Aerosol Characterization by Dual-Wavelength Polarization Lidar Measurements over Kochi, Japan during the Warm Seasons of 2008 to 2010

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Abstract

Dual-wavelength polarization lidar measurements of aerosols and clouds were conducted over Kochi (33.6°N, 133.5°E) during the warm-season field campaign of the Japanese Cloud Seeding Experiment Precipitation Augmentation project in June–July of 2008 to 2010. Lidar-derived aerosol optical properties were compared with the microphysical properties obtained with aircraft-based instruments to evaluate the utility of the lidar data for characterizing the background aerosol, which critically affects the success of cloud seeding to enhance precipitation. The results showed that the particle backscattering coefficient at 532 nm correlated well with the number concentration of aerosols with diameter (D_p) exceeding 0.3 μm (correlation coefficient, $r = 0.89$), whereas the correlation of the backscattering coefficient with the number concentration of cloud condensation nuclei (CCN), activated at a water saturation of 0.7% or 1.0%, was lower ($r = 0.74$) because of the low sensitivity of the lidar system to the small CCN particles ($D_p \approx 0.04 \mu\text{m}$). In lidar data collected on 1 July 2008, the depolarization ratio (δ) was high (20%), and the backscatter wavelength exponent ($\hat{\alpha}$) was low (< 0.5) between altitudes of 4 and 6 km, and they were low ($\delta = 2.5\%$) and moderate ($\hat{\alpha} = 0.7$) between 0.5 and 1.0 km, suggesting the presence of supermicrometer-sized, non-spherical particles in the upper altitude range and a predominance of submicrometer-sized particles and/or spherical particles in the lower altitude range. These values were consistent with aircraft measurements, indicating the presence of supermicrometer-sized mineral particles in the upper altitude range and a predominance of submicrometer-sized sulfates and supermicrometer-sized sea-salt droplets at lower altitude. Our results demonstrate the utility of lidar data for aerosol characterization, although the further improvement of CCN characterization by lidar is necessary.

Keywords aerosol; lidar; aircraft measurement

1. Introduction

Water shortage is a crucial problem in western Japan. Cloud seeding to increase precipitation in a target area is a promising technique for ameliorating this problem. However, this technique involves many uncertainties, and it is difficult to validate the effects of seeding on the augmentation of precipitation. To gain a deeper understanding of cloud seeding, the Japanese Cloud Seeding Experiment for Precipitation Augmen-

tation (JCSEPA) project was carried out over the five years from 2006 to 2011 (Murakami and JCSEPA Research Group 2011). As a part of this project, field campaigns were conducted during the warm seasons (June–August) of 2008 to 2010 on Shikoku Island, Japan, which often experiences water shortages. During the experiments, a dual-wavelength polarization lidar was deployed at Kagami supersite in Kochi Prefecture to monitor the vertical distributions of background aerosols and cloud properties. The main objective of the lidar measurement was to characterize the background aerosol's microphysical properties (number concentration, size distribution, and chemical composition), because these properties critically affect the raindrop formation rate achieved by cloud seeding.

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For example, Kuba and Fujiyoshi (2006) and Kuba and Murakami (2010) demonstrated by using a hybrid cloud microphysical model that the effect of cloud seeding on the raindrop formation rate depends the characteristics of the background aerosol. They showed that seeding with large-particle cloud condensation nuclei (CCN) increases the amount of rainfall under polluted conditions (i.e., when there is a large number concentration of small hygroscopic particles), whereas it decreases the amount of rainfall under unpolluted conditions (i.e., a small number concentration of small hygroscopic particles). Therefore, for cloud seeding, it would be helpful to be able to monitor the background aerosol by lidar and to assess the aerosol's characteristics by lidar measurements. However, obtaining the detailed microphysical properties of aerosols from only their lidar-derived optical properties is a challenging proposition. It would be possible to estimate one parameter (e.g., number concentration) if we knew (or could assume) the values of all other parameters (e.g., particle size distribution, shape, and refractive index) from the lidar-derived optical property. However, these parameter values vary with height and over time in the real atmosphere. Several studies (e.g., Catrall et al. 2005; Omar et al. 2009; Burton et al. 2012) have developed methods for characterizing aerosol types (e.g., maritime aerosols, desert dust, particulates from biomass burning, and urban/industrial pollutants) from lidar-derived extinction and backscattering coefficients and depolarization ratios at multiple wavelengths. Moreover, inversion techniques for retrieving the aerosol size distribution and the number concentration from multiwavelength lidar data have been developed (Müller et al. 1999a, b; Veselovskii et al. 2002, 2012). However, few studies have verified these retrieval techniques by using in situ data because of the difficulty of obtaining collocated measurements from both lidar and in situ instruments. Reagan et al. (1977) compared the aerosol number size distribution, complex refractive index, and mass loading inferred from bistatic lidar–solar radiometer measurements with those measured directly by instruments on board an aircraft. They found a similarity in the size distribution shape between the two measurements and agreement with the absolute values within one order of magnitude. Hoffman et al. (2012) compared particle size distributions retrieved from multiwavelength Raman lidar data with those obtained with a differential mobility particle sizer at a high-altitude mountain station and found good agreement for particle diameters exceeding $0.1\ \mu\text{m}$. Sakai et al. (2012) compared the backscattering coefficient and

depolarization ratio derived from lidar with the aerosol mass concentration and particle shape in filter samples obtained near the ground surface. They found an approximately linear relationship between the backscattering coefficient and the mass concentration and between the depolarization ratio and the abundance of nonspherical mineral dust. To validate and improve lidar measurement techniques for aerosol characterization, more comparisons of lidar results with collocated in situ measurements are needed. In this paper, we present our aerosol characterization results derived from lidar data and compare them with microphysical properties determined by aircraft-based measurements, based on data obtained during a JCSEPA field campaign of the warm cloud seeding experiments. Section 2 describes the instrumentation used. In Section 3, we present the results of the lidar measurements and compare the backscattering coefficient data with the particle number concentration data, the depolarization ratio, and the backscatter wavelength exponent with the aerosol type and size distribution. In Section 4, we summarize our results.

2. Methods

2.1 Dual-wavelength polarization lidar measurement

The lidar system used in this study (Table 1) employs a Nd: YAG laser with a frequency-doubled option, operating at 20 Hz with pulse energies of 180 and 200 mJ at wavelengths of 532 and 1064 nm, respectively. Light backscattered by atmospheric molecules, aerosols, and cloud particles is collected by two Cassegrain telescopes, one with a diameter of 20 cm for near-range signals and the other with a diameter of 35 cm for far-range signals. We used a co-axial lidar configuration for the small telescope and a bi-axial configuration for the large one to obtain an aerosol profile from the lowermost troposphere (altitude 80 m) to the lower stratosphere (altitude ~ 35 km). The lidar detects Rayleigh–Mie backscattering at 1064 nm with parallel and perpendicular components at 532 nm and Raman nitrogen backscattering at 607 nm (night-time only). The signals of the 532 and 607 nm channels are detected by photomultiplier tubes, while that of the 1064 nm channel is detected by an avalanche photodiode. The data are acquired by a transient recorder that combines photon counting and analog-to-digital conversion. The data were collected at 3-min or 20-s intervals (in coordination with the aircraft measurements) at a vertical resolution of 7.5 m.

The backscattering ratio (R) and particle backscattering coefficient (β_p) were computed from the elastic

Table 1. Specifications of the lidar used for the cloud seeding experiment.

Transmitter:				
Laser type	Nd: YAG (Continuum Surelite I)			
Wavelength (nm)	532	1064		
Energy/pulse (mJ/pulse)	180	200		
Repetition rate (Hz)	20			
Beam divergence (mrad)	0.2			
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Receiver:				
Telescope type	Cassegrain (Celestron C14)		Cassegrain (Celestron C8)	
Diameter (cm)	35		20	
Field of view (mrad)	1.0		2.0	
Measured altitude range (km)	2–35 (approximate)		0.08–10	
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Detection species:				
Detection wavelength (nm)	532		607	1064
Polarization	Parallel	Perpendicular	—	—
Detector type and number	PMT × 3	PMT × 2	PMT × 2	APD × 2
Range resolution	7.5 m			
Temporal resolution	20 sec (minimum)			
Data acquisition	Photon counting (250 MHz bandwidth) & Analog-to-digital conversion (12-bit resolution) (Licel GbR)			

backscatter signal by the method of Fernald (1984) as follows:

$$R = \frac{\beta_p + \beta_m}{\beta_m}, \quad (1)$$

where β_m is the molecular backscattering coefficient. In the computation, we assumed the extinction-to-backscatter ratio (lidar ratio) to be 50 sr at 532 nm and 45 sr at 1064 nm, based on previous studies in Asia (Chiang et al. 2008; Catrall et al. 2005). The uncertainty of β_p for these assumed lidar ratios was estimated to be 20% at 532 nm and 10% at 1064 nm by changing the lidar ratios by 30 sr. We used the molecular backscattering and extinction coefficients that were obtained from the atmospheric density measured by radiosonde at the nearest aerological observation station (Shionomisaki, 33.45°N, 135.77°E). We normalized the signal by assuming $R = 1.02$ at 532 nm and 1.13 at 1064 nm at altitudes between 10 and 12 km (Russell et al. 1982).

The total linear volume depolarization ratio (δ_v) was obtained by taking the ratio of the perpendicular to total components of the backscattering signal at 532 nm. The particle depolarization ratio (δ), which is a measure of average particle nonsphericity and depends on the particle size and its refractive index (e.g., Mishchenko et al. 1996), was computed from R and δ_v by

Table 2. Date and altitude range of the collocated measurements with lidar and aircraft.

Date	Time Period (JST)	Altitude range (km)
June 23, 2008	13:46–13:51	2.7–3.6
June 28, 2008	11:00–11:10	0.7–3.7
July 1, 2008	14:42–14:58	0.6–4.9
June 1, 2009	16:34–16:46	0.6–3.3
June 2, 2009	12:26–13:17	0.7–3.3
June 6, 2009	16:35–17:02	1.6–3.1
June 15, 2009	15:14–15:41	0.9–3.3
June 16, 2009	14:12–14:42	0.9–3.3
June 5, 2010	10:28–10:49	1.6–5.1
June 9, 2010	14:22–14:46	0.9–4.2
June 12, 2010	15:20–15:59	0.9–6.0
June 17, 2010	11:37–12:27	1.0–6.0
June 23, 2010	10:40–11:06	1.0–4.9
June 24, 2010	11:03–11:19	0.9–3.9

$$\delta = \frac{\delta_v R - \delta_m}{R - 1}, \quad (2)$$

where δ_m is the molecular depolarization ratio (0.49%) for this lidar system (Sakai et al. 2003).

The backscatter wavelength exponent ($\hat{a}$), which is a measure of particle size and type (e.g., Sasano and Browell 1989; Ansmann et al. 2002), was computed by

$$\hat{a} = -\frac{\ln(\beta_{1064}/\beta_{532})}{\ln(1064/532)} \quad (3)$$

This value generally increases as the proportion of supermicrometer-sized particles decreases (Sasano and Browell 1989). The lidar measurements were conducted from June 2008 to July 2010 at Kagami supersite in Kochi Prefecture, Japan (33.60°N, 133.48°E; 50 m above sea level). Data were acquired for 553 days during this period (acquisition rate 71%).

2.2 Aircraft-borne measurements

Samples were collected by instruments on board an aircraft (King Air B200T) for detailed vertical profiles of aerosol and cloud microphysical properties during a spiral descent from 7.5 to 0.6 km above the ground level over the observation site. The properties measured were aerosol number size distribution, aerosol sample for electron microscopic and elemental composition analyses, CCN number concentration, cloud particle images, and cloud/ice water content. Ambient air was introduced into the aircraft through an inlet into a manifold, which distributed the sampled air to the instruments. The aerosol size distribution was measured with a scanning mobility particle sizer (SMPS) (Model 3936, TSI, USA) for particles with diameter (D_p) in the range of $0.01 \leq D_p \leq 0.3 \mu\text{m}$ and with an optical particle counter (OPC) (KC-01E, RION, Japan) for particles with $D_p \geq 0.3 \mu\text{m}$. A continuous-flow CCN counter (CCNC) (CCN-200, DMT, USA) was used to measure the CCN number concentration at a water vapor supersaturation of 1% (2008) or 0.7% (2009 and 2010). The minimum particle diameter detectable with this CCNC is approximately $\sim 0.04 \mu\text{m}$ at supersaturations from 0.7% to 1%. Aerosol particles for electron microscope analyses were directly sampled with an onboard two-stage low-volume impactor. The details of the sampling method and the analysis procedure are as described by Sakai et al. (2012), except that we also used a transmission electron microscope (JEM-1400, JEOL, Japan), operated at an accelerating voltage of 120 KV, to analyze particle morphology and an energy-dispersive X-ray (EDX) silicon drift detector (X-Max 80, Oxford Instruments, UK) equipped with a microscope, operated in scanning electron microscope mode, to analyze the elemental composition. A total of 32 aircraft data sets were acquired during the period from 2008 to 2010. To compare the aerosol properties between the lidar and aircraft measurements, we selected 14 days over which both lidar aircraft measurements were conducted.

Fig. 1. Averaged particle number size distributions derived from aircraft data collected during 14 flights over Kochi, conducted during June and July from 2008 to 2010 in nine altitude ranges between 0.5 and 5.0 km (thin solid lines). Horizontal orange arrows show the diameter ranges detectable with the CCN counter ($D_p \geq 0.04 \mu\text{m}$) and the OPC ($D_p \geq 0.3 \mu\text{m}$). The thick black line shows the backscattering cross sections at the wavelength of 532 nm for particles with a refractive index of 1.53–0.008i.

3. Results

3.1 Comparison of backscattering coefficient with particle number concentrations

Monitoring number concentrations and size distributions of aerosols and CCN for hygroscopic cloud seeding is desirable, because these properties critically affect seeding efficacy. To assess the suitability of lidar measurements for monitoring number concentrations, we compared lidar-derived β_p values with aerosol and CCN number concentrations obtained with airborne instruments.

Figure 1 shows the average particle number size distribution measured in the altitude range between 0.5 and 5.0 km during 14 flights of the aircraft with the SMPS and OPC installed on board. The mode diameters of the particle size distributions were $0.1 \mu\text{m}$ or smaller over all altitude ranges, and the effective radii computed from SMPS and OPC data ranged from 0.16 to $0.39 \mu\text{m}$ over the altitude range.

Because the OPC and CCNC detected particles in different size ranges (Fig. 1), the correlations between the number concentrations of particles detected by each instrument and β_p also differed.

Fig. 2. Scatter plot of the particle backscattering coefficient at 532 nm versus (a) the particle number concentration for $D_p \geq 0.3 \mu\text{m}$ and (b) the CCN number concentration activated at water supersaturation level of 1% (2008) or 0.7% (2009/2010) over Kochi in June and July from 2008 to 2010. Error bars indicate the standard deviation. The linear regression line between β and N (solid line) and the power-law regression line (dotted line) are also shown.

Figure 2 presents scatter plots of β_p at 532 nm (β_{532}) versus the number concentration of particles with optically equivalent diameters exceeding $0.3 \mu\text{m}$ (N_a) obtained with the OPC and those of β_p at 532 nm versus the CCN number concentration (N_{CCN}) obtained with the CCNC, during the 14 flights under background aerosol conditions, when seeding particles were absent. These OPC and CCN data were simultaneously obtained at altitudes from 0.1 to 6.0 km. A regression analysis between β_{532} and the number concentrations yielded the linear regression equations $N_a = 5.3 \times 10^4 \beta_{532}$ and $N_{\text{CCN}} = 5.5 \times 10^5 \beta_{532}$ (solid lines in Fig. 2), where N_a and N_{CCN} have units of cm^{-3} and β_{532} has units of $\text{km}^{-1} \text{sr}^{-1}$. The correlation coefficient (r) between β_{532} and N_a is 0.89, and $r = 0.74$ between β_{532} and N_{CCN} . If we fit a power function to the data, we obtain the regression equations $N_a = 1.7 \times 10^4 (\beta_{532})^{0.85}$ with $r = 0.85$ and $N_{\text{CCN}} = 1.5 \times 10^5 (\beta_{532})^{0.46}$ with $r = 0.75$ (dotted lines in Fig. 2). The value of r is higher between β_{532} and N_a than between β_{532} and N_{CCN} , because lidar is more sensitive to the size range detected with the OPC ($D_p \geq 0.3 \mu\text{m}$) than to that detected with CCNC ($D_p \geq \sim 0.04 \mu\text{m}$).

To demonstrate this, we also plotted in Fig. 1 the backscattering cross section at 532 nm as a function of particle diameter (thick black line) for dust-like aerosol with a complex refractive index of $1.53-0.008i$ (Shettle and Fenn 1979). In the figure, we can see that the

particle backscattering cross sections for $D_p = 0.04-0.06 \mu\text{m}$ were approximately 10^{-3} of those for $D_p \geq 0.3 \mu\text{m}$, and the number concentration over the D_p range from 0.04 to $0.06 \mu\text{m}$, which is detectable only with CCNC, was approximately 10 times the number concentration for $D_p \geq 0.3 \mu\text{m}$. Thus, the correlation of β_{532} with N_{CCN} is lower than that with N_a , because 1) most CCN particles were smaller than $0.3 \mu\text{m}$, and 2) the backscattering cross sections of the CCN particles were much smaller than those of the large aerosol particles. As a result, the sensitivity of the lidar to the CCN particles detected with the CCNC was lower than its sensitivity to the aerosol particles detected with the OPC. It might be possible, however, to estimate the number concentration of larger CCN particles activating at a water supersaturation point lower than 0.7%, because the higher correlation with β_{532} is expected.

The vertical distributions of N_a estimated from β_{532} compared with those measured with airborne OPC (Fig. 3) showed that N_a varied by one order of magnitude below an altitude of 3 km among the 14 dates. This result indicated that it is important to monitor the aerosol concentration distribution during cloud seeding experiments. The lidar and aircraft-derived N_a values showed poor agreement in altitude ranges with abundant aerosol particles (e.g., about 2 km on 2 June 2009, between 2 and 3 km on 12 June

Fig. 3. Vertical distributions of the particle number concentration for $D_p \geq 0.3 \mu\text{m}$ obtained with the airborne OPC (gray line) and estimated from β_{532} obtained by lidar (black line) by using the linear regression equation $N_a = 5.3 \times 10^4 \beta_{532}$ (see Fig. 2a).

2010, and about 2.5 km on 17 June 2010), possibly because aerosols with optical and microphysical properties different from those of the background aerosol were present there. Another possible explanation for the poor agreement is that the lidar measured the ambient aerosols, which had become larger in the humid air, whereas the OPC measured dry aerosols

after they were introduced into the interior of the aircraft. The maximum ambient relative humidity in the layers with abundant aerosols was 45–50%, whereas the relative humidity inside the aircraft was lower than 20%.

3.2 Comparison of the depolarization ratio and backscatter wavelength exponent with aerosol shape, chemical composition, and size distribution

Aerosol chemical composition and size distribution are secondary factors that affect cloud seeding. For example, water-soluble particles act more effectively as CCN than insoluble ones (e.g., Pruppacher and Klett 1997). Furthermore, large water-soluble particles are activated at lower supersaturation levels than smaller ones. Thus, to evaluate the suitability of conditions for hygroscopic seeding, the estimation of the chemical composition (hygroscopicity) and size distribution of an aerosol from lidar measurements is desirable. To assess the capability of lidar for this estimation, we compared lidar-derived δ and $\hat{a}$ values with aerosol type and size distributions obtained by aircraft measurement.

We plotted $\hat{a}$ against δ for 1 July 2008, when aerosol samples had also been collected by aircraft over the observation site (Fig. 4) and compared the results with ranges of δ and $\hat{a}$ of aerosols reported by previous observational and model studies (Cattrell et al. 2005; Tao et al. 2008; Hess et al. 1998; Sakai et al. 2010). We estimated the uncertainties of lidar-derived δ and $\hat{a}$, caused by the assumption of a constant lidar ratio, to be $\pm 2\%$ and ± 0.1 , respectively, based on calculations in which the lidar ratios were changed by $\pm 30\%$. Between the altitudes of 0.5 and 1.0 km, δ ranged from 2% to 4% and $\hat{a}$ from 0.8 to 1.0, whereas between 4.2 and 6.0 km, δ ranged from 5% to 28% and $\hat{a}$ from 0.2 to 0.8 (Fig. 4). These results suggested that spherical and submicrometer-sized particles (e.g., sulfates) predominate in the lower altitude range, while supermicrometer-sized nonspherical particles (e.g., mineral dust) are mixed with submicrometer-sized particles in the upper altitude range. To validate these results, we examined aerosol particles collected on 1 July 2008 at altitudes ranging from 0.5 to 0.6 km and from 4.0 to 6.7 km near the observation site by transmission electron microscopy (TEM; Fig. 5). We found that the supermicrometer-sized particles collected at 0.5 to 0.6 km were mostly spherical, whereas those collected at 4.0 to 6.7 km were nonspherical; submicrometer-sized particles were mostly spherical in both altitude ranges. We classified the particle shapes observed by TEM into four categories—spherical (i.e., round), spherical with opaque inclusions, aspherical (quasi-round shape without sharp edges), and nonspherical (irregular shape with sharp edges)—and determined the number fractions of each category in the images (Fig. 6). The spherical particles are probably liquid droplets, and the

Fig. 4. Scatter plot of the backscatter wavelength exponent versus the particle depolarization ratio at altitudes of 0.5 to 1.0 km (solid circles) and 4.2 to 6.0 km (open diamonds) on 1 July 2008 along with typical ranges for aerosols from various sources.

nonspherical particles are solids. Aspherical particles might be droplets or solids with a liquid coating, and spherical particles with inclusions are likely liquid droplets with solid, internally mixed inclusions. We found that 67% (66%) of the supermicrometer-sized particles at 3.6 to 6.7 km (0.5 to 0.6 km) were nonspherical or aspherical, and 96% to 98% of the submicrometer-sized particles were spherical in both altitude ranges. This result is consistent with the lidar-derived δ and $\hat{a}$ results, which suggested that substantial numbers of large nonspherical particles are present at altitudes between 4.2 and 6.0 km and that spherical particles predominate between 0.5 and 1 km.

The EDX analysis results indicated that the major elements which compose the supermicrometer-sized nonspherical particles are Si and Al, whereas the major elements in the supermicrometer-sized spherical particles are Na, S, and Mg. These results suggested that the nonspherical particles are mineral particles and the spherical particles are composed of sea salt. The major element of the submicrometer-sized particles was S, which suggested that they are sulfates. The average relative humidity between 0.5 and 0.6 km was 82%, which is higher than the deliquescent humidity of sea salt (75.3%; Tang 1996); thus, consistent with the TEM images of the spheres (Fig. 5a), the sea-salt

Fig. 5. Transmission electron micrographs of aerosol particles collected at altitudes from (a) 0.5 to 0.6 km and (b) 4.0 to 6.7 km on 1 July 2008.

Fig. 6. Number fractions of particles of different shapes collected at altitudes between (a) 0.5 and 0.6 km and (b) 3.6 and 6.7 km on 1 July 2008: (left) supermicrometer-sized particles and (right) submicrometer-sized particles.

particles were likely present as liquid droplets. The average relative humidity from 4.0 to 6.7 km was 38%.

We compared these results with the average particle

number size distributions measured with the OPC in the altitude ranges from 0.5 to 1.0 km and from 4.5 to 5.5 km (Fig. 7). Unfortunately, SMPS data from 1 July

2008 were unavailable because of an air leakage problem in the SMPS. Submicrometer-sized particles predominated between 0.5 and 1.0 km, whereas supermicrometer-sized particles predominated between 4.5 and 5.5 km. The number concentration ratio of particles with $D_p \geq 1.0 \mu\text{m}$ to those with $D_p \geq 0.3 \mu\text{m}$ was 0.4% between 0.5 and 1.0 km and 6% between 4.5 and 5.5 km. Moreover, the effective radius, computed from the OPC data, of particles in the 0.5 to 1.0 km (4.5 to 5.5 km) altitude range was $0.19 \mu\text{m}$ ($1.48 \mu\text{m}$). These results are qualitatively consistent with the lidar-derived data for $\bar{a}$, suggesting that submicrometer-sized particles are more prevalent in the lower altitude range than in the upper altitude range (Fig. 4). Therefore, hygroscopic seeding in the lower altitude range might be the most likely to enhance raindrop formation in clouds including high concentrations of submicrometer-sized particles.

The relative standard deviation of the particle number concentration ratio $D_p \geq 1.0 \mu\text{m}$ to $D_p = 0.3\text{--}1.0 \mu\text{m}$ was 200% (mean $\pm$ standard deviation (SD), $0.040 \text{ cm}^{-3} \pm 0.083 \text{ cm}^{-3}$) between 4.5 and 5.5 km, whereas between 0.5 and 1 km, it was $\sim 100\%$ ($0.0037 \text{ cm}^{-3} \pm 0.0042 \text{ cm}^{-3}$). This result is consistent with the larger variation of δ and $\bar{a}$ in the upper altitude range than in the lower altitude range (Fig. 4), and it suggested a large variation of the external mixing ratio of supermicrometer-sized mineral dust particles to submicrometer-sized sulfate particles in the upper range. Although to assess the suitability of conditions for hygroscopic seeding, it would be best to estimate the number concentrations of each aerosol type from the lidar measurements, the values of N/β and δ (or $\bar{a}$) of each aerosol type must be known to estimate the number concentrations of two aerosol components, and the values of three parameters (N/β , δ , and $\bar{a}$) must be known to estimate the number concentrations of three components. For example, Chen et al. (2001) and Sugimoto et al. (2003) estimated the relative contributions of spherical aerosols and non-spherical dust aerosols to β_{532} from δ by assuming that δ was constant for both these aerosol components. Nishizawa et al. (2007, 2011) estimated the aerosol extinction coefficients of fine-mode (water-soluble) and coarse-mode (sea salt or dust) aerosols from β , δ , and $\bar{a}$ by making assumptions about the size distribution, shapes, and refractive indices of each of the three aerosol components. However, the uncertainties in the estimation techniques of the number concentrations are high if the aerosol particles are hygroscopic. The values of N/β , δ , and $\bar{a}$ vary depending on the ambient relative humidity, owing to changes in the phase and

Fig. 7. Average particle number size distributions in the altitude ranges from 0.5 to 1.0 km (dotted line with circles) and 4.5 to 5.5 km (thick line with diamonds) measured with the OPC on 1 July 2008. The error bars represent the standard deviation.

size of the particles (e.g., Tang and Munkelwitz 1994; Sakai et al. 2010). Moreover, these parameter values also vary depending on the internal mixing state of the particles (e.g., Lesins et al. 2002).

A 3-day back-trajectory analysis computed with the HYSPLIT model (Draxler and Rolph 2012) showed that the air parcel at 0.75 km above the observation site had been transported over the central part of Japan's main island and the Sea of Japan, whereas the one at 5.0 km had had been transported over the Asian continent 2–3 days before (Fig. 8). This result suggested that the sulfate and sea-salt particles in the lower altitude range originate over mainland Japan and the Sea of Japan, whereas the mineral particles in the upper altitude range originate in arid regions of the Asian continent.

3.3 Vertical distribution of background aerosol properties based on lidar measurements

To study the temporal and vertical variability of the type and concentration of the background aerosol, that is, properties relevant to cloud seeding, we plotted the vertical distribution of β_{532} in relation to altitude at the times of the 14 aircraft measurements in June and July 2008–2010 (Fig. 9). The values of β_{532} below the altitude of 2.5 km (0.3 to 2.5 km: $\beta_{532} = 1.1 \times 10^{-4}$ to $1.6 \times 10^{-2} \text{ km}^{-1} \text{ sr}^{-1}$; average, $2.9 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$)

Fig. 8. Three-day backward trajectories of air parcels arriving at the measurement site (33.6°N, 133.5°E) on 1 July 2008, computed using the HYSPLIT model. The solid lines with circles and dashed lines with asterisks indicate the trajectories of the air parcel arriving at altitudes of 0.75 and 5.0 km, respectively. The solid diamond indicates the observation site.

exceeded those above that altitude (2.5 to 6.0 km: $\beta_{532} = 2.7 \times 10^{-5}$ to $3.0 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$; average, $4.0 \times 10^{-4} \text{ km}^{-1} \text{ sr}^{-1}$) and also exhibited a larger variation. Using these values and the linear regression equation (see Section 3.1), we estimated N_a to range from 10 to 1000 cm^{-3} with an average of 150 cm^{-3} between 0.3 and 2.5 km, and from 1 to 200 cm^{-3} with an average of 20 cm^{-3} between 2.5 km and 6.0 km. Between 0.3 and 2.5 km, δ and $\bar{a}$ were (mean $\pm$ SD) $5 \pm 3\%$ and 1.2 ± 0.5 , respectively, whereas they were $9 \pm 10\%$ and 1.0 ± 0.5 between 2.5 and 6.0 km. These values suggested that supermicrometer-sized spherical (water-soluble) particles and/or submicrometer-sized particles are more abundant and that the average particle size is smaller in the lower altitude range than in the upper altitude range. Therefore, we suggest that to enhance precipitation, clouds with a base below 2.5 km should be seeded with hygroscopic particles, because they are likely to have a higher number concentration of small particles and a lower number concentration of large particles. However, the uncertainty is large, because it is difficult to estimate the CCN concentration from lidar-derived β_{532} , which is a parameter more sensitive to aerosols than to CCN, and because the ratio of N_{CCN} to N_a is variable.

4. Conclusion

We compared lidar-derived aerosol optical properties with aircraft in situ measurements of microphysi-

Fig. 9. Vertical distribution of the particle backscattering coefficient (β_{532}) over Kochi from June to August from 2008 to 2010. The symbol shape indicates the value range of the depolarization ratio (δ : $\circ$, $\delta < 10\%$; $\square$, $10\% \leq \delta < 20\%$; and $*$, $\delta \geq 20\%$) and its size indicates the value range of the backscatter wavelength exponent (small, $\bar{a} \geq 1$; medium, $0 \leq \bar{a} < 1$; and large, $\bar{a} < 0$). The upper horizontal axis shows the aerosol number concentration (N_a) estimated from β_{532} by using the linear regression equation $N_a = 5.3 \times 10^4 \beta_{532}$ (see Fig. 2a).

cal properties during a warm-cloud seeding experiment from June to August of 2008–2010. The backscattering coefficient at 532 nm correlated well with the particle number concentration of aerosol particles with $D_p \geq 0.3 \mu\text{m}$, whereas the correlation with the CCN number concentration at supersaturation of 0.7% or 1.0% was lower because of the low sensitivity of lidar to small CCN. The results of a case study on 1 July 2008 indicated that aerosol shape and size estimated from δ and $\bar{a}$ were consistent with those obtained from aircraft measurements. Both showed a predominance of supermicrometer-sized spherical sea-salt droplets and submicrometer-sized sulfate particles between altitudes of 0.5 and 0.6 km and a higher proportion of supermicrometer-sized nonspherical mineral particles between 4.0 and 6.7 km. The average aerosol number concentration estimated from the lidar data was 150 cm^{-3} between 0.3 and 2.5 km and 20 cm^{-3} between 2.5 and 6.0 km among the 14 dates. The mean particle size and the fraction of large nonspherical particles were higher in the upper altitude range than in the lower altitude range. Our results demonstrate the utility of lidar measurements for monitoring and characterizing background aerosol properties. However, further study is necessary to enable the lidar estimation of the number fraction of each aerosol type and the CCN number concentration, which are important for assessing the suitability of conditions for hygroscopic seeding.

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