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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Specifying light absorbing properties of aerosol particles in fresh snow samples, collected at the Environmental Research Station Schneefernerhaus (UFS), Zugspitze" *by* Claudia Linke et al.

Anonymous Referee #3

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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 parti-

cles - 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 But in its current state, the manuscript is not ready and thus is not recommended for publication at this time.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 $\sim$ 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.

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 $\sim$ 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 $\sim$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 $\sim$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.

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.

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?

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)

Line 291-293: The authors report and discuss the "enhancement" factor of the soot-equivalent mass derived from the photoacoustic spectrometer with decreasing wavelength (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).

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.

Please provide plot of fullerene standard size distribution.

Editorial comments

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

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

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.

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