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Vertical Change in Extinction and Atmospheric Particle Size in the Boundary Layers over Beijing: Balloon-borne Measurement

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ABSTRACT

Aerosol size and number concentration were observed in the atmospheric boundary layer over Beijing (from near the ground to 1,200 m) on March 15 (a clear day) and 16 (a dusty day), 2005. The results were further compared with lidar measurements in order to understand the dependency of extinction on the particle size distribution and their vertical changes. The boundary layer atmosphere was composed of several sub-layers, and a dry air layer appeared between 400 and 1,000 m under the influence of dust event. In this dry air layer, the concentration of the fine-mode particles (diameter smaller than 1.0 μm) was slightly lower than the value on the clear day, while the concentration of coarse-mode particles (diameter larger than 1.0 μm) was remarkably higher than that on the clear day. This situation was attributed to the inflow of an air mass containing large amounts of Asian dust particles and a smaller amount of fine-mode particles.

The results strongly suggest that the fine-mode particles affect light extinction even in the dusty atmosphere. However, quantitatively the relation between extinction and particle concentration is not satisfied under the dusty atmospheric conditions since laser beam attenuates in the atmosphere with high concentration of particles. Laser beam attenuation effect becomes larger in the relation between extinction and coarse particle content comparing the relation between extinction and fine particle content. To clarify this problem technically, future in situ measurements such as balloon-borne lidar are suggested. Here extinction was measured at 532 nm wavelength. Measurements of extinction at other wavelengths are desired in the future.

Key words: Extinction, Vertical distribution of aerosol, Particle size, Boundary layer, Visibility

1. INTRODUCTION

Atmospheric aerosols play an important role in climate change by affecting global cloud albedo, radiative transfer, ozone chemistry, acid rain, and visibility (Jacobson *et al.*, 2001; Seinfeld *et al.*, 1998; Charlson *et al.*, 1992). They also have an important and direct effect on the atmospheric environment and human health (Chan *et al.*, 1997). Increases in the number concentration of aerosol particles, especially the fine particulate matter (particles with diameter smaller than 2.5 μm: $PM_{2.5}$), may also have significant health effects (Donaldson *et al.*, 1998; Dockery *et al.*, 1994), reduce visibility, and concentrate large amounts of poisonous compounds in the air (An *et al.*, 2000; Prospero *et al.*, 1999). Such harmful effects have understandably caused widespread public concern (Zhang *et al.*, 2003). Indeed, particulate matter is one of the key pollutant types in most of the big cities in China (Wang *et al.*, 2000).

Reduced visibility has become a severe issue in many large cities of the world and can cause inconveniences in daily life disturbing traffic and transpor-

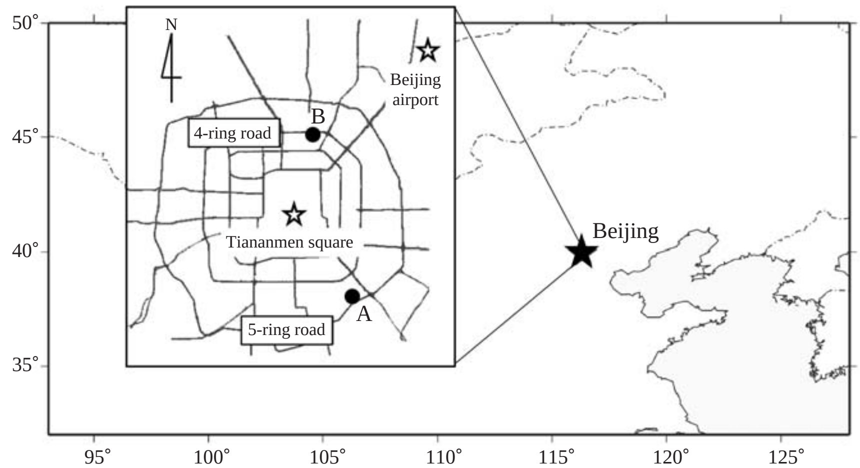

Fig. 1. Observation sites. (A) Base of the atmospheric observation; (B) China-Japan Friendship Environmental Protection Centre.

tation including delay of flights. Many studies have investigated issues of reduced visibility and particulate matter (Wang *et al.*, 2006; Song *et al.*, 2003; Yan *et al.*, 2003; Chan *et al.*, 1999; Baik *et al.*, 1996; Larson *et al.*, 1989).

These studies have mostly been based on ground observations and few measurements have investigated the particulate characteristics and visibility in the free atmosphere or boundary mixing layer over urban regions of China. Knowledge of free atmospheric particulate content and its effect on slant visibility is essential to control safely aviation system considering that activities of big cities including Beijing city are strongly depending on air transportation. Beijing, which has approximately 14.4 million inhabitants, has been experiencing rapid economic growth and urbanization for decades, and consequently faces many problems due to pollution. In addition, previous studies have focused on monitoring PM_{10} and $PM_{2.5}$ particles, without clarifying size dependences.

The results described here is the first case study discussing the vertical change in aerosol size/number concentration and extinction (at wavelength=532 nm) in the boundary layer over Beijing, China, on the basis of the measurement with balloon-borne optical particle counters and lidar.

2. MEASUREMENTS AND METHODS

2.1 Observation Sites

Tethered-balloon measurements were performed at the Atmospheric Observing Experimental Base of Institute of Atmospheric Physics (39.82°N, 116.47°E), close to the south 5th Ring Road in Beijing, south-east of the central part of the city (Matsuki *et al.*, 2005;

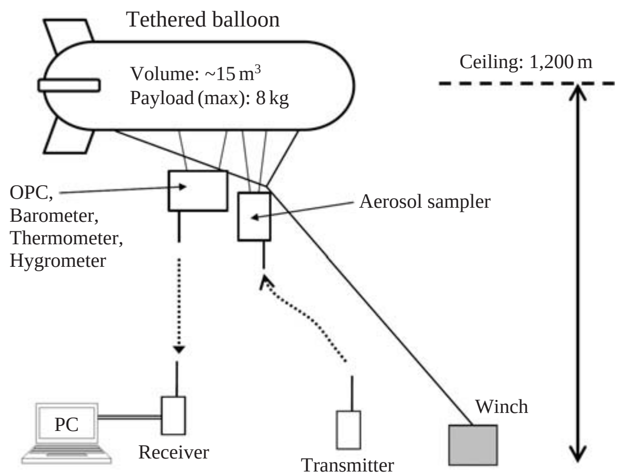

Fig. 2. Balloon train.

Fig. 1). Lidar measurements were performed at the China-Japan Friendship Environmental Protection Centre (39.98°N, 116.42°E) which is near the north-central part of the 4th Ring Road and locates 18 km north of the tethered-balloon site. The distance between the two observation sites was about 10 km. This difference, as described later, can reasonably be neglected considering the atmospheric structures of the planetary boundary layer observed here.

2.2 Instrumentation and Measurements

The tethered-balloon (TB) observation system is similar to that described by Matsuki *et al.* (2005), so we describe the specifications of the system only briefly here. Fig. 2 shows the balloon train used. A barometer was mounted on the balloon to measure the atmospheric pressure, which we used to calculate altitude. The balloon system could reach an altitude of 1,200 m above the ground within 30-40 minutes. It was therefore possible to monitor the changes in aerosols at a time resolution of about 1 hour. The size and number concentration of the atmospheric aerosols were observed in 20-second intervals with an optical particle counter (OPC), which sized particles at diameters of 0.3, 0.5, 1.0, 2.0, and 5.0 μm (Yamashita *et al.*, 2003). A detailed description of the counter was given by Hayashi *et al.* (1998). Vertical profiles of the number concentration of particles, temperature, relative humidity, and air pressure in the same air mass were obtained by balloon-borne measurements at high vertical resolution (about 0.5 m, with resolution dependent on the sounding speed). Table 1 lists the observation time and meteorological conditions near the surface at the balloon site.

Detailed specifications of the polarization lidar, which was installed at the China-Japan Friendship Environmental Protection Centre, can be found in Shimizu *et al.* (2004). The lidar measurements were made using backscattering light from atmospheric particles at 532 nm wavelength (second harmonic wavelength of Nd: YAG laser). Scattering light received by a telescope with a diameter of 20 cm, and detected with photomultiplier tubes. A detailed description of the lidar was given by Sugimoto *et al.* (2003).

3. RESULTS AND DISCUSSION

3.1 The Relationship between Visibility and Extinction

The relationship between visibility and atmospheric extinction is expressed as (Koschmieder *et al.*, 1924)

$$L_v = -\ln 0.02/b = 3.912/b_p \quad (1)$$

where L_v is the visibility in kilometres and b is the atmospheric extinction per kilometre. This formulation is widely used in the routine measurement of the meteorological observatory in the world and regarded as one of the global standards.

The atmospheric extinction can be broken into three parts: $b = b_r + b_a + b_p$, where b_r is the molecular Rayleigh scattering, b_a is the gaseous absorption, and b_p is the extinction due to aerosols. Under standard conditions (sea-surface temperature 15°C, air pressure 1013.25 hPa, air density 1.2250 kg/m^3), the value of Rayleigh scattering is 0.013 km^{-1} (Peundorf *et al.*, 1957). Some atmospheric gas species, especially NO_2, may have ab-

Table 1. Surface meteorological records and observing time.

Time	Items						
	Temperature (°C)	Relative humidity (%)	Wind speed (m/s)	Wind direction (deg.)	Cloud (0-10) (Total/Low)	Visibility (km)	Weather records
2005/03/15 16:30-18:30	14.1	18	2.5	SSW	0/0	20.0	Clear
2005/03/16 16:30-18:30	14.0	27	1.6	SSW	10/0	9.0	Smoke screen

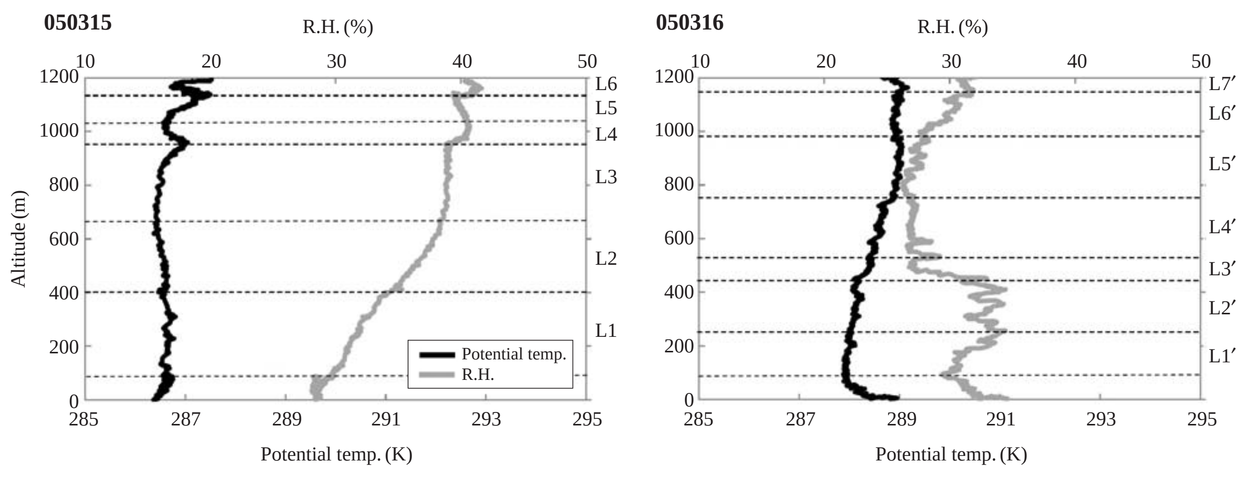

Fig. 3. Potential temperature (K) and relative humidity (R.H., %) plotted separately for the two observation days (Left: March 15, right: March 16). The black lines represent the potential temperature and the gray lines represent the relative humidity.

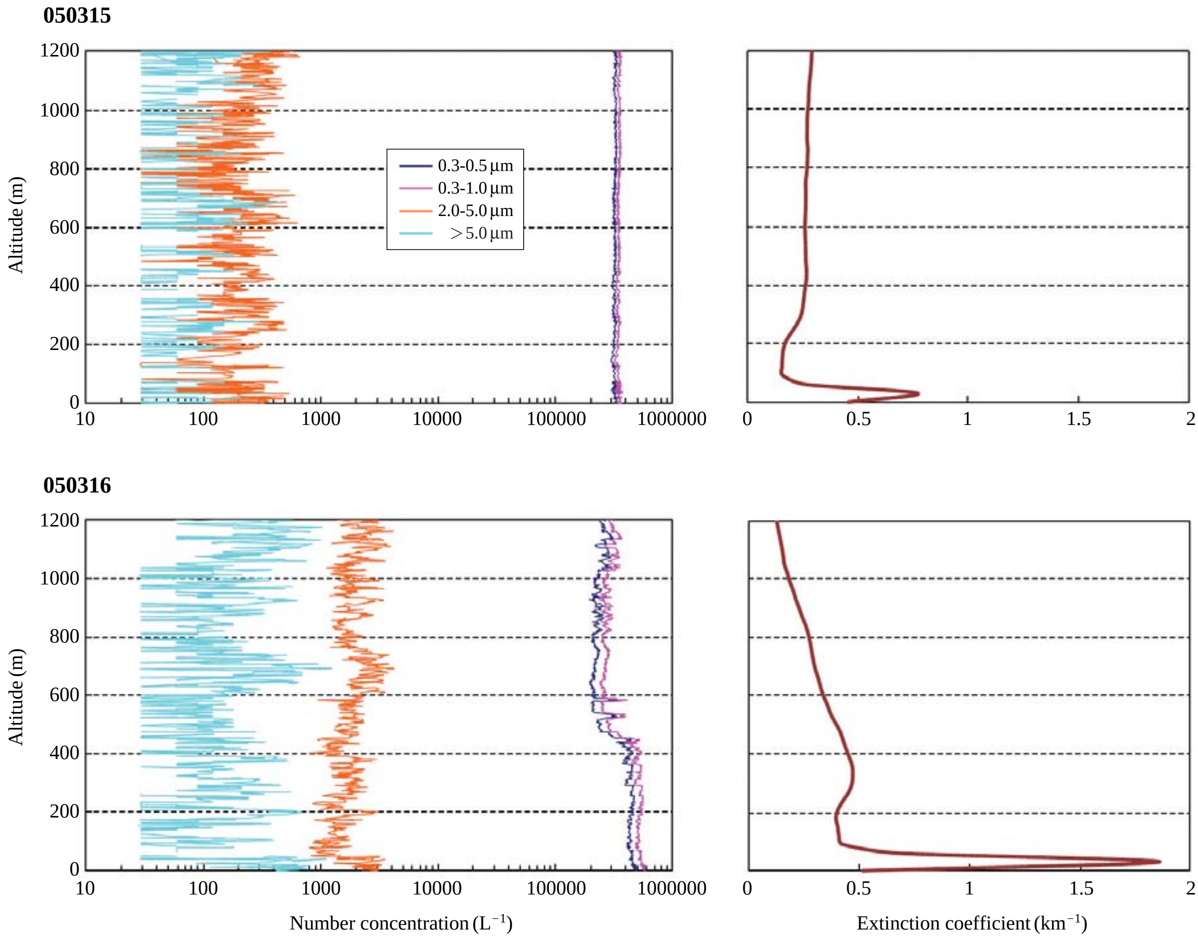

Fig. 4. Profiles of particle number concentrations (L^{-1}) in different size ranges and extinction ratio (km^{-1}) on March 15 (up) and 16 (down).

sorption properties in visible light wavelength ranges. Previous research has verified that the contribution of particulate aerosols to the extinction occupies more than 90% of the total *b* over Beijing (Liu *et al.*, 2004). Hence it is easily confirmed that the empirically determined equation (1) is roughly satisfied in the atmosphere over Beijing, and the visibility has approximately inverse correlation with the particle extinction. According to previous investigations, the size distributions and chemical compositions of atmospheric aerosols have been strongly affected by dust particles and particles of anthropogenic origin in the Beijing region (Wu *et al.*, 2009; Li *et al.*, 2008; Xu *et al.*, 2008; Yuan *et al.*, 2007; Wang *et al.*, 2000; Liu *et al.*, 1999). It is therefore interesting to analyse the relationship between extinction and the size-segregated number concentration of particles, in a case study of a large city in a developing country. However, this relationship cannot reveal which particle size controls the extinction.

3. 2 Vertical Structures of Particle Number Concentrations and Extinctions

As described above, the distance between the lidar observation and balloon sounding sites is about 10 km. Balloon-borne observations took about 1 hour to collect one dataset covering from near the ground to 1,200 m. Horizontal scale of observation field (duration time of observation × typical wind speed) is considered to be about 7-10 km taking into horizontal wind speed of 2-3 m/sec during the observation. Hence the difference in the observation locations in the present analysis is negligible. As shown in Table 1, the weather conditions of the two observing days (050315 and 050316 represent March 15, 2005 and March 16, 2005, respectively) were quite different: the former was a fine day, while the latter was more polluted, as shown by the weather report of the China Meteorological Administration. Figs. 3 and 4 present the vertical profiles of atmospheric relative humidity, potential temperature, size-segregated particle number concen-

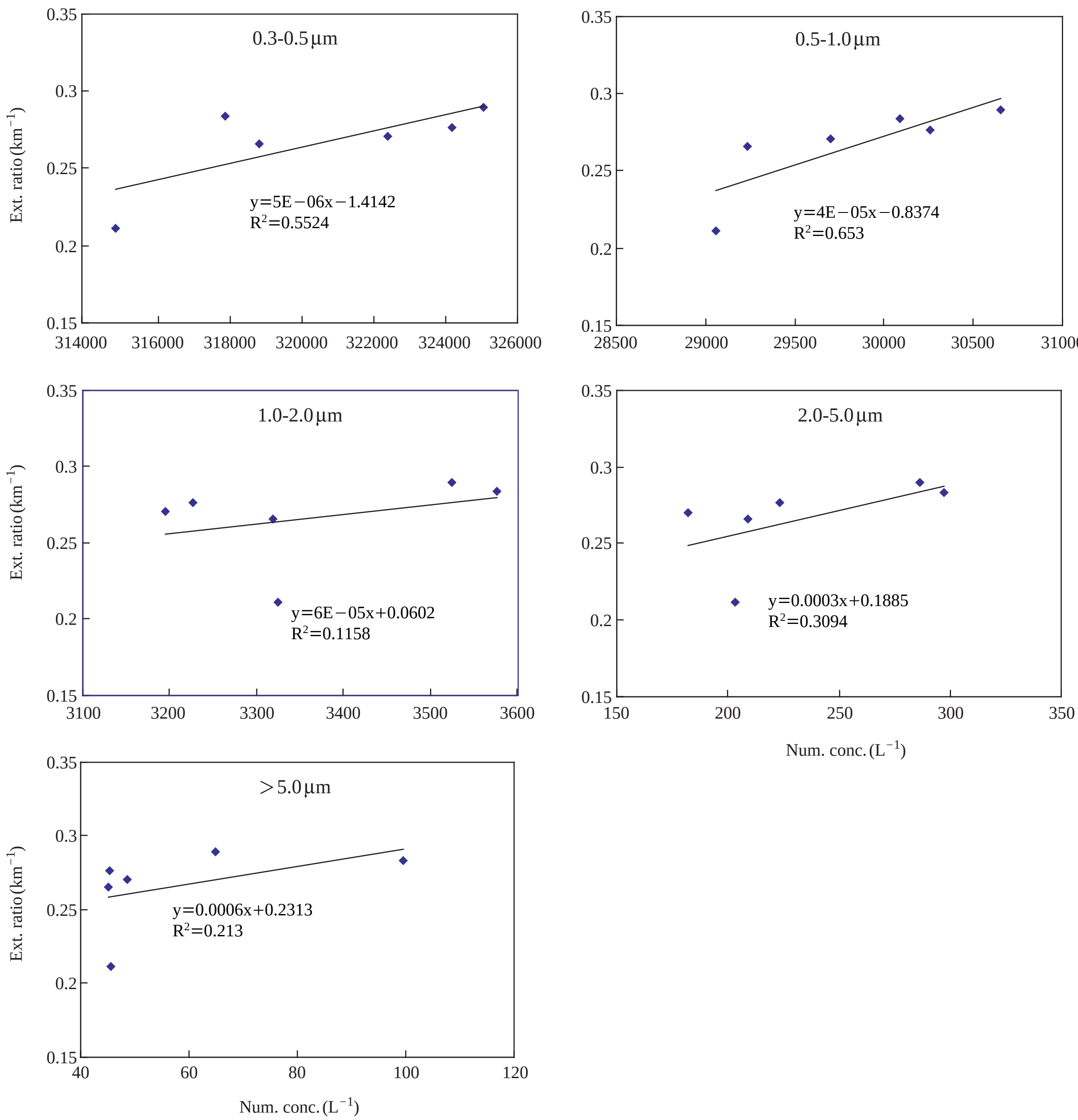

Fig. 5. Correlation between the extinction coefficients (km^{-1}) and the particle number concentrations (L^{-1}) in different size ranges in the case of 050315 (clear day).

tration, and light extinction for the respective days.

Comparing the particle concentration of March 15 with that of March 16 (Fig. 4), there is large difference in concentrations of particles with diameter $>2.0\,\mu m$, and values of March 16 (dusty day) are about 10 times higher than those of March 15 (clear day) from near the ground to about 1,200 m height. From the profiles of coarse mode particle contents, it is suggested that the dust storm disturbed aerosols not only in the boundary mixing layer (potential temperature shows clear decline slope from the ground to about 80 m and narrow decline region at about 400 m. Here we define the region from near the ground to 400 m as the boundary mixing layer of March 16) but also in the free atmosphere (Figs. 3 and 4). In the free atmosphere of March 16, relatively dry air was observed suggesting

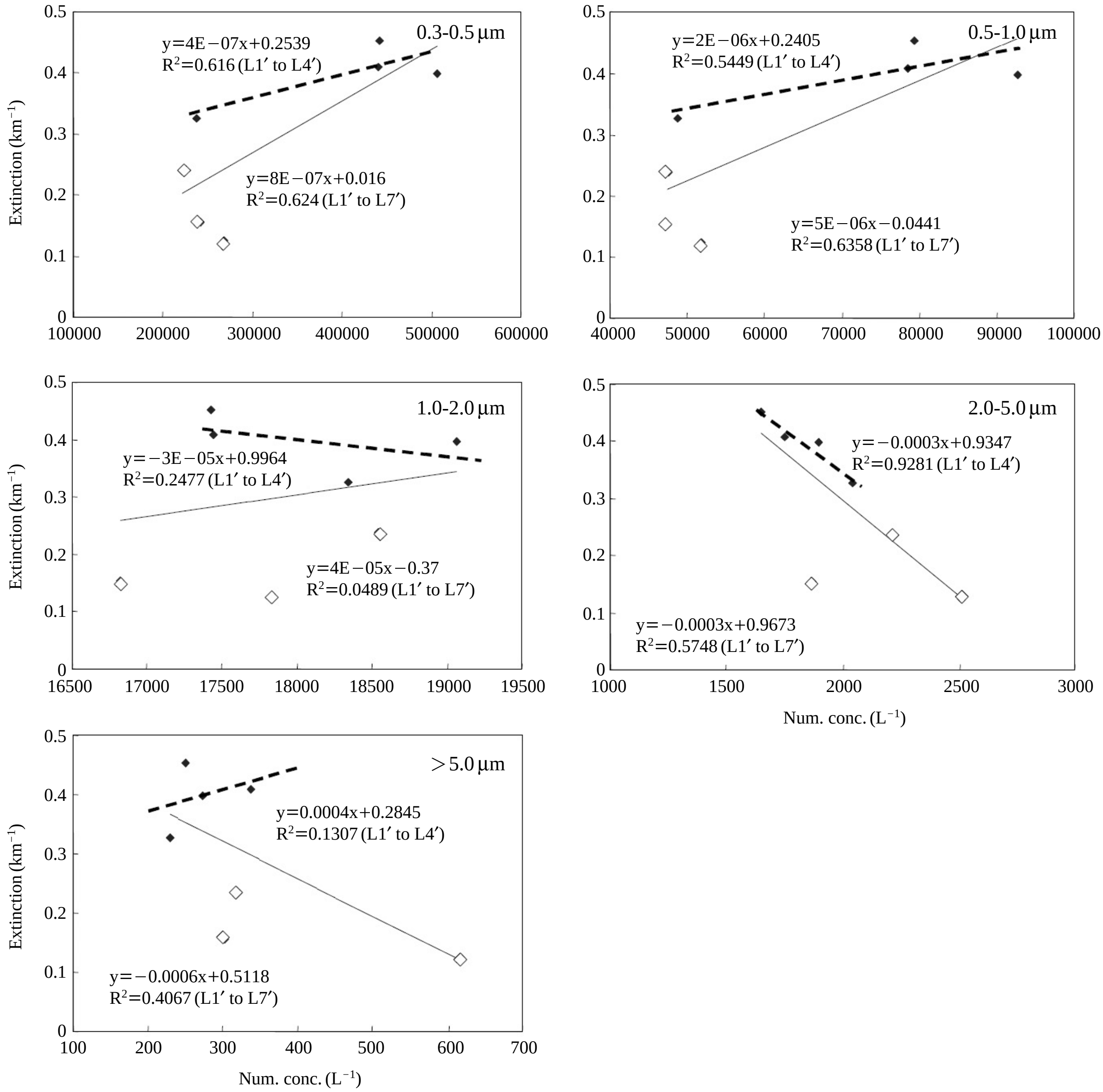

Fig. 6. Correlation between extinction coefficients (km^{-1}) and particle number concentration (L^{-1}) in each size range in the case of 050316 (dusty day). The dotted lines (L1′ to L4′) show the correlation between the extinction and number concentration, neglecting data points L5′, L6′ and L7′.

inclusion of air mass having different origins and/or histories (L4′ and L5′ in Fig. 3). Interestingly, extremely enhanced content of coarse mode particles was found in the dry air mass (Fig. 4).

Concerning with the concentration of fine particles, very interesting feature was observed in the dusty day (March 16) (Fig. 4). The fine particle concentration decreased in the free atmosphere (corresponding to the layers of L4′ and L5′), which were apparently lower than the level of the clear day (March 15). The decrease of the fine particle concentration has been, on the basis of ground-based particle sampling, frequently reported during dust episode by many investigators (e.g., Zhang *et al.*, 2008). When the air mass such as the case in the free troposphere of 16 March descends to near the ground, high concentration of dust particles is observed without effect of pollutants including fine particles formed secondly from polluted gases.

The profile of potential temperature on March 15 suggests active vertical mixing compared to that of 16 March, especially from near the ground to about 800 m height. It can be suggested that the stable thermodynamic structure of 16 March, especially from near the ground to about 80 m, caused polluted air because the pollutants could not diffuse upwards and their concentrations became higher in the low atmosphere. Here, considering vertical profiles of potential temperature and relative humidity, we divided the boundary layer into six sub-layers (L1-L6) for 050315 and seven sub-layers (L1′-L7′) for 050316 measurements. If we decide the boundary layer according to the position of the first clear inversion in the profiles of potential temperature, the boundary layer height will be about 150 m on March 15 and about 200 m on March 16 (Fig. 3). However, it is reasonable to determine the height of the boundary layer top considering humidity profiles in addition to the potential temperature profiles since the ground is large source of water vapour, and heights of about 950 m or 1,150 m of March 15 and about 430 m of March 16 would be top of the boundary layer (Fig. 3).

Fig. 4 shows the relation of the vertical profiles between the particle number concentrations in different size ranges and light extinction at the wavelength of 532 nm. The extinction (α_{sl}) and particle number concentration (C_{sl}) in each sub-layer are given by

$$\alpha_{sl} = \frac{\int_{\delta z} \alpha(z, r) dz}{L_{sl}} \tag{2}$$

and

$$C_{sl} = \frac{\int_{\delta z} C(z, r) dz}{L_{sl}} \tag{3}$$

where $\alpha(z, r)$ and $C(z, r)$ are extinction coefficient and particle number concentration as a function of altitudes (z) in each sub-layer, respectively. L_{sl} represents the layer thickness in each sub-layer (=layer top height−layer bottom height), respectively.

3.3 Correlation between the Extinction and Particle Number Concentration

Figs. 5 and 6 show the correlation between the extinction coefficients and the particle number concentrations in different size ranges in the case of 050315 (the clear day) and 050316 (the dusty day), respectively. According to (2) and (3), six data points of particle concentration and extinction were obtained for the clear day (Fig. 5) and seven data points were obtained for the dusty day (Fig. 6). Close positive correlations existed between the extinction coefficients and the particle number concentrations for particle diameters smaller than 1.0 μm in both cases.

For particles of diameter larger than 2.0 μm (050316 case), assessing their contribution is difficult using only lidar data (Fig. 6) because the intensity of the laser beam possibly decreased as the laser beam penetrated the layer of large optical depth. Therefore the extinction deduced from the lidar measurements will be underestimated above about 800 m, comparing the profiles of lidar returns of 050315 and 050316 in Fig. 4. The open symbols in Fig. 6 represent values that were considered to be disturbed by the large optical depth layer (L5′, L6′, and L7′). The correlation between fine-mode particle concentration and extinction is clearly positive even if the values higher than 800 m are neglected. However, correlation between coarse-mode particle concentration and extinction is still negative even though the values of L5′, L6′ and L7′ are neglected, and effect of laser beam attenuation is not completely avoided.

Our results suggest that the contribution of fine-mode particles (diameter smaller than 1.0 μm) to light extinction or the visibility is important not only near the ground, but also over the planetary boundary layer (or the free troposphere) in Beijing. The attenuation of lidar beam is caused in dusty atmosphere, and consequently large underestimation of extinction is made. This is important problem to be solved when we want to estimate slant visibility from lidar return. To clarify this problem technically, future in situ measurements such as balloon-born OPCs are suggested. Here, we estimated extinction only at wavelength of 532 nm. Observations at other wavelengths would also be useful to discuss the relation between extinction and aerosol concentration.

4. SUMMARY

We performed tethered-balloon observations twice in spring (on dusty and clear days) in Beijing, China. Vertical profiles of size-segregated particle concentration were obtained with the OPC. To understand the relationship between the visibility and the particulate matter suspended in the urban atmosphere, the vertical profiles of the OPC datasets and the corresponding lidar data were compared. The present observations reveal that the concentration of fine-mode particles (diameter < 1.0 μm) was positively correlated with the extinction. Consequently, fine-mode particles strongly contribute to light extinction and visibility in the boundary layer over Beijing, even under different weather conditions. However, the extinction deduced from the lidar return is certainly underestimated under the dusty atmospheric condition and correspondence between particle concentration and extinction is strongly disturbed, especially in size ranges of coarse particles due to strong lidar return attenuation.

The balloon-borne measurements showed that the aerosol layer was composed of some sub-layers. The sub-layers sometimes contained apparently higher content of particles than the value near the ground in dusty day, and therefore it will be hard to estimate slant visibility only from the lidar return data since intensity of laser pulse rapidly decreases during penetration in such high concentration particle layer. In situ measurements will be effective to correct underestimation in lidar return intensity, and the results presented here are only case study and much more observations are desired in future.

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