Response to Anonymous Referee #2

The manuscript presents a method to derive microphysical observations from lidar observations at different wavelengths. Such lidar-microphysical retrieval schemes are of great importance in studying aerosol and clouds. However, I have major concerns regarding the feasibility of the technique given the systematic effects on the retrieval of the particle backscattering coefficient (needed for your approach), which is known to be a difficult retrievable for cloudy situations, due to the lack of a reference (aerosol-free) height to calibrate the lidar. It is also difficult to see a practical use of the method, considering the limited retrievable range of sizes of the droplets (or aerosols). Furthermore, to my opinion, the different sections of the study were not developed and discussed deeply enough. Major revisions need to be made.

Response: Thank you very much for your nice comments. Your question and suggestion are very helpful for us to improve the quality of our paper. We appreciate the reviewer's thoughtful review and constructive comments. The following is our point-to-point replies.

Please see my specific comments below:

Page 1 (Introduction): You introduced the problem of getting microphysical information about aerosol and clouds, but as aerosol particles and cloud hydrometeors have been historically approached in different ways, you need to introduce the approaches separately. On the one hand, spectrally resolved information has shown a potential to retrieve microphysical information in the case of aerosol particles, in the case of water clouds, there are quite some limitations because of the larger sizes compared to aerosols.

For this reason, in the case of clouds, there have been several studies that have tried to get information using lidar/radar synergy or lidar-only approaches based on multiple scattering that can be evaluated using dual- or multiple-FOV lidar. A thoughtful literature review of current cloud-retrieval techniques is missing in the manuscript.

Answer: Thank you for your nice comments. A thoughtful literature review of current cloud-retrieval techniques has been added in the revised manuscript, and they are " For clouds, there are two methods used for the detection of cloud microphysical parameters. The first method is using lidar/radar synergy for cloud microphysical parameters [a1-a3], which can achieve the retrieval of cloud droplet with large cloud particles. For thin and sparse clouds or nascent clouds, cloud droplet particles are usually small and cannot be detected by millimeter wave cloud radar, which affects the application of this method. The second method is to use multiple scattering information in clouds detected by multi field of view (FOV) or dual FOV LiDAR to retrieval microphysical parameters of water clouds [b1]. However, in order to obtain multiple scattering signals using ground-based lidar, the larger FOV of telescope is required, which will greatly affect daytime detection. "

a1. Zhien Wang and Kenneth Sassen. Cirrus Cloud Microphysical Property Retrieval Using Lidar and Radar Measurements. Part I: Algorithm Description and Comparison with In Situ Data. Journal of applied Meteorology. 2002, 41: 218-229.

a2. Jothiram Vivekandan, Virendra P. Ghate, Jorgen B. Jensen, Scott M. Ellis, and M. Christian Schwartz. ATechnique for Estimating Liquid Droplet Diameter and Liquid Water Content in Stratocumulus Clouds Using Radar and Lidar Measurements. Journal of Atmospheric and Oceanic Technology, 2020, 37: 2145-2161.
a3. Yinchao Zhang,Su Chen,Wangshu Tan,Siying Chen,He Chen,Pan Guo, Zhuoran Sun, Rui Hu, Qingyue Xu, Mengwei Zhang, Wei Hao and Zhichao Bu. Retrieval of Water Cloud Optical and Microphysical Properties from Combined Multiwavelength Lidar and Radar Data. Remote Sens. 2021, 13, 4396.
b1. Nanchao Wang,Kai Zhang,Xue Shen,Yuan Wang, Jing Li, Chengcai Li, Jietai Mao, Aleksey Malinkad,Chuanfeng Zhao,Lynn M.Russellf,Jianping Guo, Silke Gross, Chong Liu, Jing Yang, Feitong Chen,Lingyun Wu, Sijie Chen, Ju Ke,Da Xiao, Yudi Zhou , Jing Fang, and Dong Liu. Dual-field-of-view high-spectral-resolution lidar: Simultaneous profiling of aerosol and water cloud to study aerosol–cloud interaction. PNAS, 2022, 119(10): e2110756119

Eq. 2: It is not quite clear how one gets this equality using the gamma function. Can you add some more steps and explanations to this derivation? And, how valid is it to use the gamma function definition in an integration that does not go up to infinity?

Answer： Eq.(3) in the manuscript is the definition of the Gamma function in the positive real number field in mathematics. When applied to aerosol particles or cloud droplets, although the radius is arbitrary, the particle radius interval cannot be infinitely small. The number of particles within each radius bin is also an integer. We use functions to fit the particle number concentration distribution, such as Junge Distribution, Modified Gamma Distribution, Lognormal Distribution, etc., in order to seek more intuitive ways to describe the particle number concentration distribution. Simplifying the distribution of discrete multi-channel particle numbers with radius to a Gamma distribution containing only three parameters may indeed reduce the accuracy of describing the number concentration spectrum, but it is beneficial for understanding the average characteristics and spatiotemporal distribution of particles.

In the revised manuscript, we have swapped the order of formulas (3) and (4).

Line 82: Is there a reference to cite for the cloud probe? FSSP-100-ER. You only include a reference for the aerosol probe.

Answer： FSSP-100-ER is an instrument that measures cloud droplet size and concentration using light scattering, with the measurement range of 0.5-47 μm. This has been added in the revised manuscript.

Line 79: These are quite interesting results. Could you deepen the meaning of those parameters? In principle, the b parameter is related to the width and c is related to the size. Is Figure 1 saying, there is always a linear relationship between the size and the width of the distribution?

Answer： In principle, the b parameter is related to the width and c is a slope parameter. Figure 2.1 shows the statistical relationship between b and c, which was obtained from the statistical results between the aerosol size distribution and cloud droplet size distribution observed by the aircraft.

Line 110-11: How exactly can you derive the effective radius from Eq. 14?

Answer: The detailed descriptions of the algorithm will be added in the revised manuscript. "In this algorithm, the first step is to establish a lookup table between aerosol/cloud optical parameters and microphysical parameters. 1) Assuming that aerosol particles and cloud droplets follow the Gamma distributions, calculate the extinction coefficient and backscatter coefficient at different laser wavelengths (355nm and 1064nm in this paper) based on the Mie scattering theory; 2) Calculate the ratio of backscatter coefficients for two wavelengths, which is the backscatter color ratio, or calculate the ratio of extinction coefficient to backscatter coefficient, which is the radar ratio; 3) Change the parameters of the aerosol to obtain the gamma distributions with effective radius from 0.2 μm to 3 μm, calculate the optical parameters and corresponding optical parameter ratios (radar ratio or backscatter color ratio) for each Gamma distribution, and establish the lookup table for aerosol effective radius; 4) Similar to the step 3, establish the lookup table for cloud drops (effective radius are from 0.5 μm to 5 μm). After the lookup table is completed, the microphysical parameters of aerosols or clouds are calculated based on the lookup tables and LiDAR detection data. The specific steps are as follows: 1) the dual-wavelength (355 nm and 1064 nm) Raman LiDAR need be selected for the detection of atmosphere; 2) Raman and Fernald methods are used for the retrieval of optical parameters at multi-wavelengths, and the backscatter color ratio or lidar ratio can be obtained ; 3) aerosol and cloud layers are identified based on lidar echo signals; 4) Retrieve the effective radius of aerosols or cloud droplets at different heights based on optical parameters ratios and lookup tables; 5) Calculate the parameters b and c in the Gamma distribution according to formulas (13) and (16); 6) Calculate the value of a in the Gamma distribution according to formula (17); 7) Calculate the number concentration according to formula (3). "

Fig 3, Fig 4: It is not stated how the size range limits were defined (the blue lines). How can one assume one is on this range only using, e.g., the backscattering ratio 355/1064? Smaller particle sizes (left side of the range) might also produce similar ratio values. There is no uniqueness in the parameter you propose to use.

Answer: The applicability of this algorithm is limited, and it is applicable for aerosols and small cloud droplets. For aerosols, particle diameter is usually 0.01~10 μm, while the effective particle diameter ranges from 0.3 μm to 1.2 μm for urban aerosols (This is calculated based on ground and aircraft observation data.). Usually, water droplets larger than 2 microns are called cloud droplets. Therefore, this algorithm is suitable for the detection of urban aerosols and initial cloud formation, or for cloud lateral boundary.

Line 164: How do you exactly verify the algorithms? How were the backscatter and lidar ratios calculated from the size distributions? Please provide more accurate information on what is obtained from the measurements.

Answer: Thanks for your nice comments.

The verification of this algorithm is achieved through simulation. The specific steps are as follows: 1) Calculate the effective radius and number concentration using APSD and

CDSD observed by aircraft and formula (9); 2) Calculate the backscatter coefficient at two wavelengths of 355 nm and 1064 nm according to the formula (1a), and then calculate the color ratio; 3) According to the color ratio and the algorithm described in Figure 3, the effective radius and number concentration profiles can be retrieved; 4) Compare the effective radius and numerical concentration in steps 2) and 4), as shown in Figure 6, to verify the algorithm inversion.

According to formula (1a), the backscattering coefficient can be calculated from the APSD or CDSD.

$$\alpha(\lambda) = \int_{r_{\min}}^{r_{\max}} \pi r^2 Q_{\text{ext}}(m, r, \lambda) n(r) \mathrm{d}r \quad (1a)$$

$$\beta(\lambda) = \int_{r_{\min}}^{r_{\max}} \pi r^2 Q_{\text{b}}(m, r, \lambda) n(r) \mathrm{d}r \quad (1b)$$

here, $\alpha(\lambda)$ and $\beta(\lambda)$ are the extinction coefficient and backscattering coefficient at wavelength of λ, m is the complex refractive index, $Q_{\text{ext}}(m, r, \lambda)$ and $Q_{\text{b}}(m, r, \lambda)$ are the extinction and backscattering efficient factor, $n(r)$ is the number particle size distribution. Lidar ration can be calculated from the following equation.

$$\mathrm{LR} = \frac{\alpha(\lambda)}{\beta(\lambda)} \quad (1c)$$

Line 179: Only theoretical errors are considered, what about systematic errors, such as in the retrieval of the backscattering coefficient, first needed to initialize the retrieval of microphysical properties?

Answer： More discussions and explanations about error analysis of the algorithm has be added to the revised draft.

For urban aerosols and water clouds, their particles are spherical, so the error caused by non-spherical particles can be ignored.

The error introduced by the assumption of Gamm distribution is relatively complex and difficult to accurately calculate. This study evaluates this error by numerical simulation based on APSDs and CDSDs data by aircraft observations. Actually, the error presented in Figure 7 is mainly caused by the assumption of Gamm distribution. Calculate optical parameters of over 5000 sets of APSDs and CDSDs data, and retrieve the microphysical parameters using our algorithm. The calculated standard deviations between the inversion results and the actual data are: for aerosols, the standard deviation of the effective radius is ~10%, and the standard deviation of numerical concentration is 20%; For clouds, the standard deviation of the effective radius is 15%, and the standard deviation of numerical concentration is 20%.

The deviation introduced by improper assumption of complex refractive index may be the largest term in this technique. For water clouds, the complex refractive index is stable and the deviation caused by it can be ignored. It is difficult to accurately obtain the complex refractive index of aerosols, and the deviation caused by the complex refractive index may reach over 100%. Figure 6 shows the effect of complex refractive index variation on the optical parameter ratio. From Figure 6, it can be seen that when the real part of the complex refractive index changes within the range of 0.03 and the imaginary part changes within 0.01, the effective radius deviation caused by the complex refractive index is within a controllable range. After calculation, the deviation does not exceed 40%. And it can be seen that although complex refractive index can lead to the significant change of the effective radius value, when the aerosol is constant, its monotonic characteristics remain unchanged, which means that the evaluation of particle size changes is reliable.

The above three errors are independent of each other. Considering the actual inversion ability of LiDAR, the deviation of color ratio will reach 10%. The final evaluation shows that the mean square deviation of the inversion error of aerosol effective radius is less than 45%, and the standard deviation of the inversion error of cloud droplet effective radius is 25%.

Line 222: how is the black curve defined/determined?

Answer： The authors have made modifications and added explanations for the boundary of the aerosol layer and the cloud layer in the original text's figures and descriptions. After modification, for the data from 03:00 to 04:30, the red line indicates the lower boundary of the thin aerosol layer 3.2 km before cloud formation. For the data from 04:30 to 09:40, the black line indicates the cloud base height of the cloud layer. Due to the lack of clear meaning of the black line after 09:40, it is removed. The calculation methods for thin aerosol layer base and cloud base are both based on the differential zero crossing method, while for aerosol layer base, the lower boundary is identified by selecting an appropriate threshold for the differential signal of the echo signal.

The author has made modifications and added explanations for the boundary between the aerosol layer and the cloud layer in the original text's figures and descriptions, as following:

“According to Fig. 8(a), there are signals changing from weak to strong above the black curve near 3 km. After 5:00, the echo signal gradually increased and the laser could not penetrate, suggesting that this should be a process of cloud formation. The red and black lines in the figure correspond to the lower boundaries of the aerosol layer and cloud layer of interest, respectively, calculated by differential zero crossing method. According to the temperature and humidity profiles shown in Fig. 8(b), the temperature below 3.5 km is higher than 0℃, and the relative humidity reaches over 90% at 3 km-3.2 km. Therefore, it can be determined that the strong signal appearing near 3 km in the atmosphere is water cloud. ”

Line 227: The backscattering coefficients at the different wavelengths are key for the retrieval scheme. So how do you exactly calculate the backscattering coefficient? In cloudy situations, it is well known, that is quite difficult to retrieve the backscattering coefficient (either using Klett/Fernald or even the Raman method) because of the strong attenuation in the cloud layer, which does not allow the usage of a reference (aerosol-free) height to calibrate. The strong attenuation also makes the retrieval of the extinction a major issue. On the other hand, multiple scattering will take place in the clouds as soon as it gets densely enough. So how does your approach avoid the multiple-scattering effect?

Finally, how do these underlying uncertainties affect the retrieval of the microphysical parameters, such as the effective radius and cloud droplet number concentration?

Answer：

The reviewer is quite right that the accuracy of the backscatter coefficient is crucial for the retrieval of microphysical parameters, and its inversion error directly affects the final result.

In this manuscript, the backscattering coefficient at 355nm was retrieved using the Raman- Mie method. The inversion accuracy of this method is relatively high. The retrieval of 1064nm backscatter coefficient is performed using the Fernald method. We propose a multi-wavelength aerosol backscattering ratio parameterized-calibration method (MABP-CM) based on parameterized equations for reference condition calibration of Fernald inversion, which was described in detail in Ref. (Xinhong Wang, 2022).

The ground-based lidar is used for the observation of cloud in this manuscript. The laser divergence angle is less than 0.5mrad, the FOV of the telescope is 0.5mrad, and the detected cloud height is low. Therefore, the influence of multiple scattering on the results is weak and can be ignored.

More discussions and explanations about error analysis of the algorithm has be added to the revised draft.

Line 240: How is the maximum number concentration 130 cm-3? In the exemplary profile (Fig. 9f) there are no values larger than 100 cm-3. There was also no explanation of what was done below the cloud base. Was the retrieval version for aerosols applied here, or the cloud version was used for the whole profile?

Answer： The maximum number concentration is less than 100 cm^{-3}, and this will be revised in the revised manuscript. In the process of cloud shown in Figure 8, the aerosol hygroscopicity increase plays an important role. According to Figure 9b, the relative humidity reaches 100% near 3km, and below 3km, the relative humidity is less than 100%. Therefore, the aerosol lookup table is used below the cloud base for the retrieval of aerosol profiles, and the cloud droplet lookup table is used above the cloud base (gray shaded area).

Line 250 (Fig 10f): I do not see, how can it be possible, that the concentration of droplets in the cloud layer (~2000 cm-3), is two orders of magnitude than the aerosol concentration below the cloud (~10 cm-3). Where is all that CCN coming from?

Answer： We have carefully checked our results and ultimately determined that they are reasonable. But we need to pay attention to two points: 1) The number concentration (~2000 cm-3) pointed out by the reviewer is located near the altitude of 3.1km, where the inversion error of the data is the largest. The error range of the number concentration reaches (~200 cm-3 to~2000 cm-3), as indicated by the error bar in the manuscript; 2) There may be much CCN near this location. According to Figure 8, the cloud layer should be formed after the aerosol hygroscopicity increase. On that day, the vertical structure of the atmosphere had an uneven distribution of aerosols, and there should be more aerosols around 3km. And according to Figure 8 (b), it can also be observed that there is a significant inversion layer at 3.2km, so it is normal for there to be more aerosol accumulation below the inversion layer.

Line 258 (Fig. 12): Same issue as Fig 10. And why even bother defining regions 1 and 2, where there was no cloud at all yet?

Answer: The definitions of regions 1 and 2 in the figure are distinguished based on the echo signal in Figure 8. There are no clouds in regions 1 and 2, but based on the lidar echo signal, we can see a more obvious signal growth and change process. The echo signal in Figure 8 shows three distinct stages. The first stage (Region 1) is clouds have not yet formed, but an obvious layer of aerosol can be seen; In the second stage (region 2), there is a significant trend of enhancement in the echo signal; In the third stage (Region 3), cloud droplets are generated and clouds appear.