

# ***Interactive comment on “HSRL-2 aerosol optical measurements and microphysical retrievals vs. airborne in situ measurements during DISCOVER-AQ 2013: an intercomparison study” by Patricia Sawamura et al.***

## **Anonymous Referee #1**

Received and published: 12 June 2016

Sawamura et al. present in their manuscript a comparison study of airborne lidar and in-situ measurements and retrievals. The measurements were recorded during two aircraft campaigns in 2013 in California and Texas. The in-situ recordings of particle size and light scattering had to be transformed to ambient conditions. For this, the hygroscopic growth factor was retrieved by inverting measurements of a humidified and dry nephelometer, a PSAP and two instruments that measured the particle size distribution. Mie calculations were performed in order to compare the in situ measured backscatter and extinction coefficients to the HSRL-2 retrievals. A clear correspondence between in-situ and lidar retrievals was found. However, distinct discrepancies

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were found which are discussed. Additional sensitivity studies were performed to investigate the influence of the limited size ranges of the different instruments and the influence of the used parameterizations.

The manuscript structure and presentation quality have to be substantially improved. The focus is often lost and the reader gets confused by unnecessary technical details and repetitions throughout the manuscript. All technical, retrieval related or campaign specific information should be moved to the methods part. In this way the results can focus on the actual findings. Currently, many isolated and singular sentences make up own paragraphs which makes the manuscript difficult to read. Thematically related topics can often be combined to own paragraphs. The amount of tables and figures should be limited to the important ones (additional information should be moved to the supplementary material). Heading titles and section labelling should be improved as well (e.g. avoiding single subsections without subsequent follower and unnecessary long titles). The conclusions should be more concise and should focus on the lessons learned. Substantial editorial work is therefore unavoidable.

The topic and findings are of interest to the scientific community. However, there are many clarifications and questions to the analysis and its interpretation which have to be adequately answered before publication (see detailed comments below). It is for these reasons that I recommend major revisions.

## Detailed comments

Comments are given in arbitrary order.

1. Page 1, line 10-13: Please be more quantitative here and state approximate numbers.
2. Page 1, line 19: Add 'e.g.' before McFarquhar et al. (there are many more studies

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- who emphasize this aspect).
3. Page 2, 1st paragraph: The discussion on the advantages or disadvantages of remote sensing, in-situ, ground-based or airborne should be more balanced. In-situ measurements e.g. have the advantage that they are more detailed with respect to microphysical and chemical aerosol properties while they are limited in space (point measurement). Remote-sensing can cover larger areas but their retrievals often depend on assumptions and give less microphysical detail. Airborne measurements are expensive and thus not feasible for monitoring, etc. ...
  4. Page 2, line 20: Are 13 references for the retrieval techniques really needed here? The same reference chain appears on page 4 again. The authors should focus on the important publications. No discussion and references on previous validation studies are given, which should be added here.
  5. Page 2, line 25: Most in-situ and all monitoring measurements are usually performed at dry conditions to keep them comparable. Please add this info here.
  6. Page 2, line 34: DISCOVER-AQ is not defined yet.
  7. Page 3, line 3: The Zieger et al., 2010 reference is incorrect here. The lidar in-situ comparison (using humidified nephelometer measurements) were done in Zieger et al. (2011) and in Zieger et al. (2012).
  8. Page 3, line 13-14: Why was no hygroscopic adjustment necessary?
  9. Page 3, Sect. 2: I would suggest to replace the title of this section ("DISCOVER-AQ") by something more descriptive (e.g. 'Campaign description' or something similar). The information on the number of profiles analysed (line 18) could also be moved to the campaign description section.

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10. Page 4, Sect. 3: The section heading could also be improved here. Instead of 'LaRC HSRL-2' the authors could use 'Airborne high spectral resolution lidar' or just 'Airborne lidar measurements'. Parts of the second paragraph of Sect. 3 are repetitive from the introduction (remove it here or there). Section 3.1 is not followed by a Sect. 3.2, so I would remove this subsection heading if nothing follows or restructure.
11. Page 4, line 27: Repetition and not needed here.
12. Page 5. line 15 and Table 1: This table is not really needed and could be omitted or moved to the supplement.
13. Page 5, line 25: Does the 5  $\mu\text{m}$  size cut relate to dry or to ambient RH? If the size cut relates to ambient conditions, then the effect of hygroscopic growth will influence the presented results since the actual size cut at elevated RH might much smaller. Please clarify.
14. Page 5, line 28: How was the humidified nephelometer calibrated? How did the authors determine the exact RH of the wet scattering coefficient? Were salt calibrations preformed (see e.g. recommendations given in Zieger et al., 2013)?
15. Page 5, line 16: As correctly stated, the  $\gamma$ -fit is one empirical fit among many. However, the real limitation is not the fit, it is rather the fact that the humidified scattering coefficients were only measured at one elevated RH due to experimental limitations (aircraft measurements). Therefore, it will remain unknown if phase transitions have occurred below 80% RH or not. In theory you can apply the  $\gamma$ -fit for different regimes of the humidogram (e.g. if hysteresis is present) separately (in Zieger et al., 2010, for example, the  $\gamma$ -fit was used to describe hysteresis effect due to deliquescent sea salt).
16. Page 5, line 22: To back-up the negligence of the absorption enhancement, the

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- authors should state the campaign mean and standard deviation of the single scattering albedo at this point.
17. Page 7, line 7: How were the multiple charged particles treated for the ammonium sulphate calibration of the LAS and UHSAS using the DMA?
  18. Page 7, line 10-11: Please state the mean and standard deviation of the dry and wet relative humidities (preferable in the instrument section).
  19. Page 7, Eq. 3: If  $\bar{g}$  is not constant over the size range (as done later in the sensitivity study) then the change in number distribution has to be calculated as well. See Eq. 5 in Zieger et al. (2013).
  20. Page 7, line 22: Please add that an internal mixture was assumed (correct?).
  21. Sect 5.2 and Fig. 2: Every inconsistency in the in-situ measurements (e.g. particle losses) will be balanced by the retrieved refractive index. The statement, at this point, on the consistency of the in-situ measurements can only be valid if the corresponding retrieved refractive index is shown as well. This profile should be added to Fig. 2. To save space the authors could consider to show only one wavelengths for the scattering coefficient.
  22. Fig. 3: The course of the hygroscopic growth factor is probably highly driven by the ambient relative humidity. For a better comparison, the RH profile should therefore be shown as well.
  23. Page 10, line 24-25 and Fig. 4: The authors state that the retrievals compare well to the in-situ measurements. How were these profiles chosen? Comparing Fig. 4 with Fig. 5 it seems to be that only nice examples were cherry-picked. Therefore this sentence should be rephrased.

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24. Fig. 4: The good agreement is remarkable. While the in-situ measurements of surface, volume and effective radius show clearly the same profile shape, some exceptions can be observed in the HSRL-2 retrievals. For example, in the second profile at approx. 600 m altitude (also at 1700 m), the retrieval of effective radius, surface and volume are not in correspondence. Is this due to the fact that they are independently retrieved? A consistency check could be included in the analysis (i.e. volume and surface value should give the appropriate effective radius under the assumption of spherical particles).
25. Page 10, line 27: Figure 5 is only described by one short sentence. Please be more detailed here. If the figure is not important then it should be removed.
26. Fig. 5: I find the systematic difference of the particle number concentration interesting. The HSRL-2 seems 'to see' more particles than the UHSAS. However, I would expect the UHSAS to be more sensitive to small particles which dominate the total number concentration. Is there any explanation for this? On the other hand, the surface and volume concentrations seem to agree well, while the effective radius is systematically larger for the in-situ measurements. This is surprising since the effective radius can be calculated from the surface and volume ( $r_{\text{eff}} = 3V/A$ ; see e.g. Grainger et al., 1995) and therefore should agree well. Or is it differently defined/retrieved here? Please clarify.
27. Fig. 6: Similar to Fig. 5, I find it remarkable that the bias of the effective radius is positive with hygroscopic correction and negative without hygroscopic correction. Should it not be similar to the surface and volume concentration?
28. Page 10: The third paragraph is repetitive.
29. Page 10, line 30 and abstract: I am astonished by the fact that the bias of the total aerosol volume concentration is smaller than the one of the surface concentration. The in-situ instruments have difficulties measuring large particles (as discussed

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- later in the manuscript) which on the other hand determine the total aerosol volume. How can this be explained?
30. Page 11, line 15-16: Why is this interesting? Any explanations?
  31. Page 11, line 19 and Fig. 8: Looking at the graphs (especially at the first panel) I would rather talk about 'good' or 'very good' agreement (and not excellent). How were these points averaged?
  32. Page 12, line 17-23 and Fig. 9: Although the median bias slightly decreases if the LAS measurements are used, the IQR increases for these cases. This should be added and discussed as well.
  33. Page 12, line 24-27: Particle losses was only one hypotheses among many in Zieger et al. (2011). In fact, this study also compared  $3\beta + 2\alpha$  lidar measurements to in-situ recordings that were re-calculated to ambient conditions. The lidar agreed much better to the in-situ measurements than the MAX-DOAS (especially during nighttime, see Fig. 12 in Zieger et al., 2011). For the MAX-DOAS, it was hypothesized that the lowest and compared layer was overestimated due to lofted layers (e.g. caused by ammonium nitrate partitioning). In addition, the MAX-DOAS retrieval could have been influenced by horizontal gradients in aerosol concentration. Nevertheless, in this work, the influence of coarse mode particles is definitely a hot candidate for the underestimation of the in-situ data. To further investigate this, the authors could, similarly to Zieger et al. (2015), compare their in-situ optical properties to the columnar measurements of AOD (Fig. 8). Zieger et al. (2015) also found a clear underestimation of the in-situ derived AOD and hypothesized as one possible reason that coarse particles were not sufficiently sampled (e.g. being lost in the canopy or within the inlet system) due to the pronounced wavelength-dependency. The calculated fine-mode fraction could be added to Fig. 10 which would be more convincing.

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34. No subsection is followed after 6.2.1. Therefore I would re-order and add an extra subsection or remove this heading.
35. Sect. 6.2.1 and Fig. 10: The argumentation is very speculative. The CA dataset contains much less datapoints than the TX dataset so the statistics is different. Looking at Fig. 7 again, it is hard to see a clear and significant difference between the two datasets. I would suggest to move this figure to the supplement. Alternatively, the authors could further test their hypothesis in a more convincing way, e.g. by colour-coding the points in Fig. 7 by the fine mode fraction or by plotting absolute or relative differences of the retrievals vs. the fine mode fraction of the AOD.
36. Page 13, third paragraph and Table 4: The choice and definition of the cut-off diameters is not clear to me. Sedimentation or diffusion losses should be low between 100 and 1000 nm, so I don't understand the choice of 0.7 and 0.4  $\mu\text{m}$ . In addition, the hygroscopic growth factors were much larger during the campaign (up to 1.6 at elevated RH, see Fig. A1). Therefore, the particles (if the UHSAS sampled at dry conditions) were actually much larger at ambient conditions due to hygroscopic growth and the cut-off diameters should be set to values above 1  $\mu\text{m}$ . The interpretation (see 4th paragraph) should then be adapted. I would interpret the sensitivity study that coarse mode particles above  $\sim 1.2 \mu\text{m}$  are only really relevant for the  $\beta_{1064}$ -measurement. Please clarify and adapt accordingly.
37. Sect. 7 (Discussion) and Sect. 8 (Summary and conclusions): These sections are again very repetitive and often dissipate. Please focus and discuss the main findings. Both sections can be combined. The references to previous findings are missing and should be added to the discussion. The limitations of the lidar retrieval technique are not discussed or even mentioned at all in the conclusion part, which has to be added. It would be beneficial to the paper if the authors would add a short and precise outlook and recommendation part to their work.

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38. Sect. 7.1: The phase transitions are important mainly for pure compounds. In the ambient atmosphere, clear and distinct phase transitions or hysteresis effects have been observed (using humidified nephelometers that look at the overall/integrated effect) mainly when sea salt was present (see e.g. Zieger et al., 2013, for an overview). Organic compounds and mixtures with other inorganic substances will most likely lead to a smooth hygroscopic behaviour without pronounced deliquescence. Figure 12 also shows a dominance of WSOC and  $\text{NO}_3$  and thus makes deliquescence quite unlikely. In addition, the particles in the ambient atmosphere will most likely be on the upper branch of the hysteresis curve if they have experienced an elevated RH before the time of measurement. For the ambient optical properties, which are studied here, the authors should look and discuss the related scattering enhancement factors, which is an integrated value while HTDMA's only look at distinct (and fine mode limited) dry sizes. There is no Sect. 7.2 following 7.1, so please restructure.
39. Page 26, Table 3: The linear regression and correlation coefficients should also be given for the comparison of the microphysical parameters from Fig. 5.
40. Page 26, Table 2: The choice of biases smaller than 50% seems quite arbitrary. How is this justified?
41. Figure A1 and Sect. A1: This part is to reviewer's opinion quite important since it demonstrated the validity of the presented retrieval method for  $\bar{g}$  and the good quality of the recorded in-situ data. It could be moved to the main part of the manuscript. However, it is unreasonable to show  $\bar{g}$  vs. the ambient RH because the wet scattering coefficient was always measured at a constant RH (80-85%). A comparison to  $\kappa$  is therefore not appropriate since the entire curvature of  $\bar{g}$  is predetermined by the here used  $\gamma$ -parametrization. The authors should show a distribution plot of  $\bar{g}$  at RH=80-85% (or preferable at one fixed RH by recalculating the wet scattering coefficients to one fixed RH). These values can then be

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- compared to literature values.
42. Sect. A2: This section is quite difficult to read and understand (again many paragraphs consisting of only one sentence). A flow chart could help here. It is not clear on why Fig. A2 has to be shown. It is probably sufficient to state that the simulations were done for similar conditions as the HSRL-2 measurements. Table A2 is hard to interpret as well. Why was the noise not added to the RH of the ambient and humidified nephelometer measurement? The same is true for the influence of the coarse mode, which might not have been sufficiently sampled. Both aspects will have a clear effect on the retrieval uncertainty (see Appendix A and Fig. A1 in Zieger et al., 2013). The sensitivity to the ambient RH is not discussed at all and should be added here. Were the ambient RH measurements of the two aircrafts compared?
  43. Fig. 10: How exactly was the scaling of the volume distribution to the aerosol layer height performed? And why were two different heights (1 km and 3 km) for the two campaigns chosen. Maybe it would have been easier to just normalize all volume size distributions to 1 and then calculate the average values.

### Technical comments

1. Page 5, line 5: Replace 'mum' by  $\mu\text{m}$ .
2. Throughout the manuscript: Please don't put the unit meter in italics.
3. Page 8, line 22: One web-link is probably sufficient here.
4. Fig. 2: Please use the introduced variables for the scattering and absorption coefficients (y-labels) and avoid abbreviations like Scat450 or Abs532. Instead of 'recalc' it would be better to use an abbreviation like 'retr' (retrieved).

5. Fig. 3: Please use the correct variables for the axis-labelling (see comment above). Units should be next to the numbers and not in the next row.
6. Fig. 2 and 3: To be consistent in the equations, the scattering efficiency should also depend on the dry or ambient particle diameter.
7. The section headings for 5.2 to 5.4, 6.2 are quite long and complicated. They can be shortened to be more concise (e.g. 'Retrieval of dry complex refractive index' or 'Retrieval of the effective growth factor' or 'Optical closure study').
8. Page 13, line 2: Please replace here and throughout text (where possible)  $3\beta+2\alpha$  by 'extinction and backscatter coefficients'. This will improve the reading flow since the acronym is very specific for the lidar community and not known to the majority of the readers.
9. Please replace 'optical particle counters' by 'optical particle size spectrometers'. UHSAS and LAS are not just counting particles like a CPC, they also size them.
10. Fig. 4: Please add that these profiles are for the fine mode fraction only.
11. Fig. 8: Units missing in the statistics text blocks.
12. Fig. 11: Define SZD at the beginning of the caption.
13. Page 17, line 9:  $\kappa$  can range according to Petters and Kreidenweis (2007) up to 1.3 (sea spray).
14. Page 17, line 29: The correct formula for ammonium sulphate is  $(\text{NH}_4)_2\text{SO}_4$
15. Sect. A1: Please harmonize the variable names (i.e. the refractive index is given in different ways).
16. Fig. 1: Replace 'King Air' by 'B-200' as shown in the figure (or vice versa).

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17. AOT and AOD are not used in the same way throughout the manuscript. For example, in Fig. 8 it is AERONET AOT and in Fig. 10 it is AOD for the fine fraction. Please harmonize.

## References

- Grainger R., Lambert A., Rodgers C., Taylor F., and Deshler T., Stratospheric aerosol effective radius, surface area and volume estimated from infrared measurements, *J. Geophys. Res.*, 100(D8), 16507–16518, 1995.
- Petters M. and Kreidenweis S., A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7(8), 1961–1971, doi:10.5194/acp-7-1961-2007, 2007.
- Zieger P., Aalto P.P., Aaltonen V., Äijälä M., Backman J., Hong J., Komppula M., Krejci R., Laborde M., Lampilahti J., de Leeuw G., Pfüller A., Rosati B., Tesche M., Tunved P., Väänänen R., and Petäjä T., Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study, *Atmos. Chem. Phys.*, 15(13), 7247–7267, doi:10.5194/acp-15-7247-2015, 2015.
- Zieger P., Fierz-Schmidhauser R., Gysel M., Ström J., Henne S., Yttri K., Baltensperger U., and Weingartner E., Effects of relative humidity on aerosol light scattering in the Arctic, *Atmos. Chem. Phys.*, 10(8), 3875–3890, doi:10.5194/acp-10-3875-2010, 2010.
- Zieger P., Fierz-Schmidhauser R., Weingartner E., and Baltensperger U., Effects of relative humidity on aerosol light scattering: results from different European sites, *Atmos. Chem. Phys.*, 13(21), 10609–10631, doi:10.5194/acp-13-10609-2013, 2013.
- Zieger P., Kienast-Sjögren E., Starace M., v. Bismarck J., Bukowiecki N., Baltensperger U., Wienhold F., Peter T., Ruhtz T., Collaud Coen M., Vuilleumier L., Maier O., Emili E., Popp C., and Weingartner E., Spatial variation of aerosol optical properties around the high-alpine site Jungfraujoch (3580 m a.s.l.), *Atmos. Chem. Phys.*, 12, 7231–7249, doi:10.5194/acp-12-7231-2012, 2012.
- Zieger P., Weingartner E., Henzing J., Moerman M., de Leeuw G., Mikkilä J., Ehn M., Petäjä T., Clémer K., van Roozendaal M., Yilmaz S., Frieß U., Irie H., Wagner T., Shaiganfar R., Beirle S., Apituley A., Wilson K., and Baltensperger U., Comparison of ambient aerosol extinction

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coefficients obtained from in-situ, MAX-DOAS and LIDAR measurements at Cabauw, *Atmos. Chem. Phys.*, 11(6), 2603–2624, doi:10.5194/acp-11-2603-2011, 2011.

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