

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-27

Anonymous Referee #1

Referee comment on "Spatiotemporal changes in aerosol properties by hygroscopic growth and impacts on radiative forcing and heating rates during DISCOVER-AQ 2011" by Daniel Pérez-Ramírez et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2021-27-RC1>, 2021

General Remarks:

Aerosol hygroscopicity, which describes water uptake by particles in a humid atmosphere, is a crucial feature that determines the physical properties of particles and subsequently their roles in atmospheric radiation. By integrating a series of lidar, in-situ, and radiosonde measurements from DISCOVER-AQ Washington aircraft campaign, GSFC and AERONET-DRAGON ground stations, and launched radiosondes in site, this study revealed aerosol hygroscopicity vertically in two selected humid days during DISCOVER-AQ Washington campaign. The study explained the significant increase of extinction and backscattering with height owing to aerosol hygroscopicity. The study also revealed that aerosol hygroscopicity is responsible for larger cooling effects in the shortwave range near the ground, and a larger warming near the top of PBL where aerosol hygroscopic growth was highest by using vertically-resolved aerosol extinction/backscattering measured by lidars. This is an interesting and valuable study. The paper is well written. I recommend publishing it in ACP after the authors make the following minor modifications.

Specific comments:

- Abstract line 24: Please explain radiosonde measurement before giving its application.
- Abstract lines 29-30: Need more discussion for this conclusion, see details in specific comment 15
- Page 3 line 3: A good reference you may want to cite here:

Myhre, G., B. H., Samset, M. Schulz, Y. Balkanski, S. Bauer, T. K. Berntsen, H. Bian, N. Bellouin, M. Chin, T. Diehl, R. C. Easter, J. Feichter, S. J. Ghan, D. Hauglustaine, T. Iversen, S. Kinne, A. Kirkevåg, J.-F. Lamarque, G. Lin, X. Liu, G. Luo, X. Ma, J. E. Penner, P. J. Rasch, Ø. Seland, R. B. Skeie, P. Stier, T. Takemura, K. Tsigaridis, Z. Wang, L. Xu, H. Yu, F. Yu, J.-H. Yoon, K. Zhang, H. Zhang, and C. Zhou, Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, *Atmos. Chem. Phys.*, 13, 1853-1877, doi:10.5194/acp-13-1853-2013, 2013.

- Pages 4-6: Could you summarize all the relevant measurements in a table?
- Page 6 line 191: Please also give the full name of "Nd:YAG".
- Page 7 line 226 to page 8 line 2: Why not use Raman Lidar measured backscattering at 532 nm?
- Page 8 lines 255-256: Please explain briefly how to derive aerosol single scattering albedo and asymmetry factor from the P-3B aircraft and the AERONET inversions.
- Page 9 lines 266-272: In general, the study of aerosol hygroscopicity does not depend on the atmospheric condition of well-mixed layers. This condition may be important specifically when using Lidar to study aerosol hygroscopicity. It is better to add "Lidar" somewhere in this sentence. Please also clarify the definition of well-mixed layers: the same type of aerosol with altitude, or the constant water vapor mixing ratio with altitude, or both?
- Page 10 lines 1-2: How do you calculate a new γ' ? Do you keep previous calculated $f(\text{RH})$ unchanged? How do you map RH in the Gaussian distribution in the calculation? A more detailed explanation for γ' calculated is desired.
- Page 10 line 293: The relative differences in γ should "increase", not "decrease", although the absolute γ value "decreases". Also "relative uncertainties" in the sentence should be changed to "random uncertainties".
- Page 10 line 294: Could you elaborate on this sentence? Could random uncertainties be negative? If yes, why do you discuss positive random uncertainties only? Is that because the negative random uncertainties give a symmetric result of its positive counterpart?
- Page 11 line 322-324: I'm confused by the words "maximum" and "minimum" describing relative differences in γ . The minimum bias should be zero. It may be better to use the range of biases for the discussion.
- Page 13 line 382: The approximate time should be around 17:20 UTC.
- Page 19 lines 591-592: What is the measurement evidence to support the conclusion of "with water soluble organic carbon accounting for at least 93% of the total carbon mass"?
- Page 24 lines 702-705 and Figure 11 & 12: This conclusion is not straightforward based on the hourly AOD change shown in Figure 11 & 12 as well as previous discussed aerosol hygroscopicity. The diurnal variation of aerosol dry mass and RH may also contribute to the AOD hourly change. For example, the dry sulfuric aerosol masses shown in Figure 16 start to increase around 19 UTC in both boundary layer and surface. This temporal aerosol mass enhancement also supports the AOD hourly variation. To derive the conclusion more strictly, I suggest rearranging the analysis by first discussing the specific aerosol properties (i.e., total number of particles, aerosol masses, aerosol extinction and hygroscopicity, etc). I also suggest adding the corresponding discussion for P-3B measured RH here.
- Page 26 line 768-769: Did P-3B measure aerosol absorption coefficient as well? It would be better to discuss aerosol masses (e.g., Figure 16) first to help understanding the performance of aerosol extinction in Figure 14 such as the small change between the a_{dry} and a_{amb} above 2km even though $f\text{RH}$ is pretty high. Does P-3B have RH measurement? If yes, could you demonstrate RH values in the three layers in the figure?

- Figures 15 & 16: Please switch the captions of these two figures. Also, the titles should be 2011/07/29 for a,b,c, and 2011/07/22 for d, e, f.
- Page 31 lines 883-887: The unit of the differences of ambient minus dry ARE should be $\text{mW/m}^2/\text{nm}$, not $\text{W/m}^2/\text{nm}$, at specific wavelengths (i.e., 532 nm and 355 nm).
- Page 32 lines 895-896: The impact of aerosol on solar radiative effect depends on not only AOD, but also single scattering albedo (i.e., particle absorption) and asymmetric factor (i.e., particle size and shape). Aerosol particle hygroscopicity impact on all these parameters.
- Page 32 lines 900: Please change "Differences between ..." to "The maximum differences between ...".
- Page 33 lines 942-945: Could the authors show the vertical aerosol dry mass distribution with P-3B measurement? This information helps to untangle the compounding impacts of aerosol hygroscopicity and aerosol dry mass on aerosol extinction (and backscattering).

Technique corrections:

- Page 8 line 239: Section number of "computations of aerosol radiative forcing and heating rates" should be 2.4 instead of 2.3.
- Page 9 line 264: change "hygrosocpicity" to "hygroscopicity".
- Page 12 line 1: Replace "the use of" with "the".
- Page 20 line 599: Please change "imaginary refractive index" to "real refractive index".
- Page 31 line 884: change first "July 29" to be "July 22".
- Page 31 line 885: delete the second "July" in "July 29 July".
- Page 32 line 902: change "July 29" to be "July 22".
- Page 32 line 919: missing "." after the sentence.