TOTAL SCATTER-TO-BACKSCATTER RATIO OF AEROSOL DERIVED FROM AEROSOL SIZE DISTRIBUTION MEASUREMENT

W. N. Chen, S. Y. Chang, Charles C.K. Chou, T. K. Chen

Research Center for Environmental Changes, Academia Sinica, Taipei 115, Taiwan, wnchen@rcec.sinica.edu.tw

ABSTRACT

Based on in-situ aerosol size-distribution measurements and Mie scattering theory, total scattering coefficients and backscattering coefficients were calculated to derived wavelength dependent Lidar ratio *S* for 355 nm and 532 nm. Effective radius and volume C/F ratio (coarse/fine) of aerosol are also calculated to study the relationships between Lidar ratio and particle size dependences. The results show backscatter-related scattering properties are more sensitive to coarse mode particle than total scattering. The mean values of Lidar ratio for 355 nm and 532 nm are 31.9±6.2 sr and 40.5±6.1 sr respectively, and S_{355} and S_{532} are linear correlated for S_{355}<50 sr. S_{355} is highly correlated with effective radius of aerosol, and S_{532} is highly correlated with volume C/F ratio.

1. INTRODUCTION

Light scattering by aerosols plays an important role in problem of visibility and also atmospheric radiation balance. Lidar is a powerful remote-sensing tool to obtain vertical profile of atmosphere by considering aerosol scattering as tracer of atmospheric motions and pollution sources. The aerosol extinction (total scattering and absorption) coefficient to backscattering coefficient ratio *S* (or so called Lidar Ratio) is dependent on chemical composition and size distribution but independent on concentration, therefore, Lidar ratio was treated as indicator of the characteristic of aerosols. The value of Lidar ratio is varied with characteristic of aerosols. Lidar or in-situ measurements and computer simulations show Lidar ratio for sea salt, Asian dust, Saharan dust, biomass burning, and urban aerosols are 20-30 sr, 40-60 sr, 30-40 sr, 45-65 sr, 20-80 sr respectively.

For Rayleigh/Mie Lidar inversion algorithm [1], Lidar ratio is the most important unknown factor to determine extinction and backscatter of aerosol or cloud [2] and may cause unexpected uncertainty on Lidar profiles. Moreover, a known Lidar ratio is also necessary to correct the overlapping problem caused by incomplete overlap between laser beam and field of view of telescope [3-5]. Therefore, a Lidar-independent method to obtain Lidar ratio is needed to validate Lidar observations and to improve the accuracy of Lidar measurements.

Based on aerosol size distribution measurement during March-May 2005 at Taipei basin, Taiwan, the light scattering properties of aerosol such as total scattering coefficient, backscattering coefficient, and Lidar ratio were calculated by Mie scattering theory. The effective radius and volume C/F ratio ((PM10-PM2.5)/PM2.5) of aerosol are selected to study the relationship between size distribution and Lidar ratio. The wavelengths of 355 nm and 532 nm of Nd:YAG laser was selected as basis of calculation, because 355 nm and 532 nm are commonly used as laser source of Lidar. That implies measured S_{355} or S_{532} could be applied to estimate effective radius and C/F ratio, and vice versa.

2. EXPERIMENTS

The aerosol particle size distribution experiment took place in an urban area inside Taipei Basin. The period investigated in this paper is March-May 2005. The Taipei Basin, located in the north of Taiwan, has high densities of population, vehicles and motorcycles. The sampling site (25°00'N, 121°32'E) positioned at the weather observatory of the National Taiwan University located in the southwestern part of the Taipei Basin.

The particle number size distribution was obtained with a combination of scanning mobility particle sizer (SMPS) and aerodynamic particle sizer (APS) to cover a range of particle size from 0.02 μm to 20 μm. The total scattering coefficient was measured by a nephelometer (TSI 3565). To avoid the cut-off characteristics of impactor change the sampling progresses, no impactor was used in front of the SMPS/APS systems and nephelometer [6].

Typically, PBL aerosol is combination of three modes correspond to Aitken nuclei, accumulation mode aerosols, and coarse aerosols. The shape of which is often modeled as the sum of lognormal modes. Each lognormal mode is defined by a total number concentration N_{0i}, a median radius r_{0i}, and a geometrical width *s*.

$$n(r) = \frac{N_0}{\sqrt{2\pi} r \ln s} \exp\left(-\frac{(\ln r - \ln r_0)^2}{2(\ln s)^2}\right) = \sum_i \frac{N_{0i}}{\sqrt{2\pi} r \ln s_i} \exp\left(-\frac{(\ln r - \ln r_{0i})^2}{2(\ln s_i)^2}\right) \quad (1)$$

The effective radius r_e and equivalent variance v_e had the following relationship with lognormal distribution:

$$r_e = \frac{\int r^3 n(r)dr}{\int r^2 n(r)dr} = r_0 \exp[2.5(\ln s)^2] \quad (2)$$

$$v_e = \frac{\int (r - r_e)^2 r^2 n(r)dr}{r_e^2 \int r^2 n(r)dr} = \exp[(\ln s)^2] - 1 \quad (3)$$

Since effective radius may not very sensitive to number concentration of coarse mode particles, a C/F ratio is define as ratio of the volume of total coarse mode particle (diameter>2.5 μm) and the volume of total fine mode particles (diameter<2.5 μm).

Mie scatter theory is used to compute the total scatter efficiency (Q_{sca}) and backscatter (Q_{back}) efficiency at the wavelength λ at 355 nm and 532 for single particles as functions of the radius r. The total scatter coefficient (σ_λ) and backscattering coefficient (β_λ) is defined as the follows.

$$\sigma_\lambda = \int 2\pi r^2 Q_{sca}(\lambda, r) n(r) dr \quad (4)$$

$$\beta_\lambda = \int 2\pi r^2 Q_{back}(\lambda, r) n(r) dr \quad (5)$$

The total scatter-to-backscatter ratio (Lidar Ratio) is defined as

$$S(\lambda) = \frac{\sigma(\lambda)}{\beta(\lambda)} \quad (6)$$

Fig. 1 demonstrates a sample Q_{sca} and S (Q_{sca}/Q_{back}) for 355 nm and 532 nm. Please note Q_{back} (not shown in figure) was smoothed before calculating S because Q_{back} is very scatter for particle radius larger than 0.2 μm.

Refractive index is an important parameter for calculating light scattering properties of aerosol. Since there is no available refractive index reference for common aerosol compositions in wavelength range spanning from UV to IR, in this paper, aerosol is assumed to be composite by $(NH4)_2SO_4$ and H_2O. Toon *et al.* [7] has reported the real part of refractive index of $(NH4)_2SO_4$ from 0.3 μm to 40 μm. Hale and Querry [8] has reported complex refractive index of H_2O between 0.2~200 μm. The calculation of the refractive index for the mixture of $(NH4)_2SO_4$ and H_2O follow the rule of partial molar refraction approach [9]. In addition, the way to approaching the actual refractive index of aerosol is achieved by adjust the water content to make the calculated total-scattering coefficient fit with the total-scattering coefficients measured by nephelometer. The aerosol absorption coefficient is not available during the sampling period. Hence, the effect of aerosol absorption was not included to approaching complex refractive index of aerosol. The sunphotometer (AERONET) measurements indicate the mean value of single scattering albedo (SSA) over Taipei area in spring is about 0.9. Therefore, the actual Lidar ratios S may be 10% larger than the calculated values.

Dust and particle produced by combustion are usually irregular in shape. Koepke and Hess [10] has shown that the difference between the phase functions of spherical and non-spherical absorbing fine aerosols of both urban and continental types is small for scattering angles in the 15°-170°, which implies that the Mie theory is not inappropriate for modeling light scattering on irregular urban aerosols in the accumulation mode. Simulation [9-10] shows that mixtures of irregular dust-like particles generally show a smaller backscatter than that of equivalent spheres. During the Asian winter monsoon, Asian dust oriented from main land China consists of very irregularly shaped aerosols may enhance the coarse mode aerosols. Irregular shaped particle could be easily identified by its polarization property. A depolarization and Raman Lidar system co-owned by Research Center for Environmental Changes and National Taiwan University was operated at same experimental site during sampling period. Therefore, the cases consist of many irregular particles were distinguished and would not affect the statistic results.

3. RESULTS AND DISCUSSIONS

During the sampling period, the effective radii and volume C/F ratios of aerosol vary from 0.1 μm to 0.6 μm and 0 to 2 respectively. The mean values are r_e= 0.261±0.071 μm and C/F=0.532±0.289. The refractive index n varies from 1.45 to 1.52, and the mean value is n=1.48±0.026. Fig. 2 shows a typical particle size distribution sampled at 2005/3/21 14:00 LST by SMPS/APS system. As shown in figure, the observed particle size distribution is fitted to a combination of five lognormal distributions (mode I to V). The effective radius and C/F radio for whole size spectrum are r_e=0.271 μm and C/F=0.531. The scattering properties of each mode are listed in Table 1. Refer to Table 1, about 90% of total scattering coefficients (σ_{355} and σ_{532}) are contributed by mode IV (particle diameter smaller than 1 μm). About 67% of β_{532} and 77% of β_{355} are contributed by Mode IV and about 30% of β_{532} and 19% of β_{355} are contributed by Mode V. The results show aerosol scattering properties are controlled by concentration of fine mode aerosol and coarse mode aerosol. Moreover, backscatter β would be much sensitive to the variation of coarse mode particles than total scattering σ, and β_{532} is more sensitive to coarse mode than β_{355}.

Lidar ratio is concentration independent characteristic of aerosol. Fig. 3 shows the occurrence frequency of derived Lidar ratios during the sampling period. Lidar ratio for 355 nm and 532 nm are mainly distributed in 20-40 sr and 30-50 sr respectively, and the mean values are S_{355}= 30.6±4.3 sr and S_{532}=42.94±4.8 sr. The values of S_{532} are close to urban type aerosol. Fig. 4 shows the relationship between S_{355} and S_{532}, where dash line is the running mean value and standard deviation of S_{532} ($\bar{S}_{532} \pm \delta_{S_{532}}$) and $\delta_{S_{532}}$ varies from 3 to 7. For S_{355}> 50 sr, S_{532} is less sensitive on S_{355}. For S_{355} less than 50 sr, the running mean values show S_{532} are linearly and highly correlated with S_{355} and the correlation factor is R=0.67. The linear regressive curve (solid line) is fitted to data point that S_{355}<50 sr which gives a regressive equation $S_{532}=S_{355}\times0.706+17.0$. For Nd:YAG laser based dual-wavelength Lidar system which equips with only one Raman channel for 355 nm or 532 nm. If one of the Lidar ratios (S_{355} or S_{532}) could be determined by measurement, this regressive equation would be useful to obtain another Lidar ratio S_{532} (or S_{355}) to improve accuracy of aerosol backscattering profile calculation (e.g. by Klett's method [1]).

Lidar ratio is related with aerosol size distribution [11-12]. Fig. 5 shows Lidar ratios S_{355} and S_{532} derived from particle size distributions were negatively correlated with effective radius and volume C/F ratio. As mentioned above (refer to Fig. 2 and Table 1), β_{532} is more sensitive to variation of coarse mode particles than β_{355}, therefore, higher correlations were found between S_{355} and r_e (R = -0.79) and S_{532} and C/F ratio (R= -0.86) as expected. That implies measured S_{355} could be applied to estimate effective radius and S_{532} could be used to estimate C/F ratio. Fig. 5 also indicates Lidar ratio could be obtained from surface ground aerosol measurements and applied to the correction of Lidar overlap problem. The relationship between Lidar ratio and effective radius is determined by particle size distribution and refractive index. Wandinger et al. [11] has demonstrated a similar but different relationship for stratospheric aerosol. Therefore, the relationships shown in Fig. 5 may just represent the characteristic of aerosols over Taipei area.

4. SUMMARY

Based on aerosol size distribution experiment during March-May 2005 at Taipei basin, Taiwan, the scattering properties of aerosol such as Lidar ratios were calculated by Mie scattering theory. The major results are summarized as follows: (1) The mean values of Lidar ratio for 355 nm and 532 nm are 31.9±6.2 sr and 40.5±6.1 sr respectively. (2) S_{355} and S_{532} are linear correlated for S_{355}<50 sr, which imply S_{532} (or S_{355}) could be roughly determined by measured S_{355} (or S_{532}). For some of dual-wavelength Lidar systems which equipped only one Raman channel (355 nm or 532 nm), the relationship between S_{355} and S_{532} would be useful to estimate the Lidar ratio of non-Raman channel with an uncertainty about 5 sr. (3) Lidar ratio S_{355} is highly correlated with effective radius of aerosol, which indicates Lidar measured S_{355} could be used to get the vertical profile of aerosol effective radius. (4) Lidar ratio S_{532} is highly correlated with volume C/F ratio, that imply if C/F ratio could be obtained by surface ground measurement (e.g. PM10 and PM2.5), an estimated S_{532} could be got. That would be helpful for most of non-Raman Lidar systems to improve the accuracy of Lidar inversion. (5) Backscattering coefficient was found to be more sensitive on the variation of coarse mode aerosol than total scattering coefficient. Which implies backscatter-related scattering properties (Lidar ratio, angstrom component) of aerosol may be better choices to investigate coarse mode dominated aerosol events (e.g. Asian dust).

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FIGURES AND TABLES

Table 1. Scattering properties of aerosol modes as shown in Fig. 2.

	σ_{532} (km^{-1})	β_{532} (sr^{-1}km^{-1})	S_{532} (sr)
Mode I	0.000	0.00000	8.45
Mode II	0.000	0.00000	9.02
Mode III	0.002	0.00016	13.56
Mode IV	0.198	0.00358	46.98
Mode V	0.030	0.001889	15.83
Mode I to V	0.230	0.006265	36.71

	σ_{355} (km^{-1})	β_{355} (sr^{-1}km^{-1})	S_{355} (sr)
Mode I	0.000	0.00000	8.53
Mode II	0.002	0.00000	9.89
Mode III	0.003	0.00013	23.46
Mode IV	0.089	0.00279	32.13
Mode V	0.010	0.00069	14.04
Mode I to V	0.102	0.00361	28.35

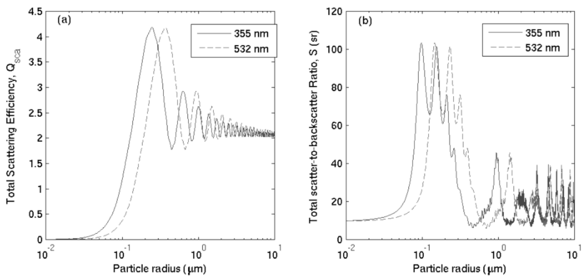

Fig. 1. Total scattering efficiency Q_{sca} and total scatter-to-backscatter ratio S for 355 nm (soild line) and 532 nm (dashed line) with refractive index of n=1.48. Note Q_{back} was smoothed before calculating S because Q_{back} is very scatter for particle radius larger than 0.2 μm.

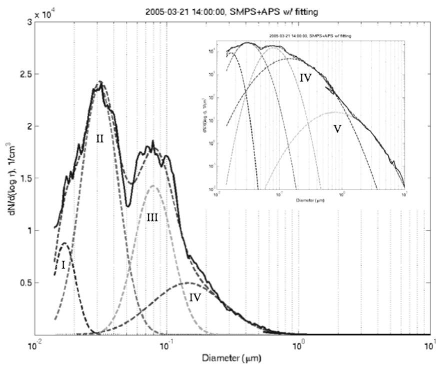

Fig. 2. Sample particle size distribution measured by SMPS/APS system at 2005/3/21 14:00LT. r_e=0.217 μm, S_{532}=36.71, S_{355}=28.35.

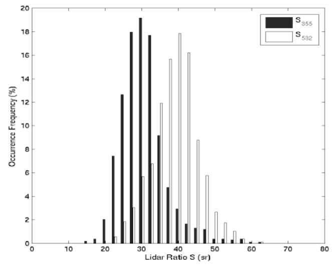

Fig. 3. The Occurrence frequencies of Lidar Ratio S_{355} and S_{532}. The mean values and standard deviations are S_{355}=31.9±6.2 sr and S_{532}=40.5±6.1 sr respectively.

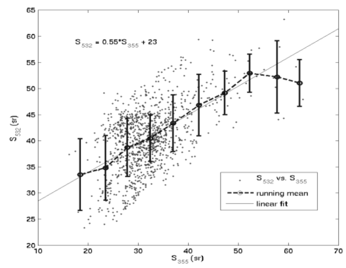

Fig. 4. Relationship between S_{355} and S_{532}.

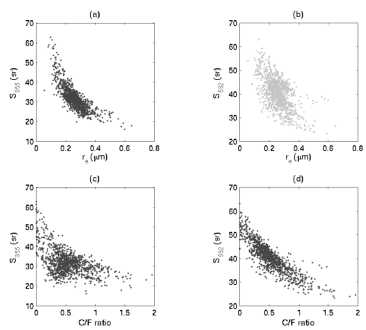

Fig. 5. (a) and (b) the relationship between Lidar Ratio and effective radius. (c) and (d) the relationship between Lidar ratio and CF/ratio.