We thank the anonymous reviewer for the helpful comments. These comments helped to substantially improve the manuscript. Below we give detailed answers to the individual reviewer comments in blue.

Manuscript: Specifying light absorbing properties of aerosol particles in fresh snow samples, collected at the Environmental Research Station Schneefernerhaus (UFS), Zugspitze (Linke et al.,)

It is often regarded that the polar regions are the "canary in the coal mine" regarding this region's sensitivity to changes in radiative forcing. This sensitivity derives, in part, from the very high surface albedos that typify these regions. However, despite the recognized importance of this subject, our quantitative understanding of the radiative contribution by light absorbing aerosols - be it atmospheric or surface-deposited particles - in these regions - and on snow/ice - is still very low. Therefore, more research is indeed needed to improve our understanding of this subject and to reduce the associated uncertainty. To this end, the manuscript by Linke and co-workers undertake an investigation of light absorbing properties of aerosols deposited on snow near the Environmental Research Station Schneefernerhaus with the goal to better quantify the aerosol types (classes) of light absorbing aerosols present in this region. To derive the optical properties, these researchers combine aerosol light absorption measured by a photoacoustic spectrometer with refractory black carbon (rBC) mass loading measurements reported by a single particle soot photometer (SP2) on re-aerosolized snow samples. The insights and data that can be gleaned from a study such as this is of value and should, eventually, be published. But in its current state, the manuscript is not ready and thus is not recommended for publication at this time.

One of the biggest issues this reviewer has centers on how these researchers are combining their light absorption and rBC mass measurements. In the atmospheric aerosol community, we take these two datasets and derive the mass absorption cross-section (MAC; m2/g). This property enables researchers to say something about the mixing state of the rBC particles as well as something about aerosol type (e.g., the presence/absence of light absorbing organic material/particles (e.g., brown carbon or BrC and dust). However, despite the ubiquity of this methodology in our community, the authors, instead, advocate using a methodology that compares the SP2-derived mass with what they term as a "fullerene soot equivalent mass" derived from the photo acoustic spectrometer. In order to use a "fullerene equivalent mass" derived from the light absorption measurements the researchers must assume that the MAC is constant - the literature is populated with many studies that show that the MAC is not constant. For example, at 550 nm, the black carbon MAC for fresh (uncoated) soot is ~ 7.5 m2/g while for thickly-coated black carbon particles this value could be 12-14 m2/g at this wavelength, thus representing a factor of 2x change. How do the authors account for the potential changes in the MAC brought about by changes in the rBC mixing state? Why not simply derive a MAC and utilize the variability in this value to infer something about the light absorbing particles deposited on the snow? The authors provide no justification as to why their methodology is preferred. If the authors feel strongly that this methodology offers an advantage not available with a MAC-based methodology, then they need to present that argument. Further, any sources of error in the interpretation of the data using this methodology is not addressed and needs to be.

The referee can be sure that the authors know the importance and the ubiquity of the MAC in the atmospheric aerosol community as they published important contribution to this field in the past (e.g. Schnaiter et al., 2003; Schnaiter et al., 2005; Schnaiter et al., 2006). The authors used the optical equivalent BC mass here because it was used in one of the most comprehensive studies on light absorbing impurities in Arctic snow by Doherty et al. (2010). Also, the authors are aware of the fact that the MAC is not constant in the atmosphere and the MAC of thickly coated soot can be enhanced by a factor of two, as they provided the first experimental proof of this effect already in 2005 (Schnaiter et al., 2005). However, the authors agree that the presentation of the absorption measurements is insufficient in the

manuscript. Therefore, we did a thorough reanalysis of the snow samples following the following strategy that also includes a MAC analysis of the data:

1. The snow mass specific absorption cross section $\sigma_{abs}$ [m$^2$/mL] was deduced from the absorption coefficient $b_{abs}$ [m$^{-1}$] using the nebulizer flow settings and the nebulizing efficiency (Eq. 1 of the revised manuscript):

$$\sigma_{abs} = 10^{-3} \cdot b_{abs} \cdot R_{neb}/R_{pp} \cdot \varepsilon_{neb}^{-1}$$

2. This snow mass specific absorption cross section is plotted as a function of the refractory BC mass concentrations in a new figure (Fig. 10 of the revised manuscript). Linear regression fits to the data then give the BC mass specific MAC of the snow samples. We found MAC values that are up to a factor of two larger than the MAC of Fullerene soot. A table is added to the manuscript contrasting the mass and optical properties (including the MAC) of Fullerene soot with those of the snow samples.

3. To be comparable with other studies (e.g. Doherty et al., 2010), we calculated the equivalent BC mass concentration $c_{BC}^{equiv}$, i.e. the amount of BC that would need to be present in the snow to account for the measured absorption, from $\sigma_{abs}$ using the MAC of Fullerene soot (Equation 2 of the revised manuscript):

$$c_{BC}^{equiv} = \sigma_{abs}/\text{MAC}_{FS}$$

4. We plotted $c_{BC}^{equiv}$ as a function of the refractory BC mass concentration in a new figure (Fig. 11 of the revised manuscript) and found a good correlation of both concentrations but with correlation coefficients of 2.0, 1.9, and 1.4 for 405, 532, and 658 nm, respectively (note that $c_{BC}^{equiv}$ is a function of the wavelength). We conclude from this that there is additional non-BC light absorbing mass in the snow, which is correlated with the BC mass and which has a strong wavelength dependence between the green and the red part of the visible spectrum. This already indicates mineral dust and organic (brown) carbon as possible carriers of this additional absorption.

5. We calculated the snow mass specific absorption cross section of the non-BC particles, $\sigma_{abs}^{nonBC}$ (Equation 3 in the revised manuscript):

$$\sigma_{abs}^{nonBC} = \sigma_{abs} - c_{BC}^{SP2} \cdot \text{MAC}_{FS} \cdot 10^{-9}$$

We added a figure (Fig. 12 in the revised manuscript) that shows the statistical analysis of the $\sigma_{abs}^{nonBC}$ for the snow samples and that compares this non-BC spectral behavior with laboratory data for Saharan dust and organic (brown) carbon (see answer to the previous comment).

Continuing, this reviewer is very concerned about the reported MAC values for the fullerene standard. Fullerene soot should be uncoated and thus should exhibit a MAC that is characteristic of an uncoated black carbon particle, namely a MAC ~ 7.5 m2/g at 550 nm. Yet, at 532 nm, the authors report a MAC of of 10.6 (+/- 2.8) m2/g - a value that is ~40% that the canonical value for denuded soot, and 65% larger than the fullerene MAC reported by Zhou (6.4 m2/g) - work cited by the authors. Additionally, it is also at odds with the recently published value by Zangmeister 6.1 m2/g at 550 nm (Carbon (2018), doi: 10.1016/j.carbon.2018.04.057). The disagreement is quite critical as it will have bearing on interpretation of data collected on the snow samples. While the authors are correct that the BC MAC is dependent on particle size, using the argument that the differences between their higher values and that reported by others

as being due to differences in the sample size distribution studied is not correct. Using a refractive index of 1.95-0.79i (Bond & Bergstrom (2006); Aero. Sci. Tech. 40:1, 27- 67) a straight forward Mie calculation reveals that the maximum MAC calculated for a BC is ~7.9 m2/g for a BC diameter of 150 nm and that the MAC only goes down with either increasing or decreasing particle size. Therefore another explanation is needed. Three potential explanations submitted here are: (i) either the fullerene soot is coated and the derived MACs reflect a lensing-enhanced value or (ii) there are some non- BC, light absorbing aerosols present in the sample, or (iii) the number concentrations used during the fullerene soot calibration are high enough that particle coincidence is occurring that the post-processing of the SP2 data does not correct for. This latter possibility is brought up because particle-resolved measurements, like the SP2, can easily find themselves in the particle-coincidence regime. Such an undercounting of the actual fullerene mass would bring the reported MACs down and thus reconcile their MAC values with others. The authors are encouraged to insure that the SP2 is not operating in this regime.

We do not agree with the Referee that the MAC of uncoated BC is canonical with a constant value of 7.5 m2/g. Only because something is continuously referenced does not necessarily mean that it is correct. A constant MAC would mean that refractive index of the material is constant. From a solid-state physical perspective this is highly uncertain in case of carbonaceous material with its variable electronic band structures. Already pure carbon can exist in five different allotropes including graphite and diamond – one is black the other transparent – so obviously completely different refractive indices. Adding now the morphological variability of fractal soot particles to the discussion with a wealth of different monomer sizes, monomer nonsphericity (irregularity), necking, and overlapping (even on a single particle) that all have an influence on the particle absorption cross section, it is conclusive to the authors that the absorption cross section of this material cannot be constant but depends on its formation conditions. Further, the MAC is calculated from the absorption cross section and the particle mass. Particle mass is measured in different ways – sometimes as a total mass (including non-refractory compounds as in the DMA-APM method), sometimes as a refractory mass only (as with the SP2). So, there comes an uncertainty also from this side into the MAC. It is correct that Fullerene soot was identified as a good standard for atmospheric BC in terms of the SP2 mass sensitivity (although with a variation of 15% from batch to batch). But does that mean that this is also the case for its spectral absorption properties given the variability in the electronic and microphysical structures discussed above? With all this, the authors do not see any reason why a laboratory to laboratory difference in published MAC values of 30-40% shouldn't be within the variability range of the material itself, the size differences and the mass measurement methods used in the different studies.
The authors took care that the SP2 wasn't operated under aerosol concentrations that would result in coincidence errors. However, it turned out that due to the upper cut size of the SP2 (560 nm), we missed about 10% of the Fullerene particle mass. In the reanalysis we therefore applied lognormal fits to the measured BC mass size distributions to account for this mass outside the SP2 sizing range (see Fig. 6 of the revised manuscript). With this correction the MAC of FS is reduced to 9.5 ± 2.2 m2/g (532 nm). Further, to be comparable with the Zangmeister et al. (2018) study who measured the MAC on size-selected FS particles, we also measured the MAC of size selected Fullerene soot particles in a separate study by adding a DMA behind the Marin-5. We found a MAC of 8.6 m2/g for the same mobility-equivalent diameter (350 nm) as used in the Zangmeister et al. study. This is still 40% larger than the 6.1 m2/g of Zangmeister et al., but given the fact that (i) a APM/DMA was used for the mass measurement in their study and (ii) they used a different batch of Fullerene soot, this difference is within the range of variability that can be expected in such a comparison.

The whole Sect. 4 was rewritten to make the discussion of the Fullerene soot MAC clearer:

"Simultaneously to the BC mass concentration measurements with the SP2, the absorption coefficients $b_{abs}$ of the Fullerene soot suspensions were measured for the three PAAS-3λ

wavelengths. Both measurements together enable the determination of the mass specific absorption cross section $\text{MAC}_{FS} = b_{abs}/c_{BC}^{SP2}$ of airborne Fullerene soot at 405, 532, and 658 nm. In **Error! Reference source not found.**, the absorption coefficients $b_{abs}$ are plotted against the SP2-derived BC mass concentrations $c_{BC}^{SP2}$ of the Fullerene soot suspension standards. Linear regression fits of the data result in $\text{MAC}_{FS}$ values of 10.5 ± 3.2 m2/g, 9.5 ± 2.2 m2/g, and 8.6 ± 3.3 m2/g for 405, 532, and 658 nm, respectively. The $\text{MAC}_{FS}$ at 532 nm is comparable to the value of 8.84 m2/g given by Schwarz et al. (2012) for Fullerene soot (lot #F12S011) deduced from ISSW measurements, but is significantly higher than the 6.1 ± 0.4 m2/g (mean ± 2σ) measured recently by photoacoustic absorption spectroscopy for size selected Fullerene soot particles by Zangmeister et al. (2018). They used a combination of a differential mobility analyzer (DMA) and an aerosol particle mass analyzer (APM) to select Fullerene soot particles within a narrow mass range from aerosol generated by an atomizer. Their $\text{MAC}_{FS}$ of 6.1 m2/g, which is given for a wavelength of 550 nm, a mobility-equivalent diameter of 350 nm, and for a particle mass of $16.6 \cdot 10^{-15}$ g, corresponds to a volume-equivalent diameter of 264 nm using a density of 1.72 g/cm3 of Fullerene soot (Kondo et al., 2011). Although, this diameter is not very different to the MMD of 228 nm of the Fullerene soot suspensions used here, part of the observed discrepancy can be attributed to the different sizes as the MAC is strongly depending on the particle diameter for particles larger than about 200 nm (e.g. Moosmüller et al., 2009). To be comparable, we measured the $\text{MAC}_{FS}$ of size selected Fullerene soot particles in a separate study by adding a DMA behind the Marin-5 in the setup shown in **Error! Reference source not found.**. A $\text{MAC}_{FS}$ of 8.6 m2/g was measured for the mobility-equivalent diameter of 350 nm, which is still ~40% larger than the $\text{MAC}_{FS}$ given by Zangmeister et al. (2018) for the same diameter. However, they used an APM to measure the BC mass, while a SP2 was used here to deduce the refractory BC mass. According to Laborde et al. (2012a), the Fullerene soot product shows a variability from batch to batch, which results in a SP2 calibration uncertainty of up to 15% (actually only two batches were compared; lot #F12S011 and lot #L18U002). They explained the differences in the SP2 response (i.e. the calibration curves) by a substantial non-refractory coating in case of the L18U002 batch that could be identified by thermodenuding the samples. Assuming that lot #W08A039 used in Zangmeister et al. (2018) has a similar coating, this would increase the APM mass measurement by about 15% compared to the SP2-derived BC mass of lot # F12S011 used in the present study. This in turn would increase the $\text{MAC}_{FS}$ from 6.1 m2/g reported by Zangmeister et al. (2018) to about 7 m2/g when using only the refractory BC mass fraction in the calculation of the $\text{MAC}_{FS}$. This assumption reduces the discrepancy between the two $\text{MAC}_{FS}$ values to 35%, which is within the uncertainty range of ± 2.2 m2/g for our 532 nm value. It is further conceivable that different batches of the Fullerene soot material have different electronic band structure (i.e. refractive index) and/or fractal aggregate structures that both change the absorption cross section of the particles at a constant particle mass (e.g. Liu et al., 2019; Zangmeister et al., 2018). **Error! Reference source not found.** shows an electron micrograph of a typical Fullerene soot aggregate sampled from the dry aerosol output of the Marin-5 nebulizer. Thus, the Fullerene soot particles do not have a simple fractal aggregate structure, but are rather complex-structured with polydisperse monomer sizes, monomer nonsphericity (irregularity), necking, and overlapping, which all have a significant impact on the optical particle properties (including the absorption cross section) compared to the idealized fractal aggregate (Teng et al., 2019). Since these microphysical details of the soot particles are very sensitive to the actual formation and subsequent treatment conditions (Gorelik et al., 2002), it is conclusive that the $\text{MAC}_{FS}$ has an even higher variability between different Fullerene soot batches compared to what is expected from the SP2 mass sensitivity only.

The wavelength dependence of the aerosol light absorption, expressed by the so-called absorption Angström exponent (AAE), was determined to be 0.46 ± 0.07 for the used Fullerene soot suspensions by analysing the $b_{abs}$ data for the 405 and 658 nm wavelengths. This AAE is close to the ~0.6 reported by Baumgardner et al. (2012) for Fullerene soot derived from multiwavelength PSAP and Aethalometer measurements and it is within the range of the 0.54 ± 0.06 determined by Zhou et al. (2017) from ISSW spectrometer

While on this subject, what were the re-aerosolized number concentrations in the snow samples studied? On lines 243-244, the authors state that ". . ..Marin-5 nebulizer was then fixed at a rate of 0.32 mL min-1, which guarantees a high enough particle mass concentration for the photo acoustic measurement. Depending upon the mass concentration and mode size of the particles, this could hint at co-incidence issues with the SP2.

The concentrations behind the Marin-5 were always low enough (< 1000 #/cc) to avoid any coincidence issues.

Other specific issues:

Do the authors worry about the loss of water soluble BrC during sample preparation that might not be reflected when re-aerosolizing their snow samples?

We do not have any information on potential soluble BrC mass loss.

Line 167: What were the PSL diameters used? While the incandescence channel of the SP2 is sensitive enough to detect <100 nm rBC particles, the scattering channel is typically limited to optical diameters > 200 nm. What is the particle number loading in the instruments (see reference to coincidence above)

The characterisation of the particle number efficiency of the nebulizer and the daily performance control was performed with monodisperse polystyrene latex (PSL) particles (Postnova Analytics GmbH, Landsberg am Lech, Germany) with nominal diameters of 240 ± 5 nm and 304 ± 5 nm. The particle number concentration within these suspensions is about $3 \cdot 10^8$ 1/mL. A diluted PSL standard suspension sample was prepared daily by pipetting 1 mL suspension into a 100 mL graduated flask filled with Nanopure water. This results in a concentration of typically a few hundred particles per cc at the output of the Marin-5.

Line 291-293: The authors report and discuss the "enhancement" factor of the soot- equivalent mass derived from the photoacoustic spectrometer with decreasing wave- length (1.65, 2.28, and 2.38 at 658 nm, 532, 405 nm, respectively) and conclude that this suggests these samples might contain mineral dust or BrC. The data might be able to say something more concrete. While the authors should conduct a more thorough literature search, mineral dust tends to absorb more in the red then the blue, whereas BrC exhibits the opposite wavelength dependence. The wavelength dependence of light absorption observed in the present study suggests that the non-rBC absorbing species is BrC. Also, it should be noted that BrC can exhibit absorption at the red wavelengths, though, again, the wavelength dependence favors more absorption in the blue (e.g., tar balls: Hoffer, et al., 2017. Brown carbon absorption in the red and near infrared spectral region. Atmos. Meas. Tech. 10(6): 2353–59).

Although we cannot directly measure the dust concentrations in the snow samples, we can draw some conclusion by analyzing the spectral signature of the snow mass specific absorption cross section of the non-BC particles, i.e. after subtraction the spectral cross section that is expected from the SP2 mass data using the MAC of Fullerene soot (Equation

3 of the revised manuscript; see Point 5 above). We added a figure (Fig. 12 in the revised manuscript) that shows a comparison of the non-BC spectral absorption cross section with laboratory absorption data for Saharan dust particles. Thus, Saharan dust is a good candidate to explain the observed non-BC light absorption in the snow samples. We did the same comparison with laboratory data of OC, which reveals that OC could also explain the observed non-BC absorption - not only in terms of wavelength dependence but also in terms of variability.

The whole paragraph on the analysis of the spectral absorption analysis has been rewritten to make our argumentation of a significant influence of non-BC particles on the light absorption in snow clearer.

line 349: "The term brown carbon is not clearly defined or characterized." This is very misleading. Simply put, brown carbon, BrC, is organic aerosol that absorbs light. While the chemical composition for any given BrC aerosol may very, as long as the organic aerosol absorbs light it is cataloged as BrC. Please reword this sentence.

Paragraph rephrased to:

"The term "brown carbon" is mainly related to a strong wavelength dependence of the visible light absorption observed in these materials. From a chemical perspective, BrC can generally be divided into humic-like substances (HULIS) and tar balls (Wu et al., 2016). HULIS can be characterised mainly as a mixture of macromolecular organic compounds with various functional groups and are expected e.g. in oxidation processes of biogenic precursors (Wu et al., 2016). Tar balls are emitted from biomass burning and are of spherical, amorphous structure and are typically not aggregated. Moreover, light absorbing organic material and HULIS can be formed from the water-soluble fraction of biomass burning aerosol compounds, and is therefore suggested as an atmospheric process for the formation of light absorbing BrC in cloud droplets (Hoffer et al., 2004). Further examination of snow samples from different locations as well as systematic investigations on the optical behaviour of biogenic particulate matter is therefore necessary to evaluate the influence of biogenic (including biological), BrC and mineral dust on the aerosol absorption properties in the visible spectral range."

Please provide plot of fullerene standard size distribution.

Added (Fig. 6 of the revised paper).

Editorial comments

There is no reason to put quotes around the word fullerene.

Agreed and changed.

Line 67: "Most Himalayan glaciers as glaciers elsewhere have retreated..." Please insert comma after first occurrence of the word "glaciers" and after "elsewhere".

Introduction has been completely reworded.

Line 311: "To get a general idea of the nature of the components that are solved and dissolved within the snow samples ionic chromatography . . ." the word "solved" does not seem right here. Please check and correct as necessary.

Reworded to:

"To further examine the nature of the particulate components that are deposited in the snow samples ion chromatography (IC) … "