the Rayleigh (total) cross section and in the Rayleigh phase function. Values of $\rho_m > 0$ result in an increase in the Rayleigh scattering cross section; details are provided below.

[1] Bunner's discussion following his Eqn 2.65 is slightly incorrect as the index of refraction should be evaluated as $n(\lambda)$ and not at a fixed wavelength of 400nm.

The variation of the index of refraction of air with wavelength is also of some (but lesser) importance..

Finally it is useful to note a variety of notation and terminology difference that appear in the literature. These include:

- the King factor [7], F_K:

$$F_K = \frac{6 + 3\rho_m}{6 - 7\rho_m}$$

  to replace the equivalent longer expression (explicitly written with the depolarization factor, ρ_m) *e.g.* in Eqn [1];

- the "γ"-factor also in place of the depolarization factor, ρ_m as *e.g.* in Eqn [2];

- and for air, where the index of refraction $n \approx 1$, for practical purposes:

$$(n-1)^2 \;\approx\; \frac{(n^2-1)^2}{4}$$

2 Current Status and Comparison with Earlier Results

Currently the best paper on Rayleigh scattering in air appears to be the work of Bucholtz [6]. Bucholtz work was confirmed by Breon [8]. Furthermore Naus and Ubachs [9] experimentally confirmed Bucholtz's findings with an accuracy of 1% for Ar and N_2, and of 3% for SF_6. We were unable to find any experimental verifications of Rayleigh predictions for air!

All recent papers start from Eqn [1]. The differences come from small differences in values used for $n(\lambda)$ and N, and from effectively larger differences in values used for $\rho_m(\lambda)$. Bucholtz [6] provides both tabular results as well as an analytic (fit) to the Rayleigh cross section in the range 200nm$\leq \lambda \leq$500nm:

$$\sigma(\lambda) \;=\; A \cdot \lambda^{-(B + C\lambda + D/\lambda)}$$

with coefficients: A = 3.01577×10^{-28} cm^2, B = 3.55212, C= 1.35579 μm^{-1} and D=0.11563 μm and the wavelength is in μm. The differences between the Bucholtz's tabular data and the analytic formula are $< 0.2\%$ for 200nm$\leq \lambda \leq$500nm.

For corrections to fluorescence data it is more straight forward to express the Rayleigh cross section as an attenuation length, $\Lambda(\lambda)$:

$$\Lambda \;=\; \frac{Molecular\ Weight}{\sigma N_0}$$

where the molecular weight of air is 28.95 gm/mole and N_0 is Avogadro's number. To check with Bucholtz's paper, we also evaluate the Rayleigh optical depth, $OD(\lambda)$, of the atmosphere:

$$OD(\lambda) = \frac{Total\ mass}{\Lambda(\lambda)} = \frac{Pressure}{\Lambda(\lambda)\cdot g}$$

where the ground level pressure is 1013 mbar and the acceleration of gravity $g = 9.81$ m/s^2. The Rayleigh attenuation length and atmospheric optical depth are recorded in Table 1; the Rayleigh attenuation length is plotted in Fig. 1.

Table 1: Rayleigh attenuation lengths and atmospheric optical depth *versus* wavelength. For comparison Bucholtz [6] gives OD(400nm) = 0.3606 or a difference of 0.3% from this Table value.

Wavelength $\lambda(\mu m)$	Attenuation Length Λ (g/cm^2)	Optical Depth OD
0.28	631.39	1.6357
0.29	735.75	1.4037
0.30	851.92	1.2123
0.31	980.50	1.0533
0.32	1123.48	0.9193
0.33	1281.28	0.8060
0.34	1453.70	0.7104
0.35	1644.11	0.6282
0.36	1850.41	0.5581
0.37	2074.83	0.4978
0.38	2321.28	0.4449
0.39	2587.39	0.3992
0.40	2873.50	0.3594
0.41	3185.80	0.3242
0.42	3519.31	0.2935

Before comparing Bucholtz's [6] results to other papers it may be of interest to summarize the historical values for the depolarization parameter, ρ_m. Baum and Dunkelman [5] mention that according to Born the depolarization parameter is 0.04 throughout the optical spectrum. Tousey and Hulbert [8] note that Rayleigh worked with $\rho_m = 0.042$ or $\rho_m = 0.05$, Cabannes with $\rho_m = 0.041$, Raman with $\rho_m = 0.0437$ and Rao with $\rho_m = 0.0415$. In the end Tousey and Hulbert [8] used $\rho_m = 0.04$. Teillet [9] mentions in his 1990 paper that Penndorf [10] in 1957 and Elterman [11] in 1968 use a depolarization parameter $\rho_m = 0.035$. Furthermore Teillet writes that Young

[12] published a series of papers treating this issue and finally recommended a value of $\rho_m = 0.0279$. Bucholtz [6] uses values between $\rho_m = 0.0318$ at 300nm to $\rho_m = 0.0284$ at 500nm.

Since there is little agreement concerning the exact value for the depolarization parameter for air it is useful to look at the consequences. This was done by varying ρ_m in Eqn 2 of Baum and Dunkelman [5] (or equivalently Tousey and Hulbert [8] ... or for that matter Eqn [1] above). For completeness this is reported here:

$$\sigma \;=\; \frac{8\pi^3}{3} \cdot \frac{(n-1)^2}{N\lambda^4} \cdot \frac{6(1+\rho_m)}{6-7\rho_m} \cdot \Big[3 + \frac{1-\rho_m}{1+\rho_m}\Big] \cdot 10^5 \quad km^{-1} \tag{3}$$

Actual values used included: $N = 2.689 \times 10^{19}$ air molecules/cc at STP, $(n-1) = 0.0002926$ for air at 589nm and STP, the wavelength dependence of the index of refraction of air from Handbook of Chemistry and Physics and the wavelength is in cm. With a depolarization parameter $\rho_m = 0.04$ input to Eqn [3] the result was:

$$\sigma \;=\; (1.316 \times 10^{-12}) \cdot \frac{(n-1)^2}{\lambda^4} \quad km^{-1}$$

at STP in agreement with Eqn 2a of Baum and Dunkelman [5]. The results for depolarization parameter $\rho_m = 0.04$ and for depolarization parameter $\rho_m = 0.03$ (reported as Rayleigh attenuation lengths, $\Lambda(\lambda)$ are compared with Bucholtz in Fig. 2.

As the differences are not large, the fractional relative differences: $(\Lambda_{BD} - \Lambda_B)/\Lambda_B$ are plotted (again) in Fig. 3. We use Λ_{BD} for the Baum and Dunkelman attenuation lengths evaluated using Eqn [3] with different values of ρ_m. Λ_B is the Bucholtz attenuation length (see Table 1). The slope (*i.e.* wavelength dependence) in the curves is related to the increased wavelength dependence in the Rayleigh cross section in Bucholtz [6] in comparison to Baum and Dunkelman [5]. In Baum and Dunkelman [5] the wavelength dependence is (only) in the factor $(n(\lambda) - 1)^2$ and not also in ρ_m. The vertical offset of these curves comes from the range of (recent) values for ρ_m in the literature.

The best agreement between Baum and Dunkelman [5] and Bucholtz [6] would be achieved for a value of ρ_m somewhat larger than 0.03. So $\rho_m \approx 0.032$ would be a good match at an average wavelength of 360nm. The actual value for $\rho_m(360nm)$ used by Bucholtz [6] was 0.030. As the literature does not limit the value of ρ_m to better than $\sim \pm 0.01$ the resulting uncertainty in the Rayleigh attenuation lengths are $\sim 1.5\%$. If we take Bucholtz [6] as the best estimator, then the Rayleigh attenuation length at 400nm is 2874 gm/cm^2, Table 1, in comparison to a typical value of 2974 gm/cm^2 used in recent HiRes analyses [13]. This corresponds to a 3.3% (systematic) shift from previously used values and would suggest that the Rayleigh correction in previous analyses was somewhat too small.

3 Implications for the Rayleigh *Phase Function*

The phase function describes the angular distribution of the Rayleigh scattered light; see Eqn [2] and discussion in Sect. 1 of this note. The γ-factor in Eqn [2] is approximately $\gamma = \rho_m/2$; thus for a depolarization factor of $\rho_m = 0.03$ $\gamma \approx 0.015$. The wavelength dependent values used by Bucholtz [6] are given in Table 2.

Table 2: γ-factor (input to Eqn [2]) variation with wavelength from Bucholtz [6].

Wavelength $\lambda(\mu\text{m})$	γ-factor
0.28	0.01672
0.29	0.01643
0.30	0.01614
0.31	0.01614
0.32	0.01586
0.33	0.01557
0.34	0.01557
0.35	0.01528
0.36	0.01528
0.37	0.01528
0.38	0.01499
0.39	0.01499
0.40	0.01499
0.41	0.01493
0.42	0.01488
0.45	0.01471

The relative fractional difference, $(PF_\gamma - PF_0)/PF_0$, is shown *versus* scattering angle in Fig. 4. By PF_γ we imply Eqn [2] evaluated with a non-zero value for γ. By PF_0 we imply Eqn [2] evaluated with $\gamma = 0$. The fractional changes are $\leq 1.5\%$ from the approximate $1 + cos^2(\theta)$ form for Rayleigh scattering.

4 Summary

Papers on Rayleigh scattering were reviewed. The recent paper of Bucholtz [6] argues for a systematic $\sim 3\%$ increase in the Rayleigh cross section in comparisons to values used until recently by the HiRes collaboration. The change in the Rayleigh phase function (from $1 + cos^(\theta))$ is $\pm 1.5\%$ and is angle dependent. Simple functional forms are provided for evaluating the Rayleigh attenuation length *versus* wavelength and the Rayleigh phase function *versus* wavelength and scattering angle.

Acknowledgments

We want to recognize that Michael Roberts encouraged this study as well as providing us with a stack of papers on Rayleigh scattering. We also want to note that Stan Thomas (HiRes) has independently come to similar conclusions.

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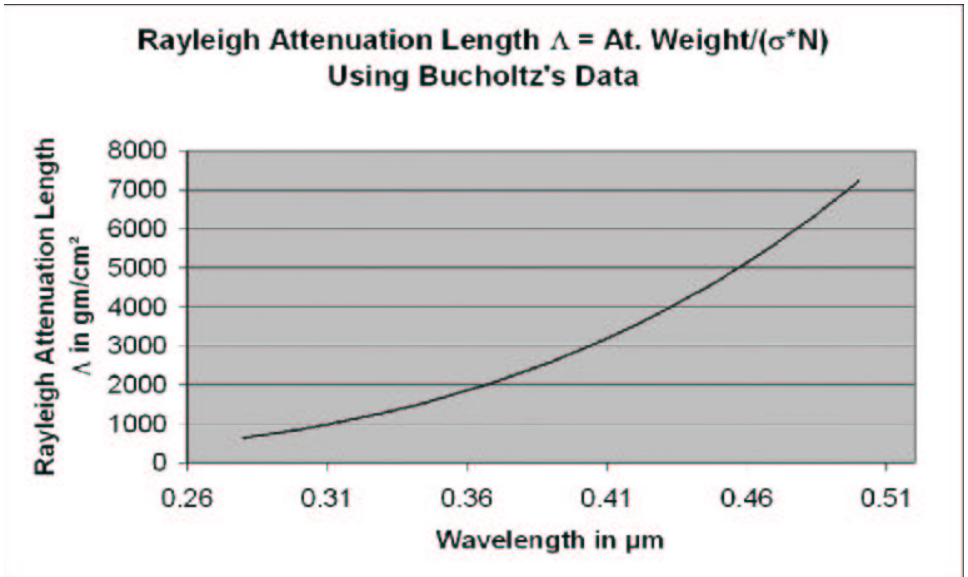

Fig. 1: Rayleigh attenuation length, $\Lambda(\lambda)$, *versus* wavelength following Bucholtz [6] (see also Table 1).

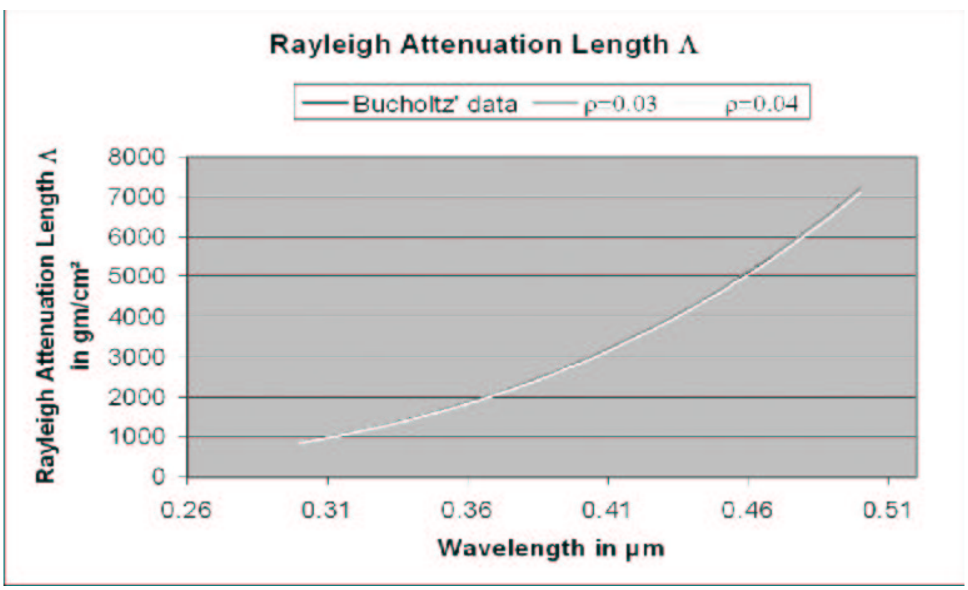

Fig. 2: Rayleigh attenuation length, $\Lambda(\lambda)$, *versus* wavelength from Baum and Dunkelman [5] compared to Bucholtz [6].

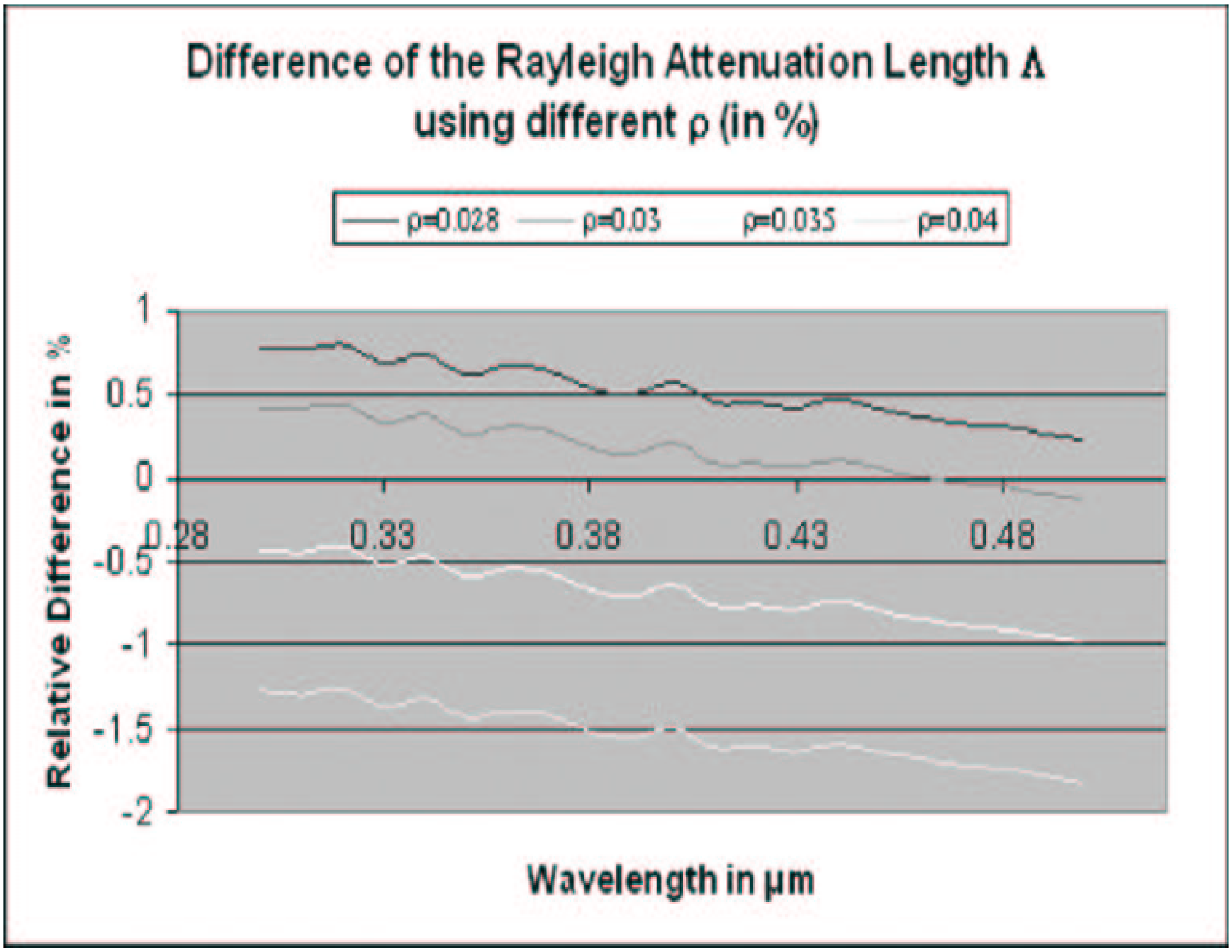

Fig. 3: Fractional relative differences (in %) between attenuation lengths based on Baum and Dunkelman [5] *versus* Bucholtz [6] for 4-different values of the depolarization parameter input to Eqn [3] (from Baum and Dunkelman [5]).

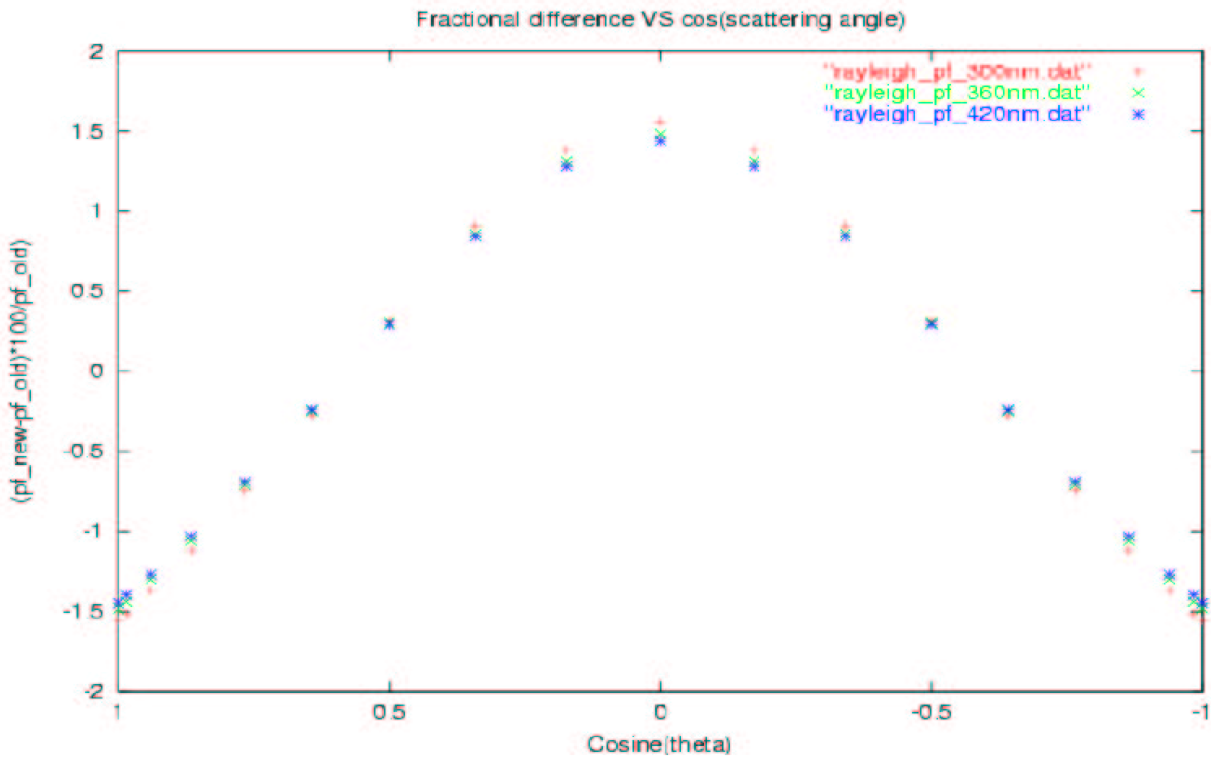

Fig. 4: Fractional relative differences (in %) between the Rayleigh phase function evaluated using $0.01614 \leq \gamma \leq 0.01488$ *versus* the phase function evaluated with $\gamma = 0$. The values of γ are from Table 2 for wavelengths between 300nm and 420nm.