Response to Editor

Dear Editor/Prof. Silke Gross,

Thank you very much for your and the reviewers' valuable and constructive comments/ suggestions. We have considered the reviewer's comments very carefully. All the comments have been responded point to point as shown below and the manuscript has been revised accordingly. We believe that the quality of the manuscript has been promoted now.

We would like to submit the revised version of the manuscript and hope that it can meet the requirement for publication in **Atmospheric Chemistry and Physics**.

Thank you again for considering our work!

Yours sincerely,

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Responses to RC1

General Remarks

This manuscript presents a valuable long-term study, using an elastic backscatter lidar (532 nm) to evaluate the influence of particle hygroscopic growth on aerosol optical depth (AOD) over central China. The authors have compiled an extensive observational dataset spanning 2010-2024, which enables an analysis of trends related to emission reduction policies. Hygroscopic growth is a critical factor governing AOD, and the authors try to quantify the difference between ambient and dry-condition AOD across different years and seasons. The manuscript provides important new results on this topic, is well and clearly written, and is suitable for publication in Atmospheric Chemistry and Physics. I have just several technical comments.

Response: We appreciate your thoughtful review and valuable comments on our manuscript. In response, we have added a discussion on the influence of hygroscopic growth on lidar ratio, and have conducted a sensitivity analysis to assess the uncertainties in AOD_{dry} introduced by variations in the lidar ratio. In addition, we have also discussed the deliquescence nature of aerosol. Point-by-point responses are provided below, and the manuscript has been revised accordingly.

Specific comments

Comment: Authors use Fernald approach for calculation of the backscattering coefficient and lidar ratio of 50 sr to obtain the extinction coefficient. However, lidar ratio strongly depends on RH, so this issue should be discussed. I recommend the recent publication:

Haarig, M., Engelmann, R., Baars, H., Gast, B., Althausen, D., and Ansmann, A.: Discussion of the spectral slope of the lidar ratio between 355 nm and 1064 nm from multiwavelength Raman lidar observations, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2025-449, 2025.

Response: Thank you very much for pointing this out. Relevant statements have been added as follows "**As mentioned in Section 2.1, the particle backscatter (or extinction) coefficient is retrieved using the Fernald method with an assumed fixed lidar ratio of 50 sr for anthropogenic aerosols. However, it should be noted that the lidar ratio varies with ambient RH and can increase notably under high RH conditions. For example, Haarig et al. (2025) reported that the lidar ratio of continental aerosols measured by Raman lidar at 532 nm was 48.1±7.4 sr at 70-80% RH, and increased to 65.3±9.9 sr at 85-92% RH.**" (please see lines 173-179) The resulting uncertainties in AOD_{dry} introduced by different lidar ratios will be assessed and discussed in the following response.

Comment: Lidar ratio even for low RH presents significant variations. Corresponding uncertainties should be discussed. May be AERONET data can be used, to support the choice of lidar ratio of 50 sr (AERONET allows to reconstructs lidar ratio).

Response: Thank you for the reviewer's valuable suggestions. However, as Wuhan does not have an AERNET site, we adopted typical lidar ratio values for anthropogenic aerosols from existing literature. Previous studies have reported a fixed LR of 50 sr as an average value derived from combined lidar and sun photometer measurements (Takamura et al., 1994) or Raman lidar observations (Müller et al., 2007) for urban aerosols in the ambient troposphere.

Nevertheless, a sensitivity analysis is conducted to assess the uncertainty of applying a fixed lidar ratio of 50 sr for all conditions. A LR-RH relationship from Zhao et al. (2017) is adopted for the sensitivity analysis:

$$\mathrm{LR} = \mathrm{LR_0} \times (0.92 + 2.5 \times 10^{-2}(\mathrm{RH} - 40) - 1.3 \times 10^{-3}(\mathrm{RH} - 40)^2 + 2.2 \times 10^{-5}(\mathrm{RH} - 40)^3) \qquad (1)$$

Since the RH in the lower troposphere over Wuhan ranges approximately from 40% to 70% (see Figure 7b of our manuscript), we set $\mathrm{LR_0}$=47 sr in Eq. (1); this assigns a LR of 50 sr to RH conditions around 50-55%. As derived from Eq. (1) (Zhao et al., 2017), if RH=40%, LR=$\mathrm{LR_0}$×0.92=43.24 sr, which will be applied at RH<40%.

Taking the case on 2 August 2023 as an example, LR can reach up to 70 sr when RH excceds 80% (Figure 1R(a)), resulting in a rapid increase in the extinction coefficient at corresponding altitudes (figure 1R(b)). After removing the influence of hygroscopic growth, the extinction coefficient is lower than that obtained using the original LR=50 sr (Figure 1R(c)). The corrected AOD$_{dry}$ is 0.082, representing a 15.5% decrease from the original value of 0.097. A sensitivity analysis was further conducted for 10 cases in 2023 (γ=0.6), covering a range of pollution levels and RH conditions (Table 1R). The results indicate that when considering the effect of hygroscopic growth on lidar ratio, corrected AOD$_{dry}$ values generally decrease by 10-25%. This sensitivity analysis and the corresponding discussions have been added as an appendix section in the revised manuscript. (please see lines 373-394)

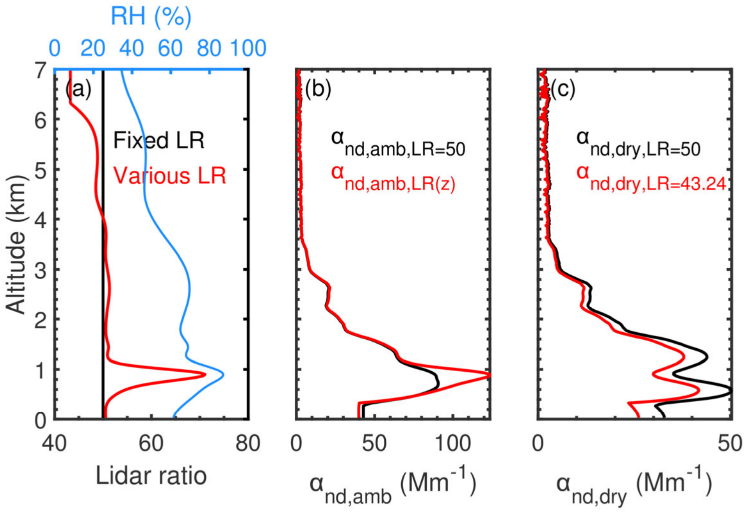

Figure 1R. Profiles of (a) lidar ratio and relative humidity; (b) non-dust extinction coefficient in ambient atmosphere; (c) non-dust extinction coefficient in dry conditions during 1639-1758 LT on 2 August 2023. The Black curves denote the results obtained using the original fixed lidar ratio of 50 sr, while the red curves denote the corrected results.

Table 1R. Comparisons of AOD$_{dry}$: original LR versus corrected LR.

Date	AOD$_{dry,LR=50}$	AOD$_{dry,LR=43.24}$	RH(%)	(AOD$_{dry,LR=50}$- AOD$_{dry,LR=43.24}$)/ AOD$_{dry,LR=50}$
2023-01-04 0400-0519 LT	0.435	0.367	29±18	15.6%
2023-02-03 1347-1417 LT	0.431	0.345	68±10	20.0%
2023-04-09 1753-1824 LT	0.105	0.092	47±26	12.4%
2023-06-08 0927-1046 LT	0.201	0.181	57±17	10.0%
2023-08-02 1639-1758 LT	0.097	0.082	55±19	15.5%
2023-08-13 0957-1114 LT	0.500	0.373	78±8	25.4%
2023-08-22 1625-1744 LT	0.138	0.120	29±24	13.0%
2023-10-24 0933-1052 LT	0.274	0.234	47±23	14.6%
2023-11-22 2008-2127 LT	0.267	0.231	38±24	13.5%
2023-11-29 1131-1250 LT	0.430	0.348	47±28	19.1%

Comment: When Fernald approach is used, backscattering is calculated only above the height of full overlap. I did not find in the manuscript discussions about extrapolation data to the ground level (sorry if I missed it).

Response: For our lidar system, the lowermost height with complete field-of-view (FOV) observation is 0.3 km. Therefore, particle extinction coefficients below 0.35 km were assumed to be equal to those at 0.35 km (Jing et al., 2025). Relevant statements have been added in Section 2.3 (please see lines 159-161)

Comment: Hanel parametrization is applicable above deliquescence point. Discussion about deliquescence nature of aerosol considered would be useful.

Response: In light of the reviewer's comments, the discussions about deliquescence nature of aerosol have been added as follows "**Note that the Hänel parametrization is applicable only above the deliquescence relative humidity (DRH), which is defined as the phase transition point at which a hygroscopic solid transform from the solid phase to the liquid phase at a specific RH (Mauer and Taylor, 2010). Generally, water-soluble inorganic salts exhibit distinct DRH values, e.g., 80% for ammonium sulfate, 75% for sodium chloride, and 61% for ammonium nitrate (Lee et al., 2001; Wise et al., 2007; Zawadowicz et al., 2015). Under ambient conditions, however, the DRH of inorganic salts trends to decrease when they are internally mixed with organic compounds (Brooks et al., 2002; Luo et al., 2020)**…." (please see lines 150-156)

Comment: Abstract, ln 11. "both parameters". Probably should be corrected.

Response: We have modified accordingly.

Comment: P.9, ln 207. The use of "annual mean hygroscopic growth parameter" definitely leads to significant uncertainties. Would be good to discuss.

Response: Relevant statements have been added as follows "**The standard deviation of the annual mean γ would affect AOD$_{dry}$, as indicated by the error bars in figure 6a, resulting in an uncertainty of approximately 10%.**" (please see lines 251-252)

Comment: P.9 ln 210. "These values suggest that hygroscopic growth on average increased AOD by 30.7%...". Keeping in mind uncertainties of approach such accuracy looks excessive.

Response: Thank you very much for your reminder. The relevant statement has been revised as follows "**The average AOD$_{dry}$ and AOD$_{amb}$ were measured to be 0.315±0.164 and 0.404±0.219, respectively, across the period considered in our study. These results indicate that hygroscopic growth on average enhanced AOD by approximately 30% under humid atmospheric conditions.**" (please see lines 231-234) Additionally, the average AOD$_{dry}$ has been updated from 0.309 to 0.315. The initial value was derived using a constant γ=0.62 for the entire period, whereas our analysis employs the annual mean γ for each respective year.

References:

Brooks, S. D., Wise, M. E., Cushing, M., and Tolbert, M. A.: Deliquescence behavior of organic/ammonium sulfate aerosol, Geophys. Res. Lett., 29, 23–1, https://doi.org/10.1029/2002GL014733, 2002.

Haarig, M., Engelmann, R., Baars, H., Gast, B., Althausen, D., and Ansmann, A.: Discussion of the spectral slope of the lidar ratio between 355 and 1064 nm from multiwavelength Raman lidar observations, Atmos. Chem. Phys., 25, 7741–7763, https://doi.org/10.5194/acp-25-7741-2025, 2025.

Jing, D., He, Y., Yin, Z., Huang, K., Liu, F., and Yi, F.: Evolution of tropospheric aerosols over central China during 2010–2024 as observed by lidar, Atmos. Chem. Phys., 25, 17047–17067, https://doi.org/10.5194/acp-25-17047-2025, 2025.

Lee, W. M. G., Huang, W. M., and Chen, Y. Y.: Effect of relative humidity on mixed aerosols in atmosphere, J. Environ. Sci. Heal. A, 36, 533–544, https://doi.org/10.1081/ESE-100103482, 2001.

Luo, Q., Hong, J., Xu, H., Han, S., Tan, H., Wang, Q., Tao, J., Ma, N., Cheng, Y., and Su, H.: Hygroscopicity of amino acids and their effect on the water uptake of ammonium sulfate in the mixed aerosol particles, Sci. Total Environ., 734, 139318, https://doi.org/10.1016/j.scitotenv.2020.139318, 2020.

Mauer, L. J., and Taylor, L. S.: Water-solids interactions: deliquescence. Annu. Rev. Food Sci. Technol., 1, 41–63, https://doi.org/10.1146/annurev.food.080708.100915, 2010.

Müller, D., Ansmann, A., Mattis, I., Tesche, M., Wandinger, U., Althausen, D., and Pisani, G.:. Aerosol-type-dependent lidar ratios observed with Raman lidar, J. Geophys. Res. Atmos., 112, https://doi.org/10.1029/2006JD008292, 2007.

Takamura, T., Sasano, Y., and Hayasaka, T.: Tropospheric aerosol optical properties derived from lidar, sun photometer, and optical particle counter measurements, Appl. Opt., 33, 7132–7140, https://doi.org/10.1364/AO.33.007132, 1994.

Wise, M. E., Semeniuk, T. A., Bruintjes, R., Martin, S. T., Russell, L. M., and Buseck, P. R.: Hygroscopic behavior of NaCl-bearing natural aerosol particles using environmental transmission electron microscopy, J. Geophys. Res. Atmos., 112, https://doi.org/10.1029/2006JD007678, 2007.

Zawadowicz, M. A., Proud, S. R., Seppalainen, S. S., and Cziczo, D. J.: Hygroscopic and phase separation properties of ammonium sulfate/organics/water ternary solutions, Atmos. Chem. Phys., 15, 8975–8986, https://doi.org/10.5194/acp-15-8975-2015, 2015.

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, https://doi.org/10.5194/acp-17-12133-2017, 2017.

Responses to RC2

General Remarks

The paper of He et al. has too much similarity with the paper of Jing et al. (2026) (https://doi.org/10.5194/amt-19-389-2026)., which is not properly disclaimed. Thus, it cannot be accepted. Thus, my decision is to reject the current manuscript, but encourage the authors to work on the structure and then re-submit. Further details are given below.

Many results are given which seem to come from the current study, but in fact they come from the paper of Jing et al. (2026), however, this it is not always clearly stated.

One example: In the abstract you state:

"During China's rapid air-cleaning period of 2010-2017, AODamb declined significantly by -0.068 yr^{-1}; in contrast, the rate of decrease of AODdry was -0.049 yr^{-1} which is 28% slower, but the decrease of the dry aerosols more accurately captures aerosol emission reductions."

But the -0.068 per year do come from the study of Jing et al. (2026) (where you are of course co-authoring) and only the -0.049 per year are the result the novel study which I review here.

This is just one example, but is valid for most of the manuscript. You need to make clear, what is new in this studyà this is investigation of the dry AOD over a longer time and the differences and so on. Everything which was resulting from Jing et al, should be clearly stated and not repeated until really needed.

You could, for example, state in the abstract:" Jing et al. (2026), found a decrease of the ambient AOD by -0.068 per year. We used the same lidar data set to estimate the dry AOD and contrast it to the ambient AOD. Doing so, we find a decrease of -0.049 per year". Or similar.

One more example:

Figure 3a of He et al. is completely similar to Fig. 5a of Jing et al. (2026), but also here this is not properly stated and also not needed for this work.

As a consequence, I would propose the authors rework on the manuscript, which has indeed interesting and new results, but clearly make it a follow-up paper from Jing et al. (2026), stating that they use the same data set and build on the results of the previous publication.

Due to these severe issues, I also did not yet review the results part, because a clear restructuring of the manuscript needs to be done first.

Response: We sincerely appreciate your thoughtful review and valuable comments on this manuscript. In fact, we have conducted a series of studies based on our lidar dataset collected in Wuhan from 2010 to 2024; they together provide a comprehensive picture of the evolution of the atmospheric environment in Wuhan, a typical megacity over central China, over the past 15 years. Please allow us to briefly outline the overall storyline here:

1) First, Jing et al. (2025) investigated the evolution of tropospheric aerosols over Wuhan and identified two distinct phases in AOD variation during 2010-2024: a declining trend (-0.077 yr^{-1}) during 2010-2017 (Stage I), followed by a fluctuating period during 2018-2024 (Stage II). We further divided the AOD contributions from anthropogenic aerosols and dust aerosols and first reported a decline rate of **-0.068 yr^{-1}** (as noted in your comments) for anthropogenic aerosol optical depth during 2010-2017.
2) Second, Jing et al. (2026) focused on the long-term hygroscopic growth characteristics of anthropogenic aerosols using the same lidar dataset collected in Wuhan. In that study, we introduced a methodology for retrieving the hygroscopic growth parameter γ based on the Hänel parameterization, presented a case study as an example, and analyzed the annual and seasonal variations of γ.
3) In the present study under open discussion (He et al., 2026), we still use the same lidar dataset (2010-2024) in Wuhan but focus on deriving the anthropogenic AOD under dry conditions (AOD_{dry}) by applying the annual γ values reported in Jing et al. (2026). By doing this, we can clearly know the respective contributions of ambient humidity and actual anthropogenic aerosol emission to the lidar-observed-ambient AOD. Consequently, the decline rate of **-0.068 yr^{-1}** for anthropogenic AOD during 2010-2017

reported by Jing et al. (2025) can be revised to **-0.049 yr^{-1}** after accounting for hygroscopic growth effect.

Therefore, each of the studies mentioned above has an independent and well-defined objective. We structured the work in this way to avoid an overly lengthy manuscript with too many focuses. We fully agree with the reviewer that the present manuscript should be clearly framed as a follow-up work to Jing et al. (2025) and Jing et al. (2026), building upon the methodology and partial findings reported in those two previous studies. Actually, we indeed stated this **'FOLLOW-UP'** somewhere in the under-review version of the manuscript, for example, in section 3.2 as below: **"Jing et al. (2025) previously reported that the 532-nm $\mathrm{AOD_{amb}}$ in Wuhan decreased rapidly with a rate of -0.068 yr^{-1} during 2010-2017 (defined as 'stage I'), followed by a fluctuating period from 2018 to 2024 (defined as 'stage II'). When accounting for hygroscopic growth in 'stage I', the rate of decline of $\mathrm{AOD_{dry}}$ was -0.049 yr^{-1}, i.e., approximately 28% lower than the rate of decline of $\mathrm{AOD_{amb}}$. This result indicates that the actual reduction in aerosol emissions was slower than that suggested by the direct lidar measurements in the ambient atmosphere."**, and **"All the lidar-derived particle backscatter coefficient profiles were modified from ambient to dry atmospheric conditions by the use of the pre-derived annual mean hygroscopic growth parameter".** Nevertheless, we really appreciate the reviewer's suggestion and will further emphasize the **'FOLLOW-UP'** nature of this work when revising the manuscript, thereby clarifying the overall storyline linking Jing et al. (2025), Jing et al. (2026), and He et al. (2026).

As a result, in the revised manuscript, we have largely modified several paragraphs, explicitly emphasizing that this study is a follow-up work of Jing et al. (2025) and Jing et al. (2026) (please see lines 13, 17-20, 85-94, 191, 193-194, 215, 229, 337, and 346). For Figure 3a, we have added a clear statement indicating that the result was adopted from Jing et al. (2026) and is retained here for the convenience of readers (please see lines 193-194). We have made every effort to clarify what were obtained from our previous studies and what are new findings in the current study. Point-by-point responses to the reviewer's specific comments are provided below, and the manuscript has been revised accordingly. We would greatly appreciate it if the reviewer can kindly reevaluate our work in the next round of review.

Specific comments

However, some few comments already:

Comment: The authors need to discuss the uncertainty introduced by applying one fixed lidar ratio of 50 sr for all aerosols and all humidity conditions.

Response: Thank you very much for the reviewer's constructive comments. Previous studies have reported a fixed LR of 50 sr as an average value derived from combined lidar and sun photometer measurements (Takamura et al., 1994) or Raman lidar observations (Müller et al., 2007) for urban aerosols in the ambient troposphere. Therefore, here we adopted typical lidar ratio values for anthropogenic aerosols from existing literature.

Nevertheless, a sensitivity analysis is conducted to assess the uncertainty of applying a fixed lidar ratio of 50 sr for all conditions. A LR-RH relationship from Zhao et al. (2017) is adopted for the sensitivity analysis:

$$\mathrm{LR} = \mathrm{LR_0} \times (0.92 + 2.5 \times 10^{-2}(\mathrm{RH} - 40) - 1.3 \times 10^{-3}(\mathrm{RH} - 40)^2 + 2.2 \times 10^{-5}(\mathrm{RH} - 40)^3) \qquad (1)$$

Since the RH in the lower troposphere over Wuhan ranges approximately from 40% to 70% (see Figure 7b of our manuscript), we set $\mathrm{LR_0}$=47 sr in Eq. (1); this assigns a LR of 50 sr to RH conditions around 50-55%. As derived from Eq. (1) (Zhao et al., 2017), if RH=40%, LR=$\mathrm{LR_0}$×0.92=43.24 sr, which will be applied at RH<40%. Taking the case on 2 August 2023 as an example, LR can reach up to 70 sr when RH exceeds 80% (Figure 1R(a)), resulting in a rapid increase in the extinction coefficient at corresponding altitudes (figure 1R(b)). After removing the influence of hygroscopic growth, the extinction coefficient is lower than that obtained using the original LR=50 sr (Figure 1R(c)). The corrected $\mathrm{AOD_{dry}}$ is 0.082, representing a 15.5% decrease from the original value of 0.097. Furthermore, 10 cases in 2023 (γ=0.6) were also analyzed, covering a range of pollution levels and RH conditions (Table 1R). The results indicate that when considering the effect of hygroscopic growth on lidar ratio, corrected AOD_{dry} values generally decrease by 10-25%. This sensitivity analysis and the corresponding discussions have been added as an appendix section in the revised manuscript. (please see lines 373-394)

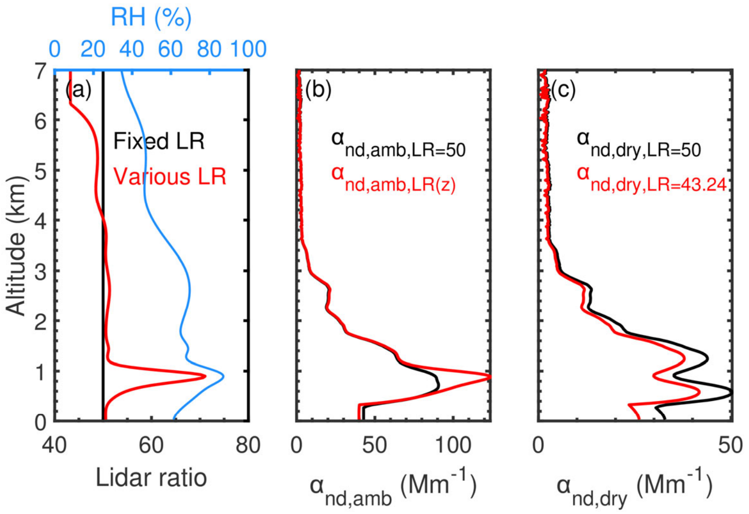

Figure 1R. Profiles of (a) lidar ratio and relative humidity; (b) non-dust extinction coefficient in ambient atmosphere; (c) non-dust extinction coefficient in dry conditions during 1639-1758 LT on 2 August 2023. The Black curves denote the results obtained using the original fixed lidar ratio of 50 sr, while the red curves denote the corrected results.

Table 1R. Comparisons of AOD_{dry}: original LR versus corrected LR.

Date	$AOD_{dry,LR=50}$	$AOD_{dry,LR=43.24}$	RH(%)	$(AOD_{dry,LR=50} - AOD_{dry,LR=43.24}) / AOD_{dry,LR=50}$
2023-01-04 0400-0519 LT	0.435	0.367	29±18	15.6%
2023-02-03 1347-1417 LT	0.431	0.345	68±10	20.0%
2023-04-09 1753-1824 LT	0.105	0.092	47±26	12.4%
2023-06-08 0927-1046 LT	0.201	0.181	57±17	10.0%
2023-08-02 1639-1758 LT	0.097	0.082	55±19	15.5%
2023-08-13 0957-1114 LT	0.500	0.373	78±8	25.4%
2023-08-22 1625-1744 LT	0.138	0.120	29±24	13.0%
2023-10-24 0933-1052 LT	0.274	0.234	47±23	14.6%
2023-11-22 2008-2127 LT	0.267	0.231	38±24	13.5%
2023-11-29 1131-1250 LT	0.430	0.348	47±28	19.1%

Comment: Figure 2 is not clear to me. For example, in my understanding beta_nd is needed also in the grey box, but this is not shown in this figure. Thus, please properly overwork the flowchart. Also, state when you use radiosonde data and when ERA5.

Response: In light of the reviewer's comments, we have added an arrow from β_{nd} (in the blue box) to the grey box. In addition, relevant references have been provided for each part of the methodology. We have also marked in the figure where radiosonde data or EAR5 reanalysis data were used. Please see the updated Figure 2 in the revised manuscript.

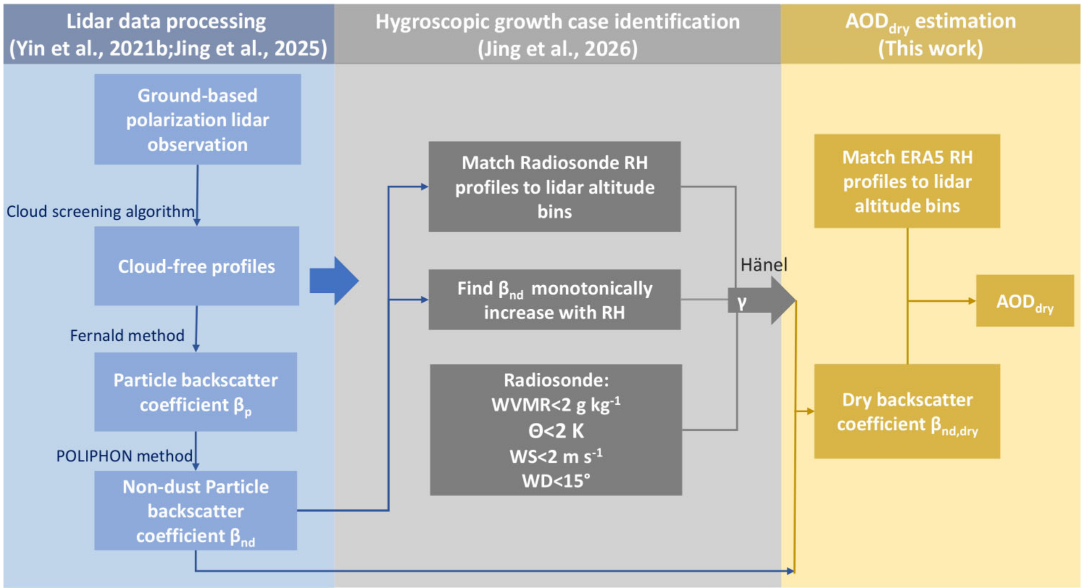

Figure 2R. Revised flowchart of data processing.

References:

He, Y., Jing, D., Yin, Z., Müller, D., Liu, F., Zhang, Y., Yi, Y., Huang, K., and Yi, F.: Hygroscopic growth obscures actual variation in anthropogenic aerosol optical depth over central China during 2010–2024, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2025-6360, 2026.

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