Mass Extinction Efficiency for tropospheric aerosols from Portable Automated Lidar and β-ray SPM counter

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ABSTRACT

Mass Extinction Efficiency (MEE) is defined as the ratio of extinction coefficient to atmospheric aerosol mass concentration. It relates the amount of mass to the optical extinction of aerosols. Knowledge of MEE is important in the estimation of aerosol contribution to the radiation budget in the Earth environment. In this study, MEE is observed using a Portable Automated Lidar (PAL), a system developed by Center for Environmental Remote Sensing (CEReS), Chiba University, together with a ground-based β-ray SPM counter. The influence of humidity is considered on the mass concentration of the SPM measured by the β-ray counter. Also, Mie scattering simulation using area aerosol size distribution models has been undertaken to explain the observed MEE values.

1. INTRODUCTION

There has been a growing concern in the global warming trend of terrestrial atmosphere. Aerosols play an important role in the scattering and absorption of solar heat [(1)]. One of the key parameters in the computation and/or simulation of Earth's radiation budget is the mass extinction efficiency (MEE) [(2)]. This parameter relates the mass amount of aerosol particles to the optical extinction, and it is defined by the following equation:

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

where $Q_{ext}(r,\lambda,m)$ is the extinction efficiency, r is the particle radius, λ is the wavelength, m is the refractive index, ρ is particle 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 particulate matter (PM) 2.5) and coarse particles (PM10 excluding the PM2.5 component), respectively.

2. METHOD AND SETUP

Extinction coefficient is derived from the lidar data with Fernald's inversion method with the lidar ratio (S_1) for 532 nm of 30 sr, as used in other lidar studies in Chiba. Normally the S_1 value changes in a range of 20-40 sr, yielding $\pm$6% error in the retrieved extinction coefficient. The PAL system [(3)], located at the Chiba Prefectural Environmental Research Center at Ichihara City (35.52N, 140.07E, about 40 km southeast of Tokyo), is a compact Mie lidar system operating at 532 nm with laser pulse energy of 15 μJ and repetition rate of 1.4 kHz. The diode-pumped, solid-state (DPSS) Nd:YAG laser sits on the side of a 20 cm Cassegrainian telescope. The system has a co-axial configuration and takes slant path measurements at 38^0 elevation. It is equipped with an automatic realignment system that adjusts laser direction every 15 min to maintain proper alignment [(3)]. The extinction value averaged for 1 h in the height range of 250 to 750 m is used to represent the aerosol extinction coefficient in the mixed boundary layer. Data under rainy and cloudy conditions have been excluded from the analysis. The values of MEE are derived for datasets that

exhibit high correlation (with correlation coefficient *R* higher than 0.9) between extinction coefficient and SPM data (here a total of 179 h out of 608 h observations).

Table 1: PAL Specification.

Transmitter	
Laser	LD-pumped Q-switch Nd:YAG
Wavelength	532 nm
Laser Pulse Width	50 ns
Repetition Rate	1.4 kHz
Laser Pulse Energy	15 μJ
Beam Divergence	50 μrad
Receiver	
Telescope Diameter	20 cm
Telescope Type	Cassegrainian
Field of View	0.2 mrad
Band pass filter	5 nm
Detector	PMT
Model	HPK-R1924P
Quantum Efficiency	10% - 25%
Range resolution	24 m

The β-ray SPM counter (Shimadzu AAMS-4160), located approximately 70 m from the lidar site, measures the mass concentration of ambient aerosols. The counter measures the total SPM in the concentration range of 0.001 to 10 mg/m^3. The apparatus is placed in an observatory (9 m X 9 m X 3 m), where a 6 m-long inlet glass tube (5.5 cm inner diameter) is located partly outside (3.9 m) and partly inside (2.1 m). The flow rate is 100 l/min. Downstream, the air flow is funneled into a 2.3 m flexible tube (14 mm inner diameter, 15 l/min flow rate) which is connected to the SPM counter. Aerosol particles in the air flow are then collected on a glass fiber filter for 50 min, then the mass concentration is determined from the β-ray transmission measurement.

The influence of humidity [4] is considered on the concentration of the SPM measured by the β-ray counter. In general, the humidity condition is different between inside and outside the observatory. During the sampling process prior to the measurement, changing relative humidity (RH) conditions result in a change of aerosol water content, hence a change in mass concentration. The temperature and humidity were measured both inside the counter and just outside the observatory. From the measured RH values, correction is made such that the SPM mass concentration in equilibrium with the ambient condition is derived. This is achieved by means of Tang's model[4] of aerosol growth with relative humidity by considering the hygroscopic effect of common aerosol species: sea salt, $(NH_4)_2SO_4$, NH_4NO_3 and $NaNO_3$. The fraction of each component is derived from air sampling measurements that are regularly conducted at the end of every month. Changes in the measured SPM mass concentration result in changes in the value of MEE.

In addition, Mie scattering simulations using the urban, maritime [5] and Chiba-area [6] size distribution models verify the dependence of MEE value on particle size. The Chiba-area size distribution model is based on measurements derived from a 9-stage Andersen sampler [6]. High and low MEE values are computed for fine and coarse dominated particles, respectively.

3. RESULTS

3.1 PAL and β-ray SPM counter

Figure 1 shows the MEE obtained from measurements conducted in September 2005. Both corrected and uncorrected values are plotted against outside RH. During this observation period, the average value of RH is 78% outside the observatory and 48% inside the counter. The corrected MEE decreases at higher RH values, mainly due to the fact that high RH results in the increase of aerosol size, and therefore the size distribution becomes dominated by coarse particles. The uncorrected MEE increases with RH since the particles are dried in the course of the measurement, leading to lower mass concentration and higher MEE.

Table 2 shows average MEE value and the corresponding standard deviation before and after the

correction.

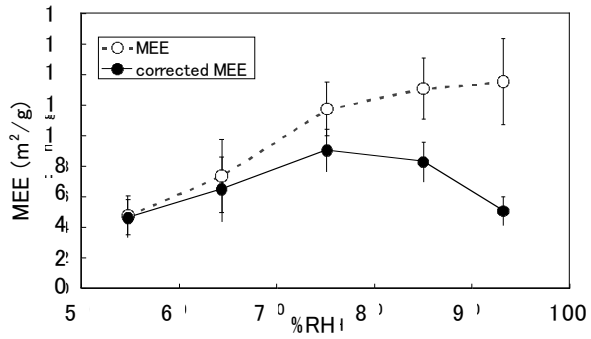

Figure 1 MEE and Corrected MEE vs. outside RH.

Table 2 MEE correction statistics (in m^2/g).

	MEE	corrected MEE
Average	11.4	7.36
Standard deviation	6.2	3.8
Maximum value	34.6	15.8

In addition to the β-ray SPM counter, a Tapered Element Oscillating Microbalance (TEOM) was used to discriminate between fine and coarse aerosols. This provides data on the particles with diameters less than 2.5 μm at 50% cut-off [4] (PM2.5). By subtracting the measurement of the TEOM from that of the β-ray counter, it is possible to obtain the mass concentration of the coarse particles. Figure 2 shows the correlation plot of MEE with the coarse particle mass concentration. As coarse particles dominate, the MEE value tends to decrease.

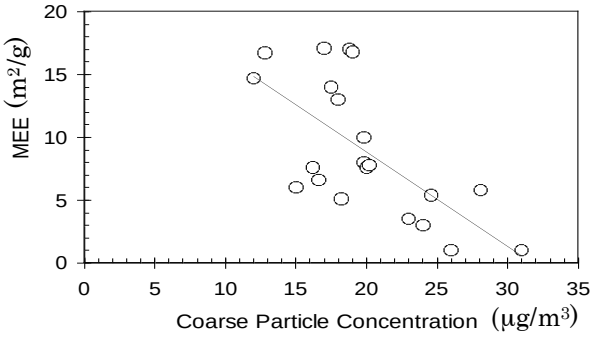

Figure 2 MEE vs. Coarse Particle Concentration.

3.2 Simulation

The MEE values are estimated for maritime, urban and Chiba-area size distribution models using Mie scattering simulation. Table 3 shows the MEE values for fine (with diameter $d < 2.1$ μm) particles, coarse (2.1 μm $< d < 11$ μm) particles and total (fine and coarse) SPM. Urban size distribution gives the highest MEE value among the three size distributions. This is due to anthropogenic aerosols including soot particles, which show high extinction, and sulfates with small sizes (masses). The MEE of Chiba-area size distribution is also computed with varying RH as shown in Fig. 3. Due to high extinction and low mass, fine particles give the highest MEE. Below 70% RH, there is no change in the value of MEE, whereas the value rapidly increases around 80% RH. On the other hand, coarse particles constantly show small MEE due to large mass and the inefficiency in light scattering, with little dependence on RH. Concerning the total SPM, the MEE value is in between the fine and coarse values, and MEE slightly decreases under humid conditions. This decrease is due to the fact that the increase in RH causes the increase in particle size, and the size distribution becomes dominated by larger sized particles. Although extinction also increases as RH is increased, it is compensated by the increase in mass of a particle such that MEE remains almost constant. This result is consistent with the observed MEE behavior shown in Fig. 2.

Table 3: MEE values (in m^2g^{-1}) simulated for fine particles, coarse particles and the total SPM (fine + coarse). Three types (maritime, urban, and Chiba) of size distributions are assumed.

r (μm)	MEE (Maritime)	MEE (Urban)	MEE (Chiba)
Fine	7.07	5.33	3.37
Coarse	0.71	0.69	0.76
Total	1.80	3.14	1.64

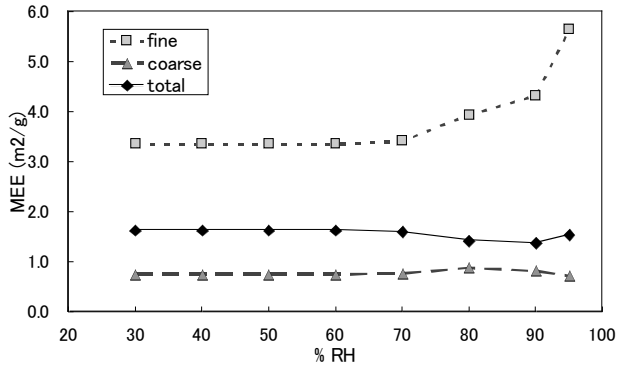

Figure 3 Simulated MEE (fine, coarse, total) vs. RH.

The MEE is also computed for individual aerosol species in their dry state with the use of the Chiba-area size distribution model. Table 4 shows the result for each aerosol component. This simulation reveals that elemental carbon (EC) exhibits the highest MEE value of 7.3 m^2g^{-1}. This result is to be expected since EC has the highest imaginary refractive index (m=1.75-0.44*i*), yielding the highest extinction.

Table 4: Simulated MEE for each aerosol type.

Aerosol Type	Computed MEE (m^2g^{-1})
EC	7.29
OC	2.09
NH_4SO_4	3.52
NH_4NO_3	1.19
Sea Salt	1.27
Soil	1.22
EC – NH_4SO_4 mixture	4.61

4. CONCLUSION

This study has shown that the information on the mass extinction efficiency of aerosols in the lower troposphere can be derived using the extinction efficiency data from the portable automated lidar (PAL) lidar and the SPM data from the β-ray SPM counter. The correlation study has shown that aerosol size distribution dominated by fine particles results in higher MEE value than distributions with more coarse particles. The simulated total MEE value is small compared to the observed MEE, presumably attributable to the fact that the MEE values are derived for 1 month, while the model is based on a 48-h air sampling measurement. Nevertheless, the general trend of the observation (as seen in Fig. 1) is still reasonably reproduced by simulating the change of humidity during the sampling process.

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