Measurement of atmospheric aerosol extinction profiles with a Raman lidar

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A method is presented that permits the determination of atmospheric aerosol extinction profiles from measured Raman lidar signals. No critical input parameters are needed, which could cause large uncertainties of the solution, as is the case in the Klett method for the inversion of elastic lidar returns.

The remote determination of aerosol concentration profiles has important applications in areas such as heterogeneous atmospheric chemistry, weather and climate research, and pollution monitoring. Until now single- or multiple-wavelength backscatter lidars have been used for this purpose. The appeal of these devices is their conceptional simplicity, ease of operation, and high speed.

The basic formalism used to determine the aerosol extinction from elastic backscatter signals is the so-called Klett inversion method.[1-3] This procedure, with all its subsequent modifications and improvements, suffers from the fact that two physical quantities, the aerosol backscatter and extinction coefficients, must be determined from only one measured lidar signal. This is not possible without assumptions about the relation between the two and an estimate of a boundary or reference value of the aerosol extinction. These data are usually hard to assess and cause large uncertainties in the aerosol extinction coefficients that are determined.

In Raman lidar, the inelastic (Raman) backscatter signal is affected by aerosol extinction but not by aerosol backscatter. Therefore analysis of the Raman lidar signal alone permits the determination of the aerosol extinction. There have been several descriptions of Raman lidar applied to aerosol backscatter, visual range, and optical depth,[4-7] but until now a general formalism for determining range-resolved aerosol extinction coefficients from Raman lidar signals has not been given.

Let the Raman lidar equation be written as

$$P(z, \lambda_L, \lambda_R) = \frac{BO(z)}{z^2}\beta(z, \lambda_L, \lambda_R) \times \exp\left\{-\int_0^z [\alpha(\lambda_L, \zeta) + \alpha(\lambda_R, \zeta)]\mathrm{d}\zeta\right\}. \quad (1)$$

Here P is the power received from distance z at the Raman wavelength λ_R if the laser pulse is transmitted at λ_L, $O(z)$ is the overlap function between the laser beam and the field of view of the receiver, α is the depth-dependent total extinction coefficient at wavelengths λ_L and λ_R, and B contains all depth-independent parameters. The backscatter coefficient β is linked to the differential Raman backscatter cross section $\mathrm{d}\sigma/\mathrm{d}\Omega$ of a gas of molecule number density N by the relation

$$\beta(z, \lambda_L, \lambda_R) = N(z)\frac{\mathrm{d}\sigma(\lambda_L, \lambda_R, \pi)}{\mathrm{d}\Omega}. \quad (2)$$

The profiles of both nitrogen and oxygen can be used since their number densities are usually well known. With $P(z) \equiv P(z, \lambda_L, \lambda_R)$, it follows from Eqs. (1) and (2) that

$$\alpha(\lambda_L, z) + \alpha(\lambda_R, z) = \frac{\mathrm{d}}{\mathrm{d}z}\left[\ln\frac{O(z)N(z)}{z^2P(z)}\right]. \quad (3)$$

$O(z)$ is equal to unity if the path of the transmitted laser beam is entirely within the field of view of the receiver, which is the case above a certain minimum height $z_{\min}$. The two extinction coefficients in Eq. (3) may be written as

$$\alpha(\lambda_{L,R}, z) \equiv s_{\mathrm{mol}}(\lambda_{L,R}, z) + s_{\mathrm{aer}}(\lambda_{L,R}, z), \quad (4)$$

where s_{mol} and s_{aer} are the extinction coefficients due to absorption and Rayleigh scattering by atmospheric gases and aerosol scattering and absorption, respectively. Equation (3) can then be written as

$$s_{\mathrm{aer}}(\lambda_L, z) + s_{\mathrm{aer}}(\lambda_R, z) = \frac{\mathrm{d}}{\mathrm{d}z}\left[\ln\frac{N(z)}{z^2P(z)}\right] - s_{\mathrm{mol}}(\lambda_L, z) - s_{\mathrm{mol}}(\lambda_R, z). \quad (5)$$

Assuming a wavelength dependence of the aerosol extinction such as

$$\frac{s_{\mathrm{aer}}(\lambda_L)}{s_{\mathrm{aer}}(\lambda_R)} = \frac{\lambda_R}{\lambda_L}, \quad (6)$$

we obtain from Eqs. (5) and (6) the unknown extinction coefficient profile

$$s_{\mathrm{aer}}(\lambda_L, z) = \frac{\frac{\mathrm{d}}{\mathrm{d}z}\left[\ln\frac{N(z)}{z^2P(z)}\right] - s_{\mathrm{mol}}(\lambda_L, z) - s_{\mathrm{mol}}(\lambda_R, z)}{1 + \frac{\lambda_L}{\lambda_R}}. \quad (7)$$

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This result holds for altitudes $z > z_{min}$ for which the Raman lidar overlap function $O(z) \equiv 1$.

For an estimate of $d/dz[\ln N(z)]$ and $s_{mol}(\lambda_{L,R})$, the air density profile must be known. Height profiles of the atmospheric density can be assessed by using one of several models available (such as the U.S. Standard Atmosphere) with measured values of the ground temperature and pressure, or they can be directly measured with radiosondes. Light extinction by ozone can also be taken into account.[8,9]

The theory developed has successfully been applied to profiles obtained with the GKSS Raman lidar. Since this system has been described in detail,[10] only the more important features are repeated here. The radiation source is a XeCl excimer laser, the transmitter and receiver are positioned side by side with the planes of all curved mirrors horizontal, and the polychromator is a 17th-order echelle-type device with a holographic grating for cross dispersion. The technical data of the system are given in Table 1.

Figure 1 shows an example of an aerosol extinction coefficient profile obtained with the Raman lidar using the procedure outlined above. The statistical error of the measurement, i.e., the error contribution from lidar signal noise, is given in the right-hand trace of Fig. 1. The number of laser shots (234,142) was obtained in 19 min of measurement time. Before Eq. (7) was solved, the profile of the range-corrected signal $P(z)z^2$ taken with 60-m measurement resolution was smoothed by forming a sliding average over 360 and 1440 m for heights $z < 3700$ m and $z > 3700$ m, respectively. s_{aer} was then calculated with the same effective resolution. This must be kept in mind for the interpretation of the measured profile.

In addition to the statistical error as plotted in Fig. 1, systematic errors must also be considered. The contribution from the molecular extinction uncertainty amounts to ≤ 0.01 km^{-1} if the ozone concentration deviates by no more than a factor of 3 from the standard ozone profile and the air density deviates by no more than 5% from the standard atmosphere; this corresponds to errors in the estimated value of temperature and pressure of $\delta T \leq 10$ K and $\delta p \leq 1$ kPa. The

Table 1. GKSS Raman Lidar Technical Data

Transmitter	
Primary wavelength	308 nm
Maximum pulse energy	270 mJ
Maximum repetition rate	250 Hz
Optics	400 mm, $f/3.75$
Receiver	
Optics	800 mm, $f/3.75$
Detectors	THORN EMI 9893 QB 350
Polychromator	
Type	Czerny–Turner
Main dispersion grating	Echelle, 316 grooves/mm
Cross dispersion grating	Holographic, 2400 grooves/mm
Data acquisition	
Type	Photon counting
Wavelength channels	3
Time channels	1024
Minimum time-bin width	100 nsec
Maximum count rate	300 MHz

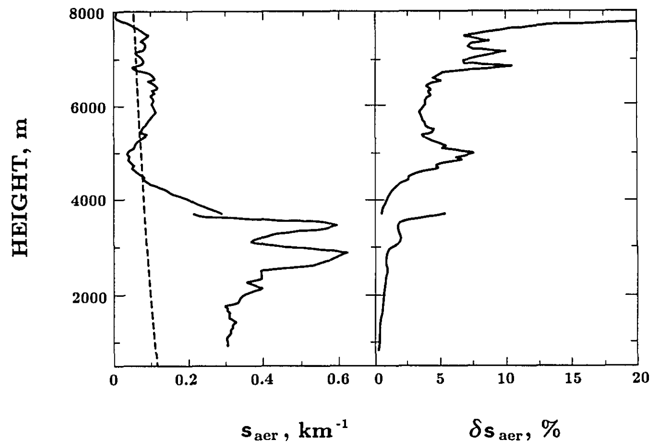

Fig. 1. Atmospheric aerosol extinction coefficient s_{aer} and statistical error δs_{aer} measured with a Raman lidar at 308-nm primary and 323-nm Raman (O_2) wavelengths. The measurement was taken on May 17, 1988, at 00:12:01 hours local time in Geesthacht, Federal Republic of Germany. Measurement time is 19 min, and the total number of laser shots is 234,142. Ground values for the standard atmosphere model are 100.8 kPa and 15°C. The discontinuity at 3700 m reflects the change from 360 to 1440 m of averaging height at this level. The molecular extinction coefficient s_{mol} is also shown for comparison (the dashed curve).

term $d/dz[\ln N(z)]$ can contribute significantly to the error of s_{aer} when standard atmosphere conditions are assumed but a temperature inversion is present. A temperature gradient dT/dz between 0 and 3 K/100 m leads to an error δs_{aer} of between 0.015 and 0.025 km^{-1}. Temperature inversions are mainly observed in the lower troposphere, especially at the top of the boundary layer. The error due to non-λ^{-1} behavior of the aerosol extinction amounts to ±1% if the right-hand side of Eq. (6) is raised to the power k and k is allowed to vary between 0.8 and 1.2 Error considerations thus show that, owing to the relatively large sensitivity of the aerosol extinction coefficient at 308 nm to errors in the air density estimate, aerosol measurements in clean, near-Rayleigh atmospheres are only possible in combination with air density determinations as can be obtained from radiosonde measurements of temperature and pressure.

The advantage of the Raman method over the Klett method lies in the closed-form solution of the former as opposed to the recursive formula of the latter. Catastrophic instabilities as are common in the Klett method cannot occur in the Raman approach; the propagation of errors of input parameters into the aerosol extinction coefficient is straightforward; and the nature of the input parameters is such that a measurement, and thus a small error in s_{aer}, appears feasible.

In conclusion, it has been demonstrated that Raman lidar measurements provide aerosol extinction data with adequate accuracy and spatial and temporal resolution. The main shortcoming of the Raman method is the greater complexity of the apparatus. The limitation of the present GKSS system to nighttime measurements, caused by the choice of the 308-nm radiation, can be alleviated by switching to shorter wavelengths, although increased ozone absorption and larger effects of air density uncertainties must be carefully investigated.

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