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 $\sigma_{\mathrm{ext}}(\lambda)$*. 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* $\gamma(355\ \mathrm{nm}) = 0.29\ (\pm 0.01)$*,* $\gamma(532\ \mathrm{nm}) = 0.48\ (\pm 0.01)$*, and* $\gamma(1064\ \mathrm{nm}) = 0.31\ (\pm 0.01)$ *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 2^{nd} part (see exemplarily below).

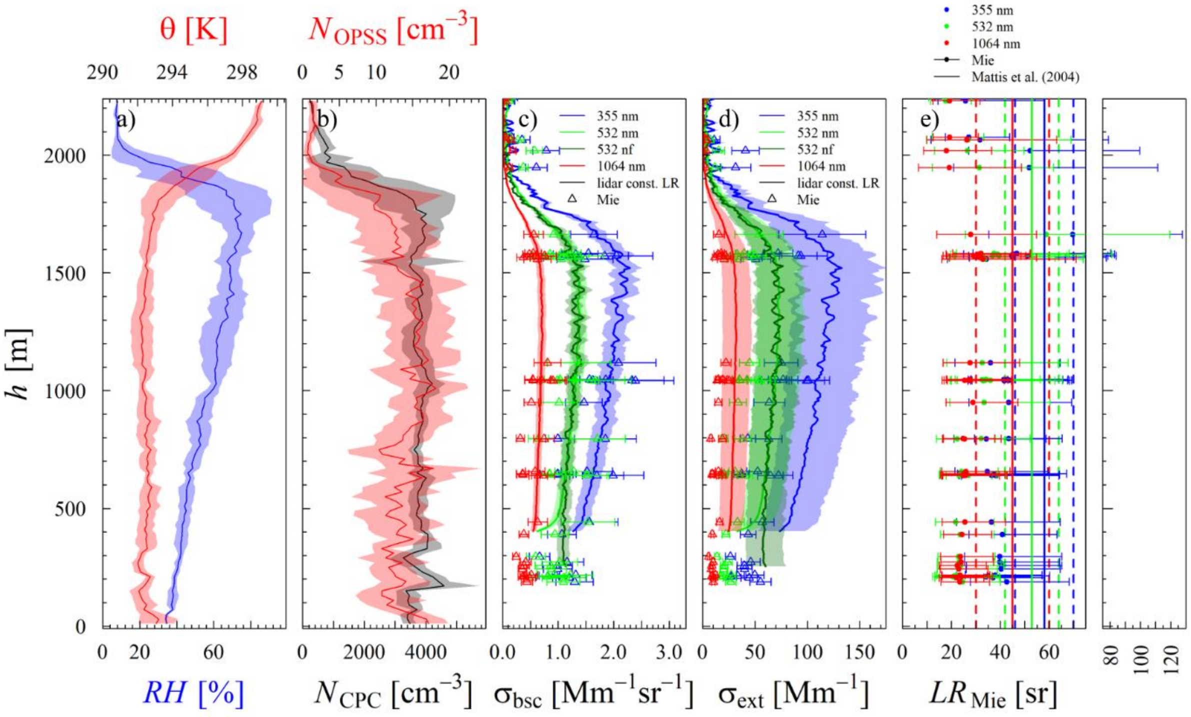

Figure 1: Updated Figure 5.

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.

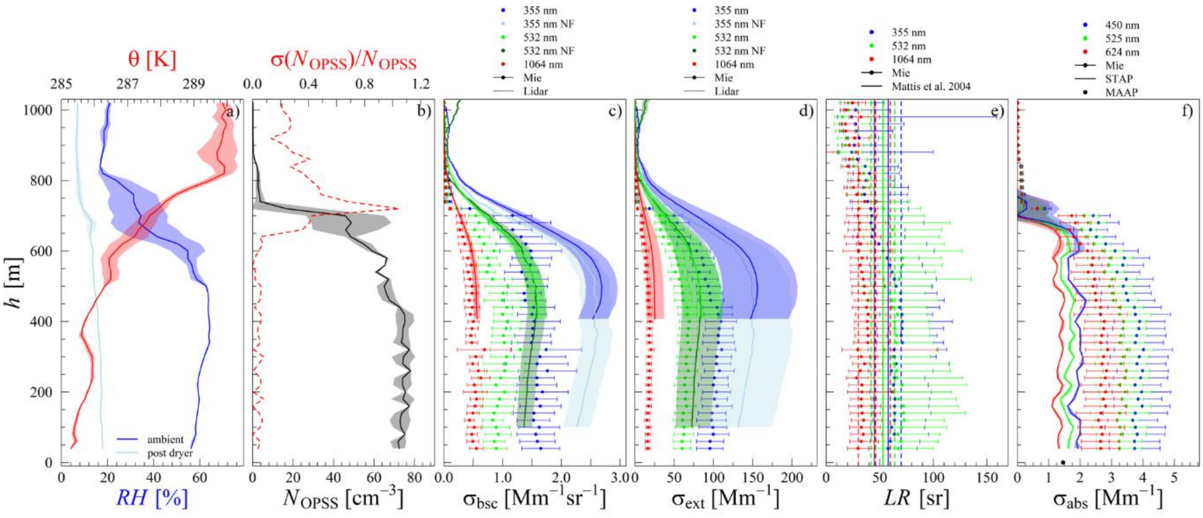

Figure 2: Updated Figure 8.

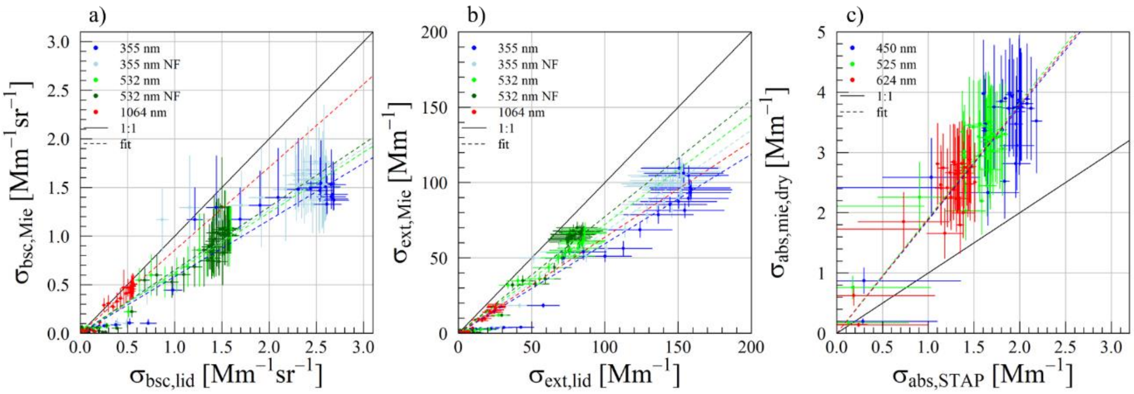

Figure 3: Updated Figure 10.