Optical properties of Asian dusts in the free atmosphere measured by Raman lidar at Taipei, Taiwan

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Abstract

The optical properties (extinction-to-backscatter ratio, backscattering, depolarization, and backscatter-related Angstrom exponent) and height distribution of Asian dusts were measured using a two-wavelength Raman/depolarization lidar at Taipei, Taiwan, during the Asian dust seasons in 2004 and 2005. Dust layers were frequently observed in the free atmosphere (1–6 km). Dust optical thickness ranged from 0.01 to 0.55; backscatter-related Angstrom exponents ranged from 0.42 to 1.47; and lidar ratios (extinction-to-backscatter ratio) for 355 nm ranged from 32 to 72 sr (steradian). The mean values of dust particle depolarization and extinction coefficient are $14 \pm 6\%$ and 0.16 km^{-1} , respectively, which are close to the moderate dust depolarizations and extinctions observed in free atmosphere in China and Japan. Backscatter-related Angstrom exponents were found correlated positively with lidar ratio and negatively with particle depolarization, indicating that the dust optical characteristics are predominated by size distribution. Dusts were found to tend to exhibit unusual low depolarization properties under moist conditions (relative humidity $\text{RH} > 70\%$), and the possible explanations are discussed.

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1. Introduction

Asian dusts play important roles in the climate system and bio-geochemical cycle in the Asia-Pacific region by affecting the radiation balance by scattering and absorbing solar radiation, or modifying the cloud properties by acting as cloud condensation nuclei and ice forming nuclei (Duce,

1995; Arimoto et al., 2006). Therefore, it is important to obtain the optical properties and height distribution of Asian dust for the studies related to long-range transport and atmospheric radiation problems. During the dust seasons, aerosols with total depolarization ratio (TDP) higher than 5% were frequently observed in the free atmosphere over Taipei, Taiwan by a Raman/depolarization lidar, indicating the existence of non-spherical particles. Regional air quality model (Chen et al., 2004; Tsai et al., 2004) simulation and HYSPLIT back-trajectory analysis show that

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most of the depolarization layers oriented from northwestern China and outflow of Asian dusts are the most possible sources of the observed non-spherical particles.

The intensive optical properties (independent on concentration) of aerosol such as Angstrom exponent, depolarization, and lidar ratio (extinction-to-backscatter ratio) are complicated functions of aerosol size distribution, composition, and shape (Anderson et al., 2000; Clarke et al., 2004; Carrico et al., 2003; Sakai et al., 2006). Angstrom exponent α is a good indicator of the size of particles (Angstrom, 1964). Higher values ($\alpha > 2$) are typically observed for accumulation mode particles (Reid et al., 1999; Eck et al., 1999) and lower values (α near 0) have been observed for coarse mode particles such as Saharan dusts and Asian dusts (Eck et al., 1999; Sakai et al., 2002). The value of lidar ratio varies with the characteristic of aerosols. Lidar or in situ measurements and computer simulations show that lidar ratios for sea salt, Asian dust, Saharan dust, biomass burning, and urban aerosol are 20–30, 40–60, 30–40, 45–65, and 20–80 sr, respectively (Ansmann et al., 1992a; Takamura et al., 1994; Anderson et al., 2000; Liu et al., 2002). The depolarization can be considered as an indicator of the non-sphericity of the particle and had been widely applied to study the irregularly shaped particle such as cirrus cloud (Sassen, 1991; Chen et al., 2002). Lidar measurements reveal that particles such as Asian dust, biomass burning, and sea salt would exhibit clear depolarization properties owing to irregular shape. Depolarization is close to zero for spherical particle. For dry or crystallized sulfate, depolarization ratio is about 2% (Cooper et al., 1974; Sassen et al., 1989). For Asian dust, biomass burning, and sea salt, the depolarization ratios are about 5–30% (Liu et al., 2002; Sakai et al., 2002; Iwasaka et al., 2003; Kim et al., 2004; Shimizu et al., 2004), 6–11% (Wandinger et al., 2002; Murayama et al., 2004), and 8–22% (Murayama et al., 1999), respectively.

The inter-relationship between intensive scattering properties of Asian dust had not been well studied. For dusts in the free atmosphere, simultaneous measurements of such scattering parameters are necessary to understand the micro-physical properties of dust. This paper reports the vertical distribution of free tropospheric dusts measured using a two-wavelength Raman/depolarization lidar over Taipei, Taiwan, during the Asian dust seasons in 2004 and 2005. In Section 2, we describe the

Raman lidar system and the derived atmospheric parameters. Three dust cases are selected to demonstrate the vertical profile of optical properties and to see how they are correlated. Optical properties of dust episodes such as backscatter coefficient, particle depolarization, lidar ratio, Angstrom exponent, and optical thickness are summarized and discussed. We note that most of the dust Angstrom exponents are negatively correlated with particle depolarizations and positively correlated with lidar ratios, implying that the dust scattering characteristics were predominated by particle size. The relative humidity (RH) may significantly affect depolarization properties of dusts and is discussed in Section 4.

2. Lidar system and methodology

2.1. Lidar system description

The lidar is a two-wavelength Raman and depolarization lidar system employing the second and third harmonics of Nd-YAG laser at 532 and 355 nm. This lidar is funded by Research Center for Environmental Changes (RCEC), Academia Sinica and Department of Atmospheric Sciences, National Taiwan University (NTU) and is installed in the weather observatory (25.014°N, 121.54°E) of the National Taiwan University, which is located in the southwestern part of the Taipei Basin. Fig. 1 represents the Taipei basin on the northern part of Taiwan, and the location of the lidar site together

Fig. 1. Geographical location of the observations in Taipei Basin (NTU: Weather observatory in National Taiwan University, CWB: Banchiao meteorological station).

with the location of Banchiao meteorological station (25.00°N, 121.43°E, about 9.9 km away from NTU) of Central Weather Bureau (CWB), where radiosoundings are systematically performed twice per day at 00:00 and 12:00 UTC (08:00 and 20:00 LT, local time).

The lidar system measures the vertical distribution of aerosol backscattering, extinction, and depolarization (532 nm only) by detecting the Rayleigh/Mie backscattering by atmospheric gases and aerosol particles and the Raman backscattering by atmospheric nitrogen (at nighttime and only for 355 nm). Detailed specification of this lidar is given in Table 1. The measurements were carried out routinely under thick-cloud free condition to probe the atmosphere in the height range between 0.3 and 8 km. Lidar signals were acquired for every 1 min and were usually averaged for every 15 min to improve the signal-to-noise ratio. During the sampling period (February–June, 2004 and January–May, 2005), there were 109 partial-cloudy days available for free atmosphere measurement. However, owing to the weather condition and signal quality, Raman signals were not available for most dust episodes.

The incomplete overlap between the laser beam and the receiver field of view significantly affects lidar observations of particle optical properties in the near-field range. Following the overlap correction method proposed by Wandinger and Ansmann (2002), the percentage of signal reductions caused by incomplete beam overlap at altitudes of 1.5 and 0.5 km are found about 2% and 20%, respectively.

Table 1
Specifications of Raman lidar at National Taiwan University (25.01°N, 121.54°E)

Transmitter	
Laser type	Nd:YAG
Wavelength (nm)	532/355
Pulse energy (mJ)	65/60
Repetition rate (Hz)	20
Receiver	
Telescope type	Cassegrain
Diameter	40 cm
Focal length	160 cm
Detector	PMT × 4
Signal detection	Analog/photon counting
Range resolution	7.5 m
Measured channels	
Rayleigh/Mie scattering	355 and 532 nm
Depolarization	532 nm
Nitrogen Raman scattering	355 nm (nighttime only)

2.2. Methodology

The Raman lidar equation can be expressed as

$$P_{\lambda}(z) = P_{\lambda L} \frac{A}{z^2} \beta_{\lambda}(z) T_{\lambda L}(z_0, z) T_{\lambda}(z_0, z), \quad (1)$$

where λ_L is the laser wavelength, $P_{\lambda L}$ is the laser power, $P_{\lambda}(z)$ is the measured lidar signal at wavelength λ returned from altitude z , A is the system calibration factor, $\beta_{\lambda}(z)$ is the volume Rayleigh/Mie or Raman backscattering coefficient of the scattering media at λ , and $T_{\lambda}(z_0, z)$ is the atmospheric transmission at λ between lidar at altitudes z_0 and z that is given by

$$T_{\lambda}(z_0, z) = e^{-\int_{z_0}^z [\sigma_{a,\lambda}(z') + \sigma_{m,\lambda}(z')] dz'}, \quad (2)$$

where $\sigma_{a,\lambda}(z)$ and $\sigma_{m,\lambda}(z)$ are the volume extinction coefficients of the aerosols or air molecules. Aerosol backscattering ratio R_{λ} is defined as

$$R_{\lambda}(z) = 1 + \frac{\beta_{a,\lambda}(z)}{\beta_{m,\lambda}(z)}, \quad (3)$$

where $\beta_{a,\lambda}(z)$ and $\beta_{m,\lambda}(z)$ are volume backscattering coefficients of aerosols and air molecules at λ , respectively. In order to derive an analytical solution for the lidar equation, it is common to assume that these parameters are related in the form of the aerosol extinction-to-backscatter ratio or the so-called lidar ratio S_{λ} .

Solving the lidar equation requires knowledge about how lidar ratio varies along the light path (Klett, 1981; Fernald, 1984). In this study, R_{355} and S_{355} were retrieved by the combined Raman/Rayleigh–Mie inversion algorithm (Ansmann et al., 1992b; Whiteman et al., 1992). The air density profile is derived from closest radiosonde measurement. Since Raman signal is not available for 532 nm, the calculation of backscatter uses the Klett's method (Klett, 1985) by setting lidar ratios of 532 nm S_{532} equal to S_{355} (Sugimoto et al., 2002). The tail-up value for the inversion is usually set as $R_{532} = 1.05$ at 6 km. In order to minimize the uncertainty caused by the initial value, only cases that aerosol backscattering ratios are less than $R_{532} < 1.1$ for a continuous range about 500 m above dust layers are selected in this study. For further analyses, the height resolutions of aerosol backscattering profiles are reduced to match the aerosol extinction profiles before computing profiles of S_{355} . To further reduce the random errors, Raman and Rayleigh/Mie signals are integrated for every 30 min.

Table 2

Optical properties (lidar ratio S_{355} , backscattering coefficient β_{a355} , particle depolarization δ_p for 532 nm, backscatter-related Angstrom exponent α_{back} , and optical depth AOD for 355 nm) of dusts observed in free atmosphere

Date	Heigh (km)	RH (%)	$\delta_p - \alpha_{\text{back}}$ slope	$\beta_{a,355}^{\text{mean}} \times 10^{-6}$ ($\text{m}^{-1} \text{sr}^{-1}$)	δ_p^{mean} (%)	δ_p^{max} (%)	$\alpha_{\text{back}}^{\text{mean}}$	S_{355} (sr)	AOD
19/02/2004	1.05–2.02	58	–12	7.78	6	7	1.3	49	0.37
15/03/2004	2.54–2.99	73	–313	3.60	7	5	1.2	62	0.20
	2.99–3.75	49	–16	2.43	6	11	1.2	53	
06/04/2004	1.26–2.48	62	–12	2.80	7	18	0.8	41	0.14
19/04/2004	0.83–1.43	20	–11	3.92	12	15	0.5	48	0.19
	2.25–3.37	24	–12	1.40	9	12	1.2	46	
21/04/2004	2.04–3.06	72	34	1.81	6	24	0.4	32	0.06
25/04/2004	1.21–2.05	93	–292	4.64	4	7	1.4	51	0.39
	2.05–3.38	75	–12	3.06	8	11	1.2	47	
09/05/2004	1.16–1.72	85	–85	3.55	14	15	0.7	49	0.24
	1.72–2.55	6	–7	2.84	12	15	1.3	62	
14/03/2005	1.84–3.76	44	–8	0.69	17	21	0.5	42	0.05
03/04/2005	1.45–2.90	62	–7	4.42	9	16	1.0	45	0.55
	3.53–4.16	62	–9	1.04	15	18	0.7	36	
04/04/2005	1.10–2.79	35	–14	9.34	9	11	1.0	42	0.47
15/04/2005	1.57–2.88	57	–9	5.14	7	10	1.2	48	0.32
	2.88–3.00	78	–24	4.03	11	12	1.5	70	
18/04/2005	2.47–4.23	52	–6	3.04	13	11	1.1	54	0.29
21/04/2005	1.06–2.42	54	–11	4.36	14	24	0.4	36	0.21
26/04/2005	3.00–3.48	62	–10	0.75	15	24	0.7	37	0.01
18/05/2005	3.41–4.16	51	–8	1.09	13	17	0.7	49	0.04
Average	–	–	–	3.41 ± 2.18	11 ± 4	14 ± 6	1.0 ± 0.3	47 ± 9	0.23 ± 0.17

The uncertainty of lidar ratio is $\Delta S_{355} \approx 5\text{--}10\text{ sr}$. The bottom and top of dust layers are determined by threshold $\delta_p = 3.5\%$.

The backscatter-related Angstrom exponent α_{back} is calculated from β_a at 355 and 532 nm and is defined as the negative of the slope of β_a with the wavelength in logarithmic scale:

$$\alpha_{\text{back}} = -\frac{\text{dln } \beta_a(\lambda)}{\text{dln } (\lambda)}. \quad (4)$$

The value α_{back} approximately decreases as the size increases and vice versa. Generally, Angstrom exponent provides information on the aerosol size distribution, with values greater than 2.0 corresponding to accumulation mode particles, such as fresh biomass burning smoke, and values closer to zero for coarse mode particles, such as dust.

As a key factor for solving lidar equation, the difference between S_{355} and S_{532} had not been well studied. By using dual-wavelength Raman lidar, Murayama et al. (2004) reported that the lidar ratios for Asian dust with particle depolarization about 20% are 49 and 43 sr for 355 and 532 nm, respectively. For aerosol at RH < 95%, Ackermann (1998) showed that the difference is less than 5–10 sr. In this study, the possible uncertainty of α_{back} owing to uncertainty of S_{532} was approached by recalculating all R_{532} and α_{back} , assuming S_{532}

were 15% over-estimated. The recalculated R_{532} (for dust episodes listed in Table 2) are found being under-estimated by 0.8–8.5% with a mean value of $6.0 \pm 2.9\%$ and the recalculated α_{back} are over-estimated by 3–40% with a mean value of $17 \pm 10\%$. Such uncertainties may not influence the discussions about dust optical properties.

The total (aerosol + molecule) depolarization ratio, TDP, is defined as the ratio of the return light of perpendicular to parallel polarization:

$$\text{TDP} = \frac{P_{\perp}}{P_{\parallel}}, \quad (5)$$

where $P_{\parallel}$ and $P_{\perp}$ are the integrated return power for parallel and perpendicular directions relative to the outgoing laser beam. To eliminate the depolarization components produced by the air molecules, the particle depolarization δ_p is expressed by the following equation:

$$\delta_p = \frac{R \times \delta - \delta_m}{R - 1}, \quad (6)$$

where $\delta = \text{TDP}/(1 + \text{TDP})$ (Cairo et al., 1999; Murayama et al., 1999) and the molecular

depolarization ratio δ_m is given as 1.4% (Weber et al., 1967; Young, 1980).

Note that we have not taken into account the multiple scattering while calculating the dust optical properties from the lidar signals, so that the values of extinction coefficient and lidar ratio could have been underestimated because the forward-scattered light reduces the apparent attenuation of light detected with lidar (Sakai et al., 2003a). Multiple scattering may significantly affect both the estimations of the cloud optical depth from lidar return signals and the discrimination between the liquid and the solid phases of cloud layers by using the polarization lidar data. When the optical depth of the atmospheric target is not negligible or the phase function of the scattering particles is strongly forward peaked, as in cirrus clouds, or the observation is from a long distance, the multiple scattering contribution to the lidar signals cannot be neglected (Wang et al., 2005) and could be expressed as an exponential function of optical thickness (Eloranta, 1998). For clouds, Wandinger (1998) had shown multiple-scattering errors of measured extinction and backscatter coefficients may be up to the order of 50% and 20%, respectively. For tropospheric aerosols, Ackermann et al. (1999) indicated that multiple scattering for aerosol is expected to be less important for ground-based lidar systems because the forward-scattering peak of the phase function is less pronounced than that of the cloud. Ackermann et al. (1999) also showed that for optical depths of typical atmospheres (extinction coefficients less than 1.96 km^{-1}) the multiple-scattering contribution to the backscattered signal is less than 5% and the relative error of the retrieved aerosol extinction profile is less than 3%. For our system, since the observed dusts' optical thicknesses and extinction coefficients at 355 nm are 0.01–0.55 (Table 2) and $0.03\text{--}0.39 \text{ km}^{-1}$, the multiple scattering contributions for dust backscatter might be less than 5%. However, the multiple-scattering-induced error on dust optical properties is not taken into account in this study.

Depolarization is also strongly depended on the concentration of particles owing to multiple scattering effects. On the basis of laboratory experiments, Zaccanti et al. (1993) and Gai et al. (1996) had shown that if the cloud optical thickness is less than 1 then the effect of multiple scattering would be insignificant on the depolarization, but if the cloud optical depth exceeds 3 then multiple scattering induced depolarization might be up to 20–30%.

In Section 3.2.3, we will show a liquid-cloud-induced depolarization. Since all of the optical depths of dust layers shown in this study are less than 0.6, the depolarization introduced by multiple scattering can be ignored.

3. Results

3.1. Height distribution

TDP-enhanced aerosols were frequently observed in the free troposphere at northern Taiwan in the Asian dust seasons. During the sampling period, depolarization layers were found existing in the free troposphere in 91 of the 109 operation days, and to locate inside the boundary layer in 28 of the operation days. Aerosol TDPs ranged from 7.5% to 25%. The height distribution for aerosols with $\text{TDP} > 3.5\%$ is shown in Fig. 2, where TDP is considered as the “maximum” TDP in each lidar operation day. As shown in Fig. 2, most of the depolarization layers are located at heights 1–5 km, and occasionally we could find multiple layered depolarization aerosols.

By comparing lidar depolarization measurements with aerosol water-soluble ions, our previous studies (Chen et al., 2007) had shown that the lidar depolarization ratios were highly correlated with the water-soluble calcium ion, which is one of the major indicators of mineral dust (Mori et al., 1999; Kim and Park, 2001). For aerosol layers in the free atmosphere, a regional air quality model (TAQM) (Chen et al., 2004; Tsai et al., 2004) and HYSPLIT back-trajectory analyses were applied to investigate the possible origin of those depolarization layers listed in Table 2. Fig. 3 shows the 5 days HYSPLIT back-trajectories for the three dust episodes demonstrated in Section 3.2. The starting dates were 3/4/2005 (Fig. 5), 9/5/2004 (Fig. 7), and 15/4/2005 (Fig. 8) and the initial heights are 2.5, 2, and 2 km, respectively. Both TAQM and HYSPLIT have shown that the most possible origins of depolarization layers are dust source areas in Mainland China, implying that Asian dusts are the major sources of non-spherical particles. Dusts usually were uplifted due to strong winds generated by surface low-pressure systems over the source regions, such as Inner Mongolia, developed through frontal-genesis and then transported in free atmosphere to Taiwan following the front movement (Chen et al., 2004). However, more detailed synoptic, meteorological, or regional air quality model analyses are necessary

Fig. 2. Height distributions for aerosol with total depolarization ratio TDP > 3.5% in the dust seasons of 2004 and 2005.

for understanding the long-range transport problem of Asian outflow, but that is outside the purpose of this paper.

3.2. Case study

3.2.1. 3–4 April 2005

A depolarization episode occurred at 3–4 April 2005 is shown in Fig. 4 to demonstrate the typical optical properties of lidar observed Asian dusts. The

signal strengths are not good enough because lidar signals were occasionally attenuated by clouds during daytime of April 3. An aerosol layer with maximum total depolarization of TDP $\approx 25\%$ could be figured out at heights 1–4 km, indicating that this layer is predominated by non-spherical particles. On April 3, Taipei was situated at the outskirts of a high-pressure system. The center of the high-pressure system was located at the Yellow Sea and then shifted to East China Sea (between

Fig. 3. NOAA HYSPLIT 5 days back-trajectories for depolarization episodes demonstrated in this paper. The starting dates are 3/4/2005 (Fig. 5), 9/5/2004 (Fig. 7), and 15/4/2005 (Fig. 8) and the initial heights are 2.5, 2, and 2 km, respectively.

Fig. 4. Temporal and spatial cross sections of (a) range corrected signal for 532 nm and (b) total depolarization ratio obtained by lidar on 3–4 April, 2005. The temporal resolution is reduced to 15 min to increase signal-to-noise ratio.

Japan and Taiwan) on April 4. HYSPLIT back-trajectory (refer to Fig. 3, starting from 3/4/2005 12:00 LT) shows that air parcel had passed Inner Mongolia, China, which is one of major dust sources, indicating that the most possible origin of non-spherical particle is Asian dust. From 02:00 to 23:00 LT on April 4, a range corrected signal and a TDP show that the dust top descended from 4 to 1 km. Whereas temperature and RH profiles (launched at 8:00 LT, Fig. 5(b)) show a subsidence temperature inversion appeared at 2.5 km, which is close to the height of dust top at 08:00 LT, implying that the descending of dust layer might be caused by subsidence associated with the traveling high-pressure system.

Fig. 5 shows lidar measured vertical profiles of optical properties (R_{532} , R_{355} , S_{355} , α_{back} , TDP, and δ_p) obtained at April 3 06:00 LT (upper panel) and April 4 06:00 LT (lower panel). Temperature and RH obtained by closest radiosonde measurements (launched at 08:00 LT) are shown for comparison. On April 3 at 06:00 LT, two depolarization layers with R_{532} /TDP of the order about 3.36/5% and 1.75/7% were found at 0.8–3.4 and 3.4–4.2 km, respectively. The optical properties for the lower and upper depolarization layers are particle depolarization ratios $\delta_p \approx 9.3\%$ and 16.2%; Angstrom exponents $\alpha_{\text{back}} \approx 1.01$ and 0.75; lidar ratios $S_{355} \approx 45$ and 36 sr. On April 4 at 06:00 LT, the upper layer disappeared and the lower dust layers were found located between 1.1 and 2.8 km with R_{532} about 5, δ_p about 9%, S_{355} about 42 sr, and α_{back} about 1.06. The relatively small Angstrom exponents (0.5–1.2) imply that the dust layers are dominated by coarse mode particles compared with urban aerosols (Holben et al., 2001). The average values of S_{355} for dust layers shown in Fig. 5 are about 36, 42, and 45 sr, which are close to the value of dust lidar ratios observed in Japan (Liu et al., 2002; Sakai et al., 2003a; Murayama et al., 2004).

Fig. 6(a) shows the scatter plot of δ_p as a function of α_{back} for this dust episode along with some other selected episodes listed in Table 2. The values of δ_p decreased approximately linearly from 26% to 5% with increasing α_{back} from -0.2 to 1.8, suggesting that the aerosol depolarization or non-sphericity is more prominent for large aerosol. Distinct negative correlations were found between α_{back} and δ_p for each episode and most of the α_{back} and δ_p are correlated with similar linear relationship. A linear regressive line with a slope about $y \approx -9$ fitted for depolarization episodes is shown in Fig. 6(a). Since

dust depolarization property is related to its large size and irregular shape, this kind of negative correlations can be explained by the mixing of irregularly shaped coarse mode dusts and spherical-like fine mode particles. Nevertheless, since fine mode particles might not cause significant depolarization at 532 nm even if they are nonspherical (Mishchenko and Sassen, 1998), it is not appropriate to conclude that such fine mode particles are non-spherical dust particles or spherical-like anthropogenic aerosols at the current stage.

To further examine the dust $\delta_p - \alpha_{\text{back}}$ correlation, three lidar and in situ surface ground coincident dust measurements (16/2/2004, 5/3/2005, and 19/3/2005) are also shown in Fig. 6(a) for comparison. These surface ground episodes had been verified as Asian dust events by aerosol water-soluble ions (Chen et al., 2007) and it could be easily figured out that their $\delta_p - \alpha_{\text{back}}$ also correlated in a similar way as dusts in free atmosphere.

3.2.2. 9 May 2004

Aerosol vertical distribution is found associated with vertical RH variations. Radiosonde measurements show that RH values in most dust layers are lower than 60% (refer to Table 2), probably because they mainly originated from the dry regions in Asian continent. Nevertheless, high RH (RH up to 90%) was occasionally observed at depolarizing regions, possibly as a result of mixing with moist air caused by a strong front activity. In some of the moist dust layers, we note that δ_p and α_{back} might not correlate with the same relationship as dry episodes. Fig. 7 shows a dust layer with maximum $\delta_p \approx 15\%$ observed at 9 May 2004 20:30 LT distributed at a height below 3.1 km. Referring radiosonde launched at 20:00 LT, relative humidity at a height above 1.3 km was about RH = 10–15% and at heights 1.3–0.7 km was RH = 70–95%.

The scatter plots of δ_p versus α_{back} for dusts in dry region (1.3–3 km, marked as $\times$) and in moist region (0.5–1.3 km, marked as $\circ$) are shown in Fig. 6(b). In dry region, $\delta_p - \alpha_{\text{back}}$ correlation is similar to cases shown in Fig. 6(a). In moist region, $\delta_p - \alpha_{\text{back}}$ correlation is much different from dusts in the dry region. The slope of the regression lines fitted for the dry and moist regions are $y = -7$ and -85 (as shown in figure), respectively, which implies that dust depolarization property might be changed via physical or chemical process related to water vapor. Similar correlation reported by Sakai et al. (2002) is shown in the figure for comparison, but it is to be

Fig. 5. Vertical profiles of aerosol backscattering ratio for 355 and 532 nm, lidar ratio for 355 nm, particle depolarization for 532 nm and backscatter-related Angstrom exponent measured on (a) 3 April 06:00 LT and (b) 4 April 06:00 LT of 2005. Temperature and relative humidity were measured by radiosonde launched at 08:00 LT and 20:00 LT.

Fig. 6. Scatter diagram of particle depolarization ratio as a function of backscatter related Angstrom exponent for (a) dust episode observed on 3–4 April, 2005 (Fig. 4) along with other episodes selected from Table 2 and (b) dust episodes observed on 9 May, 2004 and 15 April, 2005. Similar result by Sakai et al. (2002) is shown for comparison.

Fig. 7. Vertical profile of aerosol backscattering ratio for 355 and 532 nm, lidar ratio for 355 nm, particle depolarization, and backscatter-related Angstrom exponent measured on 9 May 20:30 LT, 2004. Temperature and relative humidity were measured by radiosonde launched at 20:00 LT.

noted that the laser wavelengths used by Sakai et al. are 1064 and 532 nm, which are different from Our's.

3.2.3. 15 April 2005

Pollutant-coated dust particles could serve as CCN, which may enhance the collision and coalescence of droplets (Rosenfeld et al., 2001). A liquid cloud inside a dust layer observed on 15 April 22:30–23:30 LT, 2005 is shown in Fig. 8. To make it clear, each profile was progressively offset. Dust layer with backscattering ratio of about $R_{355} \approx 2$

could be found distributed below 3.2 km. The maximum δ_p is about 10% and is negatively correlated with α_{back} as dry dusts cases. Radiosonde measurement indicates that (not shown in figure) the relative humidities at 20:00 LT for altitudes above 3.2 km, 2.7–2.9 km, and 1.5–2.7 km were about 31%, 79%, and 57%, respectively. At 22:30 LT, a cloud with a geometric thickness of about 250 m could be found located at 2.7 km. The maximum backscattering ratio for this cloud layer is about $R_{355} \approx 3$ and the corresponding cloud δ_p and α_{back} are 0.5% and 1.1, respectively. Lower

Fig. 8. Vertical profiles of optical parameters measured at local time of 22:30, 23:00, and 23:30 on 15 April, 2005. Each profile was progressively offset to make the figure clear. The offset value for R_{355} , S_{355} , δ_p , and α_{back} are 1, 5 sr, 5%, and 0.5, respectively. (a) Backscattering ratio, (b) lidar ratio, (c) particle depolarization ratio, (d) Angstrom exponent.

depolarization and smaller Angstrom exponent indicates that this cloud is mainly composed of large spherical droplets.

From 22:30 to 23:00 LT, cloud R_{355} increased from 3 to 12, α_{back} decreased from 1.1 to 0.25, lidar ratio S_{355} decreased from 43 to 24 sr, but δ_p increased from 0.5% to 6%. This cloud disappeared at 23:30 LT and the minimum δ_p decreased to about 1.4%. The scatter plots of δ_p versus α_{back} for dusts in dust (solid square) and in cloud region (hollow square) are shown in Fig. 6(b). The δ_p – α_{back} correlation for this cloud is obviously different from dusts' correlations. Since spherical droplet should exhibit low polarization, the increasing of δ_p should be resulted by multiple scattering (Zaccanti et al., 1993; Gai et al., 1996). We note that both the lidar ratio (~ 24 sr) and the Angstrom exponent (~ 0.2) of this cloud are significantly lower than

those of dusts that might be also caused by multiple scattering effect.

4. Discussions

Measurements of optical properties of Asian dusts acquired in the dust seasons in 2004 and 2005 are examined here. Average dust optical properties including lidar ratio S_{355} , backscattering coefficient β_{a355} , particle depolarization δ_p for 532 nm, backscatter-related Angstrom exponent α_{back} , and optical depth AOD at 355 nm are shown in Table 2. The average values along with the standard deviations of the observed dust backscattering coefficient, lidar ratio, and optical thickness are $\beta_{a355} = 3.41 \pm 2.18 \times 10^{-6}$ ($\text{m}^{-1} \text{sr}^{-1}$), $S_{355} = 47 \pm 9$ sr, and $\alpha_{\text{back}} = 0.23 \pm 0.17$, respectively. The average value of extinction coefficient

is about $1.6 \times 10^{-1} \text{ km}^{-1}$ ($S_{355} \times \beta_{a355}$), which is close to the moderate values of dust extinction observed in free atmosphere in China and Japan (Zhou et al., 2002; Murayama et al., 2004; Shimizu et al., 2004).

Particle depolarization, Angstrom exponent, and lidar ratio are intensive optical properties that are highly related to aerosol composition, incident light wavelength, and fraction of coarse and fine modes. As indicator of particle size distribution, observed dust α_{back} ranges from 0.4 to 1.5 which is remarkably lower than that for the accumulation mode aerosols. The observed dust lidar ratios range from 32 to 70 sr and were found to be positively correlated with α_{back} with correlation $R^2 = 0.48$. Such positive correlation is similar to that reported by Anderson et al. (2000) and Ferrare et al. (2001) when comparing aerosol lidar ratios with Angstrom exponents measured by 180° backscatter nephelometer or sun photometer. Ferrare et al. (2001) show S_{355} (derived from Raman lidar) increases as the volume ratio (ratio of fine to coarse particle mode) increases. By combining a Rayleigh/Mie lidar, a sun photometer, and an optical particle counter (OPC), Takamura et al. (1994) also found larger values of S_{532} appear when the contribution of small-mode aerosols ($0.31\text{--}0.44 \mu\text{m}$ in nominal radius) is relatively large. These two observations imply that the lidar ratio for the fine mode is greater than that for the coarse particle mode. Therefore, increasing the ratio of fine to coarse mode particles would make both Angstrom exponent and lidar ratio increase.

As mentioned above, dust optical characteristics were found influenced by particle size and relative humidity. Particle depolarization of dust is known to be dominated by its large size and irregular shape. But for dust episodes listed in Table 2, δ_p are not well correlated with α_{back} ($R^2 = 0.27$). The effect of relative humidity is examined more closely here. Table 2 lists averaged relative humidities and slopes of $\delta_p\text{--}\alpha_{\text{back}}$ (as demonstrated in Fig. 6) for each dust layer, where RHs are obtained by closest radiosonde measurements. For dust layers with slopes of $\delta_p\text{--}\alpha_{\text{back}}$ close to dry dusts ($y = -6$ to -16), the relative humidities ranged from 6% to 75%. And for abnormal cases ($y = 34$, -24 to -313), the relative humidities ranged from 72% to 93%. This result implies the effect of relative humidity on dust depolarization might be more significant than on the particle size when RH is higher than 70%. Fig. 9 shows the scatter

Fig. 9. Scatter diagram of mean dust particle depolarization ratios as a function of mean backscatter-related Angstrom exponents for dusts at dry or moist environments. Values of δ_p and α_{back} are listed in Table 2.

diagrams of δ_p^{mean} versus $\alpha_{\text{back}}^{\text{mean}}$, where “dry” dusts ($\text{RH} < 70\%$) and “moist” dusts ($\text{RH} > 70\%$) are marked as solid circles and hollow squares. It can also be easily figured out that the correlation between $\alpha_{\text{back}}^{\text{mean}}$ and δ_p^{mean} is improved to $R^2 = 0.46$ for dusts at dry condition.

The change of size distribution and refractive index of aerosol due to variation in the RH would cause lidar ratio to change (Ackermann, 1998). Anderson et al. (2000) and Ferrare et al. (2001) had found aerosol lidar ratios tend to increase as relative humidities increase. In this study, we do not note significant influence of relative humidity on dust lidar ratio. For “dry” dusts, lidar ratios are correlated with Angstrom exponents by $R^2 = 0.42$, which is close to the correlation for “dry”+“moist” data set ($R^2 = 0.48$). By numerical simulation, Ackermann (1998) had shown that lidar ratio of desert aerosols could be assumed to be independent of relative humidities if RH is less than 90%. In Table 2, we find the correlation between observed dust lidar ratios and relative humidities is $R^2 = 0$, which is consistent with Ackermann’s conclusion. More measurements are necessary for further investigation.

Tropospheric aerosol particles usually consist of several chemical components and are mixed both externally and internally. The effect of relative humidity on observed depolarization might be explained by external mixture of irregularly shaped dusts and spherical-like particles. The negative correlations (Fig. 6a) between particle depolarizations and Angstrom exponents imply that the

observed dust depolarizations are determined by mixing ratios of non-spherical dust particles in the coarse mode and spherical-like particles (anthropogenic aerosols or mineral particles smaller than the laser wavelength of 532 nm) in the fine mode. The fraction of coarse mode dusts in size distribution might be changed owing to gravitational settling (dry deposition) during long-range transportation from dust sources to Taiwan. When dusts were mixed with moist airs, the observed depolarization could decrease and the Angstrom exponent could not change owing to external mixing with spherical particles whose sizes were close to mineral particles. One possibility of these large spherical particles is anthropogenic aerosols in which size increases due to hygroscopic properties. The other possibility is these large spherical particles are the mineral particles coated with aqueous solution (Sakai et al., 2002). Mineral dust that passes over polluted areas had been frequently observed to be internally mixed (Seinfeld and Pandis, 1998) or coated with anthropogenic aerosols or sea salts (Levin et al., 1996; Maxwell-Meier et al., 2004; Trochine et al., 2003; Clarke et al., 2004; Kim et al., 2003; Niimura et al., 1998; Zhang et al., 2003). Okada et al. (1990) have reported that these coated mineral particles are frequently present in the surface atmosphere in Japan during Asian dust events. In free atmosphere, airborne sampling (Sakai et al., 2003b) showed that 77% of the observed mineral particles were coated with a solution at a height about 1.8 km and the fraction of coated particles decreased with increasing height. For dusts coated with pollutants, the heterogeneous reactions of mineral dust particles are important because they could change the particle surface properties, which can therefore affect the hygroscopic properties of dusts (Laskin et al., 2005) and cause morphological changes in the dusts' shape (reducing depolarization).

Nevertheless, mineral particles with spherical shape have been recently identified in dust particles. Matsuki et al. (2005) had observed spherical Ca-rich particles at Beijing, China during dust events and concluded that those particles might be transformed from irregular calcite (which is one of the major components of Asian dust particle (Okada and Kai, 2004)) owing to heterogeneous chemical process (Krueger et al., 2003, 2004; Vlasenko et al., 2006). For such a condition, δ_p should be expected to exhibit abnormal correlation with α_{back} as "dry" dust particles.

5. Conclusions

We reported case studies of the Raman and Depolarization lidar measurements of optical properties of Asian dusts in the free troposphere during dust seasons in 2004 and 2005 over Taipei, Taiwan. During the sampling period, aerosols with TDP larger than 3.5% were frequently observed at altitudes below 6 km. For episodes shown in this study, a regional air quality model was applied to investigate the possible origin of depolarization layers and the results show that Asian dusts are the most possible origin of non-spherical particles.

Dust optical thickness ranged from 0.01 to 0.55 (mean value of 0.23 ± 0.17); lidar ratio S_{355} varies from 32 to 70 sr (47 ± 9 sr); backscatter-related Angstrom exponent α_{back} ranged from 0.4 to 1.5 (1.0 ± 0.3); mean value of particle depolarization is 14 ± 6 , but maximum δ_p may be up to 24%. Observed lidar ratios and particle depolarizations are close to lidar ratios of Asian dusts measured in China and Japan. We also note that the lidar ratio and particle depolarization are well correlated with the Angstrom exponent, indicating that the dust optical characteristics are predominated by size distribution. The relationship between the aerosol non-sphericity and the aerosol size was investigated by seeking the correlation between particle depolarization δ_p and backscatter-related Angstrom exponent α_{back} . By studying δ_p – α_{back} correlation, we found that dusts tend to exhibit abnormal depolarization properties under moist conditions ($\text{RH} > 70\%$). The distortion of dust δ_p – α_{back} correlation may be caused by change in the dust shape owing to external mixing of irregularly shaped dusts and spherical-like particles, where the spherical-like particles might be the internally mixed mineral particles. However, for most dust episodes, particle depolarizations and Angstrom exponents are well negatively correlated by similar tendency, indicating that the dust layers are composed by irregularly shaped coarse mode dusts and spherical-like fine mode dusts or anthropogenic aerosols.

Suspending particles such as cirrus cloud, dust, crystallized sea salt, and biomass burning smoke are non-spherical particles that could be found in free atmosphere. More measurements and studies for other irregular-shaped particles are necessary to clarify the relationship between physicochemical properties of particle such as size distribution, shape, and chemical composition.

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