# Effects of mixing state on optical and radiative properties of black carbon in the European Arctic

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. While the referee comments are given in **black bold,** our answers are given below in blue letters. Additionally, we added the changes we made in the revised manuscript in **blue bold** letters.

**Answers of the authors to anonymous Reviewer #2**

Anonymous Review of Manuscript acp-2018-45 GENERAL REMARKS

This paper uses in situ observations of black carbon from a surface site in the arctic to explore the mixing state and optical properties and then carry this data forward into a radiative transfer model to explore sensitivities. Given that the arctic is a very sensitive region in this regard and not very well studied, this is an important piece of work and well within ACP's remit. While the results aren't particularly dramatic, given the current uncertainties, it is vitally important that these treatments are given an observational basis. The paper is generally very well written in terms of language. I have only a few reservations, but these are mainly of a general nature and will not likely affect the paper's conclusions, therefore I recommend publication after minor revisions.

Specific comments of Reviewer#2

**When comparing with other arctic SP2 datasets, DOI: 10.5194/acp-15-11537-2015 will likely be of relevance, as this made airborne observations in a similar region.**

The reference of Liu et al. (2015) was implemented in the text in Section 3.2.1. Now the text reads:

**[…] Recently, three studies also used the SP2 to investigate BC in the Arctic. Raatikainen et al. (2015) reported a mean rBC mass concentration of 26 ng $m^{-3}$ at the Pallas Global Atmosphere Watch station (68°N, Finland) during winter 2011-2012. Taketani et al. (2016) investigated the spatial variability of rBC at sea level between the Northern-Pacific and Arctic oceans during September 2014. At latitudes higher than 75°N the rBC mass concentration was highly variable (0-60 ng $m^{-3}$ on one-minute time resolution), with an average of 1 ± 1.2 ng $m^{-3}$. In addition, Liu et al. (2015) presented rBC measurements performed in the low and middle troposphere in the European Arctic in spring 2013, when the rBC mass concentration varied between 20 and 100 ng $sm^{-3}$. While observations in the present study are similar to those by Raatikainen et al. (2015) and Liu et al. (2015) the much lower rBC mass concentrations reported by Taketani et al. (2016) can most likely be attributed to the facts that they sampled a different season and that an intense stagnation event occurred over Svalbard in 2012. The rBC mass size distribution peaked at $D_{rBC}$ =240 nm with 33% of rBC mass in the BC core diameter range $D_{rBC}$ =200 - 300 nm (Figure3a; Table 2). This is similar to BC core sizes reported from previous observations in the Arctic region (Liu et al., 2015; Raatikainen et al., 2015; Taketani et al., 2016), […].**

**Generally, there is a tendency in this paper to not introduce certain variables properly. One example would be the various definitions of RF, but there are others. The authors should take care to make sure that each variable or parameter is properly defined when it is first used, in particular the use of subscripts.**

The complete and proper definition of all variables, acronyms and abbreviations was verified:

- The acronym PRF, standing for sun precision filter radiometer, was removed.
- The abbreviation $m_{rBC}$ is now introduced in Section 2.2.1. The original sentence was modified accordingly:
  **[…] Hereafter we follow the recommendation of Petzold et al. (2013) and use the term refractory black carbon (rBC) whenever referring to black carbon properties quantified with laser-induced incandescence and use the term $m_{rBC}$ whenever referring to the refractory black carbon mass concentration […]**.
- The acronym for standard deviation (SD) is introduced in Section 3.1 as:
  **[…] The AOD was 0.097 on average with a standard deviation (SD) of 0.022 […].**
- The meaning of $D_{Shell}$ is now explicitly defined in Section 3.2.3:
  **[…] Alternatively, the BC mixing state can be expressed as the ratio of the total diameter of the BC-containing particle ($D_{Shell}$) over the diameter of the BC core ($D_{rBC}$ for SP2 measurements and $D_{Core}$ for optical calculations), and univocally called shell-to-core diameter ratio ($D_{Shell}/D_{Core}$). […].**
- Following the previous point, the statement now reads:
  **[…] This is a result of relatively large size parameters approaching the geometric optics regime in which MAC $\propto D_{Core}^{-1}$ […]**.
- $AOD_{Obs}$ is now properly defined in Section 3.4.1:
  **"[…] the observed AOD ($AOD_{Obs}$) […]"**.

The nomenclature of radiative forcing and related terminology is addressed in the following comment.

**The authors refer to 'radiative forcing', but this is inconsistent with the IPCC definition of the term, which is the anthropogenic perturbation compared to a preindustrial base case. I would recommend that the authors refer to this differently, for instance 'radiative contribution'**

The reviewer raised an important nomenclature issue, which was already topic of discussion in previous publications (Bond et al., 2013). Lacking a better alternative, we kept the term "radiative forcing", while explicitly explaining in how far our term agrees with / differs from the IPCC definition.

- Text in Section 2.4.2 now reads:
  **[…] Note, this definition of radiative forcing agrees with the definition by Stamnes et al. (2017) whereas it differs from the definition of the Intergovernmental Panel on Climate Change (Myhre et al., 2013). More details on this topic will be provided in Section** Error! Reference source not found.**. […]**
- Text in Section 3.4.2 was modified accordingly:
  **[…] $\Delta RF_{ARI}$ specifies the absolute change of the aerosol radiative forcing compared to our best guess scenario in order to quantify the effect of the absorption enhancement induced by different mixing degrees of BC (bare BC, thin, medium and thick coatings) on the $\Delta RF_{ARI}$. Our definition of radiative forcing refers to the gross effect on the radiative balance at present-day conditions, similar to Stamnes et al., (2017); but it differs from the definition by the IPCC (Myhre et al., 2013), which refers to the net effect on the radiative balance induced by anthropogenic emissions with using pre-industrial aerosol loadings as reference. However, these definitions become essentially identical for the radiative forcing by BC under the assumption that present-day BC concentrations in the Arctic region are much greater than corresponding preindustrial values. If this assumption was not applying, then our results for $\Delta RF_{ARI}$ by BC would need the anthropogenic BC mass fraction at present-day as additional scaling factor to make them consistent with the IPCC definition. […]**

**The authors use Mie to model the optical properties of the aerosol, but this is based on Mie-based data of the SP2 LEO inversion, which may suggest circular reasoning. However, I would consider this legitimate; Liu et al. (2017) present an experimental case that this applies for the 'thickly coated' particles observed by the SP2. I would suggest that the authors refer to this work to justify their method.**

We thank the reviewer for this suggestion. We now put the degree of coating in context of the findings by Liu et al. (2017):

**[…] More recently, Liu et al. (2017) confirmed that Mie theory with assuming spherical core-shell geometry realistically describes the optical behavior of embedded BC cores when the coating mass is greater than around three times the mass of BC core. In our work, the volume of the coating material was converted to mass using a density of 1100 kg $m^{-3}$, similar to Liu et al. (2017). On average, a factor of 4.15 was found between the mass of coating and BC core, supporting the assumption of core-shell as mixing geometry for our optical simulations. […] .**

**The authors used an Aethalometer to derive the absorption coefficient, but this is based on an assumed C value during the correction process. The authors should comment on the presumed accuracy of this.**

The technique and data treatment of the aethalometer is reported in Section 2.3. The authors agree with Reviewer #2, on the importance of the *C*-value (*C*) in absorption determination via filter-based photometry. We extended the discussion of uncertainties related to assumptions on the C-value:

**[...] The *C*-value, which depends on aerosol properties in a complex manner, can be determined if the true absorption coefficient is known. As no absorption reference instrument is available in this study, we use a fixed *C*-value of 3.1 as determined by Backman et al. (2016) for aethalometers operated at multiple low-elevation sites in the Arctic region[1]. The applied C-value of 3.1 might not represent the actual *C*-value at the Zeppelin site during our campaign, thus potentially causing systematic errors. Choosing a *C*-value of 3.5 instead, as recommended by the Global Atmosphere Watch Program based on data from multiple European background sites (WMO, 2016; Zanatta et al., 2016), would result in a systematic reduction of resulting absorption coefficients and MAC-values by 13%. Furthermore, the variability of the *C*-value at the European background sites was reported to be ±25% around the recommended mean. Assuming that this range matches the potential variations of the actual *C*-value at the Zeppelin site, provides uncertainties of -29%/+18% associated with the aethalometer derived absorption coefficients. […].**

**[1] Note, an updated C-value of 3.25 specific to the site and instrument used in this study is provided in the final published manuscript by Backman et al. (2017). However, the difference between these two values is well within uncertainty)**

**Furthermore, they use the Virkkula method to correct for the loading artefact, which is useful when there are no collocated measurements of BC or absorption, however, given that there is collocated SP2 data, the (more accurate) Weingartner method (or the improved method in DOI: 10.5194/amt-3-457-2010) may be possible. The authors should justify the use of the Virkkula method better.**

There are a few algorithms introduced for compensating the aerosol loading effect on filter based absorption measurements. The authors have used more than one method including the Weingartner method (Weingartner et al. 2003) and the Virkkula method (Virkkula et al., 2007) for sensitivity

analysis among those included in the DOI: 10.5194/amt-3-457-2010 article cited by the reviewer. It is admitted that the reference to Virkkula et al. (2007) is not needed, because the final data were actually treated with the Weingartner method. This reference was accidentally introduced in a revised version of the manuscript and is now removed. It is maybe relevant to add that the loading effect has a minor correction impact (< 3%) in arctic absorption data for the higher wavelengths employed here. This section becomes:

**[…] where *R* accounts for the loading correction as a function of attenuation (Weingartner et al., 2003). Although applied here, the loading correction is not a significant source of uncertainty for absorption determination at long wavelengths and observations in the Arctic (Backman et al., 2017). […]**

**The authors refer to an SP2 method to determine 'attached' particles but do not adequately describe it. They should give more detail here. Also, anecdotal evidence suggests that this indicator is not consistent between instruments, so it would be useful to demonstrate that the unit used here is capable of detecting these.**

Potential inconsistency between instruments can indeed be a critical issue and warrants attention. It would be important to know whether this quoted "inconsistency" must rather be associated with inconsistency of data analysis approaches and/or selection of particle size ranges considered, or indeed with true intrinsic imprecision of this type of information retrieved from SP2 signals. Anyway, the main reason for looking into this was to exclude presence of a substantial fraction of BC particles with "attached type" coatings, which would result in larger errors in the SP2 based coating thickness analyses and the Mie calculations which are based on "embedded type" morphology. We removed the comparison of the number fraction of attached type BC found in this study, with corresponding number fractions reported in the literature as this is not central to our manuscript. Sect. 3.2.3 now reads:

**[…] The SP2 makes it possible to distinguish two distinct types of particle morphology for individual internally mixed BC particles (Sedlacek et al., 2012; Dahlkötter et al., 2014; Moteki et al., 2014): i) BC is only a minor volume fraction and fully embedded in the coating material somewhere near the particle center, and ii) BC is attached to or at least near the surface of the coating material. We used the method introduced by Moteki et al. (2014) to show that at Svalbard, where the dominant fraction of BC-containing particles was found to have a small BC volume fraction, only around 2% of the particles containing BC-cores in the mass range of 6-10 fg exhibited the SP2 signal features corresponding to the attached geometry. While the exact value is subject to uncertainty, it is a robust result that the embedded type morphology clearly dominates over the attached type morphology for the BC particles. The fact that the dominant fraction of BC particles has substantial coatings with embedded type morphology supports using the simplified assumption of concentric core-shell geometry for inferring the mixing state based on SP2 data and for estimating the effect of the coatings on particle properties. […]**

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