Response to Referee #2:

*Blue italic font means the authors' answer,* standard, black *text symbolizes the reviewer's comments, and red italic font means manuscript changes, while black italic font symbolizes the original content of the manuscript.*

***Response:***

*We gratefully thank the referee for the spent effort and comments. We respond on the individual points below.*

This study compares lidar optical properties to those computed with Mie calculations in function of RH. The topic is important but the work suffers of important lacks in the method section probably biasing the obtained results and related considerations. Thus a deep major revision is required based on the following major issues:

Lines 290-292: "Also, the residual layer containing some aerosol layer aloft the top of the planetary boundary layer (PBL) between 1250 m and 2300 m is visible indicated by greenish colors." Given the description above and Figure 1 it is clear that ACTOS also sampled in the residual layer between ~1300 and ~2000m. I suggest to correct the sentence at line 292-293 ("The payload, therefore, was sampling in the free troposphere as well as within the planetary boundary layer and was sampling different aerosol populations") and ALL the related discussion and interpretation later in the results.

*Thanks for the comment. This part is transferred to the supplementary part of the manuscript. However, we added in line 34 - 35 in the supplementary material: "The payload, therefore, was sampling in the free troposphere as well as within the planetary boundary layer and was sampling different aerosol populations."*

*Of the four shown investigated flights of the summer campaign, two were conducted during a fully developed planetary boundary layer (flight 20150617b and flight 20150628b). Residual layers are observed for flights 20150626a and 20150628a (top right, and bottom left figure below). Below, the flight patterns of ACTOS during the measurement days are shown in the figures below. To raise the awareness of the audience, we added in line 482 - 490: "The flight was conducted in the early morning from 08 to 10 UTC. During this daytime, the PBL is usually still developing due to thermal convection. Hence, most of the data were collected within the residual layer. The residual layer is an aged layer of aerosol, and the aerosol sampled on the ground should not represent the layer aloft the PBL. However, the model calculates aerosol particle light backscatter and extinction within 35% compared to the lidar with the best agreement at 532 nm, reproducing the extinction within 12%, much smaller than the approximated lidar uncertainty. Within the PBL, presumingly up to an altitude of 600 m, the model significantly calculates larger $\sigma_{ext}(\lambda)$ and $\sigma_{bsc}(\lambda)$. Surprisingly, the assumptions within the model capture the conditions within the residual layer better than the aerosol conditions within the PBL. Maybe the more aged aerosol within the residual fits better the core-shell mixing assumption within the model."*

*And more in line 536 – 539:*

"*Above the PBL, within the free troposphere, the model is significantly larger than the lidar estimates. However, ACTOS was not flying directly above the lidar; hence, small scale differences in the PBL height could explain the difference. These variations in the PBL height are also visible in Figure S1, with distinct variations of the aerosol load within a short period.*"

*However, the overall contribution to the total data set is small, and most of the data is collected within the PBL.*

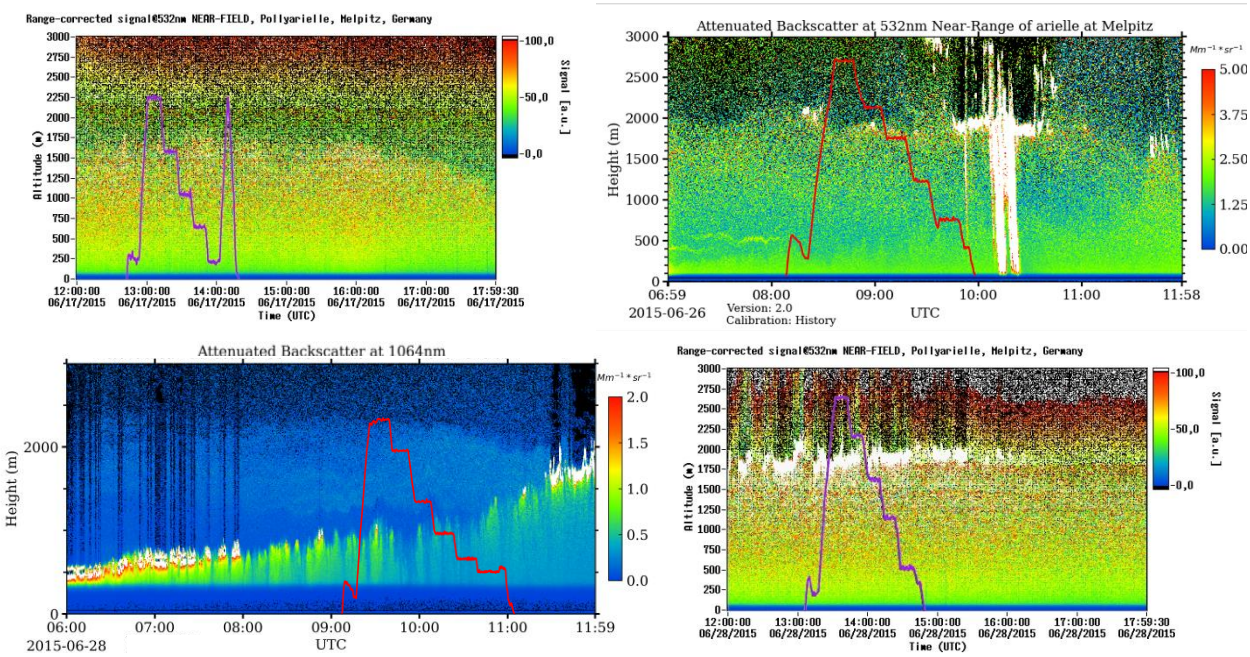


*Figure 1: ACTOS flight track and attenuated backscatter coefficient measured by the Polly$^{XT}$ lidar Arielle during the flight period of ACTOS on the measurement days of the summer campaign.*

Lines 296-297: how much below 40% RH the aerosol was sampled? Consider that aerosol efflorescence (or crystallization) can occur at RH lower than 40%, even below 30% RH in function of the aerosol chemical composition (nitrate to sulfate ratio, degree of acidity, presence of ammonium chloride etc…) (Martin, S. T.: Phase Transitions of Aqueous Atmospheric Particles., Chemical reviews, 100(9), 3403–3454, 2000). Please add a deep discussion based on this point as the manuscript aims at closure in function of RH, but the aforementioned consideration poses an important issues to the capability to reach this goal.

*We express many thanks for the comment. Although the efflorescence of hygroscopic aerosol particles is known, the effect is only observed in Melpitz during westerly inflows characterized by marine air masses, as Zieger et al. (2013) showed for multiple European sites, including Melpitz. During the winter campaign, between February 1 and March 15, 2017, a mean volume fraction of organic matter of 0.48 (median=0.74, IQR from 0.39 to 0.54) was observed, during the summer campaign period from June 1 to June 30, a mean volume fraction of 0.58 (median=0.59, IQR from 0.47 to 0.69). Due to these relatively high volume fractions, the hysteresis effect of scattering enhancement is not observed and therefore has not to be considered in the calculations.*

*We observed a maximum of 35.8% RH at all but one day downstream of the dryer. However, we will point out that the found parameterization is only applicable for non-marine air masses.*

*We added in lines 641 - 646: "Zieger et al. (2013) have shown the scattering enhancement due to hygroscopic growth for different European sites. In all but marine airmass-influenced cases, no hysteresis effect was observed at Melpitz, and they stated that these might occurs due to high fractions of low hygroscopic organic material. Hence, the effects of the aerosol efflorescence can be neglected since the volume fraction of the organic material within the aerosol population was relatively large during the summer campaign period. A mean volume fraction of 0.58*

*(median=0.59, IQR from 0.47 to 0.69) was estimated based on the chemical composition and assumed material densities within June 1 and June 30, 2015.*", *and in lines 682: "Nevertheless, the presented results provide good first estimates of the RH-induced LR(λ) enhancement factor based on in-situ measured PNSD for the observed RH range for the aerosol conditions at Melpitz. Although Ackermann (1998) …"*

*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, 10609–10631, https://doi.org/10.5194/acp-13-10609-2013, 2013.*

Lines 306-307, Figure 2 and Lines 316-321: The missing refractive index correction of the OPSS represents a lack of the manuscript in the way as it is actually presented. This section needs an improvement. For example, the inner "detailed geometry of the optical cell inside the instrument" should be asked to the manufacturer (or at least asking the equivalence with that reported in: Heim, M., Mullins, B. J., Umhauer, H., and Kasper, G.: Performance evaluation of three optical particle counters with an efficient "multimodal" calibration method, J. Aerosol Sci., 39, 1019–1031, doi:10.1016/j.jaerosci.2008.07.006, 2008).

*Thanks for the suggestion and input. Yes, Heim et al. (2008) provide insights on the geometry of the 1.109 optical particle sizer. Although with the given geometries, a Mie-based correction would be feasible, another reason prevents the correction of the refractive index. According to the manual, the GRIMM skyOPC is calibrated to a PSL calibrated mother device using polydisperse mineral dust (dolomite). This calibration was not reproducible within TROPOS.*

*Also, the results of Walser et al. (2017) indicate broad measurement spectra of mono-disperse PSL aerosols. These broad sizing spectra are not helpful to create a high valid refractive index correction. Moreover, for a refractive index correction, the polarization of the laser is needed but is unknown to our knowledge.*

*We updated the part in the manuscript as follows in line 407 - 410:" The manual of the skyOPC (v. 2.3) states that each offspring OPSS unit is calibrated to a mother instrument with an in-house standard using polydisperse mineral dust (dolomite). Walser et al. (2017) show broad sizing spectra of monodisperse polystyrene latex particle aerosols measured by the skyOPC. Also, the polarization of the used laser with a wavelength of 655 nm is unknown but is needed to calculate the response OPSS response curve. ~~The detailed geometry of the optical cell inside the instrument is unknown. Hence,~~ Because of these reasons, a correction regarding the complex aerosol refractive index ($n = n_r + in_i$) could not be applied to the data set. The OPSS in-situ measurements were quality checked by comparing the average PNSD of the lowermost 200 m with the ground in-situ measurements (see Figure 2)."*

*Walser, A., Sauer, D., Spanu, A., Gasteiger, J., and Weinzierl, B.: On the parametrization of optical particle counter response including instrument-induced broadening of size spectra and a self-consistent evaluation of calibration measurements, Atmos. Meas. Tech., 10, 4341–4361, https://doi.org/10.5194/amt-10-4341-2017, 2017.*

Mie calculation should be biased using the OPSS optical equivalent diameters, thus affecting a part of section 3 (Modeling optical properties with Mie), discussion and all conclusions. The later (line 326) altitude correction factor in eq. 6 does not correct the OPSS optical equivalent aerosol size-bin (i.e. the size of particles) which is, instead, the right parameter needed for proper Mie calculations. It is required to clarify this point for the reader. Moreover, the above approach generate an inconsistency with lines 359-363 ("The OPSS PNSD was corrected in

terms of the complex aerosol refractive index. Here, a complex aerosol refractive index of 1.54 + i0 was used since this resulted in OPSS PNSD with a good overlap to the MPSS PNSD. The imaginary part of the complex aerosol refractive index was forced to 0 because it leads to a significant overestimation of the coarse mode in the PNSD when the imaginary part of the complex aerosol refractive index is above 0 (see Alas et al., 2019). Note, that this complex aerosol refractive index is not the refractive index used in the Mie model") and an inconsistency with lines 368-369 (Particles larger than 800 nm have not been replaced by the PNSD measurements at ground since the refractive index correction was applied to the OPSS data where different methods were used. I suggest to improve the discussion of the Mie methodology (and related approximations) from line till line 498 to make it clearer and more consistent.

*Thanks for the suggestion. First of all, to clarify and resolve some inconsistencies, the usage of the altitude correction factor is updated within the manuscript in lines 439 - 446: "In both cases, the instrumentation onboard the payloads did not cover the entire aerosol particle size range from 10 nm to 10 µm. Since the in-situ instrumentation at the ground is quality-assured, the ground-based measurements are the reference and are utilized to correct the airborne measurements. The missing size range is addressed as follows: The size range of the corresponding PNSD from the ground fills the missing size range; from 10 nm up to 326 nm, in the winter case, in the summer case, all sizes larger than 800 nm in optical diameter. Advantageously this addresses the unaccounted underestimation of larger particles by the skyOPC in the summer case and also provides volume-equivalent diameters for the Mie calculations in that size range. To account for vertical variability within the atmosphere, the ground-based PNSD is corrected for altitude, establishing a non-fixed altitude-correction factor $f_h$.". $f_h$ corrects the missing part of the airborne PNSD and also the number concentration of particles, which is also needed to proper model aerosol optical properties.*

*In the case of the summer, yes, the uncorrected OPSS size distribution biases the Mie model results. The estimate of the bias induced by not correcting the skyOPC PNSD in this size range is challenging and cannot be determined easily. However, the airborne extinction is reproduced by the model sufficiently. In the analysis of the data, we also tackled that issue in lines 556-558: "Moreover, as the refractive index correction of OPSS tends to shift the particle towards a larger diameter, at least partially, that could explain some of the underestimations, although the used size range of the skyOPC is limited between 356 and 800 nm.". Moreover, as clarification, we added in line 428 - 429: " Contrary to the PNSD derived with the sykOPC, this OPSS PNSD is corrected with in-house software in terms of the complex aerosol refractive index." We assume a small impact since the uncorrected sykOPC mean PNSD of the lowermost 200m (Figure 2) is partially smaller than and partially larger than the PNSD at derived ground level.*

*In the winter campaign, the OPSS is corrected with a refractive index of 1.54, and calculations with 1.56 can not explain the difference of both approaches (see lines 855 - 857). "However, using the ZSR-based real part of the complex refractive index of 1.56 during both days cannot explain the lidar and Mie model differences. Applying this real part to the data of February 9, the slope of the correlation changes within absolute values of -0.055 to 0.045 compared to a real part of 1.54."*

*Biased refractive indices for the TSI OPSS correction have been addressed in the manuscript in lines 433 - 438:" For the investigated days of the winter campaign, a median complex refractive index of the aerosol of 1.56+i0.11 is found for February 9 and 1.56+i0.06 for March*

*9, respectively. However, these refractive indices are based on the ZSR mixing of homogeneously mixed particles but, a) we assumed a core-shell mixing of the aerosol particles and b) the shape of the aerosol particles is essential as well for the refractive index correction. Therefore, the used complex refractive index for correction is more an effective refractive index to match the OPSS PNSD to the PNSD derived at ground level with the MPSS and APSS."*

Lines 350-351: "truncation error of the scattering coefficient was not corrected". Please, add also the uncertainty of scattering and not only that of extinction.

*Thanks for the suggestion. The estimation of the scattering uncertainty depends on the truncation and calibration error of the CAPS. The truncation error depends on the aerosol morphology, the aerosol particle number size distribution, and the aerosol refractive index.*

*Within the CAPS a PMT (photomultiplier tube) including an integrating sphere is installed to measure the scattered light. Integrating sphere nephelometers like the one used in this study at the ground station measure the scattered light similarly and the measurement uncertainty due to calibration and truncation is usually not more than 10%. See line 203 – 205: "These measurements were completed by a Nephelometer (mod. 3563, TSI Inc., Shoreview, MN, USA), which measures the $\sigma_{sca}(\lambda)$ at 450, 550, and 700 nm with a relative uncertainty by calibration and truncation of about 10% (Müller et al., 2009)."*

*Since the airborne measured aerosol particle light scattering coefficient was not used within this study, there is no point in providing the measuring uncertainty. However, Modini et al. (2021) recently provided a detailed characterization of the CAPS PMssa instrument. Truncation correction factors were provided with uncertainties of 4% and 9% for fine and coarse mode dominated aerosols, respectively.*

*We added to the manuscript in lines 391 - 395: "The measured aerosol particle light scattering coefficient is not used within this study and therefore ~~T~~the truncation error of $\sigma_{sca}$(630 nm) is not corrected. Moreover, ~~; therefore, within this study,~~ we focus on $\sigma_{ext}$(630 nm) estimated with a 5% accuracy. However, a detailed characterization of the CAPS PMssa monitor is provided by Modini et a. (2021). Truncation and scattering cross-calibration correction factors were provided with uncertainties of 2%, and 4% to 9% for fine and coarse mode dominated aerosol, respectively."*

*Modini, R. L., Corbin, J. C., Brem, B. T., Irwin, M., Bertò, M., Pileci, R. E., Fetfatzis, P., Eleftheriadis, K., Henzing, B., Moerman, M. M., Liu, F., Müller, T., and Gysel-Beer, M.: Detailed characterization of the CAPS single-scattering albedo monitor (CAPS PMssa) as a field-deployable instrument for measuring aerosol light absorption with the extinction-minus-scattering method, Atmos. Meas. Tech., 14, 819–851, https://doi.org/10.5194/amt-14-819-2021, 2021.*

Lines 360-363: OPSS model 3330 of TSI only accept real part of refractive index. The use of 1.54 + i0 is mandatory, not a decision. Moreover, this can generate problems if "this complex aerosol refractive index is not the refractive index used in the Mie model" as reported. Please comment and clarify.

*We have to clarify that to correct the optical diameters of the OPSS, we used in-house software in which the real part and imaginary part of the complex refractive index can be varied.*

*During the intensive period between February 1 and March 16, the median real part of the aerosol particles was 1.558, the imaginary part 0.08. On both investigated days, a mean*

*complex refractive index of 1.56+i0.109 on February 9 was observed, 1.56+i0.06 on March 9, respectively. The issue is addressed in the comments above.*