Correlation study between suspended particulate matter and portable automated lidar data

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Abstract

Continuous data of the atmosphere monitored using a portable automated lidar are correlated with the concentration of ground-measured suspended particulate matter (SPM). When the boundary layer is well mixed, a high correlation is found between the lidar and the ground SPM data, making it possible to calculate the mass extinction efficiency of the aerosols in the atmosphere. It is found that within the ground layer, the value of mean mass extinction efficiency changes in a range of 4–12 m\textsuperscript{2} g\textsuperscript{−1}, with smaller and larger values occurring for size distributions dominated by coarse and fine particles, respectively.

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1. Introduction

Lidar is an instrument suitable for monitoring spatial and temporal structure of the atmosphere. When a lidar is operated continuously, the received signals provide the time–height indication of the atmosphere on the basis of aerosol optical parameters such as the backscattering and/or the extinction coefficients (Measures, 1984). These parameters are of importance in understanding the local aerosol behavior, as well as in modeling the radiation energy budget of the local atmosphere. Furthermore, optical depths of aerosols and thin clouds can readily be measured from lidar observations. Recently, we reported on a

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portable automated lidar (PAL), which is capable of obtaining aerosol/cloud data in the lower troposphere (Lagrosas et al., 2004).

When lidar data are combined with the data from other instruments, more detailed optical properties of aerosol particles can be studied. For example, when a sunphotometer is operated with a lidar system, the surface, volume and extinction-to-backscatter ratio of stratospheric aerosols can be estimated (Gobbi, 1995) with the help of the Mie-scattering analysis (Van de Hulst, 1962). By further exploring this concept, the effects of particle nonsphericity can also be observed (Gobbi, Barnaba, Blumthaler, Labow, & Herman, 2002). When multi-wavelengths are utilized, other quantities such as the mass concentration and median size of aerosols can be estimated from lidar measurements. Del Guasta (2002) measured these quantities using a two-wavelength lidar system. Yabuki, Kuze, Kinjo, and Takeuchi (2003) presented a method of deriving the aerosol refractive index and size distribution by using multi-wavelength lidar data. In general, these techniques contribute to establishing linkage between the lidar data and the aerosol micro-properties.

In the lower atmosphere, aerosols come from a number of different sources. In addition to ubiquitous soil and sea salt aerosols (coarse particles), urban aerosols have anthropogenic origins such as dust from paved or unpaved road (coarse particles), or primary or secondary particles from exhaust of automobile combustions and industrial processes (fine particles). In the context of urban air pollution, the terminology of suspended particulate matter (SPM) is used to specify aerosols causing human health problems. These are solid or liquid particles with varying sizes (less than 10 μm in diameter) and shapes whose origins are often ascribed to industrial activities. The Mie-scattering arising from these particles can be detected by lidars. In fact, when lidar backscattered signals are correlated with the SPM amount, a strong correlation can often be found between the two quantities. Munkel, Emeis, Mueller, and Schaefer (2003) observed this phenomenon during dry weather situations, finding a correlation of more than 80% between the dust concentration and the aerosol backscatter observed using a ceilometer. Also, Lagrosas et al. (2004) reported the correlation between the range-corrected signal of a PAL and SPM data from a β-ray detector. These observations can lead to the calculation of the conversion factor between the backscattered lidar signal and the SPM concentration. In order to relate this correlation to the optical conditions of the atmosphere, a more relevant quantity can be obtained if the extinction coefficients (expressed in units of m$^{-1}$) are extracted from the lidar data and compared with the SPM concentration (in units of gm$^{-3}$). This conversion factor is often called the mass extinction efficiency (MEE) (in units of m$^{2}$ g$^{-1}$), and this is an essential parameter linking the mass concentration to light scattering. A detailed explanation of MEE, including the survey of existing data, will be given in a later section (Section 3).

The aim of this paper is threefold. First, we examine the PAL data during one year to study the atmospheric conditions under which the extinction data exhibit good correlation with the SPM data. Second, it is shown from simulation and experiment that MEE values generally decrease due to the increased concentration of aerosols with larger diameters. Lastly, the MEE values are computed using the PAL data and the ground sampling data, and the relationship between the MEE parameter and aerosol size distributions is discussed.

2. Experiment

2.1. The lidar system

The PAL system is placed at the Chiba Prefectural Environmental Research Center (CERC) in Ichihara city (35.52N, 140.07E, about 40 km southeast of Tokyo), an industrial area south of Chiba city, along the
Table 1
Specification of the PAL system

<table>
<thead>
<tr>
<th>Transmitter</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Laser</td>
<td>LD-pumped Q-switch Nd:YAG</td>
</tr>
<tr>
<td>Wavelength</td>
<td>532 nm</td>
</tr>
<tr>
<td>Laser pulse width</td>
<td>50 ns</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>1.4 kHz</td>
</tr>
<tr>
<td>Laser pulse energy</td>
<td>15 µJ</td>
</tr>
<tr>
<td>Laser beam divergence</td>
<td>50 μrad</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Receiver</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Telescope diameter</td>
<td>20 cm</td>
</tr>
<tr>
<td>Telescope type</td>
<td>Cassegrain</td>
</tr>
<tr>
<td>Field of view</td>
<td>0.2 mrad</td>
</tr>
<tr>
<td>Detector</td>
<td>PMT</td>
</tr>
<tr>
<td>Model</td>
<td>HPK-R1924P</td>
</tr>
<tr>
<td>Quantum efficiency</td>
<td>10–25%</td>
</tr>
</tbody>
</table>

east coast of Tokyo Bay. This location is an ideal site for studying urban atmospheric pollution, since a significant part of the aerosol particles is considered to originate from anthropogenic sources. The PAL system is installed indoors at a height of about 4.5 m from the ground level. The beam is pointed toward the north sky with an elevation angle of 38°. The horizontal distance between the lidar location and the seashore (east coast of the Tokyo bay) is about 2.5 km in the laser beam direction.

The main components of the PAL system are a pulse laser, a 20 cm diameter telescope, a photomultiplier detector, a signal scaler and a personal computer (PC) for data acquisition and system control. Table 1 summarizes the system. A detailed description can be found elsewhere (Lagrosas et al., 2004). In order to reduce the background due to the skylight during daytime, a narrow field-of-view angle of 0.2 mrad is used. The laser beam is reflected by two prisms so that its axis agrees with the optical axis of the telescope. The alignment of the system is checked every 15 min, and if necessary, corrected automatically by adjusting the orientations of one of the prisms. The backscattered signal is collected by the telescope, detected by the photo-multiplier tube in the photon-counting mode, accumulated for 20 s, and stored in the PC.

2.2. SPM measurements at the ground level

The hourly average value of SPM concentration is obtained from the β-ray absorption instrument (Shimadzu AAMS-4160A) at the CERC ground station, 70 m from the lidar site. Because of a relatively high suction speed (100 L/min) and no application of pre-heating, it is likely that this instrument measures a quantity close to the ambient (i.e. not dried) aerosol mass concentration. The data from this station (named as Iwasaki-nishi station) are accessible from an Internet site (http://www.soramame.nies.go.jp) provided by the Ministry of Environment, Japan. In addition, a tapered element oscillating microbalance (Rupprecht and Patashnick TEOM Series 1400a) (Guidance for Using Continuous Monitors in PM2.5 Monitoring Networks; http://www.epa.gov/ttn/amtic/files/ambient/pm25/r-98-012.pdf) has been operated at the lidar site since 24 March 2003, providing the data on the particles with diameters less than 2.5 µm at 50%
cut-off. The $\beta$-ray and the TEOM instruments have minimum detection limit of $1 \mu g \cdot m^{-3}$ and 0.01 $\mu g$ (mass transducer), respectively.

For the discussion of aerosol properties, the information on aerosol components is required. Ground sampling measurements were performed in the Chiba area from September 1998 to February 2002 (Yabuki, 2003). The sampler is a low volume Andersen sampler (Tokyo Dylec AN-200) with an intake of 28.3 L min$^{-1}$. Three (out of nine) stages of the sampler were used to separate aerosol particles into the following three classes: with diameters less than 2.1 $\mu m$, between 2.1 and 11 $\mu m$, and larger than 11 $\mu m$.

3. Mass extinction efficiency

In this section, the general properties of the MEE parameter are described. The MEE is the ratio of the extinction coefficient and the mass concentration:

$$\text{MEE} = \frac{\alpha(\lambda)}{\frac{4}{3} \pi \rho \int_{r_1}^{r_2} r^2 n(r) \, dr}.$$  (1)

where $\alpha(\lambda)$ is the extinction coefficient, $\lambda$ the wavelength, $\rho$ the density, $r$ the radius ($r_1$ and $r_2$ are the lower and upper limits, respectively), and $n(r)$ the particle size distribution. The extinction coefficient can be written as

$$\alpha(\lambda) = \pi \int_{r_1}^{r_2} r^2 Q_{\text{ext}}(r, \lambda, m) n(r) \, dr,$$  (2)

where $Q_{\text{ext}}(r, \lambda, m)$ is the extinction efficiency, and $m$ is the refractive index. In the present simulation of MEE, we assume the dry mass density. Since experiments yield the ambient values of the extinction coefficient and mass concentration, a separate consideration must be given especially for high relative humidity conditions (Feczkó, Molnár, Mészáros, & Major, 2002).

The MEE parameter is often used in the study of radiative forcing to connect the mass of the scatterers to the extinction coefficient of light. For example, Di Girolamo et al. (1999) used an MEE value of 5 m$^2$ g$^{-1}$ to approximate the water content in the planetary boundary layer. The work of Pitchford and Green (1997) showed that PM2.5 aerosols and aerosols from organic species have MEE of 7 and 3 m$^2$ g$^{-1}$, respectively. Stratiform aerosols can have MEE values ranging from 1.2 to 3.4 m$^2$ g$^{-1}$ at high altitudes (15–20 km) (Di Girolamo et al., 1995). Husar and Falke (1996) showed that the MEE for fine particles (PM2.5) varied from 4 to 12 m$^2$ g$^{-1}$ for several different sites with an average of 7.4 m$^2$ g$^{-1}$. Feczkó et al. (2002) measured the MEE of ammonium sulfate, ($\text{NH}_4$)$_2\text{SO}_4$, to be 6 m$^2$ g$^{-1}$, and used this value in the estimation of regional climate forcing of aerosols using box model in a rural site in Central Europe. Mallet, Roger, Despiau, Dubovik, and Putaud (2003) calculated the MEE value for ($\text{NH}_4$)$_2\text{SO}_4$ in wet state to be 2.6 m$^2$ g$^{-1}$. Their work also showed that 40% of the light extinction from anthropogenic sources was due to elemental carbon (EC) and organic particles.

For pure water droplets, Dubinsky, Carswell, and Pal (1985) performed a laboratory measurement on the MEE values with different size distributions. They found that aerosols with large modal radius ($r_m = 6 \mu m$), corresponding to a size parameter $x_m = 2 \pi r_m / \lambda$ of 73, exhibited a smaller MEE value of 0.2 m$^2$ g$^{-1}$. Aerosols with smaller modal radius ($r_m = 0.01 \mu m$ and $x_m = 0.12$) showed a larger value of 10.8 m$^2$ g$^{-1}$, and the MEE of aerosols with modal radius of 2.5 $\mu m$ was 0.6 m$^2$ g$^{-1}$.
Table 2
MEE values from other works and from the present simulation using Mie scattering theory

<table>
<thead>
<tr>
<th>Aerosol type</th>
<th>Size parameter $x$</th>
<th>Modal radius (μm)</th>
<th>MEE ($m^2 g^{-1}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$_2$O</td>
<td>73.3</td>
<td>6</td>
<td>0.2</td>
<td>Dubinsky et al. (1985)</td>
</tr>
<tr>
<td></td>
<td>30.6</td>
<td>2.5</td>
<td>0.6</td>
<td>Dubinsky et al. (1985)</td>
</tr>
<tr>
<td></td>
<td>0.126</td>
<td>0.01</td>
<td>10.8</td>
<td>Dubinsky et al. (1985)</td>
</tr>
<tr>
<td>(NH$_4$)$_2$SO$_4$</td>
<td>2.17</td>
<td>0.2</td>
<td>6</td>
<td>Tang (1996)</td>
</tr>
<tr>
<td></td>
<td>3.26</td>
<td>0.3</td>
<td>13</td>
<td>Tang (1996)</td>
</tr>
<tr>
<td>NH$_4$NO$_3$</td>
<td>2.2</td>
<td>0.2</td>
<td>4.5</td>
<td>Tang (1996)</td>
</tr>
<tr>
<td></td>
<td>3.2</td>
<td>0.3</td>
<td>14</td>
<td>Tang (1996)</td>
</tr>
<tr>
<td>EC</td>
<td>1.0</td>
<td>0.09</td>
<td>7.3</td>
<td>Dillner et al. (2001)</td>
</tr>
<tr>
<td></td>
<td>15.46</td>
<td>1.35</td>
<td>1.7</td>
<td>Dillner et al. (2001)</td>
</tr>
<tr>
<td>(NH$_4$)$_2$SO$_4$</td>
<td>1.16</td>
<td>0.1</td>
<td>1.61</td>
<td>Watson et al. (2001)</td>
</tr>
<tr>
<td></td>
<td>2.3</td>
<td>0.2</td>
<td>4.2</td>
<td>Watson et al. (2001)</td>
</tr>
<tr>
<td>Nitrates</td>
<td>11.46</td>
<td>1</td>
<td>1.24</td>
<td>Mallet et al. (2003)</td>
</tr>
<tr>
<td></td>
<td>1.8</td>
<td>0.15</td>
<td>2.6</td>
<td>Mallet et al. (2003)</td>
</tr>
<tr>
<td></td>
<td>0.3</td>
<td>0.028</td>
<td>8–9.65</td>
<td>Mallet et al. (2003)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Aerosol type (simulation)</th>
<th>Size parameter $x^a$</th>
<th>MEE ($m^2 g^{-1}$)</th>
<th>Size parameter $x$</th>
<th>MEE ($m^2 g^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(NH$_4$)$_2$SO$_4$</td>
<td>2.1</td>
<td>5.1</td>
<td>21.3$^b$</td>
<td>0.47</td>
</tr>
<tr>
<td>NH$_4$NO$_3$</td>
<td>2.1</td>
<td>5.2</td>
<td>21.3</td>
<td>0.48</td>
</tr>
<tr>
<td>EC</td>
<td>1.1</td>
<td>7.9</td>
<td>21.3</td>
<td>0.49</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>3.2</td>
<td>5.4</td>
<td>21.3</td>
<td>0.83</td>
</tr>
<tr>
<td>Seasalt</td>
<td>—</td>
<td>—</td>
<td>29.5$^c$</td>
<td>0.52</td>
</tr>
<tr>
<td>NH$_4$NO$_3$</td>
<td>—</td>
<td>—</td>
<td>29.5</td>
<td>0.35</td>
</tr>
<tr>
<td>Soil</td>
<td>—</td>
<td>—</td>
<td>29.5</td>
<td>0.30</td>
</tr>
</tbody>
</table>

$^a$Linear regime.
$^b$Constant regime with $r_e = 1.8 \mu m$.
$^c$Constant regime with $r_e = 2.5 \mu m$.

Pure EC particles are known to exhibit high MEE values occasionally. Experimentally, Jennings and Pinnick (1980) observed an MEE value of 9.5 $m^2 g^{-1}$ for EC particles. Dillner, Stein, Larson, and Hitzenberger (2001) observed this type of MEE increase when they measured the MEE of EC particles in rural aerosols by sampling and spectrometric method. Their results showed that very fine carbon particles with diameters of 0.09 μm exhibited MEE values as high as 7.3 $m^2 g^{-1}$ (for the particles with diameters 2.7 μm, the value was 1.7 $m^2 g^{-1}$). The works of Martins et al. (1998) showed that the MEE of EC particles reached 19.3 $m^2 g^{-1}$. Such high MEE values are ascribable to the presence of internally mixed aerosols with the EC particles as the core (Martins et al., 1998), when the particles undergo the process of aging. Naoe and Okada (2001) reported a sampling analysis of urban atmosphere in Tsukuba, Japan. Their result showed that the fraction of internally mixed soot particles increased with increasing radius; in polluted cases, soot particles dominated in the larger radius range of 0.1–0.2 μm. In this situation, light is effectively absorbed and this increases the MEE.

In Table 2, the values of the MEE parameter are summarized. The upper part of Table 2 shows the values from literatures, and the lower part the results of our present simulation (Section 4.2). It is noted that here the citations from literatures are limited to those with explicit specification of the wavelength and particle sizes. The results in Table 2 will be discussed in Section 5.2.
4. Method of analysis

4.1. Correlation analysis

Here we describe the method of computing the correlation between the PAL data and the ground sampling SPM data. The PAL system in Ichihara city has been continuously operated since December 2002 up to the present. The data from January 2003 to January 2004 have been analyzed to study the correlation, and this analysis has yielded 67 cases (one case represents a continuous 12-h time span) that can be used to evaluate the MEE values, as explained in the following.

Every hour, the PAL system produces approximately 148 backscattered profiles (20 s accumulation time for each signal), each of which is inverted to give an extinction coefficient profile using Fernald’s method (Fernald, 1984) with an assumed value of extinction-to-backscatter coefficient of 30 sr. The resulting profiles are then averaged so that one profile represents the data for the whole 1 h. Considering the plausible range of the extinction-to-backscatter coefficient, an error range of ±6% is estimated for this retrieval process. The sampling time of 160 ns and elevation angle of 38° of the PAL system lead to a vertical resolution of 0.0148 km. For each 0.0148 km step in an altitude range of 0.1–1.8 km, the 1-h average of the extinction coefficient is correlated with the ground-measured, hourly value of SPM concentration. The daytime data (13 points for 06:00–18:00) are correlated separately from the nighttime data (13 points for 18:00–06:00). From the 1-year data set, we found 67 cases that showed the correlation coefficient, $R$, of greater than 0.7. This process, which is consistent with the algorithm that we adopted in our previous study (Lagrosas et al., 2004), ensures the well-mixed feature of the atmosphere near the ground level.

In the next step, a value of MEE is calculated for each of the 67 cases. Obviously, the lower altitude range is preferable since the extinction coefficients are correlated with the ground SPM values. Below 0.3 km, the PAL data suffer from the incomplete overlap between the telescope field-of-view and the laser beam. In this study, we have chosen an altitude range of 0.3–0.5 km. In this range, the MEE values are calculated for each altitude step (note that for each altitude step of 0.0148 km, we have 13-h data of the extinction coefficient and they can be matched with the 13-h data of SPM concentration). The average over 13 ($=0.2/0.0148$) steps gives us the final MEE value for that particular case.

Within a well-mixed boundary layer, the optical properties of the scatterers are relatively stable with height. Examination of PAL data has shown that in Chiba area, boundary layer height can reach as high as 2 km during high-pressure conditions and as low as below 0.3 km during rainy situations. For the 67 cases mentioned above, the value of the extinction coefficient is more or less stable in the 0.3–0.5 km region, regardless of the boundary layer height. This is due to the condition of $R > 0.7$, which ensures the synchronous change of the atmosphere for altitudes near the ground level. For the most part (49 cases) of the 67 cases, the boundary layer height is higher than 0.5 km, while for the remaining cases, the boundary layer is not clearly detected, or the height is below 0.5 km.

4.2. Simulation of MEE in the linear and constant regimes

The values of MEE can easily be estimated for small or large values of the particle diameters (Dubinsky et al., 1985). When the radius, $r$, is large, and the size parameter $x = 2\pi r/\lambda$ is greater than 20, the extinction...
Table 3
Values of the particle size parameter, radius and slope $s$ for different aerosol components in the linear regime

<table>
<thead>
<tr>
<th>Aerosols</th>
<th>Refractive index $m$</th>
<th>Size parameter $x_m$</th>
<th>Particle radius $r_m$ (µm)</th>
<th>Slope $s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_4$NO$_3$</td>
<td>1.53–0.005i</td>
<td>4.3</td>
<td>0.36</td>
<td>1.01</td>
</tr>
<tr>
<td>(NH$_4$)$_2$SO$_4$</td>
<td>1.53–0.005i</td>
<td>4.3</td>
<td>0.36</td>
<td>1.01</td>
</tr>
<tr>
<td>EC</td>
<td>1.9–0.6i</td>
<td>2.2</td>
<td>0.19</td>
<td>1.32</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>1.33</td>
<td>6.6</td>
<td>0.55</td>
<td>0.61</td>
</tr>
</tbody>
</table>

efficiency $Q_{\text{ext}}$ nearly equals to 2 (hereafter this is referred to as “constant regime”). Thus, we obtain

$$MEE = \frac{3}{2\rho} \frac{\int_{r_1}^{r_2} r^2 n(r) \, dr}{\int_{r_1}^{r_2} r^3 n(r) \, dr} = \frac{3}{2\rho} \frac{1}{r_e},$$

where $r_e$ stands for the effective radius of the aerosols. When the radius is small, on the other hand, there is a region where $Q_{\text{ext}}$ is nearly proportional to $x$ (“linear regime”). This is valid when $x < x_m$, where $x_m$ is the size parameter corresponding to the first peak of the $Q_{\text{ext}}$ curve. In this case, we have

$$MEE = \frac{3\pi s}{2\rho \lambda},$$

where $s$ is a dimensionless constant representing the slope, i.e.,

$$s = \frac{Q_{\text{ext}}}{x}.$$  

Eqs. (3) and (4) are useful for studying the behavior of MEE as a function of the aerosol size parameter.

The linear regime is examined for the cases of completely dry aerosols (NH$_4$NO$_3$, EC and (NH$_4$)$_2$SO$_4$) and wet aerosol (H$_2$O). These are the main components of the fine particles as observed in ground sampling measurements explained in Section 5.1. Table 3 shows the corresponding values of $x_m$, $r_m$ and $s$. Note that the wavelength $\lambda$ is 532 nm for the present case. From the table, it is clear that EC and H$_2$O give the smallest and highest $x_m$ values, respectively. Depending on the type of aerosols, the value of $r_m$ can change from 0.2 to 0.5 µm. Thus, a large portion of the aerosols classified as the fine particles (with diameters less than about 2.5 µm) belong to the linear regime, though the remaining part exhibit size parameters slightly larger than $x_m$. The results of the present simulation are summarized in the lower part of Table 2 (for both linear and constant regimes), and will be discussed in Section 5.2.

5. Results and discussion

5.1. Correlation study

The characteristics of the aerosols in Chiba area were studied by means of the ground sampling from September 1998 to February 2002. According to these measurements, the dominant components of fine
particles are ammonium sulfate ((NH₄)₂SO₄), elemental carbon, and ammonium nitrate (NH₄NO₃), contributing, respectively, to about 35%, 25% and 19% of the total fine particle concentration (Yabuki, 2003). In the case of coarse particles, the dominant components are sea salt, ammonium nitrate and soil, giving 37%, 20% and 20% of the total coarse concentration, respectively. Thus, it is likely that the PAL signals are due to backscattering from these types of aerosol particles, and the MEE values are representative of the combination of these aerosol components. As for the fine EC aerosols in urban areas in Japan, relatively high mass concentrations from 17% to 25% were also reported by Höller, Tohno, Kasahara, and Hitzenberg (2002), Ma, Kasahara, Tohno, and Hwang (2001), Ohta, Hori, Yamagata, and Murao (1998) and Saitoh, Sera, Hirano, and Shirai (2002) on the basis of sampling measurements.

The PAL data from January 2003 to January 2004 were reviewed and analyzed. The results were compared with the ground-measured SPM concentration, as explained in Section 4.1. Data with optically thick clouds were excluded from the analysis. Throughout the year, a good correlation has been found between the two parameters when the PAL data showed either of the following characteristics: (i) heavy backscatter signal near the ground level, due to the presence of a distinctive ground layer, and (ii) a relatively clear atmosphere during the nighttime of autumn and winter. In case (i), the boundary layer develops under the conditions of high relative humidity or an onset of rain event, and generally, the transition between nocturnal/daytime boundary layers is not very conspicuous. In case (ii), on the contrary, this type of transition of the boundary layer is often found. Typical cases are discussed in the following subsections.

5.1.1. Relatively clear atmosphere

Fig. 1a shows the range-corrected PAL signal observed during the nighttime on 13–14 December 2003. The lower atmosphere was relatively clear. During this time interval, the SPM concentration decreased from 0.05 to 0.002 mg m⁻³. From 18:00 to 20:00 (local time) on December 13, the relative humidity increased from 47% to 79%. It decreased to 43% from 20:00 to 22:00 and gradually increased to 51% up to 06:00 of the next day (Fig. 1b). The temperature in this time interval ranged from 6.7 to 11.4 °C. From these ground-measured data, it is likely that aerosol growth due to water vapor was insignificant in the lower atmosphere. Thus, this represents a case wherein the observed aerosol is relatively dry. As seen from Fig. 1c, large values are found for the correlation coefficient between the extinction coefficient and SPM concentration in the height range of 0.1–1.8 km. The good vertical mixing is brought about by the strong wind (1.7–6.8 m s⁻¹) coming from the N, NW, and NNW (from industrial areas) directions. The average MEE value in the 0.3–0.5 km region is 11.4 m² g⁻¹.

5.1.2. High backscattering case

Fig. 1d is the daytime lidar data on 1 April 2003, showing a high backscattering from the lower atmosphere. An arbitrary criterion of signal level of 200 (see the scale attached to Fig. 1d) is employed here to categorize a range-corrected signal into the high-backscattering case. The relative humidity during this time interval dropped from 87% to 28% from 06:00 to 12:00 and increased to 66% in the evening (Fig. 1e). The temperature increased from 10.7 to 17.3 °C from 06:00 to 12:00 and gradually decreased to 10.6 °C at 18:00. The wind directions in this 12-h interval were E, NE and ESE. The wind speed ranged from 2 to 5.9 m s⁻¹. Due to the low humidity, it is likely that fairly dry aerosols caused the high backscatter signal near the ground level. In this case, a high correlation is found between the extinction
Fig. 1. Observed results for two types of typical cases: (a)–(c) are for the data on 13–14 December 2003, and (d)–(f) on 1 April 2003. (a) Range-corrected PAL data (13–14 December 2003) with a low backscatter in the lower atmosphere (clear atmosphere situation). (b) Relative humidity for the 12-h interval. (c) Correlation coefficient $R$ between ground-measured SPM concentration and extinction coefficient for each altitude between 0.1 and 1.8 km. For the 0.3–0.5 km region, the average MEE value is $11.4 \text{ m}^2 \text{ g}^{-1}$. (d) Range-corrected PAL data on 1 April 2003 showing a high backscatter in the lower atmosphere, (e) temporal variation of the ground-measured relative humidity, and (f) correlation coefficient $R$ between the ground-measured SPM concentration and the extinction coefficient. The resulting MEE is $8.1 \text{ m}^2 \text{ g}^{-1}$.
Fig. 2. Plot of the MEE values from the present simulation and other works (shown in Table 2) as a function of the size parameter. In the framework of the size effect, high and low MEE values can be attributed to fine and coarse particles, respectively.

coefficient and the SPM concentration (Fig. 1f), together with a relatively constant extinction coefficient profile in the 0.3–0.5 km height range. The average MEE value in this height range is 8.1 m² g⁻¹.

5.2. Simulation results of MEE

The results of the present simulation on the MEE values are summarized in the lower part of Table 2. For all the chemical species considered here, larger MEE values (5–8 m² g⁻¹) are found for the linear regime (0 < x < xₘ), and smaller values (0.3–0.8 m² g⁻¹) for the constant regime (x > 20). For the intermediate case between the linear and constant regimes, our simulation indicates that intermediate values are found also for MEE. The MEE values show only limited variations with regard to the aerosol chemical species. Thus, roughly speaking, high and low MEE values can be associated with the dominance of fine and coarse particles, respectively. This is clearly seen from Fig. 2, where MEE values from both the present simulation and previous works are plotted against the size parameter, x. The inverse correlation between x and MEE can be referred to as the “size effect”, and this scheme applies when the dynamic effect of “rapid growth” (see below) is not dominant.

According to Schwartz (1996), as the particle radius becomes larger, i.e., r > 1 μm, the efficiency of light scattering decreases, resulting in low MEE values. This is consistent with our results in the constant regime. Also, it is noted that the experimental results observed by Dubinsky et al. (1985) are in agreement with the present results, in both the linear and constant regimes.

In Fig. 2, deviation from the “size effect” trend is found for MEE values of ammonium nitrate and ammonium sulfate. These are the data derived from the theory of Tang (1996) that dealt with “rapid growth” mainly within the linear regime. This growth is caused by the water condensation onto hygroscopic particles, and the MEE value shows rather steep increase as the size parameter increases only slightly. Although most of the correlations between the PAL and SPM data can be explained in the framework of the “size effect” mentioned above, the daytime/nighttime statistics give some indication of the manifestation of this “rapid growth” effect (Section 5.4).
Fig. 3. (a) Average daytime MEE values from January to December 2003. During summer, MEE values are relatively small compared to other seasons. (b) Correlation between the MEE values and the coarse particle mass concentration measured by the β-ray and TEOM systems from June 2003 to January 2004. In this time interval, the TEOM data are selected so that all the PAL data used to compute the MEE value have the corresponding TEOM data.

5.3. One-year statistics of MEE

Within the 1-year time interval, a total of 67 cases satisfying either of the two conditions mentioned above were found to have a good correlation with SPM concentration. Here, “one case” indicates the PAL data for a continuous time span of 12 h. Of the 67 cases studied here, high (MEE > 5 m² g⁻¹) and low (MEE < 5 m² g⁻¹) MEE values comprise a total of 45 and 22 cases, respectively. In terms of the monthly average of the daytime MEE values, Fig. 3a shows that during summer, MEE values are lower compared to the winter values. According to the sampling measurements described above, the months of summer were the times when coarse particles dominated in the lower atmosphere. During this time of the year, the volume ratio of coarse particles ranged from 50% to 61%. During winter, the volume ratio could reach as low as 17%. This result is consistent with the data from the Angstrom parameter in Chiba area measured using a ground-based sun photometer (Fukagawa, Yabuki, Kuze, & Takeuchi, 2002). During summer, the values of the parameter are as low as 0.5, while during winter, the values are as high as 2, suggesting the dominance of coarse and fine particles for summer and winter, respectively. In addition, an inverse relationship is found between the MEE value and the coarse particle concentration from June 2003 to January 2004 (Fig. 3b). This also supports the idea that the dominance of coarse particles leads to the decrease in the MEE value.

5.4. Rapid growth due to high relative humidity

In the troposphere, changes in the aerosol size distribution are normally caused by condensation and coagulation processes, which have timescales of less than 10 h (Seinfeld & Pandis, 1998). In particular, relative humidity in the lower atmosphere is an important factor governing the particle sizes. Tang’s visibility model (1996) computed the light scattering by hygroscopic aerosols in the linear regime. According to the theory, the phase transformation, growth in size, and evaporation take place when the relative humidity changes, leading to an order of magnitude change in the scattering coefficient.
Table 4
Average mass extinction efficiency (in m$^2$ g$^{-1}$) and relative humidity (RH) from January 2003 to January 2004

<table>
<thead>
<tr>
<th></th>
<th>Relatively clear</th>
<th>Average RH</th>
<th>High backscatter</th>
<th>Average RH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Daytime</td>
<td>5.7 (2.2)$^a$</td>
<td>56%</td>
<td>4.0 (2.6)</td>
<td>71%</td>
</tr>
<tr>
<td>Nighttime</td>
<td>12.1 (6.0)</td>
<td>74%</td>
<td>9.4 (6.1)</td>
<td>81%</td>
</tr>
</tbody>
</table>

$^a$One standard deviation.

In our case, this type of dynamic “rapid growth” is suggested from the average extinction coefficient and MEE data. The daytime data (31 cases) show that the peak values of the extinction coefficient below 1.8 km are in a range of (2.5–5) $\times$ 10$^{-5}$ m$^{-1}$, while those of the nighttime (36 cases) are in a range of (1–5) $\times$ 10$^{-4}$ m$^{-1}$. As a result, the nighttime MEE values are larger than the daytime values. Table 4 summarizes the average and standard deviations of MEE for the 67 cases mentioned above. The statistics of relative humidity indicate that the daytime averages (56% and 71% for relatively clear case and high backscatter case, respectively) are smaller than the nighttime averages (74% and 81%). The MEE data in Table 4 show that the nighttime averages (12.1 and 9.4 m$^2$ g$^{-1}$) are more than twice that of the daytime data (5.7 and 4.0 m$^2$ g$^{-1}$). This increase in MEE is consistent with Tang’s work (1996), i.e., a small increase in radius (1.25–1.75 times the dry radius) brought about by an increase in relative humidity (40–90%) causes increase in MEE values by a factor of 2 to 3. The high standard deviations of MEE (shown in parenthesis in Table 4) suggest the presence of aerosols of different types and/or sizes in the ground layer.

In the present study, the MEE values are derived by studying the dataset in a 12-h time period. This rather long time period is unavoidable since the $\beta$-ray SPM data are provided for every hour. In order to study the dynamic behavior of the aerosol growth, it is desirable to have a ground-sampling instrument that can be operated in a shorter time scale.

5.5. Case studies for high and low MEE values

High MEE values are observed when the atmosphere has a clear and good development of either daytime or nocturnal boundary layer. In this condition, young aerosols with smaller diameters are likely to dominate in the layer. Presumably these are the aerosols with MEE values in the linear regime described in Section 4.2. The extremely high values (MEE > 10 m$^2$ g$^{-1}$) can be attributed to the presence of very fine particles in the form of the internal mixture. If EC is present in the core, the value of MEE can be as high as 30 m$^2$ g$^{-1}$ (Martins et al., 1998). Also, when sulfate particles have other components (e.g. nitrates) with them, the MEE can reach as high as 16 m$^2$ g$^{-1}$ (Ten Brink, Veefkind, Waijers-Ijpelaan, & Van Der Hage, 1996). An example of this high MEE situation is shown in Figs. 4a–c, from PAL data on 7 October 2003. The wind directions on this day were from NE and NNE, the locations of the major highways. The wind speed was relatively low, ranging from 0 to 1.9 m s$^{-1}$. From the wind directions, it is likely that relatively fine particles from vehicle emissions in the Chiba city area were observed. During this time interval, the measured coarse SPM concentrations were low, ranging from 6 to 22 $\mu$g m$^{-3}$ (Fig. 4b). The altitude dependence of the MEE value is shown in Fig. 4c, resulting in a relatively large MEE value of 17.5 m$^2$ g$^{-1}$. The clear formation of the boundary layer shows that the atmosphere is characterized by a good vertical transport and mixing. In this case, fine particles can reach higher altitudes and can remain there for a long time.
In the case of low MEE values, the prevailing feature of the atmosphere observed from the lidar data is the absence of clear transition from daytime to nocturnal boundary layers. At the same time, the values of the relative humidity are often higher than those in the high MEE cases. This means that the aerosol is likely to be composed of aged, coarse mode particles, and their growth due to relative humidity leads
to the decrease in light scattering efficiency (this is a case for the “size effect”, and not explained by
the “rapid growth” scheme given in Section 5.4). In terms of the simulation results, aerosols with MEE
values below 1 m$^2$ g$^{-1}$ belong to the constant regime. Aerosols with MEE values below 5 m$^2$ g$^{-1}$ belong
mainly to the intermediate region between the linear and constant regimes.

Figs. 4d–f show an example of the low MEE case. The PAL data show that between 06:00 and 08:00
on 26 September 2003, the boundary layer is not well defined. This was presumably associated with the
high relative humidity, which was around 94%. During this time interval, the wind speed varied from
1.1 to 3.1 m s$^{-1}$. Ground-measured rainfall was 0 mm, indicating that the main composition of air was
uncondensed water vapor or mist. In the middle of the day, the boundary layer became evident, implying
that vertical mixing height was below 1 km. The wind speed and the relative humidity varied from 0.6 to
2.6 m s$^{-1}$ and from 72% to 88%, respectively. Near the sunset, the replacement of the daytime boundary
layer with the nocturnal boundary layer was not evident. In this time interval, the relative humidity
increased from 69% to 77% and the wind speed ranged from 1.5 to 1.7 m s$^{-1}$. The wind directions
during the 12-h interval were a combination of ESE and E (from rural areas), NE, WNW, W (from sea
areas), suggesting that the detected aerosols were a combination of maritime, soil, and soot. The altitude
dependence of MEE is shown in Fig. 4f. The small average value of 0.74 m$^2$ g$^{-1}$ suggests the dominance
of coarse particles. This is supported by the coarse SPM concentration as shown in Fig. 4d. When the
TEOM value is subtracted from the SPM concentration, the difference (representing aerosols between
2.5 and 10 μm) is 65% of the total mass concentration.

Figs. 4b and e show that coarse SPM concentrations on 26 September 2003 (Fig. 4d) are higher than
the coarse concentrations on 7 October 2003 (Fig. 4b). In fact, the average coarse SPM concentration of
the former (48 μg m$^{-3}$) is almost four times that of the latter (12.5 μg m$^{-3}$). This evidently supports the
interpretation that low MEE values are the results of high concentration of coarse particles.

6. Conclusions

This study has shown that the continuous operation of the portable automated lidar (PAL) system has
the capability of profiling atmospheric aerosols. When lidar data are correlated with ground-measured
SPM data, good correlations are obtained when the PAL signal shows a heavy backscatter near the ground
level or when there is a relatively clear atmosphere. The correlation study using 12-h datasets shows that,
on the average, the MEE values change in the range of 4–12 m$^2$ g$^{-1}$. The average daytime MEE value
is smaller than the nighttime MEE value. On the basis of the simulation study, it is likely that high
MEE values indicate the dominance of fine particles, while low MEE values suggest the dominance of
coarse particles (size effect). These findings are supported by both the ground-sampling data (especially
the TEOM data) and Angstrom coefficient from sun-photometer observations. It is also pointed out that
under high relative humidity conditions, the dynamic scheme of “rapid growth” may play a role, leading to
increase in the MEE values with the slight increase in particle diameters. For this “rapid growth” scheme,
further work is required to study the dynamical behavior of the aerosol particles in the lower troposphere.
As a whole, we have demonstrated the usefulness of the MEE data, as obtained by combining lidar data
with ground-sampling data, in deriving the information on the size of aerosol particles.

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References


