

## Response to Referee #2

Thank you for carefully reading the manuscript and providing useful suggestions to improve the paper. The replies to the referee comments are given below. The referee comments are highlighted in blue with our responses in black. The sentences in the manuscript are between the quotation marks, with the modifications in the revised manuscript in red.

**Authors provide comparison of the particle mass concentration obtained from Raman lidar and from ceilometer measurements, which is important topic. The manuscript is well written and can be published after minor revision.**

We are grateful to the referee for positive assessment of our work.

**Ln 159. “calculated from the relative humidity and the temperature profiles from GDAS1 data”**

**GDAS profiles may differ significantly from real profiles of the water vapor. This should be kept in mind when using these for correction. Did you compare GDAS with profiles obtained from Raman measurements?**

Thank you for the comment. We agree with the referee that GDAS profiles may differ from real ones. Unfortunately, radiosonde measurements were not collocated with lidar observations. The nearest sounding station locates at Jokioinen, which is ~300 km away from our measurement site. The Raman lidar relative humidity measurements are only available at nighttime. Nevertheless, we compared GDAS profiles with Raman lidar profiles, good agreements were found.

We have added in section 2.1 in the revised version for the clarity:

“  
Temperature and pressure profiles from the GDAS (Global Data Assimilation System, <https://www.ready.noaa.gov/gdas1.php>, last access: 19 March 2021) database were used for the correction of Rayleigh extinction and backscattering effects for lidar data analysis. **Aiming at the observations of water vapor profiles, the most used and well-established measurement method is radiosonde sounding. However, the closest available radiosonde data are from Jokioinen (Finland), located ~ 300 km away from the measurement site. Filioglou et al. (2017) reported the inadequate vertical representation of water vapor due to the non-stable atmospheric conditions between two sites, when using a radiosonde 100 km away. Thus, the relative humidity profiles from GDAS data were used for the water vapor number densities estimations.**  
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**Ln.166. What can we conclude from using both forward and backward Klett methods? Forward method is very sensitive to the choice of lidar ratio. The lidar ratio of smoke can vary in significant range, so use of just one value obtained from rotational Raman lidar is risky. Did you compare it with lidar ratio provided by AERONET?**

Thank you for the comments. We agree that the extinction retrieval is sensitive to the assumed lidar ratio. In this study we have chosen the reference height as close as to the layer of interest, so that the error propagation could be minimized. This was clarified in the revised version. We have also added description about forward and backward method in the revised version.

In section 2.3:

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The retrieval methods for deriving the backscatter coefficient from ceilometers are quite mature (Wiegner et al., 2014; Wiegner and Geiß, 2012). **Under favourable conditions, a relative error of the backscatter coefficient on the order of 10 % seems feasible with a careful calibration by applying the forward integration. On the contrary, significant temporal averaging of ceilometer data is required for performing a Rayleigh calibration, as the detection of molecular signals is intrinsically very difficult. Binietoglou et al. (2011) propose a two-step approach, resulting promising agreement comparing to their lidar PEARL (Potenza EARLINET Raman lidar). The uncertainty of the backscatter coefficient could be in the range of 20–30 % using the backward integration. The advantage of the forward algorithm is that calibration is required only occasionally, and it is not affected by the low SNR in the upper troposphere. However, the accuracy in deriving extinction coefficients is limited due to**  
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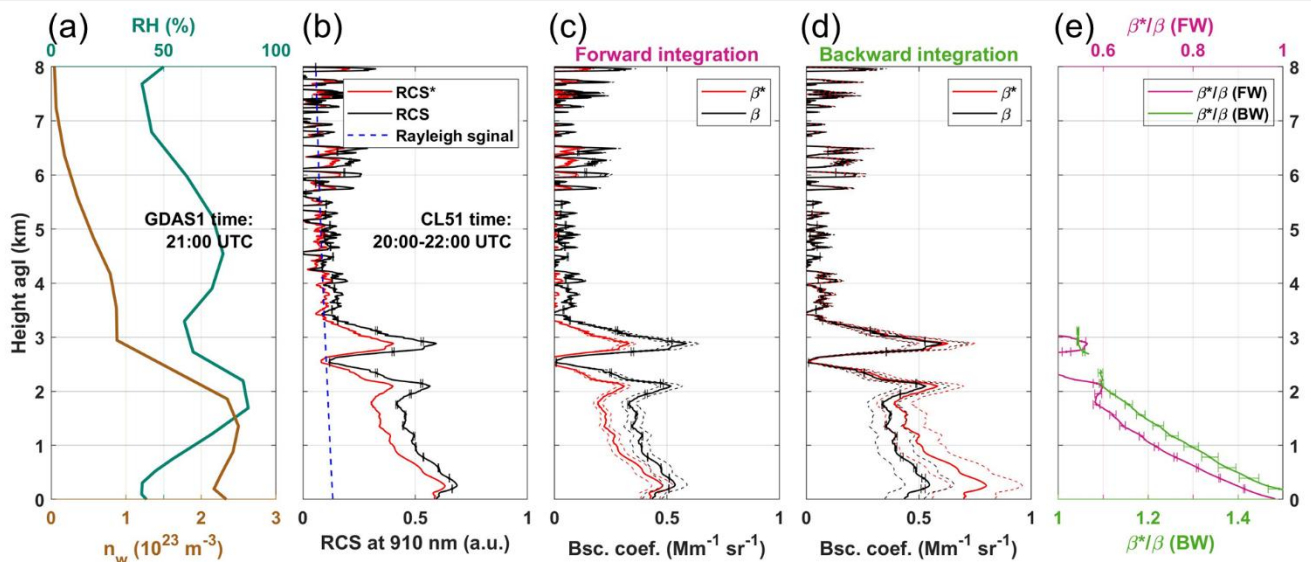
the unknown LR at 910 or 1064 nm and its uncertainties. In particular the presence of multi-layered aerosol distributions (with different aerosol types) may introduce more uncertainties. In addition, the uncertainty due to the neglecting the water vapor increased with the distance from the chosen reference height. In this study, we applied the Klett method (Wiegner et al., 2014) by defining the reference height as close as to the layer of interest, so that the error propagation (due to uncertainties of LR and water vapor transmission) would be minimized for that layer.

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 The lidar ratio provided by AERONET is for the total atmospheric column (as we mentioned in section 2.1), during the period, there were multi aerosol layers. Thus, it is not fully comparable using AERONET LR and PollyXT LR of smoke layer. Besides, AERONET level 2.0 LR were not available on 5 June (the day of the SPoI - smoke layer).

**Fig.1c. Signal above 3 km is very noisy so results probably depends on choice of the reference height.**

We agree, so we chose the reference height as close as to the layer of interest to minimize the error. We have added the uncertainties due to wrong assumptions of  $\lambda_0 \pm 2$  nm and due to the analytical solution in the Fig. 1.

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**Figure 1. Example of water vapor corrections on 2 h averaged ceilometer data on 5 June 2019 (20:00–22:00 UTC). (a) Relative humidity (RH, teal) and water vapor number density ( $n_w$ , brown) from GDAS1 data at 21:00 UTC. (b) Range-corrected signal at 910 nm, without (RCS\*, red) or with (RCS, black) water vapor correction, and the hypothetical Rayleigh-signal at 910 nm (dashed blue). (c) Retrieved particle backscatter coefficients:  $\beta^*$  without (red) and  $\beta$  with (black) water vapor correction, using forward (FW) integration Klett solution. (d) Same as (c) but application of the backward (BW) integration. (e) Ratio of the retrieved  $\beta^*$  and  $\beta$ , when using forward integration (magenta), or backward integration (green). The horizontal lines illustrate the uncertainties range due to wrong assumptions of the central wavelength  $\lambda_0 \pm 2$  nm. The uncertainties in backscatter coefficients of the analytical solution were shown by dashed lines.**

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**Fig.5d. I am a little confused. What means “contribution of BrC”? Please specify.**

It is the brown carbon (BrC) contribution to the absorption coefficient (at 370 nm).  
 But we have removed this figure and also the text about in situ measurements in the revised version.

**Fig.6. The demonstration of the temporal evolution of the profiles is useful. Still would be good to quantify the difference between lidar and ceilometer. May be provide standard deviation?**

Thank you for the suggestion, we have added a new sub-figure (Fig.5d) to show the difference on the estimated mass concentrations as follows.

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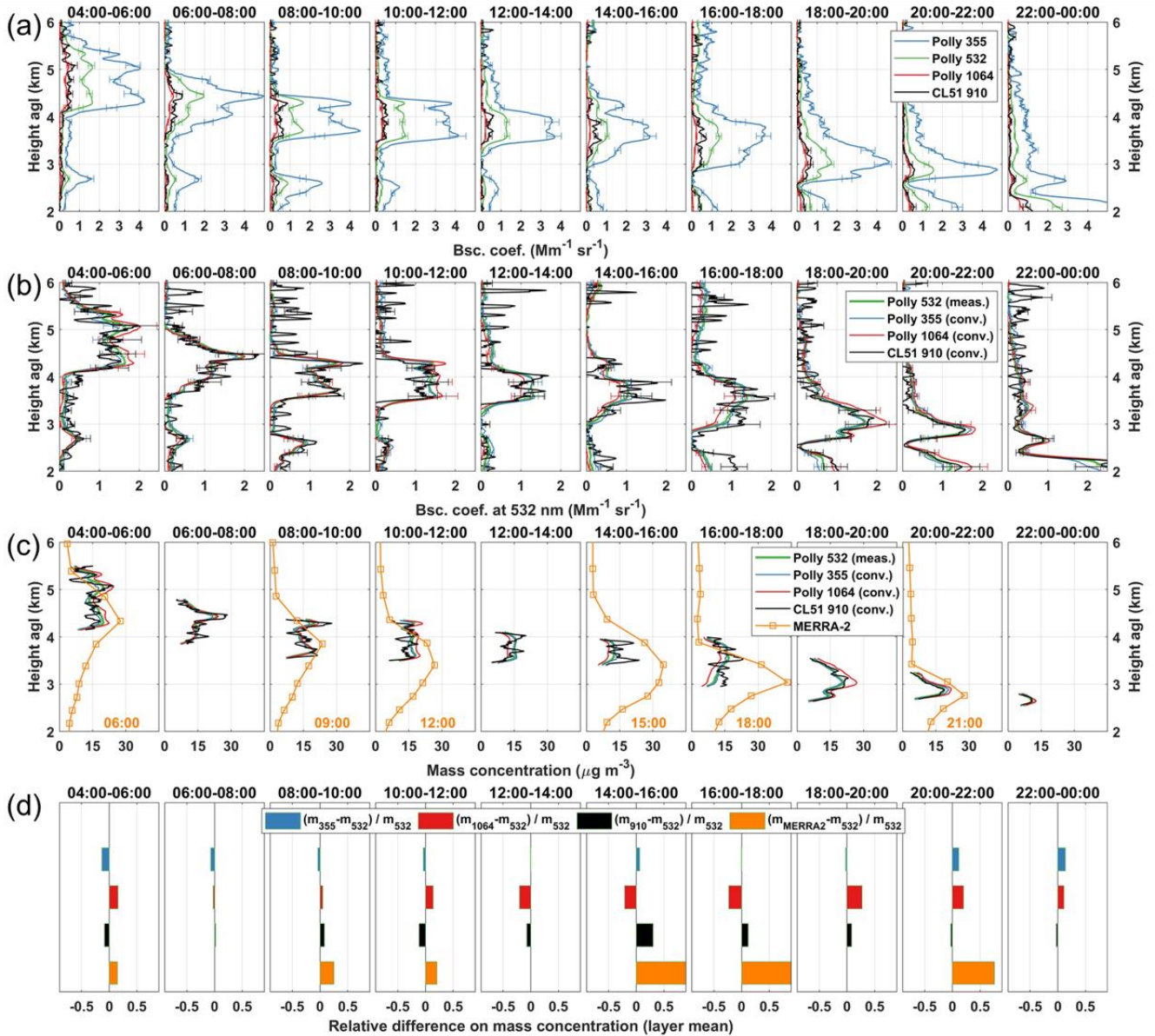


Figure 6. (a) Lidar-derived backscatter coefficients (BSC) at 355 (blue), 532 (green), and 1064 nm (red) from Polly<sup>XT</sup>, and at 910 nm (black) from CL51. (b) BSCs at 532 nm: measured at 532 nm (meas.), or converted (conv.) from measured BSCs at other wavelengths in (a). (c) Estimated mass concentration profiles for the SPoI (Smoke Plume of Interest) from BSCs in (b), based on parameters in Table 2-method #1. Mass concentrations from MERRA-2 model are also shown in orange color with corresponding time given on the bottom right of each panel. (d) Relative differences on the mass concentrations (denoted as  $m$ ) estimated from measured/converted BSCs, and from MERRA-2 model, using the one estimated from measured BSC at 532 nm as the reference. 2 h time-averaged lidar profiles are used, with the time slot (UTC) on 5 June 2019 given on top of each panel. The horizontal lines (in a, b) illustrate the uncertainties range. The uncertainties in mass concentrations (in c) are discussed in Sect. 3.2.

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I agree with the first reviewer, that numbers should be used instead expressions like “good agreement”.

Thank you for the suggestion, we have carefully checked the manuscript and made modifications for the quantitative description on ‘mass concentration value’, ‘good agreement’ or ‘large discrepancies’, in the revised version:

In section 3.2.1 “Method #1: based on BAE & the conversion factor from literature”:

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The peak value of the mass concentrations was found at 6–8h UTC, of  $\sim 23.5$  ( $27.5$ )  $\mu\text{g m}^{-3}$  estimated from the backscatter coefficients at 532 nm (910 nm). If we take the mass concentration estimated from the BSC at 532

nm as the reference, good agreements are found between the mass concentrations estimated from BSCs at different wavelengths (Fig. 5 d). The mean values of the relative differences were around 8 %, 12 %, and 18 % for the estimations from BSCs at 355, 910 and 1064 nm, respectively. Comparing 532 and 355 nm mass estimates, better agreements were found during daytime (8–20h UTC), with a difference <6 %. Nonetheless, considering 532 and 910 nm estimates, the best agreements were found at 6–8 and 20–24h UTC, with a difference <3 %, whereas the worst agreement of ~ 30 % was found at 14–16h UTC. Larger differences between 910 and 1064 nm estimates were found, with a mean relative difference of ~ 28 %, and a highest value of ~ 64 % at 14–16h UTC.

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In section 3.2.2 “Method #2: BSC at each wavelength & conversion factors from site”:

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The peak value of the mass concentrations estimated from the BSCs at 532 nm reached ~ 38  $\mu\text{g m}^{-3}$  at 6–8h UTC, higher than the one estimated from method #1 because of the bigger conversion factor. The relative differences on the mass concentrations estimated from the BSCs at different wavelengths were analysed (Fig. 7 b). Similarly, we take the mass concentration estimated from the BSCs at 532 nm (which is the wavelength most often used in earlier studies) as the reference, and found an underestimate when using BSCs at 355 nm, with a mean bias of ~ 15 %, and a peak bias of ~ 25 % at 4–6h UTC; the best agreement was found for night-time measurements (20–24 h UTC) with a bias <5 %. Nevertheless, an overestimate was found for the mass concentration estimated from the BSCs at 910 nm, with a mean bias of ~ 36 %, a peak bias of ~ 68 % at 14–16h UTC, and a minimum bias of ~ 14 % at 10–12h UTC. The overestimate for CL51-derived mass concentrations could be due to an overestimate of LR at 910 nm, since we used LR at 1064 nm in the calculations. In addition, big differences (with a mean value of ~ 42 %) were found between the CL51-derived mass concentrations and the ones estimated from the Polly<sup>XT</sup>-derived BSCs at 1064 nm; highest discrepancy were found of ~ 95 % at 14–16h and ~ 75 % at 16–18h UTC, whereas better agreements were found at 4–6h, 10–12h, and 18–24h, with bias <7 %.

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In section 3.3 “Comparison with MERRA-2 model - wildfire smoke and dust aerosol mixture”:

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In this section, the MERRA-2 mass concentrations were compared with the mass concentrations estimated from the Polly<sup>XT</sup> backscatter coefficients at 532 nm (from method #1-Fig. 5 c, and method #2-Fig. 7 a). Note that the main difference on Polly<sup>XT</sup>-estimated mass concentrations from two methods are due to the different conversion factor values (Table 2), thus the mass concentrations estimated from BSCs at 532 nm using method #1 are ~ 40 % lower than method #2. When the Polly<sup>XT</sup> estimates from method #1 were used as the reference, good consistencies were found in the morning (at 6h, 9h, and 12h UTC), with overestimations (<30 %) of MERRA-2 mass concentrations; whereas large discrepancies were found in the afternoon, with high overestimations of ~ 160 % at 15h UTC and ~ 90 % at 18h UTC. If the Polly<sup>XT</sup> estimates from method #2 were used as the reference, good consistencies were also found in the morning (at 6h, 9h, and 12h UTC), but with underestimations (<30 %); and a large overestimation of ~ 63 % was found at 15h UTC. At 15h UTC, the MERRA-2 simulated dust mass concentration fraction is more than half of the MERRA-2 simulated total mass concentration. It is good to keep in mind that both observations and simulations have significant uncertainties. The presence of cirrus cloud in the upper atmosphere during the day may also have some impacts on MODIS AOD, which is assimilated by the MERRA-2 model.

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