Response to Referee #1:

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 anonymous referee for his comments and input. The mentioned points are addressed below.

The article is still poorly written and tedious. Data from various measurements were used in this article including surface instrumentations like D-MPSS, Q-ACSM and lidar system. However, there are only lengthy analysis and few merits. Lidar has its advantage in measuring aerosol vertical hygroscopic growth in ambient environment, which is not evident in this article. In addition, errors due to the different observation methods and uncertainties from optical parameters from lidar should be considered.

We shifted some parts into the supplementary to shorten the lengthy analysis. We updated the scope of the article to highlight some findings. For details on this modification, we refer to the answer to referee #3.

However, the scope of the article is not to show the lidar capability of deriving the aerosol hygroscopic growth (HG), although it would be helpful within this study to simulate the HG of the in-situ measured dry aerosol particles. However, comparing modeled optical parameters based on an HG simulation using lidar HG estimates would be somewhat circular. Nevertheless, the capability of lidar to inspect the hygroscopic behavior of aerosol is included in the introduction:

"Previous studies have focused on the dependence of $\sigma_{ext}(\lambda)$ on ambient RH (Skupin et al., 2013; Zieger et al., 2013). Navas-Guzmán et al. (2019) utilized these effects to investigate the aerosol hygroscopicity with lidar. LR(λ) is based on the RH-dependent $\sigma_{bsc}(\lambda)$ and $\sigma_{ext}(\lambda)$, and calculations by Sugimoto et al. (2015) indicated that LR(λ) is RH-dependent as well. Ackermann (1998) provided a numerical study based on pre-defined aerosol types with distinct size-distribution shapes to establish a power series to describe the LR(λ) as a function of RH. Salemink et al. (1984) found a linear relationship between the LR(λ) and the RH. Intensively discussed is the LR-enhancement due to hygroscopic growth in Zhao et al. (2017). They reported a positive relationship between LR and RH, but their study lacks information on vertically resolved aerosol particle number size distributions and other wavelengths. However, their simulations have shown that utilizing RH-dependent LR to retrieve aerosol particle light extinction from elastic backscatter lidar signals results in significantly different values than the constant LR approach."

Uncertainties of the lidar have been specified - 10% measurement uncertainty in terms of backscatter. Extinction estimates by the lidar are derived by LR provided by Mattis et al. (2004) and corresponding *uncertainties. We also refer to the answer to referee #3, in which we described the uncertainty of the lidar-based extinction based on Gaussian error propagation.*

Errors by the in-situ observations have been tackled utilizing a Monte-Carlo simulation.

Ongoing with the additional report of lidar-based studies investigating hygroscopic behavior of aerosol, we added Zhao et al. (2017) as an additional source for an LR(RH) parameterization and updated Figure 6 with the corresponding curve.

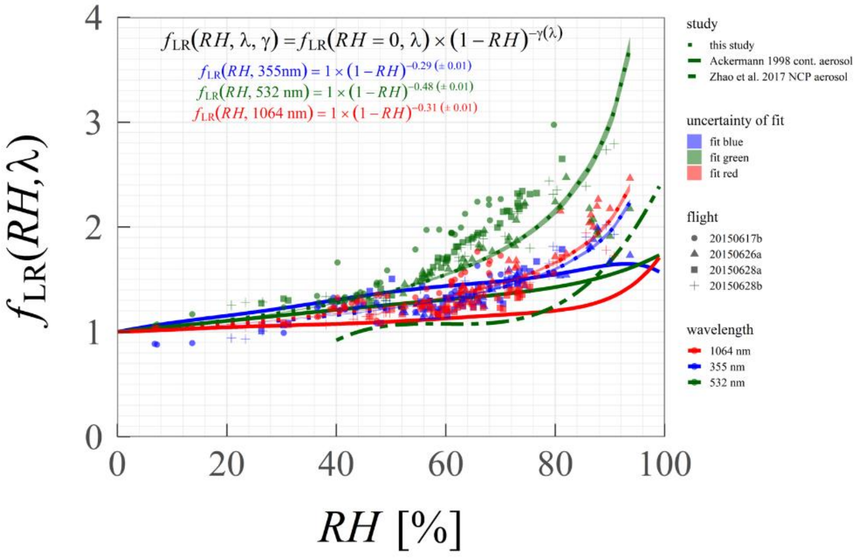

Figure 1: Updated Figure 6.

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.

The abstract is too long-winded and then innovation is difficult to find. Only one paragraph is suggested for the abstract.

We updated the abstract as requested and refer to the answer to referee #3.

Line 68: replace "does exsits" by "does exsit"

Line 101: replace "were" by "was"

Line 110: replace "is" by "are", Line 112: replace "is" by "are", Line 113: What is "This"?

Due to structural changes in the manuscript these sentences do not exist anymore (see answer to referee #3).

Line 128: delete "is"

We cannot find anything wrong with the sentence: "*Melpitz Observatory (51° 31' N, 12° 55' E; 84 m a.s.l.) is located in Eastern Germany in a rural, agriculturally used area 44 km northeast of Leipzig.*"

Line 230: replace "underestimates" by "underestimate"

Thanks for the suggestion. The sentence in line 294 is now: "*However, the bulk Q-ACSM approach might over- or underestimate~~s~~ the hygroscopicity of aerosol particles ~~lower~~smaller or larger than 165 nm in diameter*."

Line 290: replace "of" by "for"

We changed accordingly. This part is shifted to the supplementary material.

Line 343: replace "were" by "was"

Since multiple filters were used within the study we changed "filter" to "filters".

Line 535: Poor writing

Thanks for the suggestion. We in the supplementary material lines 63- 65: " *Figure S2b) displays the time series of the number concentration of all aerosol particles up to a size of 800 nm in diameter*."

Line 578: replace "led" to "lead"

We changed as requested.

Line 618: replace "oppoding to" by "opposed to"

We changed as requested: We changed in line 722 : "*~~Opposing to~~ Compared to* ... "

Line 702: "solely based on…"

We changed as requested in line 665: "…*above 90% RH which we could not observe in this study ~~because of a~~ solely based on the small number of cases and the observed RH range." ()*

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.

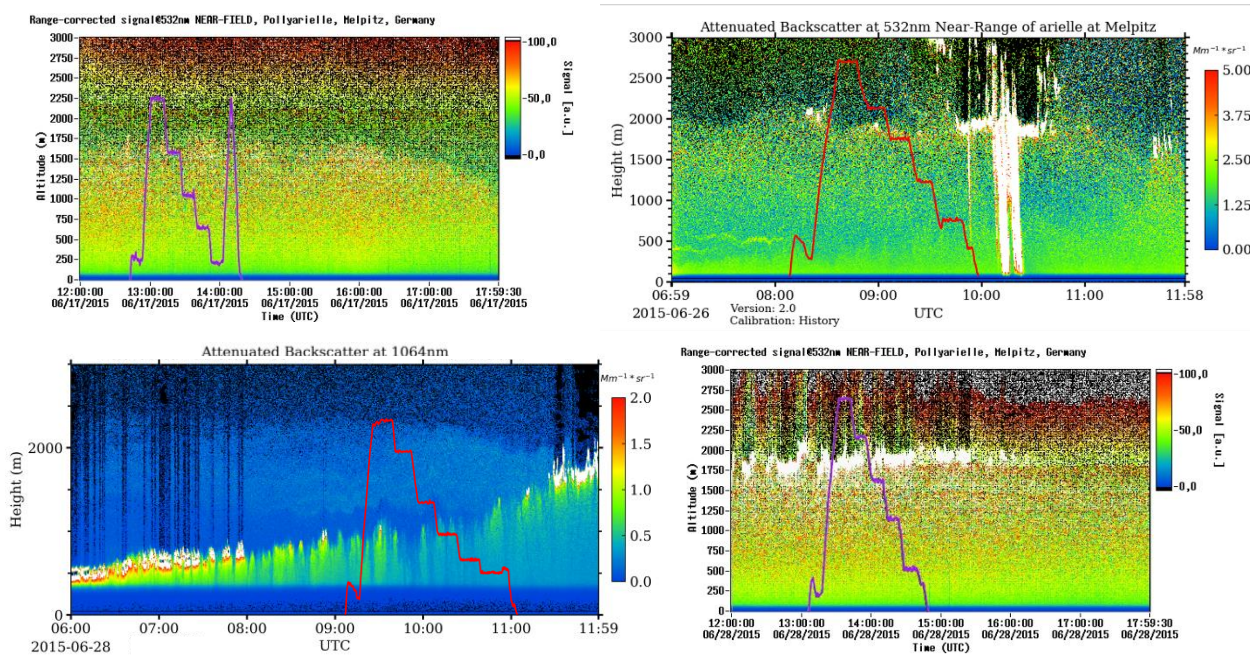

Figure 2: ACTOS flight track and attenuated backscatter coefficient measured by the Polly^{XT} lidar Arielle during the flight period of ACTOS on the measurement days of the summer campaign.

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 $LR(\lambda)$ *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.

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.

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 σ_{sca}(630 nm) is not corrected. Moreover, ~~; therefore, within this study,~~ we focus on σ_{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.

Response to Referee #3:

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 thank the referee for his fruitful comments, suggestions and questions. Below, we answered them point by point.

Referee #3:

This study compares lidar measured optical properties to those computed with Mie calculations that used airborne in-situ based inputs over two different field campaigns. In general, the manuscript is unfocused and lengthy. The tedious amount of detail makes is difficult to completely comprehend and judge the merits of their analysis. I strongly suggest that the authors only include the details of the measurements that aren't described in other publications or are essential to the analysis performed in this work.

The authors state that their study is both "unique" and "complex", which it is, but why is it important? What science is advanced by this work? There are only a few lines of background/motivation given that mentions radiative forcing and cites the IPCC, but it is not clear how this work reduces uncertainty in radiative forcing.

Do the Mie-based calculations reproduce the optical properties well enough to meaningfully reduce radiative forcing uncertainty? The authors need give more a clear science motivation in the introduction and then revisit their goals in the conclusions to discuss what has been learned from this work.

We removed the radiative forcing part of the motivation since it is not within the scope of the manuscript to improve radiative transport or radiative forcing estimates of aerosol. Instead, the study also aims to highlight the complexity of comparing multiple aerosol optical properties with in-situ and remote sensing techniques at once, particularly at the ambient state. Especially considering a high spatio-temporal resolution, such studies are often limited to payloads with a limited mass. Therefore, lightweight instruments are used preferably but with drawbacks in, e.g., the observed aerosol particle size range. Moreover, the determination of the composition of aerosol particles, which is necessary to determine their refractive index and hygroscopicity, is only possible with correspondingly expensive extensions of the load capacity of airborne systems. We updated the introduction and motivation towards the need for a comprehensive comparison study and what we can learn from the belonging challenges. Also, we think the title of the manuscript utilizing a closure study is misleading. We updated the title to " Comparison of airborne in-situ measured, lidar-based, and modeled aerosol optical properties in the Central European background – identifying sources of deviations"

The introduction states now:" *Aerosol particles can sensitively influence the Earth's radiation budget by scattering and absorption of solar radiation. The aerosol impact is described by means of the wavelength-dependent aerosol particle scattering coefficient ($\sigma sca(\lambda)$) and particle absorption coefficient ($\sigma_{abs}(\lambda)$) as well as the sum of both, denoted as particle extinction coefficient ($\sigma_{ext}(\lambda)$). In-situ aerosol measurements with unmanned aerial vehicles (UAV; Altstätter et al., 2018), helicopter-borne payloads, e.g., with the Airborne Cloud and Turbulence Observations System (ACTOS; e.g., Siebert et al., 2006, Ditas et al., 2012, Wehner et al., 2015; Düsing et al., 2018), tethered-balloon payloads (e.g., Ferrero et al., 2019, Brunamonti et al., 2020), and zeppelins (e.g., Rosati et al., 2016a) are important experimental approaches to provide vertically resolved insight into the relationship between aerosol microphysical properties, chemical composition, optical properties, and related radiative effects. Remote sensing techniques such as light detection and ranging (lidar) allow profiling of aerosol optical properties with high vertical and temporal resolution in a complementary way (Weitkamp, 2005). All these different experimental approaches are needed to improve our knowledge about the role of aerosols in the climate system and, at the same time, to reduce the uncertainties in the applied aerosol observations. Direct in-situ aerosol measurements are helpful to validate remote sensing techniques and vice versa. Lidar-based aerosol particle light backscatter coefficient ($\sigma_{bsc}(\lambda)$) profiles have been compared with balloon-borne in-situ measurements (Brunamonti et al., 2020) and Mie-modeling results (Ferrero et al., 2019). However, the airborne in-situ aerosol measurements provide the vertically resolved aerosol information (Rosati et al., 2016a, Düsing et al., 2018, Tian et al., 2020), usually for dried conditions. Lidar, on the other hand, monitors the aerosol under ambient conditions. Therefore, the effect of the RH must be considered when comparing in-situ measurements and modeling approaches with remote-sensing retrievals. Lidar systems have been previously utilized to investigate hygroscopic processes (e.g., Zhao et al., 2017; Navas-Guzmán et al., 2019; Dawson et al., 2020). Modeling aerosol optical properties can also account for the ambient state of the aerosol by simulating the hygroscopic growth of the aerosol particles utilizing, e.g., the semi-empirical parameterization of Petters and Kreidenweis (2007). Also, they can be used for the validation of lidar-based retrievals of, e.g., the absorption.*

However, modeling, remote sensing, and in situ measurements are subject to individual uncertainties that must be considered to compare these approaches. Raman-lidar systems, for instance, such as the Polly^{XT} lidar (Engelmann et al., 2016), can measure the aerosol particle light extinction and backscattering coefficients at several wavelengths λ throughout the entire troposphere, but only during nighttime hours. The standard backscatter lidar technique is applied to derive aerosol backscatter and extinction height profiles in the daytime. The required estimates for the unknown extinction-to-backscatter ratio, also lidar ratio (including its wavelength dependence, LR(λ)), can introduce large uncertainties in the obtained spectral particle backscatter and extinction profiles. Note that LR(λ) is a function of the wavelength of incoming light, the shape of the aerosol particles, the aerosol particle *number size distribution (PNSD), and aerosol chemical composition. LR(λ) estimates during daytime have been derived via a combination of direct lidar σbsc(λ) and columnar sun-photometer measurements (Guerrero-Rascado et al., 2011 A sun-photometer measures the columnar integral of $\sigma_{ext}(\lambda)$, the aerosol optical depth (AOD). An effective columnar LR(λ) can then be estimated by minimizing the difference between measured AOD and the integrated lidar-based $\sigma_{ext}(\lambda)$ derived with an assumed, best matching LR(λ). When the Klett-Fernald method (Klett, 1982, Fernald et al., 1972) is used to derive $\sigma_{ext}(\lambda)$ and $\sigma_{bsc}(\lambda)$ with lidar, the LR(λ) is kept height-constant, and this assumption introduces significant uncertainties because the lidar ratio varies with height, i.e., with changing aerosol layering and aerosol type conditions (Guerrero-Rascado et al., 2011).*

Previous studies have focused on the dependence of $\sigma_{ext}(\lambda)$ on ambient RH (Skupin et al., 2013; Zieger et al., 2013). Navas-Guzmán et al. (2019) utilized these effects to investigate the aerosol hygroscopicity with lidar. LR(λ) is based on the RH-dependent $\sigma_{bsc}(\lambda)$ and $\sigma_{ext}(\lambda)$, and calculations by Sugimoto et al. (2015) indicated that LR(λ) is RH-dependent as well. Ackermann (1998) provided a numerical study based on pre-defined aerosol types with distinct size-distribution shapes to establish a power series to describe the LR(λ) as a function of RH. Salemink et al. (1984) found a linear relationship between the LR(λ) and the RH. Intensively discussed is the LR-enhancement due to hygroscopic growth in Zhao et al. (2017). They reported a positive relationship between LR and RH, but their study lacks information on vertically resolved aerosol particle number size distributions and other wavelengths. However, their simulations have shown that utilizing RH-dependent LR to retrieve aerosol particle light extinction from elastic backscatter lidar signals results in significantly different values than the constant LR approach. The studies above have shown an inconclusive dependence of the LR(λ) to the RH and corroborate that further research is needed, e.g., a quantification based on vertically resolved in-situ measurements. On the other hand, modeling is based on a large number of aerosol input parameters regarding particle size distribution and chemical composition as a function of height which is usually not available in the required density, e.g., because of airborne platform and payload limitations. Details are illuminated in the article.

In the following, we present two field experiments conducted in June 2015 and Winter 2017 at the regional central European background measurement facility at Melpitz, about 50~km northeast of Leipzig in eastern Germany. In both field studies, ground-based and airborne in-situ aerosol measurements and accompanying remote sensing were performed as measurements were performed during various atmospheric and aerosol conditions.

This study has three goals. Of central importance is the comparison of $\sigma_{bsc}(\lambda)$ and $\sigma_{ext}(\lambda)$ profiles obtained with lidar with respective modeling results based on airborne in-situ aerosol measurements. In this context, we want to highlight the challenges that have to be faced when instrumental limitations regarding airborne payloads do not determine the complete set of physicochemical aerosol properties. The second goal deals with the dependence of the lidar ratio on relative humidity. The humidity-related

LR enhancement at the three lidar wavelengths of 355, 532, and 1064 nm is modeled with input from the in-situ aerosol measurements. Finally, the study evaluates the ability of the Mie-model to reproduce measured $\sigma_{abs}(\lambda)$ values at different wavelengths. The goal is to provide a tool for the validation of lidar-photometer-retrieved $\sigma_{abs}(\lambda)$ estimates, as Tsekeri et al. (2018) show. The presented study, which includes modeling of $\sigma_{bsc}(\lambda)$, $\sigma_{ext}(\lambda)$, and $\sigma_{abs}(\lambda)$ in the ambient and dried state based on ground-based and vertically resolved in-situ measurements of aerosol properties as well as remote sensing with state-of-the-art photometers and multiwavelength aerosol lidar, is unique in its complexity.

The study is structured as follows. First, a general overview of the methodology is presented. Subsequently, the measurement site and the deployed instrumentations are described. Afterward, the comparison of Mie-modeled with the measured aerosol optical properties is presented and discussed separately for the summer and winter field observations. Meteorological and aerosol conditions and Mie-model validation efforts are presented in the supplementary material. The quantification of the RH-induced lidar ratio enhancement is discussed for the summer case. Finally, a summary and concluding remarks are given."

As a means to reduce the scope/length of the manuscript, I would suggest that the authors consider removing the lidar parameterization analysis. For this analysis, it is hard to tell if the comparisons are a bit circular at times with in-situ inputs into a Mie model being used to derive the lidar ratio parameterization. That parameterization is then used derive the lidar extinction. Then the lidar extinction is compare to the in-situ measured extinction. It would make for the more straightforward comparison if the authors just made comparisons to the lidar backscatter coefficients and avoided the assumptions/parameterizations needed to get the extinction altogether. This would also help shorten the manuscript. Plus, the lidar ratio parameterization is more a necessity because of the limitation of the Raman system to nighttime only and is less relevant to a general audience than the closure exercise.

First of all, to address the amount of detail, we restructured the manuscript and shifted some parts to the supplementary material (Aerosol and atmospheric conditions during the campaigns, Mie model validation). Also, we wrapped the section of the used instrumentation merging the summer and winter campaign. Also, the scope of the manuscript is to provide additional information about the LR-RH-dependency and is, in our opinion, a valuable contribution, especially considering the non-conclusive finding of previous studies. Due to the number of details, it was probably misleading that we used a constant lidar ratio (values and uncertainties by Mattis et al. (2004) for central European haze aerosol) to derive the lidar-based aerosol light extinction. We agree that using the modeled LR for deriving extinction would be circular. We will emphasize that in the manuscript. We added in lines 334- 335: "… derived $\sigma_{ext}(\lambda)$. Later, the LR derived with the Mie model in the ambient state is compared with the LR provided by Mattis et al. (2004)."

Moreover, the shown results indicate that, especially for large RH ranges within the planetary boundary layer, it can be important to account for a humidity effect if the LR, and consequently the *extinction, is enhanced by 2 to 3. Also, in the Fernald-Klett (Klett, 1985 and Fernald, 1984) algorithm, an inversion algorithm to derive aerosol optical properties from lidar, the lidar ratio is included, e.g., to estimate molecular extinction a certain altitude. The consideration of a relative humidity effect would thus contribute to an improvement.*

The overall conclusions and taken home messages of the study are completely lost in the in the details of the comparison. The goal of the study is the demonstrate the closure of aerosol optical property measurements, but fairly large differences remain. The authors speculate on several different reasons as to why the modeled and lidar-measured optical properties differ, but no real definitive answer is provided. The study would benefit from further analyses that are more focused on achieving closure to within a meaningful degree of certainty and a clear motivation/definition of what is "meaningful".

Yes, to some extent, the significant differences remain. However, we only can provide some speculations of the reasons for the differences due to the interdependent underlying parameters. However, since we updated the motivation and title, the detailed analysis of various reasons is valuable for future studies.

Before even attempting the closure exercise, it would be useful to discuss how good a closure one can expect given the uncertainties both the lidar and in-situ measurements. The uncertainties appear quite large at times which would suggest improved measurements techniques and methodologies are need before a useful closure exercise could be performed.

At first, the uncertainty of the lidar system: Backscatter measurements were estimated with an assumed uncertainty of 10% following Wandinger et al. (2016). The aerosol particle light extinction derived from the lidar contains two uncertainty sources, the underlying backscatter measurements and the uncertainty of the LR used for the transformation. The latter was taken from Mattis et al. (2004) with given uncertainty estimates for central European haze aerosol. These are 58 (±12; 21%) sr for 355 nm, 53 (±11, 21%) sr for 532 nm, and 45 (±15, 33%) sr at 1064 nm. At best, the uncertainty of lidar is then:

$$\delta\sigma_{\mathrm{ext}} = \sqrt{\left(\frac{LR*\sigma_{\mathrm{bsc}}}{dLR} * \Delta LR\right)^2 + \left(\frac{LR*\sigma_{\mathrm{bsc}}}{d\sigma_{\mathrm{bsc}}} * \Delta\sigma_{\mathrm{bsc}}\right)^2} = \sqrt{(\sigma_{\mathrm{bsc}} * \Delta LR)^2 + (LR * \Delta\sigma_{\mathrm{bsc}})^2}.$$

This equation translates into relative uncertainties of 23% at 355 nm, 23% at 532 nm, and 35% at 1064 nm. To provide uncertainties of the lidar extinction we updated the manuscript:

*We added in lines 322- 323: "*LR is an intensive aerosol property. *The estimates of $\sigma_{ext}(\lambda)$ hence are subject of uncertainties arising from the LR uncertainty and the $\sigma_{ext}(\lambda)$ and $\sigma_{bsc}(\lambda)$.", and later in lines 333 - 337: "... with the LR provided by Mattis et al. (2004). With the uncertainty range of the LR by Mattis et al. (2004) and applying Gaussian error propagation, the uncertainty of the lidar-based $\sigma_{ext}(\lambda)$ is at best 23% at 355 nm, and 532 nm, and 35% at 1064 nm, respectively."*

Uncertainties of the lidar measures are displayed with shaded areas around the vertical profiles and with error bars in the scatter plots.

Uncertainties of the modeled parameters are not as simple as for the lidar measurements. Like the density of volume fraction of eBC, many interdependent input parameters are fed into the model.

Therefore, to some extent, we established the Monto-Carlo simulation to estimate the uncertainties of modeled parameters capturing the spatial and temporal variability of the input. So yes, with the used instrumentation and assumptions, a closure is hard to achieve. However, showing that is useful as well, especially if one plans a similar study in the future.

Other comments:

The abstract needs to be considerably shorter. As is, it is a detailed summary of the entire paper and largely repetitive of the material in section 5.

We rephrased the entire abstract:

" A unique data set derived from remote sensing, airborne, and ground-based in situ measurements is presented. The study highlights the complexity of comparing multiple aerosol optical parameters examined with different approaches considering different states of humidification and atmospheric aerosol concentrations. Mie-theory-based modeled aerosol optical properties are compared with respective results of airborne and ground-based in-situ measurements and remote sensing (lidar, photometer) performed at the rural central European observatory at Melpitz, Germany. Calculated extinction-to-backscatter ratios (lidar ratios) are in the range of previously reported values. However, the lidar ratio is not only a function of the prevailing aerosol type but also of the relative humidity. The particle lidar ratio (LR) dependence on relative humidity was quantified and followed the trend found in previous studies. We present a fit function for the lidar wavelengths of 355, 532, and 1064 nm with an underlying equation of $fLR(RH, \gamma(\lambda)) = fLR(RH=0, \lambda)\times(1-RH)-\gamma(\lambda)$*, with the derived estimates of* $\gamma(355\text{ nm}) = 0.29\ (\pm0.01)$*,* $\gamma(532\text{ nm}) = 0.48\ (\pm0.01)$*, and* $\gamma(1064\text{ nm}) = 0.31\ (\pm0.01)$ *for the central European aerosol. This parameterization might be used in the data analysis of elastic-backscatter lidar observations or lidar-ratio-based aerosol typing efforts. Our study shows that the used aerosol model was able to reproduce the in-situ measurements of the aerosol particle light extinction coefficients (measured at dry conditions) within 13%. Although the model reproduced the in situ measured aerosol particle light absorption coefficients within a reasonable range, we identified a number of sources for significant uncertainties in the simulations, such as the unknown aerosol mixing state, brown carbon (organic material) fraction, and the wavelength-dependent refractive index. The modeled ambient-state aerosol particle light extinction and backscatter coefficients were found to be smaller than the measured ones. However, depending on the prevailing aerosol conditions, an overlap of the uncertainty ranges of both approaches was achieved."*

line 247: what "other studies"?

Exemplarily, we added Höpner et al. (2016), Omar et al. (2009), Kim et al. (2018), and Rosati et al. (2016) and updated the references accordingly. The part is now:

"Therefore, in this and other studies, e.g., Omar et al. (2009), Kim et al. (2018), Rosati et al. (2016a), and Höpner et al. (2016), the $\sigma_{bsc}(\lambda)$ *have been converted to* $\sigma_{ext}(\lambda)$ *utilizing the extinction-to-backscatter ratio, also known as lidar ratio (LR, in sr), with:"*

Höpner, F., Bender, F. A.-M., Ekman, A. M. L., Praveen, P. S., Bosch, C., Ogren, J. A., Andersson, A., Gustafsson, Ö., and Ramanathan, V.: Vertical profiles of optical and microphysical particle properties above the northern Indian Ocean during CARDEX 2012, Atmos. Chem. Phys., 16, 1045–1064, https://doi.org/10.5194/acp-16-1045-2016, 2016.

Kim, M.-H., Omar, A. H., Tackett, J. L., Vaughan, M. A., Winker, D. M., Trepte, C. R., Hu, Y., Liu, Z., Poole, L. R., Pitts, M. C., Kar, J., and Magill, B. E.: The CALIPSO version 4 automated aerosol classification and lidar ratio selection algorithm, Atmos. Meas. Tech., 11, 6107–6135, https://doi.org/10.5194/amt-11-6107-2018, 2018.

Omar, A. H., Winker, D. M., Vaughan, M. A., Hu, Y., Trepte, C. R., Ferrare, R. A., Lee, K., Hostetler, C. A., Kittaka, C., Rogers, R. R., Kuehn, R. E., and Liu, Z.: The CALIPSO Automated Aerosol Classification and Lidar Ratio Selection Algorithm. J. Atmos. Ocean. Tech., 26, 10, 1994-2014, 2009.

Rosati, B., Herrmann, E., Bucci, S., Fierli, F., Cairo, F., Gysel, M., Tillmann, R., Größ, J., Gobbi, G. P., Di Liberto, L., Di Donfrancesco, G., Wiedensohler, A., Weingartner, E., Virtanen, A., Mentel, T. F., and Baltensperger, U.: Studying the vertical aerosol extinction coefficient by comparing in situ airborne data and elastic backscatter lidar, Atmos. Chem. Phys., 16, 4539–4554, https://doi.org/10.5194/acp-16-4539-2016, 2016a.

line 357: add space after "campaign"

Due to the rearrangement of the paper content, this part does not exist anymore.

line 523: biasing -> attenuate

We changed accordingly. Part is in the supplementary material now.

line 863: remove "In Mie-theory"

We changed as requested.

Labels the panels in each figure (e.g. a, b, c) and use those labels when referring to specific panels in the text.

We updated all Figures and corresponding references within the text accordingly.

Figure 6, last panel: suggest addition a scale break in the x axis. As is, the x limits are too wide to discern any differences.

We changed Figures 6 and 8 (now 3 and 5) accordingly by adding a break and squeezing the scale of the 2nd part (see exemplarily below).

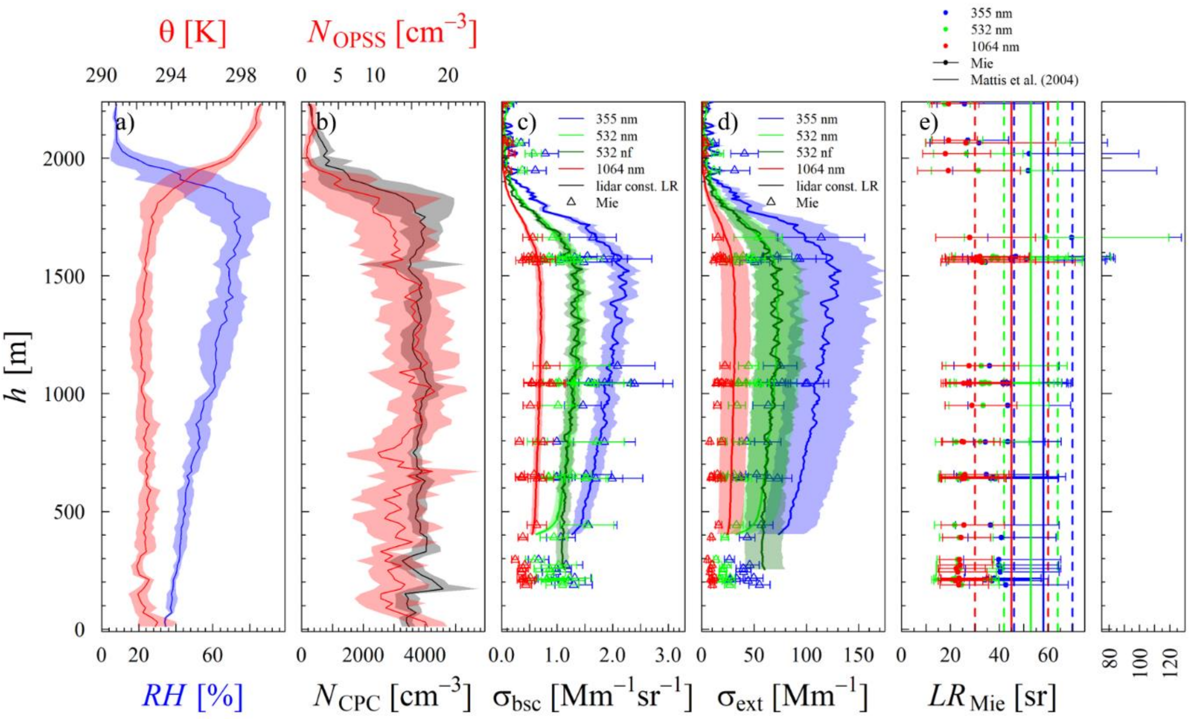

Figure 3: Updated Figure 5.

Figures 13, 14: the large amount of overlap in data points and errors bars make it difficult to see anything quantitative from these plots. These may be better plotted using some type of density-based plot.

Thank for the comment. Since 1 Hz data are not providing any helpful information, we decided to reprocess the data of the STAP on a 30-second basis, which significantly reduced the noise. Moreover, we added a correction in terms of scattering and updated all estimates of depending parameters like AAE and the correlation. We plotted the lines thinner, and exemplarily the graphs for March 9, 2017, are shown below.

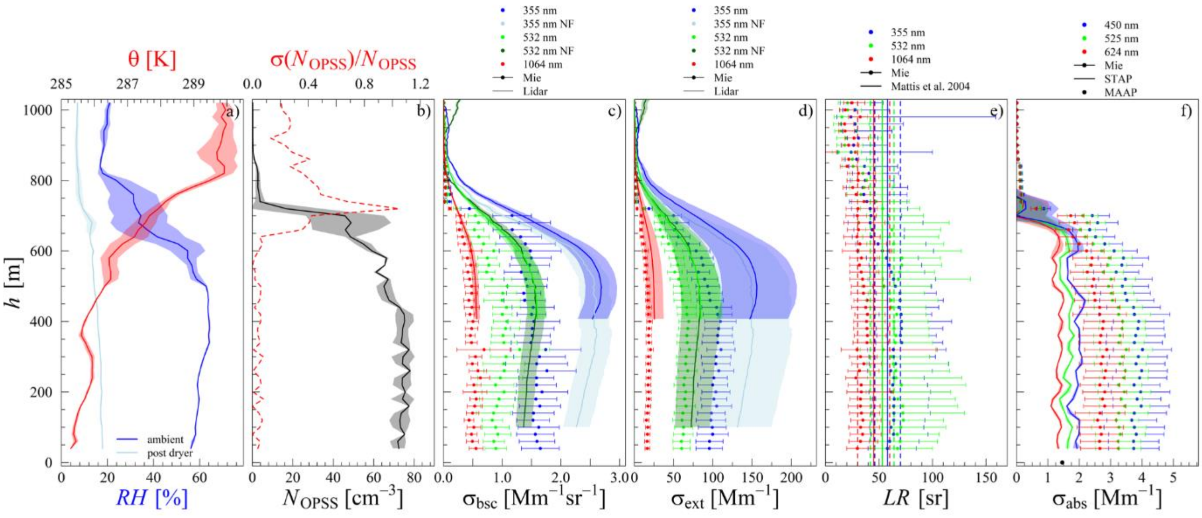

Figure 4: Updated Figure 8.

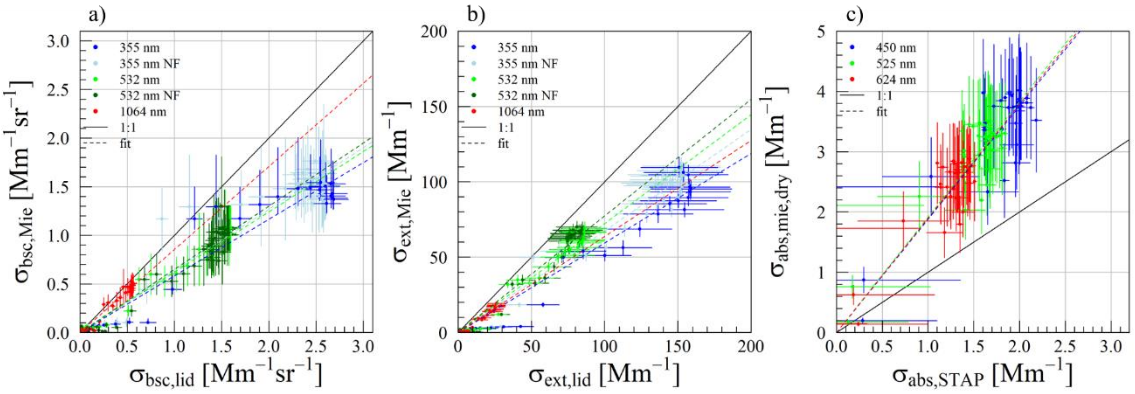

Figure 5: Updated Figure 10.