Mie-scattering simulation and measurement of mass extinction efficiency from portable automated lidar and suspended particulate matter measurements

Gerry Bagtasa [a],Nofel Lagrosas [a], Hiroaki Kuze [a], Nobuo Takeuchi [a], Shunsuke Fukagawa [a], Yotsumi Yoshii [a], Suekazu Naito [b], Masanori Yabuki [c]

[a] *Center for Environmental Remote Sensing, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan*

[b] *Chiba Prefectural Environmental Research Center, 1-8-8 Iwasaki-nishi, Ichihara 290-0046, Japan*

[c] *National Institute of Polar Research, 9-10 Kaga 1-chome, Itabashi-ku, Tokyo 173-8515*

Abstract

The continuous operation of the portable automated lidar (PAL) and measurement of suspended particulate matter (SPM) results in the observation of the diurnal changes of the mass extinction efficiency (MEE). MEE is the ratio of the extinction coefficient to the SPM concentration which relates the amount of mass to the optical extinction of aerosols. Our results show that in the height region of 300 – 500 m, the daytime MEE values are smaller than the nighttime MEE values. This can be attributed to the aerosol growth (rapid growth) due to the higher relative humidity during nighttime.

Using the Mie scattering theory, a simulation study of MEE is also undertaken by computing the extinction coefficient from the 6-year averaged air sampling measurements in Chiba University. Hygroscopic effects on aerosols are considered in the simulation and results show that MEE increases with RH, in agreement with the observation stated above. When this type of direct effect of RH (rapid growth) is irrelevant, high and low MEE values can be ascribed to fine and coarse particles, respectively, for both measurement and simulation.

In addition, MEE values are computed for individual aerosol components (elemental carbon (EC), organic carbon, NH_4SO_4, NH_4NO_3, sea salt, soil, EC-NH_4SO_4 mixture), with EC having the highest computed MEE of 7.3 m^2g^{-1} and NH_4NO_3 with the lowest MEE of 1.2 m^2g^{-1}.

Keywords: Mass Extinction Efficiency, Lidar, Tropospheric aerosol, Mie scattering theory

I. INTRODUCTION

Aerosols and clouds play an important role in the radiation budget of the atmosphere. Atmospheric sensing using a compact, continuously operated lidar can lead to the real-time observation of the dynamics of the aerosols and clouds. One of the candidates of this type of remote sensing is the portable automated lidar (PAL) system, which is capable of real-time and unattended monitoring of the atmosphere.[1)] In this paper, we discuss the application of the PAL data to the monitoring of tropospheric aerosols.[2)] The inversion of the PAL data yields the vertical profile of the aerosol extinction coefficient. In the ground sampling, the aerosol quantity is often measured by the β-ray method as the suspended particulate matter (SPM) concentration. If the extinction coefficient values from the PAL data are matched with the ground-based SPM concentration measurements, the mass extinction efficiency (MEE) of the aerosols can be calculated.

II. Mass Extinction Efficiency

The MEE parameter links the mass amount to the optical extinction of the aerosols. Mathematically, the MEE is given by

$$MEE = \frac{\pi \int_{r_1}^{r_2} r^2 Q_{ext}(r,\lambda,m) n(r) dr}{\frac{4}{3}\pi\rho \int_{r_1}^{r_2} r^3 n(r) dr} \qquad (1)$$

where $Q_{\rm ext}(r,\lambda,m)$ is the extinction efficiency, r is the radius, λ is the wavelength, m is the refractive index, ρ is density, and $n(r)$ is the particle size distribution. The high and low values of the MEE are generally attributed to the presence of fine (roughly equivalent to PM2.5) and coarse particles (PM10 except the PM2.5 components). However, when the diameters of fine aerosols grow with relative humidity (RH), the efficiency of the particles to scatter light increases and this also leads to the increase in the MEE.

Figure 1 shows MEE values of different aerosol types as a function of size parameter x taken from various literatures.[2)]

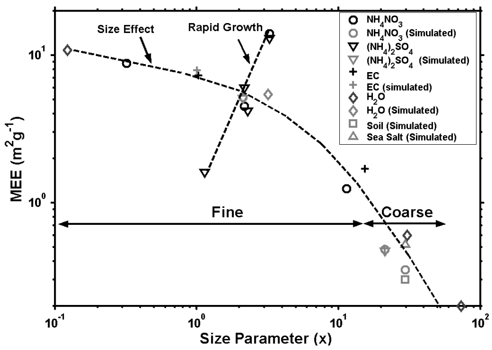


Fig. 1 MEE values as a function of size parameter from literature

It can be seen that MEE values decrease with increasing size parameter, hence the terminology of size effect. It has been pointed out that the lens effect associated with the particles of internal mixture greatly enhances the value of MEE.[2)] This is especially important for the soot aerosols surrounded by water (solution) droplet. On the other hand, MEE of a particular aerosol component may change due to a change in RH, and this is called the rapid growth. This latter effect is related to the rapid increase of $Q_{\rm ext}(r,\lambda,m)$ in a particular range of the size parameter (approximately $1<x<5$).

III. Setup

a) PAL System

The PAL system has been developed by CEReS, Chiba University, and operated at the Chiba Prefectural Environmental Research Center at Ichihara city (35.52N, 140.07E, about 40 km southeast of Tokyo). It is a compact Mie lidar system that can operate continuously and unattended for long periods of time. The system is equipped with an automatic re-alignment system that readjusts the laser direction every 15 minutes so that the proper system alignment is always maintained. Table 1 shows a more detailed specification of the PAL system.

Table 1: PAL Specification

Transmitter	
Laser	LD-pumped Q-switch Nd:YAG
Wavelength	532 nm
Repetition Rate	1.4 kHz
Laser Pulse Energy	15 μJ
Laser Beam Divergence	50 μrad
Receiver	
Telescope Diameter	20 cm
Telescope Type	Cassegrain
Field of View	0.2 mrad
Detector	PMT
Model	HPK-R1924P
Quantum Efficiency	10% - 25%

b) Low volume impactor

A three-stage low volume impactor (LVI) is used to characterize the local aerosol size distribution used for the present simulation. The air sampler is located at Chiba University and is able to measure mass concentration of elemental carbon (EC), organic carbon (OC), ammonium sulfate (NH_4SO_4), ammonium nitrate (NH_4NO_3), sea salt, and soil with aerodynamic sizes of less than 2.5 μm (PM2.5), 2.5 μm to 10 μm (PM10 excluding PM2.5) and greater than 10 μm. Only the measurements at PM2.5 and PM10 stages are used in the simulation.

IV. RESULTS

a) PAL - SPM counter

Each data set is composed of 12 points (one point per hour): the daytime data set is taken from 0600H to 1800H and nighttime data from 1800H to 0600H the following day. Data from January of 2003 to January of 2004 are analyzed. When there is a good correlation between the lidar-derived extinction coefficient and the ground-based SPM concentration measurements, the MEE for that data set is computed. High correlation coefficient ($R>0.7$) is usually obtained when there is a relatively clear atmosphere or high backscattered signal is detected.

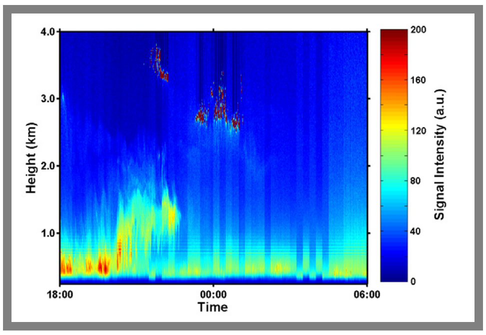


Fig. 2 Clear atmosphere range corrected Lidar data (13-14 Dec2003)

Figure 2 shows the range corrected PAL data taken on 13-14 of December 2003: this is considered clear data since no optically thick clouds are present near the ground. Fig. 3 shows the computed extinction coefficient from the same data set plotted against the SPM concentration measurement. The observed MEE value is 11.3 m^2g^{-1}.

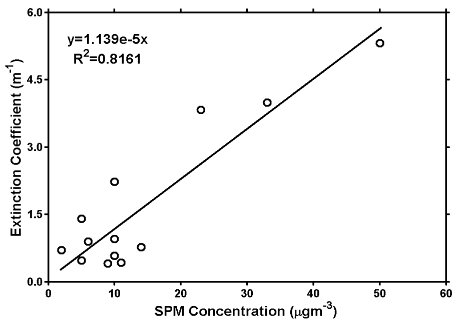
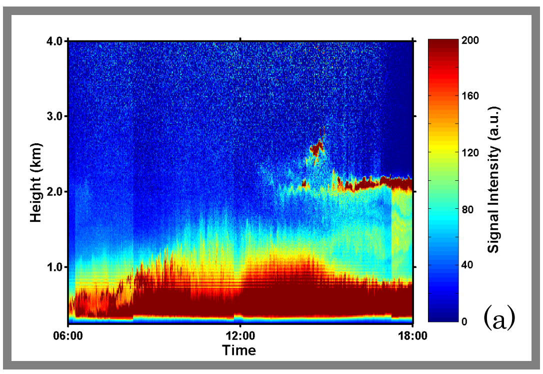


Fig. 3 Extinction coefficient vs. SPM concentration (13-14 Dec2003)

Figure. 4(a) shows lidar data of 1 April 2003. This data represents a high backscatter case due to the high signal intensity detected near the ground. Fig. 4(b) shows the extinction coefficient plotted with SPM concentration giving an MEE value of 8.1 m^2g^{-1}.



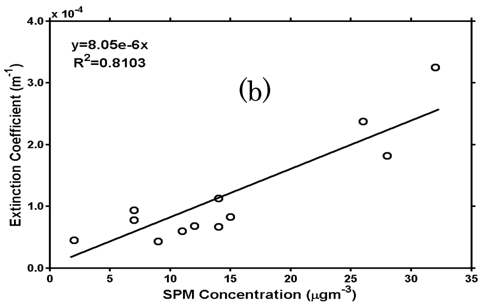


Fig. 4 (a) Range corrected PAL data, and (b) extinction coefficient vs. SPM concentration (1April 2003)

Table 2 shows the average MEE values and average relative humidity for all the analyzed data sets.

Table 2 Average (standard deviation) mass extinction efficiency (in m^2g^{-1}) and relative humidity (RH)

	Daytime	Nighttime
Relatively Clear	7.9 (5.2)	13.0 (6.2)
Average RH	56%	74%
High Backscatter	5.4 (4.0)	11.0 (7.6)
Average RH	71%	81%

Our results show that MEE values range from approximately 4-13 m^2g^{-1}. Generally, nighttime data sets yield a higher MEE values than the daytime data: this can be attributed to the Rapid Growth, due to the higher observed average relative humidity during nighttime.

Figure 5 shows the variation of the average extinction coefficient with relative humidity. Below around 70% RH, the lidar-derived extinction coefficient values appear not to change very much, whereas above 70% RH, it is seen that the extinction coefficient increases with RH. This is attributed to the aerosol growth (rapid growth).

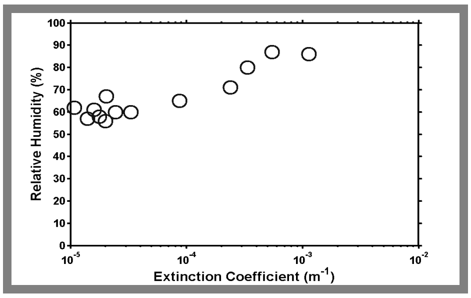


Fig. 5 Relative Humidity vs. Extinction Coefficient

Figure 6 shows the observed MEE plotted against the coarse particle concentration. The MEE value decreases with increasing amount of coarse particles, which correspond to the size effect shown in fig. 1.

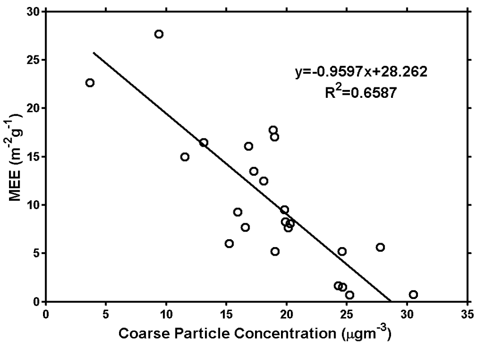


Fig. 6 MEE vs. Coarse SPM concentration

b) Simulation based on Mie scattering theory

The 6-year data of mass concentration measurements from the LVI is used to model the local aerosol size distribution in the Chiba city area for each of the following aerosol components: EC, OC, NH_4SO_4, NH_4NO_3, sea salt, soil and EC-NH_4SO_4 mixture.

From the derived size distributions, MEE value for each aerosol component can be computed using Eq. (1). Table 3 shows the result for each aerosol component. It is noted that although the MEE of a specific aerosol component remains constant regardless of the size distribution, the total MEE varies with the size distribution.

Table 3: Simulated MEE for each aerosol type

Aerosol Type	Computed MEE (m^2g^{-1})
EC	7.290
OC	2.092
NH_4SO_4	3.516
NH_4NO_3	1.194
Sea Salt	1.273
Soil	1.224
EC – NH_4SO_4 mixture	4.606

Figure 7 shows the total MEE of the 6-year average mass concentration measurement by the LVI with varying amounts of RH. There is a significant increase in the MEE value when the RH reaches 70%, which is the point where aerosols start to be coated with water. MEE value increases up to almost 300% when RH reaches 90%. This result verifies the higher MEE values observed during nighttime.

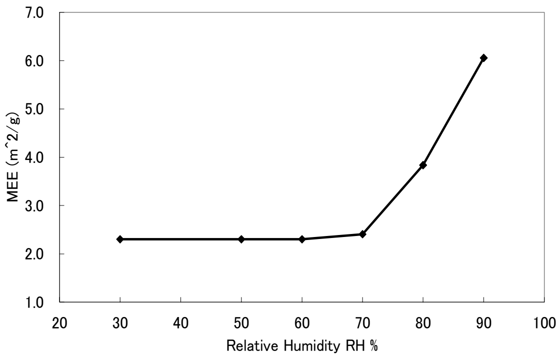


Fig. 7 MEE vs. RH Simulation

V. CONCLUSION

This study has shown that the continuous operation of the portable automated lidar (PAL) system has the capability of observing the dynamics of atmospheric aerosols. The correlation study shows that MEE observed during nighttime is generally higher than MEE observed during daytime, suggesting the influence of higher average relative humidity during nighttime. Simulations using the Mie Scattering theory support this observation: by varying the amount of relative humidity, it is shown that MEE increases with RH. Also, our simulation reveals elemental carbon (EC) as one having the highest MEE value of 7.3 m^2g^{-1}: this result is to be expected since EC has one of the highest imaginary refractive index, consequently the highest extinction.

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