

Measurement of Aerosol Parameters with Altitude by Using Two Wavelength Rotational Raman Signals

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Aerosol size distribution provides good information for predicting weather changes and understanding cloud formation. Aerosol extinction coefficient and backscattering coefficient are measured by many scientists, but these parameters depend not only on aerosol size but on aerosol concentrations. An algorithm has been developed to measure aerosol parameters such as Ångström exponent, color ratio, and LIDAR ratio without any assumptions by using two wavelength rotational Raman LIDAR signals. These parameters are good indicators for the aerosol size. And we can find Ångström exponent, color ratio, and LIDAR ratio under various weather conditions. Finally, it can be seen that the Ångström exponent has an inverse relationship to the particle size of the aerosol and the color ratio is linearly dependent on the aerosol size. An Ångström exponent from 1.2 to 3.1, a color ratio from 0.28 to 1.04, and a LIDAR ratio 66.9 sr at 355 nm and 32.6 sr at 532 nm near the cloud were obtained.

Keywords : LIDAR, Rotational Raman, Backscattering, Extinction, Color ratio

OCIS codes : (280.3640) Lidar; (290.5860) Rotational Raman; (290.1350) Backscattering; (290.2200) Extinction; (010.1100) Aerosol detection

I. INTRODUCTION

Optical properties of an atmospheric aerosol are very important parameters for predicting climate change. An aerosol has an indirect effect on the radiation budget of atmosphere providing condensation nuclei for cloud formation, because scattering and absorption of solar radiation and longwave radiation are dependent on atmospheric aerosol particle concentration, position, orientation, shape, and size. Sometimes the specific aerosol, Asian dust, impacts the Korean Peninsula or travels through the sky on the prevailing westerlies in varying degrees[1]. The occurrence rate of Asian dust has been rapidly increasing in the 2000s, and this specific Asian dust also affects the climate.

If there is fog, visibility is strongly dependent on the

amount and size of water droplets. So, to predict weather changes, water droplet size change and other parameters such as temperature must be measured. Moreover, the aerosol backscattering coefficient and extinction coefficient cannot give enough information for identifying weather changes, so it is necessary to measure other parameters which are connected with aerosol size, such as Ångström exponent, color ratio, and LIDAR ratio[2, 3].

Many scientists have tried to measure simple aerosol properties such as backscattering coefficient, and extinction coefficient[4-6]. Moreover, they have obtained these backscattering and extinction coefficients with some assumptions such as Ångström exponent, color ratio, and LIDAR ratio[6, 7]. The Ångström exponent, k , value is 4 for molecules and varies between 0 and 2 for particles by the

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Ångström's empirical formula $\alpha \propto \lambda^{-k}$ [8]. This assumption causes systematic errors, because the Ångström exponent of an aerosol is dependent on the type and size of the aerosol. The high spectral resolution LIDAR system does not need any of these assumptions, but the high spectral resolution LIDAR requires an expensive optics and laser system to separate the Mie scattering signal from the Rayleigh scattering signal, because the received LIDAR signal is a combination of the Doppler-broadened molecular backscatter spectrum and the un-broadened aerosol spectrum[4].

In this study, the Ångström exponent, color ratio, and LIDAR ratio were measured by using two wavelength rotational Raman LIDAR signals to obtain the aerosol size distribution. The backscattering coefficients, which have a close relation to atmospheric aerosol concentration were also calculated. Finally, the cloud and aerosol size characteristics were analyzed by using the Ångström exponent, color ratio, and LIDAR ratio.

II. METHOD

In rotational Raman LIDAR, the two return signals can be expressed as Eqs. (1) and (2)[9, 10].

$$P_R^{\lambda}(z) = \frac{C_1}{z^2} O(z) \beta_R^{\lambda} \exp\left\{-2 \int_0^z [\alpha_m^{\lambda}(z') + \alpha_a^{\lambda}(z')] dz'\right\}, \quad (1)$$

$$P_E^{\lambda}(z) = \frac{C_2}{z^2} O(z) (\beta_a^{\lambda} + \beta_m^{\lambda}) \exp\left\{-2 \int_0^z [\alpha_m^{\lambda}(z') + \alpha_a^{\lambda}(z')] dz'\right\}, \quad (2)$$

where P is the LIDAR signal, λ is the scattering wavelength, β is the backscattering coefficient, α is the extinction coefficient, $O(z)$ is the overlapping function, z is the altitude, C is a constant that is independent of the altitude, and E , R , a , and m represent the elastic, rotation, aerosol, and atmospheric molecule, respectively. The aerosol backscattering coefficient can be obtained from these two signals as shown in Eq. (3)[11].

$$\frac{P_R^{\lambda}(z)}{P_E^{\lambda}(z)} = \frac{C_2 \beta_a^{\lambda}(z) + \beta_m^{\lambda}(z)}{C_1 \beta_R^{\lambda}(z)} = \frac{C_2 N(z) d\sigma_m^{\lambda} / d\Omega + C_2 \beta_a^{\lambda}(z)}{C_1 N(z) d\sigma_R^{\lambda} / d\Omega} = \frac{C_2}{C_1} C_{m/R} + \frac{C_2 \beta_a^{\lambda}(z)}{C_1 \beta_R^{\lambda}(z)}, \quad (3)$$

where $C_{m/R}$ is the ratio of the scattering cross section between the Rayleigh and the total rotational Raman, N is the number density of molecules (oxygen and nitrogen), Ω is the solid angle, and σ is the scattering cross section of the atmospheric molecule. Two unknown values

$\left[\frac{C_2}{C_1} \times C_{m/R}, \frac{\beta_a^{\lambda}(z)}{\beta_R^{\lambda}(z)} \right]$ can be obtained by using the method of Kim and Cha[11]. The constant $\frac{C_2}{C_1} \times C_{m/R}$ can be obtained experimentally on a clear day by using a large range of LIDAR signals at a high altitude. And the

experimental constant C_2/C_1 can be calculated within statistical errors by using the theoretical scattering cross section of rotational Raman scattering and the elastic scattering of nitrogen and oxygen. Thus the aerosol backscattering value is $\beta_a^{\lambda}(z)/\beta_m^{\lambda}(z)$ normalized with the molecular rotational backscattering coefficient[11]. The

aerosol backscattered color ratio $[CR = \frac{\beta_a^{532}(z)}{\beta_a^{355}(z)}]$ can be obtained from the ratio of the aerosol backscattering coefficients between two wavelengths. The aerosol color ratio value provides good information about the relative size change of the aerosol, which does not directly correspond to the absolute value of the aerosol size[12].

The aerosol extinction coefficient can be obtained from Eq. (1). The aerosol extinction coefficient can be expressed as Eq. (4) from Eq. (1), by substituting $k(z)$ for a constant $C_1 \times O(z)$.

$$\alpha_a = -\frac{1}{2} \frac{d}{dz} \left(\ln \frac{z^2 P_R}{k(z)} \right) - \alpha_m, \quad (4)$$

From Eq. (3), the aerosol Ångström exponent can be obtained. As mentioned above, the two unknown values

$\left[\frac{C_2}{C_1} \times C_{m/R}, \frac{\beta_a^{\lambda}(z)}{\beta_R^{\lambda}(z)} \right]$ in Eq. (3) can be obtained experimentally. So $\frac{\beta_a^{\lambda_1}(z)}{\beta_R^{\lambda_1}(z)}$ can be calculated from the two unknown values. The ratio of backscattering coefficients, $\varrho(z) = \frac{\beta_a^{\lambda_1}(z)}{\beta_R^{\lambda_1}(z)}$, at two (355 nm and 532 nm) wavelengths are given by

$$P(z) = \frac{\beta_a^{532}(z)}{\beta_R^{532}(z)} \bigg/ \frac{\beta_a^{355}(z)}{\beta_R^{355}(z)}. \quad (5)$$

Since $\beta_R^{\lambda} = C_0 \lambda^{-4}$, it can be rewritten as

$$P(z) = \frac{\beta_a^{532}(z) (355)^{-4}}{\beta_a^{355}(z) (532)^{-4}}. \quad (6)$$

The Ångström exponent k , which describes the wavelength dependence of the particle backscattering coefficient, can be expressed by Eq. (7).

$$\frac{\beta_a^{532}(z)}{\beta_a^{355}(z)} = \left(\frac{355}{532} \right)^k \quad (7)$$

Consequently, from Eq. (6), (7), the Ångström exponent can be expressed as Eq. (8).

$$k = \frac{\log(A)}{\log\left(\frac{355}{532}\right)}, \quad (8)$$

where A is $P(z) \times \frac{(532)^{-4}}{(355)^{-4}}$.

III. EXPERIMENTAL SETUP AND RESULTS

In this study, the two wavelength Raman LIDAR system was installed by using a 355 nm and 532 nm laser beam of a Nd:YAG laser. The pulsed energy of the laser beams were 130 mJ and 300 mJ at 355 nm and 532 nm, respectively. Figure 1 shows a schematic diagram of the installed LIDAR system. To decrease laser beam divergence, a beam expander was used in the transmitting system, which can expand a laser beam by 10 times. The receiving system consisted of 4 channels, namely rotational Raman channels at 355 nm and 532 nm, and two elastic channels at 355 nm and 532 nm[10, 11].

Figures 2(a) and 2(b) show the change of aerosol backscattering coefficients measured using the two wavelength LIDAR signals on June 10, 2010 (cloudy day) at different times. In the figures, it can be seen that even though the boundary layer is at the same altitude (2000 m), the backscattering coefficient changes dramatically from $2.24 \times 10^{-4} \text{ m}^{-1}$ to $1.81 \times 10^{-4} \text{ m}^{-1}$ (355 nm) within the boundary layer. At the other wavelength (532 nm), the change of the backscattering coefficient with altitude is not the same as that of 355 nm. But the cause of the backscattering coefficient variance is unknown, because the backscattering coefficient is not solely dependent on the aerosol size and concentration. To obtain the quantitative information, the color ratio was calculated at two different times. Figure 2(c) shows the color ratios that have a close relation to the aerosol size distribution. In Fig. 2(c), it can be seen that the aerosol size distribution value decreased from 0.25 to 0.14 and from 0.26 to 0.15 in the boundary layer with the altitude at 21:50 and 23:19[12, 15]. In this case, the color ratios are similar to each other at all altitudes.

Figures 3(a) and 3(b) show the change of the aerosol backscattering coefficients on Dec. 4, 2007 (clear day) at different times. In Figs. 3(a) and 3(b), it can also be seen that even though the boundary layer is the same altitude (1300 m), the backscattering coefficient changes by a factor of 7 from $3.22 \times 10^{-5} \text{ m}^{-1}$ to $2.15 \times 10^{-4} \text{ m}^{-1}$ (355 nm) within the boundary layer. Figure 3(c) shows the change of the color ratio measured at these two different times. At 1000 m altitude, the color ratio value was 0.83 at 22:17, and 1.04 at 23:18, respectively. Compared with Fig. 2(c),

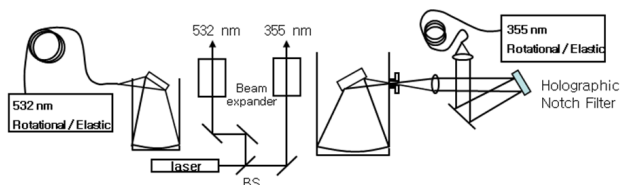


FIG. 1. Schematic diagram of the experimental system.

these figures show a strong change in the color ratio within the boundary layer according to the altitude. In particular, the change of color ratio value at the boundary layer (1300 m) was dramatic even in one hour, even though the change of color ratio value within and above this boundary layer was small. It means that aerosol dynamics usually start at the surface of the boundary[13,

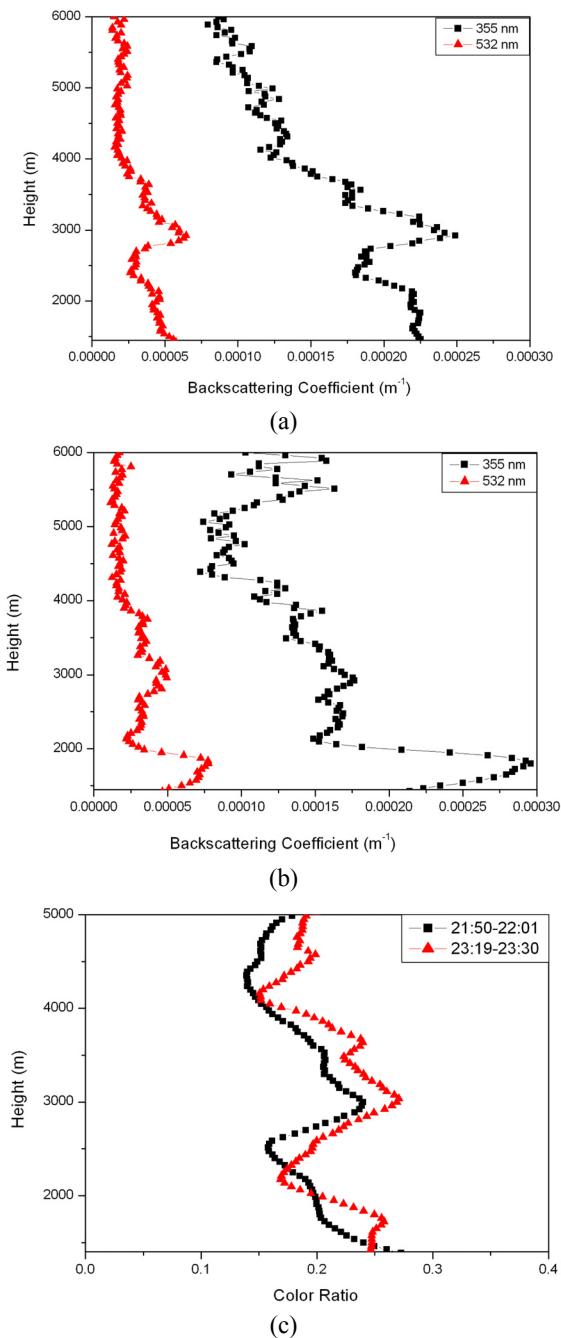
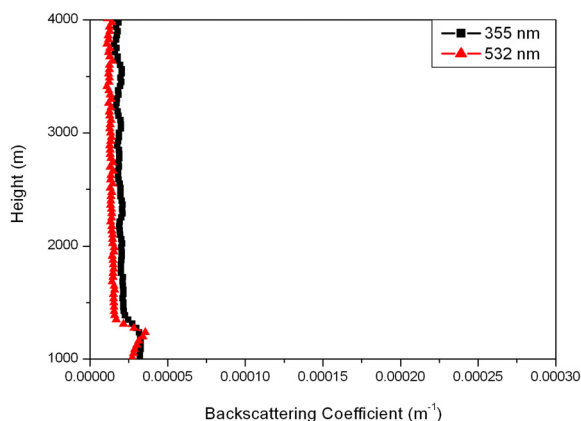
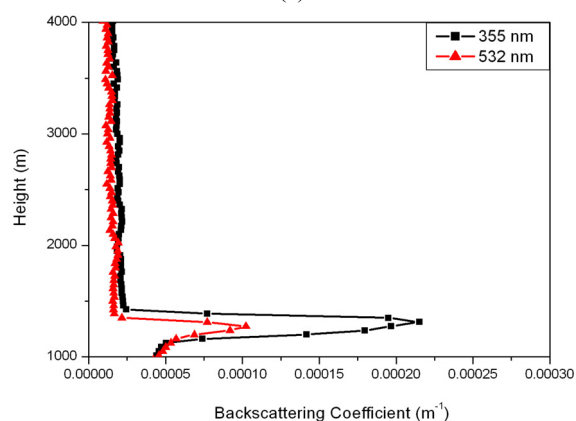


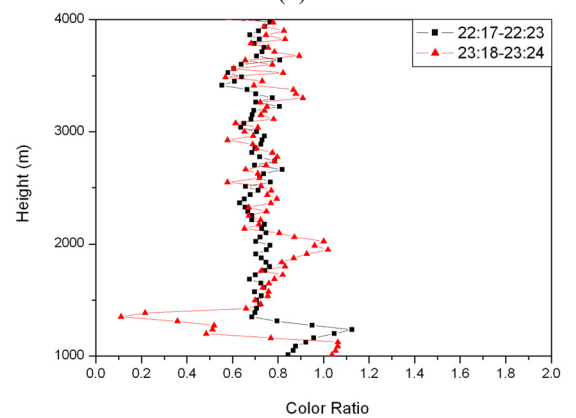
FIG. 2. The backscattering coefficients and the corresponding color ratio measured on June 10, 2010 (cloudy day) at different times; (a) the backscattering coefficients by using two wavelength LIDAR signals at 21:50-22:01 (KST), (b) the backscattering coefficients at 23:19-23:30 (KST), (c) color ratio by using the backscattering coefficients.



(a)



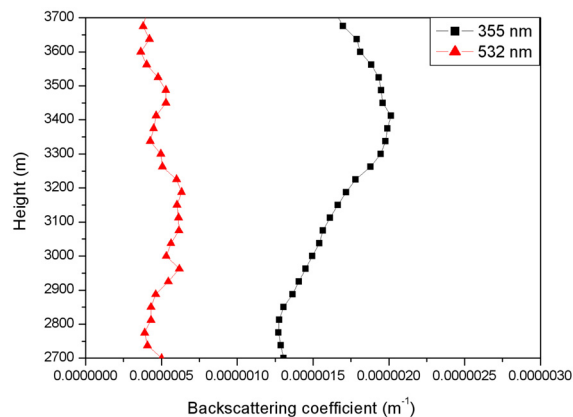
(b)



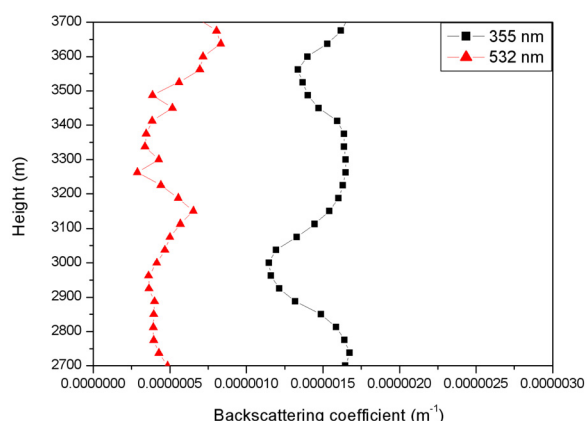
(c)

FIG. 3. The backscattering coefficients and the corresponding color ratio measured on Dec. 4, 2007 (clear day) at different times; (a) the backscattering coefficients at 22:17-22:23 (KST), (b) the backscattering coefficients at 23:18-23:24 (KST), (c) the color ratio by using the backscattering coefficients. It can be seen that the color ratio sharply changed within the boundary layer according to the altitude.

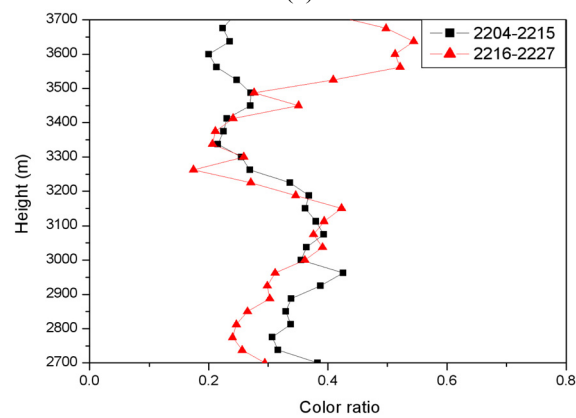
16]. In conclusion, the aerosol color ratio changes within the boundary layer and near the cloud, even in a short time period in this case.



(a)



(b)



(c)

FIG. 4. The backscattering coefficients and the corresponding color ratio measured on May 30, 2010 (cloudy day) at different times; (a) the backscattering coefficients at 22:04-22:15 (KST), (b) the backscattering coefficients at 22:16-23:27 (KST), (c) the color ratio by using the backscattering coefficients. The change of color ratio value between 3500 m and 3700 m was dramatic, even in 11 minutes.

Figures 4(a) and 4(b) show the change of the aerosol backscattering coefficients on May 30, 2010 (cloudy day) at different times. In Fig. 4(a), it can be seen that the

backscattering coefficient increased from $1.3 \times 10^{-6} \text{ m}^{-1}$ to $2.0 \times 10^{-6} \text{ m}^{-1}$ at 355 nm between 2700 m and 3400 m. The backscattering coefficient showed no change at 532 nm at the same altitudes (2700-3400 m). Figure 4(b) shows that the backscattering coefficient varied sharply at the same altitudes (2700-3400 m). On the other wavelength (532 nm), the change of the backscattering coefficient with altitude was not the same with the change of 355 nm. Figure 4(c) shows the change of the color ratio measured at these two different times. At 2700 m altitude, the color ratio value was 0.38 at 22:04, and 0.29 at 22:16, respectively. The average value of the color ratio was 0.23 at 22:04 and 0.45 at 22:16 between 3500 m and 3700 m. In particular, the change of color ratio value between 3500 m and 3700 m was dramatic, even in 11 minutes. This figure shows that the aerosol concentration and aerosol size distribution change, even in such a short time period.

Figure 5 shows the change of the Ångström exponent, the color ratio, the Ångström exponent versus color ratio, and the LIDAR ratio at 355 nm and 532 nm measured on

May 31, 2010 (cloudy day). In Fig. 5, it can be seen that the Ångström exponent changed from 2.5 to 3.1 and the color ratio changed from 0.28 to 0.42 at altitudes between 2000 m and 3000 m. There were no noticeable changes in the Ångström exponent and color ratio. So, a clear relationship between the Ångström exponent and color ratio is not known. The average value of the LIDAR ratio was 66.9 sr at 355 nm and 32.6 sr at 532 nm at the same altitude (2000-3000 m). These values are thought to be reasonable also by other studies[14, 17].

Figure 6 shows the change of the Ångström exponent, the color ratio, the Ångström exponent versus color ratio, and the LIDAR ratio at 355 nm measured on June 11, 2010 (rainy day). It can be seen that the Ångström exponent increased from 1.2 to 2.7 and the color ratio decreased from 0.64 to 0.32 at altitudes between 2000 m and 3000 m. These values are also thought to be reasonable also by many other studies[3, 12, 15]. It can be seen that the Ångström exponent and color ratio varied sharply with the corresponding altitudes when we comparing Fig. 5 with

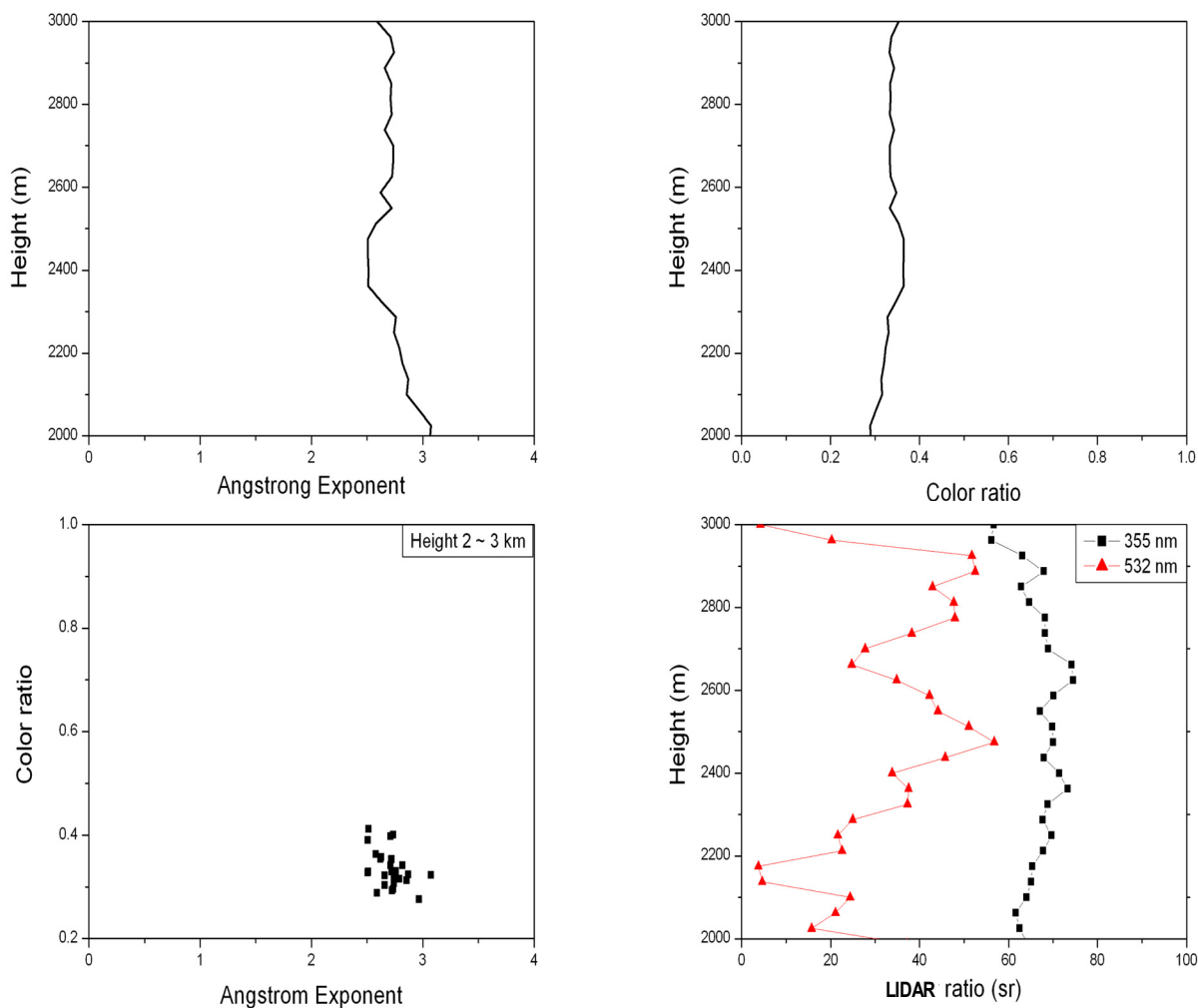


FIG. 5. The Ångström exponent, the color ratio, the Ångström exponent versus color ratio, and the LIDAR ratio at 355 nm and 532 nm measured on May 31, 2010 (cloudy day) at 21:58-22:04 (KST).

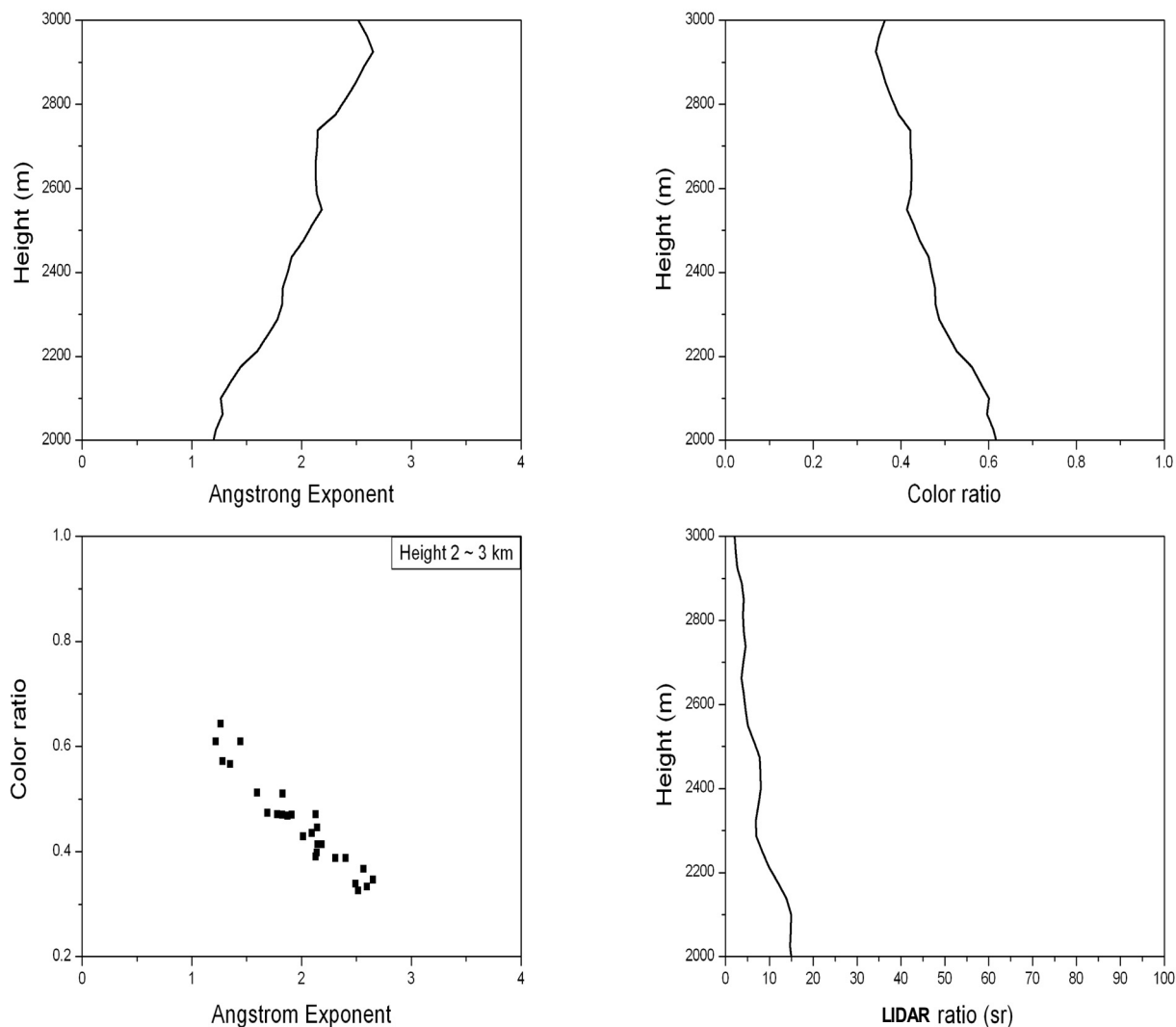


FIG. 6. The Ångström exponent, the color ratio, the Ångström exponent versus color ratio, and the LIDAR ratio at 355 nm measured on June 11, 2010 (rainy day) at 24:04-24:10 (KST). A change of Ångström exponent and color ratio could be detected when the altitude changed from 2000 m to 3000 m. So an inverse relation can be seen between the color ratio and Ångström exponent.

Fig. 6. Also, in Fig. 6, an inverse relation can be seen between the color ratio and Ångström exponent, as was mentioned in Eq. 7. The average value of the LIDAR ratio depends on the weather conditions[14, 17] it has changed from 60 sr(Fig. 5) to 7.0 sr(Fig. 6) at 355 nm at the same altitude (2000-3000 m). On a rainy day, a change of Ångström exponent and color ratio could be detected when the altitude changed from 2000 m to 3000 m. These changes according to altitude come from the change of aerosol size because these two parameters, the Ångström exponent and color ratio, simultaneously indicate an increase of aerosol size as indicated in the introduction. That is to say, the Ångström exponent k value is 4 for molecules and varies between 0 and 2 for particles[3]. This means that the aerosol size increases when the altitude approaches to the cloud or the humidity increases due to the inverse relation of the color ratio and Ångström

exponent[12]. It is certain that these aerosol size changes on a rainy day come from the change of humidity, because rainfall was observed 2 minutes after this measurement. From the experimental results, the color ratio, LIDAR ratio, and Ångström exponent can give information about the relative size change of an aerosol.

IV. DISCUSSION AND CONCLUSION

In conclusion, a two wavelength rotational Raman LIDAR system was installed that can measure aerosol parameters. The spatial/temporal aerosol extinction coefficient, color ratio, LIDAR ratio, and Ångström exponent was measured without any assumption by using two wavelength rotational Raman LIDAR signals. An inverse relation between the color ratio and Ångström exponent was observed, as was mentioned in Eq. 7. This means that the Ångström exponent

has an inverse relationship to the particle size of the aerosol and the color ratio is linearly dependent on the aerosol size. To measure the aerosol size distribution more correctly, the backscattering and extinction coefficient cannot give any information, measurement of the several parameters (color ratio, LIDAR ratio, and Ångström exponent) can provide much better results than one parameter measurement. By using these several parameters, a difference in the aerosol concentration and aerosol size distribution in the boundary layer can be observed. The boundary layer is physically very important for heat transfer, free convection currents and aerodynamics etc, but there is not enough data when using only the extinction and backscattering coefficient of the boundary layer and an aerosol dynamic study which is dependent in a complex way on the meteorological parameters. In the future, the change in the boundary layer will be measured more quickly and correctly.

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