

## #Referee1

I congratulate the authors for their high-quality work developing a theoretical approach to estimate the lidar ratio values for CALIPSO aerosol models. The research results can be used to evaluate the extinction profiles of atmospheric aerosols by using CALIPSO data. The paper is well written with an excellent logical presentation sequence. The methodology is clearly outlined and based on valid assumptions. The authors describe the approach limitations and expected sources of uncertainties. The authors compared their results to ground observations and results from other consolidated and published results, demonstrating the research's accuracy and contribution to atmospheric science.

However, there are some issues that the authors should work on before the paper's publication:

a) There are a few typo errors that I found and highlighted in the attached file;

**Answer: We thank you for your comment. These are corrected in the revised manuscript.**

b) I'm afraid I have to disagree with the statement in line 300 when the authors discuss the increase of the backscattering coefficient with relative humidity. Figure 3 shows that the backscattering coefficient sensitivity to relative humidity is higher for clean continental aerosols than urban and polluted continental aerosols. Maybe the line patterns in the figure are hard to read in black and white, making it hard to read the information from the plot. I suggest preparing the figure using colors or a continuous line for the clean continental and marine continental for better identification;

**Answer: We thank you for highlighting this point. The backscattering sensitivity with respect to clean continental and clean maritime aerosols was higher as compared to polluted continental, urban and polluted maritime aerosols. This is corrected in the revised manuscript.**

c) I suggest that the authors modify the statement in line 308. According to Figure 6(e), both wavelengths show an increase in LIDAR ratio, but 532 nm has a more significant increase than 1064 nm. Also, the LIDAR ratio values are lower at 1064nm than at 532nm;

**Answer: We thank you for this suggestion. Corrections are incorporated in the revised manuscript.**

d) I recommend using (a), (b), and (c) instead of (c), (d), and (e) in Figures 4 and 6.

**Answer: We thank you for this recommendation. Corrections are incorporated in the revised manuscript.**

e) The Figures 1 to 6 look like the graphs are in low resolution. Their presentation quality can be improved.

**Answer: We thank you for this recommendation. Corrections are incorporated in the revised manuscript.**

Once again, congratulations for the good work.

### #Referee3

Please refer to the attached file.

Chipade et al. developed a theoretical algorithm for lidar ratio estimation for covering all types of aerosol models and analyzed the effect of relative humidity (RH) on lidar ratio. This is a good work and recommended to be published in AMT after concerning some weaknesses.

Major comments:

1. I think the lidar ratio for polluted and continental and polluted maritime aerosols shown in Figure 5 is declined rather than increased. Could you please explain it in more detail? (L313-315)

**Answer: The explanation in L313-L315 is with respect to purely the lidar ratio values. With respect to relative humidity as well the lidar ratio values are uniformly increasing for polluted continental and polluted maritime aerosols as compared to clean continental and clean maritime aerosols i.e. polluted vs the clean aerosol types. This is mainly due to the soot particles present in the polluted aerosols. This is clearly explained in the manuscript.**

2. I'm confused about why the lidar ratios of aerosols indicate the opposite trends when RH is lower or larger than 80% if the water soluble particles dominate aerosols in both RH ranges.

More specifically, why does the lidar ratio of water soluble particles increase after the RH reaches 80%? (L315-322)

**Answer: As mentioned in L309-L311, increase in backscattering coefficient with relative humidity at 532 nm and 1064 nm will cause increase or decrease in lidar ratio with respect to relative humidity depending upon the rate at which the extinction and backscattering coefficients are increasing or decreasing. When the relative humidity increases from 0 to 80% there is significant decrease in imaginary part of refractive index leading to decrease in absorption. As a result, the rate at which extinction coefficient increases is either less than or equivalent to the rate at which backscattering coefficient increases. This results in the decrease in lidar ratio of aerosols when RH is increased from 0 to 80%. The increase in lidar ratio from 80% to 99% is primarily due to increase in size of water soluble particles. Corrections in the statement are incorporated now in the revised manuscript.**

3. Please cite the reference when trying to use some results from other studies to explain the phenomenon. For instance, L349-351.

**Answer: We thank you for this suggestion. These lines refer to Antarctic aerosol model. The references for the Antarctic aerosol model and size distribution of its components are cited in the results and also presented in the Table 3 and 5 of the paper. However, as per our knowledge, not much literature exists for this type and this manuscript would be first to report the lidar ratios of the Antarctic aerosols at 532 nm and 1064 nm. This is the reason, there is no reference cited for the lidar ratios of the Antarctic aerosols in L349.**

Language suggestion:

- 1) There should be a comma before 'respectively', please change it throughout the whole paper;
- 2) Line 309/313/341: 'figure' should be 'Figure'.

**Answer: This is incorporated in the revised manuscript.**

## #Referee5

(1) The authors claimed that they developed a physics-based theoretical approach to estimate lidar ratio values for CALIPSO aerosol models. In my point of view, this study employed the Mie theory to simulate lidar ratios using different aerosol models but did not develop any new methodology.

**Answer: Mie theory is used to compute the lidar ratio for CALIPSO and OPAC Aerosol models in this paper. Although new methodology is not developed, the theoretical derivation of Lidar Ratio for CALIPSO aerosol models and OPAC aerosol models considering their composition is reported for the first time. The development of physics-based theoretical approach to estimate the lidar ratio is the development of framework or derivation of lidar ratio using Mie theory. This is not reported earlier in the literature.**

(2) The paper is not well written, especially for the poor punctuation.

For example, Line 99, ‘this study, attempts to’ -> ‘this study attempts to’; Line 163, ‘Bohren, and Huffman’ -> ‘Bohren and Huffman’.

**Answer: We thank you for your suggestion. Corrections are incorporated in the revised manuscript.**

(3) “backscattering coefficient” rather than “backscatter coefficient”, for example in Line 164, please change it throughout the whole paper.

**Answer: This correction is incorporated in the revised manuscript.**

(4) Line 121-150, The geometric standard deviation should be dimensionless.

**Answer: We thank reviewer for bringing out this error to our notice. This correction is incorporated in the revised manuscript.**

(5) In Line 166 and 169, it was declared that  $n_r$  and  $n_i$  were used for real part and imaginary part of the refractive index, respectively. However, the symbols  $m_r$  and  $m_i$  were used instead in Table 1, 2, and 4.

**Answer: We thank you for bringing this to our notice. The notation is now uniformly mentioned in the revised manuscript.**

(6) In Line 113 and 200, it pointed out that  $r_m$  represented median radius. But ‘mean radius’ was used instead throughout the paper.

**Answer: We thank you for highlighting this.**

**If X has a lognormal distribution with median  $\mu^*$  and geometric standard deviation  $\sigma^*$  then its probability density function,  $f(x)$  is given by**

$$f(x) = \frac{1}{\sqrt{2\pi} x \ln(\sigma)} \exp \left[ -\frac{\ln^2(x/\mu^*)}{2 \ln^2 \sigma^*} \right], \quad 0 < x < \infty, \sigma^* > 0, \mu^* > 0$$

**Then natural logarithm of X i.e.  $\ln(X)$  has Normal distribution with mean  $\mu$  and standard deviation  $\sigma$**

**Where  $\mu = \ln(\mu^*)$  and  $\sigma = \ln(\sigma^*)$ . Normal distribution is symmetric around its mean. Thus, for normal distribution mean, median and mode are all equal.**

Thus, the radius of the aerosol particle distribution can be termed as mean or median or mode. d'Almeida et al. (1991) and Hess et al. (1998) termed the particle radius as mode radius, Ackermann (1998) termed the particle radius as median and Omar et al. (2005 and 2009) termed it as mean radius. Different authors have used different notation for particle radius. However, in this manuscript we have termed the particle radius as median radius.

**Reference:**

Limpert et al. (2001), Lognormal distributions across the sciences: Keys and Clues, Bioscience, 51(5), 341-352.

(7) The unit of  $r_m$  was not specified in Table 1, 2, and 3.

**Answer: This correction is incorporated in the revised manuscript.**

(8) What do 'Nr' and 'Dr' mean in Figure 1 and 2.

**Answer: Nr is extinction coefficient and Dr is Backscattering coefficient. This modification is incorporated in the revised manuscript.**

(9) I disagree with the authors that the simulated lidar ratios were consistent to the in-situ values from AERONET. As can be seen from Table 7 to Table 11, the lidar ratios were underestimated compared to the in-situ values in most cases, especially for the cases of Category-1 and Category-3.

**Answer: We thank you for the suggestion. We have already attempted to address this comparison in detail, in the manuscript between L226 to L242 (kindly refer following paragraph from the manuscript for your kind perusal).**

Tables 7-12 show the statistics of the lidar ratio for different AERONET stations belonging to different categories. The daily averages of the lidar ratios were obtained using the AERONET single scattering albedo and phase function values and were compared with the Mie theory estimated values. The Mie theory estimated values were observed to comply with the observed values of lidar ratios using AERONET data as the theoretically estimated values were lying in between the minimum and maximum of the daily lidar ratio values. The differences in the theoretical values estimated using Mie theory and those observed using AERONET data were primarily due to the refractive indices of the different aerosol types present at the different AERONET stations. Omar et al. (2005) had classified the different aerosol types mentioned in section 2 using cluster analysis and the geographical location of these AERONET stations was also considered to be an important factor while classification. Thus, the composition of the aerosols observed over a period of time varied resulting in the variation of the refractive indices. The theoretically computed lidar ratios were based on the refractive index of the centre of the cluster analysed using AERONET data before 2002 (Omar et al. 2005) whereas the AERONET stations data used in this study spanned over 1998 to 2021 leading to the differences in the refractive indices of the aerosol types. The shape of the aerosol particles, their size distribution and their particle density present in the atmosphere may be the secondary reasons for the differences between the theoretically estimated values of lidar ratio using Mie theory and the lidar ratio computed using AERONET stations data which needs further investigation.

(10) Table 13, the simulated lidar ratio of dust at 1064 nm is much smaller than those in CALIPSO operational algorithm; the simulated lidar ratio of Clean Continental at 532 nm and Clean Marine at both wavelengths are much larger than those in CALIPSO operational algorithm, why?

**Answer: We thank you very much for the comment. We have improved the manuscript by incorporating detailed discussion about the comparison between estimated values and reported values of literature. Following paragraphs have been added now in the revised manuscript. Once again we thank the reviewer for valuable comment.**

**Table 13 shows the lidar ratio values estimated for the CALIPSO aerosol models specified in Omar et al. (2009) and its comparison with the lidar ratio values selected in various versions of CALIOP-CALIPSO operational algorithms. It was observed that the lidar ratio values estimated using Mie theory in present study comply with the lidar ratio values reported in literature for CALIPSO operational algorithms. Omar et al. (2006) reported that the lidar ratio for dust aerosols vary between 10 sr to 146 sr when AERONET stations data was classified using cluster analysis. However, the lidar ratio value for dust aerosols proposed in this study at 1064 nm is lower than that used in the CALIPSO V4 operational algorithm. In case of desert dust particles at 1064 nm the variation up to 31 sr was allowed in CALISPO V4 operational algorithm, whereas the present study proposed lidar ratio value of 20 sr for desert dust aerosols at 1064 nm. The desert dust lidar ratio at 1064 nm proposed for CALIPSO aerosol model was observed to be consistent with OPAC desert aerosol model in which case lidar ratio was observed to be centred on 23 sr. These results for desert aerosols at 1064 nm comply with those reported by Ackermann (1998) where dry desert aerosol lidar ratio was lying just under 20 sr. The results for OPAC aerosol models are discussed in detail in the subsequent section. The dust aerosol lidar ratio values at 532 nm and 1064 nm were defined using discrete-dipole approximation (DDA) technique in CALIPSO operational algorithm initially (Omar et al. 2009). The DDA technique considers the non-sphericity of the dust particles (Kalashnikova and Sokolik 2002), whereas Mie theory is quite applicable to spherical homogeneous particles. Thus, the lidar ratio value at 1064 nm was observed to be underestimated using Mie theory, which was also reported by Cattrall et al. (2005). Shin et al. (2018) have reported that the dust lidar ratio at 1020 nm was centred at 44 sr, 40 sr, 54 sr, 36 sr, and 35 sr at Gobi, Arabian, Saharan, Great Basin and Great Victoria deserts, respectively. The dust lidar ratio at 1064 nm has thus showed a large variation temporally and geographically, and thus encouraging the utility of proposed value of dust lidar ratio for retrieval of aerosol optical properties using CALIPSO data.**

**The lidar ratio proposed for clean continental model at 532 nm in CALIPSO V4 operational algorithm was  $53 \pm 24$  sr, allowing the variation up to 77 sr. The Mie theory estimate for clean continental model at 532 nm was centred on 85 sr considering the refractive index of the centre cluster as provided in Omar et al. (2009). This lidar ratio value for clean continental aerosol model was observed to be consistent with those reported in literature. Omar et al. (2006) have reported that the clean continental lidar ratio value varied between 10 sr to 149 sr when estimated using AERONET stations data and Nehrir et al. (2011) have reported the variation in clean continental lidar ratio of 55 – 95 sr at 532 nm observed at Bozeman, Montana. The high value of lidar ratio at 532 nm for clean continental aerosols was observed due to high absorption by fine sub-micron (particles with radius  $< 0.5 \mu\text{m}$ ) particles. The variation in refractive index will also affect the lidar ratio value, which was evident when compared to OPAC aerosol models where the lidar ratio of clean continental aerosols was centred on 53 sr. Similar results were observed in case of clean marine aerosols at 532 nm.**

The theoretically proposed value in the present study for clean marine aerosols at 532 nm was 57.31 sr. The absorption by the fine particles at 532 nm leads to the high value of lidar ratio. The theoretically estimated lidar ratio for clean marine aerosols at 532 nm was observed to be consistent with that reported in the literature. Masonis et al. (2003) have measured the clean marine aerosol lidar ratio as 60.1 sr at 532 nm during Shoreline Environment Aerosol Study (SEAS) experiment. Dawson et al. (2015) have reported a variation of 10 – 90 sr in the lidar ratio of clean marine aerosols. Li et al. (2022) reported the median value of lidar ratio for clean marine aerosols of 60 sr at 532 nm. Li et al. (2022) have measured a peak value of 55 sr at 532 nm over Bay of Bengal. CALISPO operational V3 algorithm allowed variation up to 68 sr in lidar ratio of clean marine aerosols at 1064 nm whereas the present study estimated the value of 71 sr for clean marine aerosols at 1064 nm. This high lidar ratio value for clean marine particles at 1064 nm was due to scattering by coarse super-micron (particles with radius > 0.5  $\mu\text{m}$ ) particles, which was observed to be consistent as reported in Masonis et al. (2003). Thus, the Mie theory estimated lidar ratio values can provide the physical basis for the CALISPO operational algorithms and can be used as look-up table to derive the vertical extinction and backscatter particulate profiles using satellite data.

(11) Why did the lidar ratios decrease when the relative humidity was between 0~80% while increase when the relative humidity was between 80~99% ?

**Answer:** As mentioned in L309-L311, increase in backscattering coefficient with relative humidity at 532 nm and 1064 nm will cause increase or decrease in lidar ratio with respect to relative humidity depending upon the rate at which the extinction and backscattering coefficients are increasing or decreasing. When the relative humidity increases from 0 to 80% there is significant decrease in imaginary part of refractive index leading to decrease in absorption. As a result, the rate at which extinction coefficient increases is either less than or equivalent to the rate at which backscattering coefficient increases. This results in the decrease in lidar ratio of aerosols when RH is increased from 0 to 80%. The increase in lidar ratio from 80% to 99% is primarily due to increase in size of water soluble particles. This is explained clearly in the revised manuscript.

(12) Significant progress has been made in the research community for improving the aerosol optics modeling. This manuscript is mostly relied on Hess (1998) that was published 25 year ago. We all know that lidar ratios are sensitive to the particle nonsphericity, heterogeneity, and the absorption. Relevant discussions related to these issues and the weakness of the present study should be included.

**Answer:** We thank you for your suggestion. Relevant discussion related to nonsphericity of the particles is already addressed in the current manuscript between L385-L388 (following paragraph is presented for your perusal from the manuscript).

The method presented in this study to estimate the lidar ratio using Mie theory is valid only for spherical, isotropic, non-hygroscopic particles and thus there can be possible errors occurring in the lidar ratio values especially when the aerosols are anisotropic and hygroscopic in nature. Thus, there is future scope for the present study to extend it to theoretical estimation of lidar ratio in case of hygroscopic and anisotropic non-homogeneous particles.

## #Referee2

### General Comments:

This manuscript applies Mie scattering theory to estimate the lidar ratio for different CALIPSO aerosol models. From a technical perspective, this study lacks significant innovation. Additionally, the analysis of the impact of relative humidity on the lidar ratio, based on the estimation results, holds some scientific value. Therefore, it is recommended to reconsider the acceptance of this study after the following issues have been well solved.

### Specific Comments:

1) Many studies have already computed the Lidar ratio based on Mie scattering theory and analyzed the influence of relative humidity on the lidar ratio, such as Zhao et al. (2017). However, there are relatively few studies that apply these computed results to the retrieval of CALIPSO data. The author should, upon calculating the Lidar ratio, conduct an in-depth analysis by combining CALIPSO observational data to investigate the impact of relative humidity or aerosol hygroscopic growth on the retrieval of CALIPSO data.

**Answer: We thank you for your comment. We also agree with you that there are studies that have reported before the Mie theory for estimation of Lidar ratio. However, as per our knowledge, this study is one of the firsts that reports the theoretically estimated Lidar ratio for CALIPSO aerosol models. The Lidar ratios proposed in the study can be used in the retrieval of particulate extinction profiles using CALIPSO data. However, retrieving the extinction and backscatter profiles using the CALIPSO data is a three-step process, which is as follows (Young et al. 2009):**

**Step1: Feature Detection Algorithm to identify the features like aerosol and clouds using the CALIPSO Level 1 data**

**Step2: Classification of identified features as either aerosol or clouds and further sub-classification in to various aerosol types and cloud phases.**

**Step3: Hybrid Extinction Retrieval Algorithm (HERA) to retrieve extinction and backscatter profiles using CALIPSO data.**

**In our opinion, carrying out in-depth analysis by using CALIPSO data to investigate the impact of relative humidity or aerosol hygroscopic growth on the retrieval of CALIPSO data will involve all these three steps and outcome of each of these steps will bear the potential of a separate research paper. In addition, this work is beyond the scope of the present paper.**

2) Mie scattering theory is only applicable to spherical particles. In the CALIPSO aerosol models, several are predominantly non-spherical, and it's evident that Mie scattering theory cannot be applied to them. Some of the results in section 4.1 of this manuscript also confirm this. The author should, considering the results from section 4.1, distinguish between CALIPSO aerosol models for which Mie scattering calculations are applicable and focus the subsequent analysis only on those aerosol models where Mie scattering is applicable. It would be inappropriate to simply summarize the comparison results for various CALIPSO aerosol models in section 4.1 as "good agreement."

**Answer: We have provided the details about the applicability of the proposed approach for spherical particles in the revised manuscript. The CALIPSO aerosol models reported in the**

present study have been used in CALIPSO's operational algorithm (Omar et al. 2009, Young et al. 2009). These aerosol models have been obtained by classifying the AERONET data using cluster analysis (Omar et al. 2005, Young et al. 2009). Mie theory results given by AERONET level 1 data have been used to classify the data into different aerosol models (Omar et al. 2005). Thus, in the present study Mie theory is used to derive the lidar ratio for all the CALIPSO aerosol models.

3) The calculation results regarding the impact of relative humidity on lidar ratio and backscattering coefficient offer many details for further exploration. For instance, lidar ratio appears to exhibit opposite trends with relative humidity variations at low and high relative humidity levels. Furthermore, if the vertical axis of Figures 3-6 could be presented in relative terms, it might allow for a better comparison of gradient differences between different curves.

**Answer: As mentioned in L309-L311, increase in backscattering coefficient with relative humidity at 532 nm and 1064 nm will cause increase or decrease in lidar ratio with respect to relative humidity depending upon the rate at which the extinction and backscattering coefficients are increasing or decreasing. When the relative humidity increases from 0 to 80% there is significant decrease in imaginary part of refractive index leading to decrease in absorption. As a result, the rate at which extinction coefficient increases is either less than or equivalent to the rate at which backscattering coefficient increases. This results in the decrease in lidar ratio of aerosols when RH is increased from 0 to 80%. The increase in lidar ratio from 80% to 99% is primarily due to increase in size of water soluble particles. This is explained clearly in the revised manuscript.**

**The figures are updated in the revised manuscript.**

Reference:

Zhao, G., Zhao, C., Kuang, Y., Tao, J., Tan, W., Bian, Y., Li, J., and Li, C.: Impact of aerosol hygroscopic growth on retrieving aerosol extinction coefficient profiles from elastic-backscatter lidar signals, *Atmos. Chem. Phys.*, 17, 12133-12143, 10.5194/acp-17-12133-2017, 2017.