

Response to reviewer#1

Thanks for the reviewer's helpful suggestions! The comments are addressed point-by-point and responses are listed below.

Comment 1: The lidar ratio is an aerosol-type dependent parameter. Values for different aerosol types have been measured in very different environments under varying relative humidity. From these measurements, the lidar ratio of a certain aerosol type can generally be defined with a rather low standard deviation. This would not be the case if humidity effects would be as strong as described in the manuscript. Also, lidar observations are always at ambient conditions and rarely at the low relative humidity that are used by in-situ instruments.

Reply: The reason that lidar ratio (LR) is an aerosol-type dependent parameter is that it is a synthetic parameter that depends on the aerosol particle number size distribution (PNSD), aerosol composition and aerosol shape. Different types of aerosols can have different micro-physics properties and lead to different lidar ratio.

There are many works that measure lidar ratio in very different environment, and lidar ratio of different aerosol type can vary at a wide range. Ansmann et al. (2001) reported that the lidar ratio ranged from 30 to 80 sr in the polluted continental air at wavelength of 532nm. Müller et al. (2005) observed the lidar ratio of 532nm can vary from 26 to 87 sr at different height. Similar ranges of lidar ratios of the biomass burning aerosols are reported (Ulla et al., 2002).

Lidar ratios can be significantly influenced by relative humidity (RH). Salemink et al. (1984) reported the measured lidar ratios at different RH and found that the lidar ratios can increase linearly from 20 to 70 sr when the RH change increase from 40% to 80%. (Ferrare et al., 1998) also found that the lidar ratios can vary from 60 to 90 sr when the RH increases from 40% to 90%. Further research found that lidar ratio is likely to change significantly due to the substantial variation of RH in the mixed layer (Ferrare et al., 1998).

Lidar observations are always at ambient conditions and the ambient RH values vary significantly from day to day.

Comment 2: The authors extrapolate the height profile of the dry particle number size distribution from measurements at the surface. In my opinion, the authors are merely replacing any uncertainty that might be introduced by using a constant lidar ratio with the much more complex uncertainty of extrapolating dry surface measurements to a height profile, humidifying these size distributions and transforming them to optical data (i.e. lidar ratios).

Reply: The motivation of this paper is to theoretically analyze the impacts of aerosol hygroscopic growth on the LR and propose a feasible method to derive the aerosol extinction coefficient profile. At the same time, sensitivity studies are carried out to study the uncertainties of PNSD, AOD, and hygroscopicity. Many factors that are

unrelated to our research are parameterized in the vertical direction. There are many works carried out to parameterize the vertical distribution of aerosols for convenience. Furthermore, our work concentrates on the well-mixed atmospheric vertical structures as mentioned at line 93 in the manuscript. There are some other works that use the similar assumptions in the vertical direction to study the aerosol optical properties and the corresponding influence (Ferrero et al., 2014; Kuang et al., 2016; Kuang et al., 2015). Finally, we compare the retrieved results of the real-time measurement elastic-backscatter lidar signals at section 4.4 and these results show good agreements with the aerosol vertical distribution assumptions.

Comment 3: Independent profile measurements of the particle number size distribution, the extinction coefficient, and indeed the lidar ratio are needed to properly assess the merits of this work.

Reply: We agree with the reviewer's views about more working should be carried out to assess the merits of this work. However, so far these works remains a worldwide challenge to match the lidar measurement results and the independent profile measurements results. At the same time, we are now considering carrying out some works recommended by the reviewer and some results may show in other works.

Comment 4: The finding that increasing relative humidity increases the lidar ratio is not intuitive. While the increased particle size is producing a larger fraction of forward scattering compared to the dryer particles, it is the ratio of extinction coefficient (scattering plus absorption) to backscatter coefficient that determines the lidar ratio. In fact, the backscatter coefficient increases stronger in relation to the extinction coefficient when particles grow in size by taking up humidity. This manifests for instance in the low lidar ratio of 20 sr for water droplets. The highest lidar ratios are usually related to highly absorbing particles, rather than humidified ones.

Reply: These comments are lack of common knowledge of aerosol optics and totally unacceptable. Salemink et al. (1984) reported the measured lidar ratios at different RH and found that the lidar ratios can increase linearly from 20 to 70 sr when the RH change increase from 40% to 80%. (Ferrare et al., 1998) also found that the lidar ratios can vary from 60 to 90 sr when the RH increases from 40% to 90%. By

definition, $LR = \frac{\sigma_{ext}}{\beta_{sca}}$, where σ_{ext} is the extinction coefficient and β_{sca} is the

backscattering coefficient at 180. β_{sca} can be written as $\beta_{sca} = \frac{\sigma_{ext} \times SSA \times PF(180)}{4 \times \pi}$,

where the SSA is single scattering albedo, which is defined as the ratio of extinction coefficient and scattering coefficient. PF(180) is the scattering phase function at the scattering angle of 180°.

Thus, $LR = \frac{\sigma_{ext} \times 4 \times \pi}{\sigma_{ext} \times SSA \times PF(180)} = \frac{4 \times \pi}{SSA \times PF(180)}$. When particle grows, the phase

function at the scattering angle of 180° is smaller, and the SSA doesn't match the variation of the phase functions.

The variation of LR with diameter of core-shell mixing state and the mean distribution of PNSD measured at Hachi Campaign are shown in fig. 1. From fig.1, LR of single aerosol particle can varies from 20 to 300 sr with the increase of the particle diameters. At the same time, aerosol PNSD decreases with the diameter. Mean LR value is 45.4sr, which corresponds to the LR value with a mean diameter of 238nm. When particles get hygroscopic growth, the mean diameter grows, and then the mean LR grow larger. At the same time, most of the particles distribute at the range of 100 and 300nm. When these particles grow larger, they tend to have a larger value of LR.

As for the LR of water droplets, it varies at a large range from around 10 to larger than 400sr according to the calculated results of Mie scattering theory shown in fig.2. For those water droplets larger than 1000nm, they tend to have a low lidar ratio at the range of 10sr to 50sr. So we cannot understand what this comment "This manifests for instance in the low lidar ratio of 20 sr for water droplets" means.

Finally, we agree with the reviewer's opinion that the highly absorbing aerosols usually relates to high LR from the definition of the LR.

Comment 5: It is not described how the lidar ratio has been obtained. Also, it is not clear from the figures which results are simulated and which measured.

Reply: The definition of LR is detailed at line 41 in the manuscript: "article extinction-to-backscatter ratio, which is usually termed as the lidar ratio (LR), is required when retrieving σ_{ext} profiles". Correspondingly, the author added some information at line 109: "The results of Mie model contains the information of the σ_{ext} and β_{sca} , which can be used to derived the LR directly." With the information, LR can be calculated.

We added some information to clarify the simulated lidar signals at line 262: "Fig. 4 provides an example of the retrieved σ_{ext} profile by using the variable LR profile method and that by using the constant LR profile method from simulated lidar signals" and at the caption of figure 4. The measured lidar signals in section 4.4 are already marked with real-time measurement ones.

Ansmann, A., Wagner, F., Althausen, D., Müller, D., Herber, A., Wandinger, U. (2001) European pollution outbreaks during ACE 2: Lofted aerosol plumes observed with Raman lidar at the Portuguese coast. *Journal of Geophysical Research Atmospheres* 106, 20725 – 20733.

Ferrare, R.A., Melfi, S.H., Whiteman, D.N., Evans, K.D., Poellot, M., Kaufman, Y.J. (1998) Raman lidar measurements of aerosol extinction and backscattering: 2. Derivation of aerosol real refractive index, single-scattering albedo, and humidification factor using Raman lidar and aircraft size distribution

measurements. *Journal of Geophysical Research: Atmospheres* 103, 19673-19689.

Ferrero, L., Castelli, M., Ferrini, B.S., Moscatelli, M., Perrone, M.G., Sangiorgi, G., D'Angelo, L., Rovelli, G., Moroni, B., Scardazza, F., Mocnik, G., Bolzacchini, E., Petitta, M., Cappelletti, D. (2014) Impact of black carbon aerosol over Italian basin valleys: high-resolution measurements along vertical profiles, radiative forcing and heating rate. *Atmospheric Chemistry and Physics* 14, 9640-9663.

Kuang, Y., Zhao, C.S., Tao, J.C., Bian, Y.X., Ma, N. (2016) Impact of aerosol hygroscopic growth on the direct aerosol radiative effect in summer on North China Plain. *Atmospheric Environment* 147, 224-233.

Kuang, Y., Zhao, C.S., Tao, J.C., Ma, N. (2015) Diurnal variations of aerosol optical properties in the North China Plain and their influences on the estimates of direct aerosol radiative effect. *Atmos. Chem. Phys.* 15, 5761-5772.

Müller, D., Mattis, I., Wandinger, U., Ansmann, A., Althausen, D., Stohl, A. (2005) Raman lidar observations of aged Siberian and Canadian forest fire smoke in the free troposphere over Germany in 2003: Microphysical particle characterization. *Journal of Geophysical Research Atmospheres* 110, 2333-2340.

Salemink, H.W.M., Schotanus, P., Bergwerff, J.B. (1984) Quantitative lidar at 532 nm for vertical extinction profiles and the effect of relative humidity. *Applied Physics B* 34, 187-189.

Ulla, W., Detlef, M., Christine, B., Dietrich, A., Volker, M., Jens, B., Volker, W., Markus, F., Manfred, W., Andreas, S. (2002) Optical and microphysical characterization of biomass - burning and industrial - pollution aerosols from - multiwavelength lidar and aircraft measurements. 107, LAC 7-1 - LAC 7-20.

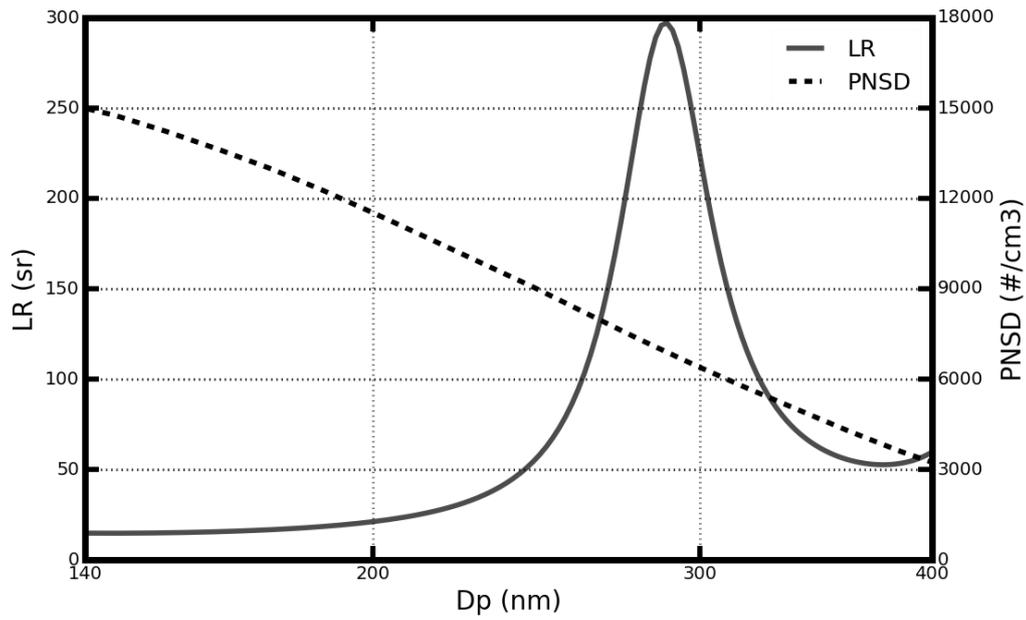


Figure 1. Solid line shows the distribution of aerosol lidar ratio values with different diameters. Dotted line shows the measured mean PNSD at the Hachi Campaign. The lidar ratio values are calculated by using the Mie scattering theory, and the complex of the aerosol is set to be $1.53+10^{-7}i$.

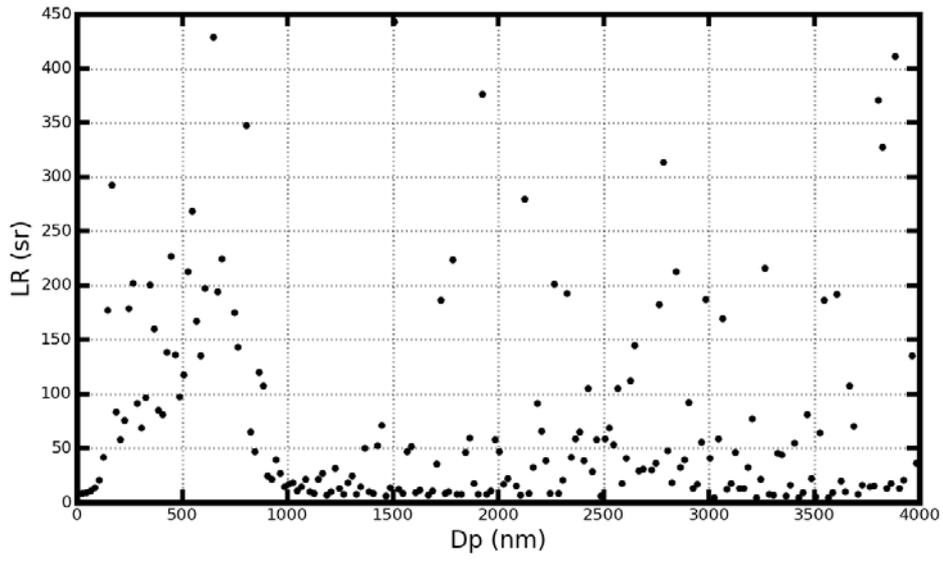


Figure 2. Variation of the LR values of the water droplets at different diameters.