Investigating the effect of hygroscopicity of aerosols on optical profiles of PBL observed by Dual-wavelength Lidar

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ABSTRACT

The light scattering and radiation properties of aerosols are mainly dominated by hygroscopicity. In this study, the relationship between the wavelength dependent ratio of lidar scattering signals (color ratio) and relative humidity and the application of the color ratio to identify the cloud base is examined.

1. INTRODUCTION

Changes in aerosol scattering properties can highly influence the Earth's climate by affecting the Earth's radiation budget. The knowledge of the direct and indirect interaction of atmospheric aerosols on climate is still quite low compared to other atmospheric constituents, not only due to high spatial and temporal variability of aerosols but also due to hygroscopicity. In the ambient atmosphere, the aerosol particles may absorb moisture from the surrounding air and increase their size causing significant change in their size distribution and their associated optical and microphysical properties[1].

In previous studies we already noticed that the inter-relationship between the aerosol Ångström exponent and particle depolarization can reveal information on particle size and shape [2]. In that study, it was reported that the aerosol hygroscopicity might be the reason for the observed distortion in the linear correlation between Ångström exponent and particle depolarization. In this study, we try to combine Mie scattering theory and Köhler theory to study the effect of aerosol hygroscopicity on PBL profiles. We also try to use the lidar color ratio (**CR**) as indicator of particle size to investigate the inter-relationship between color ratio, hygroscopicity (κ), and relative humidity.

2. METHOD

2.1. Color Ratio

The particle size can be inferred from size-dependent light scattering properties such as the Ångström exponent and color ratio [2; 3; 4] (and references therein). The Ångström exponent had been wildly used to understand the particle size. However, the Ångström exponent might be highly uncertain or unavailable because the value of the Ångström exponent highly depends on the accuracy of lidar inversion. Therefore, the ratio of lidar backscatter at different wavelengths (color ratio) might be a simple indicator and more suitable than the Ångström exponent for Planetary Boundary Layer (PBL) studies.

In this study, vertical profiles of lidar backscatter measured by Research Centre for Environmental Changes / Academia Sinica and National Taiwan University (RCEC/ASNTU) Lidar installed in National Taiwan University, Taipei, Taiwan is used for a case study. The RCEC/ASNTU Lidar is a dual wavelength depolarization lidar operated at 355 nm and 532 nm designed to measure aerosols and clouds up to 10 km during day and 20 km at night[2]. The particulate backscatter color ratio **CR** is defined as

$$CR = \frac{P_{532}}{P_{355}} \tag{1}$$

where P_{532} and P_{355} are particulate backscatter at 532 and 355nm respectively.

2.2. Hygroscopicity

The saturation ratio of a liquid aerosol droplet in aerosol in its volume equivalent diameter, S_{eq}[5], over an aqueous solution droplet can be calculated from

$$S_{eq} = a_w \times e^{A/D} \tag{2}$$

$$a_w = \frac{n_w}{n_w + in_s} \tag{3}$$

$$A = \frac{4\sigma_w M_w}{\rho_w R_v T} \tag{4}$$

where a_w is the activity of water in solution, ρ_w is the density of water, M_w is the molecular weight of water, σ_w is the surface tension of the solution/air interface, R is the universal gas constant, T is temperature, and D is the diameter of the droplet, D_d is the dry diameter.

For a multicomponent system (multiple solutes + water) at equilibrium, Eq. 2 yields the equation defining κ–Köhler theory

$$S_{eq}(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1-\kappa)} \times \exp\left(\frac{4\sigma_w M_w}{\rho_w R_v T D}\right) \tag{5}$$

Where κ can then be understood as expressing the volume (or mass or moles, with appropriate unit conversions) of water that is associated with a unit volume of dry particle.[5] The overall value for κ is given by the simple mixing rule

$$\kappa = \sum_i \varepsilon_i \kappa_i$$

The wavelength dependent backscattering coefficient of grown particles can be calculated using

$$\beta(\lambda) = \frac{\pi D^2}{4} N_d Q_{back}(\lambda) \quad (6)$$

where N_d is volume number density of particle and Q_{back} is the backscattering efficiency calculated from Mie theory with complex refractive index. In this study, $m = 1.44 + 0.43i$ is used for most of the simulation.

In general, the value of κ for soluble inorganic aerosols is in the range 0.4-1.4; for organic aerosols it is in the range of 0.01-0.5 and for black carbon is less than 0.01.

Our previous studies found κ is around 0.2 for aerosols in Taipei City and 0.1- 0.2 for aerosols in Kaohsiung City [6].

3. RESULTS

3.1. Hygroscopic Growth

Figure 1 shows the typical particle size distribution (solid black) observed in Taipei. The hygroscopic growth size distributions from *dry* to RH=98% for aerosols with κ=0.2 (solid blue) and 0.61 (dashed black) are shown for comparison. Particles with κ=0.61 are about two times larger than particle with κ=0.2 when RH=98%.

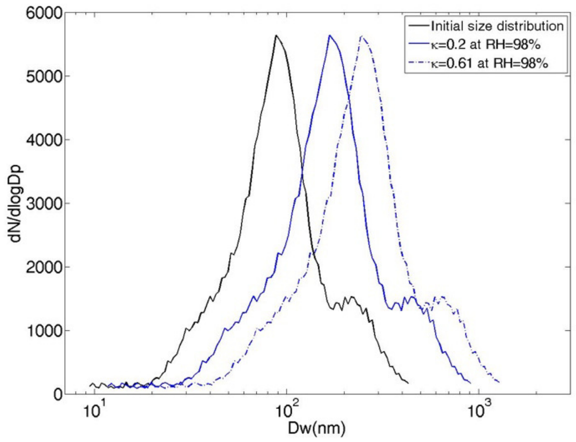

Figure 1: Dry particle size distribution (solid black) and hygroscopic particles growth from “dry” to RH=98% for particles with κ=0.2 (solid blue) and 0.61 (dashed blue).

The corresponding backscattering coefficient as a function of RH for aerosols with κ=0.2 is shown in Fig. 2. The complex refractive index of aerosol is set to $m = 1.44 + 0.043i$. Here the Particle Backscatter Coefficient (PBC) or β_{par} increases slowly with increase in RH up to RH<80%. After RH is higher than 80%, the β_{par} increases rapidly with increase in RH. At RH ≈98% the backscatter is about 10 times larger than the backscatter at RH=40%.

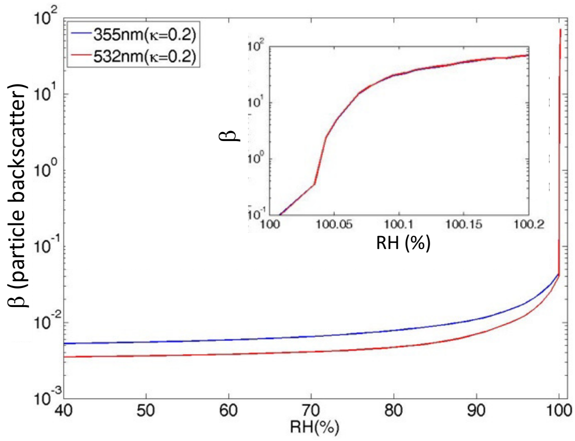

Figure 2: Hygroscopic growth of backscatter with varying RH for aerosols with m=1.44+0.043i and κ=0.2.

3.2. Color Ratio vs. RH

The calculated color ratio **CR** for different RH is shown in Fig. 3. It can be noticed here that the color ratio shows opposite tendency at lower and higher RHs, respectively. In general, the value of the color ratio is about 0.5 for dry particles. The color ratio slightly decreases with increase in RH from “dry” condition to a critical RH_T about 50% (κ=0.2), 60% (κ=0.4), or 75% (κ=0.61). If RH is larger than RH_T, the color ratio turns to increase rapidly with increase in RH and reaches a maximum *CR* of ∼0.7-0.8 at RH≈95%. This result implies a κ-dependent turning point RH_T might be useful as an indicator of κ.

3.3. Case Study

A case observed on 13 January 2009 is selected to simulate and investigate the impact of hygroscopic growth. Fig. 4 shows the time-height evolution of range corrected signal ($P \times z^2$) for 532 nm observed by RCEC/ASNTU lidar. The height of boundary layer is about 1.5 km throughout the day. The height of mixed layer was determined using gradient algorithm [7] and is around 1 km from 00Z - 06Z and decreased to 700m - 500m after 06Z.

A radiosonde was launched on 13 January 2009 at 00Z to measure the vertical profile of atmospheric parameters. The profile of measured RH is shown in Fig. 5. Based on this RH profile, the relative backscattering coefficients for 355 nm and 532 nm are calculated and shown in Fig. 6. Aerosols are assumed being well mixed and hygroscopic growth follow the κ-Köhler theory. The initial size distribution is shown in Fig. 1. The complex refractive index of dry aerosol is set to $m = 1.44 + 0.043i$ and κ is 0.2 (the typical value of κ observed in Taipei [6]).

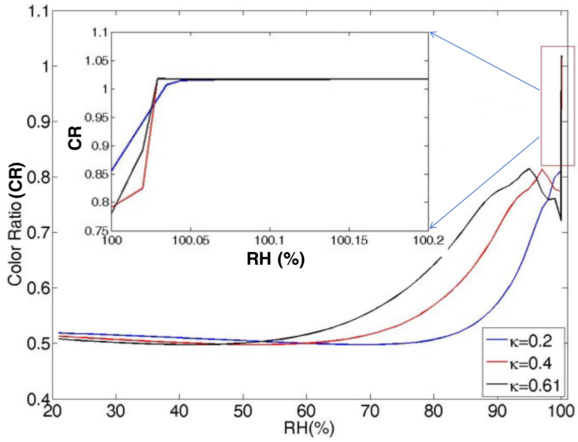

Figure 3: Calculated Color Ratio for different RH for aerosols with κ=0.2 (black), 0.4 (red), 0.61 (blue) and complex reflectivity m=1.44+0.043i.

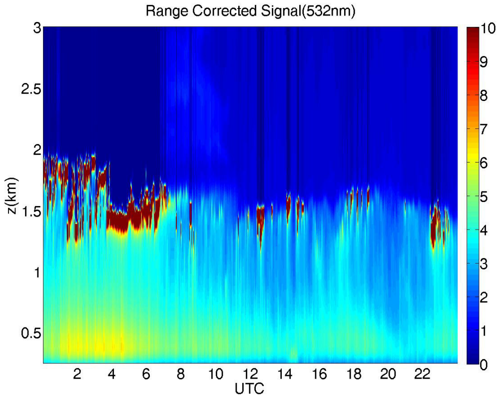

Figure 4: Time-height variation of RCEC/ASNTU Lidar range corrected signal (RCS) at 532 nm observed on 13 January 2009 at Taipei

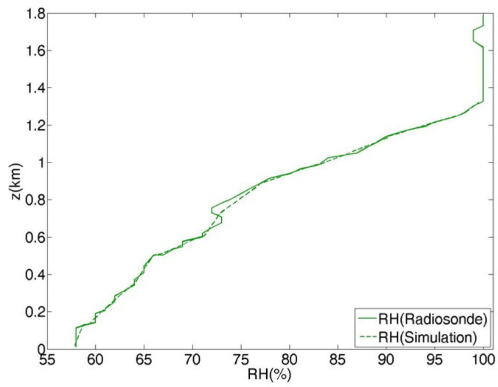

Figure 5: Radiosonde measured (solid green) RH profile on 13 January 2009 at 00Z. Smoothed RH profile (dashed green) used for hygroscopic simulation.

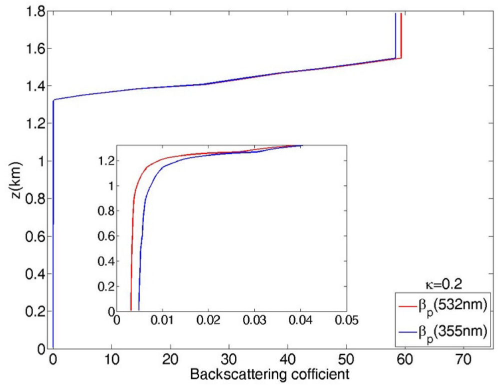

Figure 6: Simulated lidar backscattering coefficients for 355 nm (blue line) and 532 nm (red line) with κ=0.2 and m=1.44+0.043i. The initial size distribution is shown in Fig. 1 and RH profile is shown in Fig. 5.

As shown in Fig. 5, RH was about 55% at ground level. RH gradually increased with height and reached RH=98% at height $z\approx$1.25 km. A small inversion in RH was found at $z\approx$0.8 km, which has been smoothed (dashed green) for hygroscopic simulation. The simulated lidar backscattering coefficients for two wave lengths are shown Fig. 6. Backscattering coefficients for 355 nm (β_{355}) and 532 nm (β_{532}) at ground level (z=0) are about 0.005 and 0.003 m^{-1}sr^{-1}, respectively. β_{355} and β_{532} slightly increase with height up to $z\approx$1 km or RH$\approx$90%. β_{355} and β_{532} turn to rapidly increase to 0.04 m^{-1}sr^{-1} when RH$\approx$98%. Beyond RH=98%, aerosols are activated as cloud droplets and the corresponding β_{355} and β_{532} increase to 55 at $z\approx$1.4 km, which are about 1000 times higher than β_{355} and β_{532} at surface level.

The height distribution of observed (solid red) and calculated (solid blue) color ratios are shown in Fig. 7. The observed **CR** decreases from 0.6 to 0.4 with height increasing from surface level to 1.3 km. **CR** turns to rapidly increases from 0.4 to 1.4 with height (or RH) increasing from 1.3 km to 1.4 km. The calculated **CR** slightly increases from 0.42 to 0.6 with height increasing from surface level to 1.2 km. The calculated **CR** turns rapidly increases from 0.6 to 1 with height increasing from 1.2 km to 1.3 km.

The similarities and discrepancies between observation and simulation can be noticed here. The calculated **CR** starts to rapidly increase at lower RH than observed **CR**. The vertical structures of observed and calculated **CR** are much different between 1.3 km and 1.5 km. This discrepancies might be owing to incorrect κ value or RH. Fig. 8 shows the calculated **CR** profiles based on the RH profile (dashed blue) modified from radiosonde measurement (solid green) to match the observed **CR**. This result shows, a small uncertainty would cause large difference from reality. Another implication from Fig. 7 and Fig. 8 is κ might be over estimated. It can be noticed that the simulated **CR** profile with κ=0.1 is more closer to lidar observation.

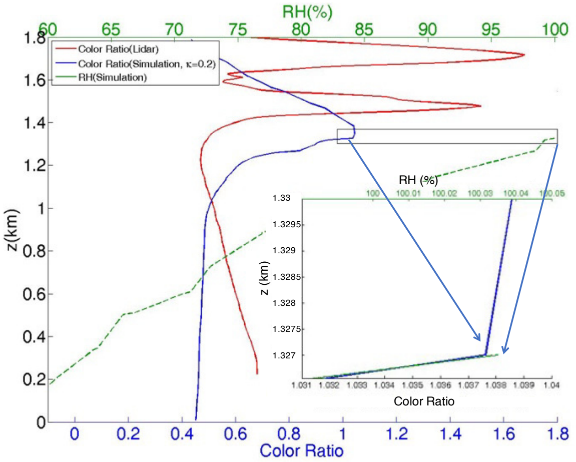

Figure 7: Observed (red) and simulated (greed) vertical profile of Color Ratio (**CR**) with κ=0.2 and RH profile (blue) observed by radiosonde.

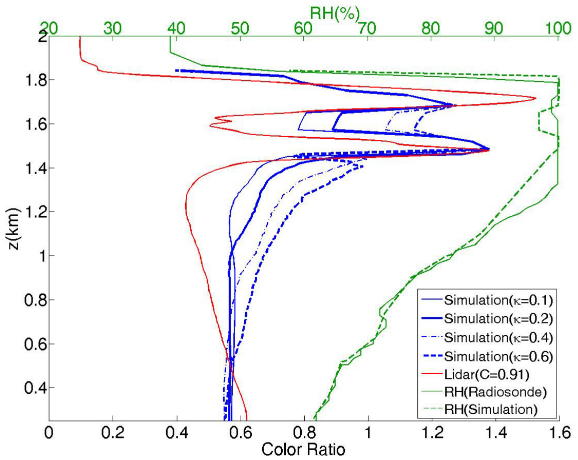

Figure 8: Same as Fig. 7 but with various κ and modified RH profile (blue).

4. SUMMARY

- In general, **CR** of "dry" particle slightly decreases with increase in RH from 0% to a critical RH_T about 60% (κ=0.2), 70% (κ=0.4), or 80% (κ=0.61). Color ratio turns to increase rapidly with increase in RH when RH is larger than RH_T.
- The discrepancies might be owing to incorrect κ value or RH. Measured RH is found highly uncertain when RH higher than 95%.
- Considering the value of κ about 0.1 can make the simulation and observation more closer but it is lower than typical value of κ observed by CCN counter. More study is needed to reveal the possible reasons. Full investigation will be presented in the conference.

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