We thank Reviewer 2 for the constructive comments and suggestions. We have addressed all comments in our response and feel these revisions improve the manuscript. The reviewer comments are copied here in italics, our response is in plain text, and revised statements from the manuscript are underlined. We also submit a "marked up" version of the revised manuscript.

Reviewer 2

The manuscript presents a case study examining radiative forcing by black carbon in the Arctic and its sensitivity to assumed mixing state. It uses observations made by an SP2 (black carbon and associated coatings) and a UHSAS (all aerosol) to constrain mixing state applied in the GEOS-Chem-TOMAS model and examines response of the calculated direct radiative effect. They find different mixing state assumptions lead to differences in DRE on the order of 0.3 W m-2, with observationally-constrained values falling within two bounding cases (complete external versus internal mixtures). The analysis is thorough and well within the scope of ACP, and will be of value to the community. I recommend its publication once the following minor points have been resolved.

General comment

Both instruments used in this study are optically based, so they have a refractive index dependence. There should be a little more discussion on the uncertainties related to converting the optical measurements to a size comparable with the model that will be used in the DRE calculations. Further, an assumed RI is provided for the SP2 coating analysis. Could the UHSAS measurements be adjusted to have an RI consistent with this assumption (e.g., 1.5 for coating species applied to all non-rBC containing particles?

The referee raises important points here. To address the referee's comments on the uncertainties related to converting optical measurements to size, we have added additional information to Sections 2.2.2 and 2.3. In these sections, we discuss the relative impact of the refractive indices used for the rBC coating material and the UHSAS measurements. While these do affect the results, their impact is minor in comparison to the refractive index used for the rBC core.

Section 2.2.2, paragraph 2 has been modified to read:

With the scattering amplitude determined by the leading-edge technique, and the measured rBC core diameter, a core-shell Mie model can be used to determine the optical diameter of the rBC-containing particles. In the Mie model we used a refractive index of 2.26–1.26i for rBC (Moteki et al., 2010), which is appropriate for rBC from fossil fuel combustion. Other studies have used this value in analyses of particles from urban areas (Laborde et al. 2013) and from biomass burning (Taylor et al. 2014). In the Mie model we used a refractive index of 1.5-0.0i for the coating species. The value of 1.5-0.0i is appropriate for dry sulfate and sodium chloride (Schwarz et al., 2008a, 2008b). Previous studies have shown that varying the refractive index

used for the rBC coating material from 1.45 to 1.59 had a minor impact on coating thickness (Taylor et al. 2015).

The following has been added to Section 2.3, paragraph 1:

The UHSAS was calibrated using polystyrene latex spheres (PSLs), which have a refractive index of n=1.59. Kupc et al. (2018) investigated the size dependence of UHSAS measurements on the assumed refractive index of the particles being measured. For particles with diameters less than 600 nm and real refractive indices of n= 1.44–1.58, the diameter measured by the UHSAS varied by +4/−10% (relative to the refractive index of ammonium sulfate, n=1.52). For this study, although significant, a shift in the UHSAS size distribution by 10% would change the fraction of particles containing rBC by less than 3% which has a negligible impact on the DRE calculations.

Specific comments

Page 5, 16-22 - Were any laboratory tests performed to verify the lower limit for detection or is this number based on literature values?

We have added the following to address the reviewers comment.

Both SP2s were calibrated against size selected external BC standards. The lower limit for mass was set at a point where detection efficiency for particles compared to a CPC was close to 100%.

Page 7 - some mention of refractive index impacts on sizing for both instruments (and a statement regarding how consistent the refractive index assumptions are between the two instruments (SP2 and UHSAS) is needed here.

As discussed above, we have added statements discussing the refractive index used for the UHSAS to Section 2.3, and statements discussing the refractive indices used for the SP2 to Section 2.2.2.

Section 3.4 - While it may be obvious to most readers, I think it is worth pointing out the limitation of coating information being available only for a subset of the BC particles in this section in addition to the other limitations listed.

This is certainly a worthwhile point to emphasize. We have added the following statements to this section:

Fourth, our measurements of BC mixing state only apply to BC particles in a limited size range. The size distributions in Figure 4 imply a substantial fraction of BC and non-BC number concentration exist outside the size range of our measurements. We attempt to account for this limitation in our model analysis through consideration of upper and lower bounds on coating and BC-containing particles

Figure 2 - I was a little confused by the wording in the caption: "rshell-constrained mixing state used SP2 measurements of BC core diameter and shell thickness to constrain BC mass". My understanding was that BC mass was always taken from the TOMAS simulation output. Does the caption mean BC mass per particle? Please clarify

We agree this caption was worded poorly. Reviewer 2 is correct, we did intend for this to read "BC mass per particle"; however, this seems unnecessary and confusing. We have re-written this sentence to be more direct:
The r_{shell}-*constrained* mixing state uses SP2 measurements to constrain BC core diameter and shell thickness.

Figure 5 caption - should be "fraction of BC aerosol mass relative to total aerosol mass"? Also, could you provide an "average level" for the observations based on typical flight levels during the study? Same for Figure 6 as well?

Yes, thank you, we have corrected this sentence.

Overall, we do not find substantial vertical variability in our measurements of BC mixing state. The three model layers provided in Figures 5 and 6 is meant to show the respective comparisons across the relevant range of altitudes from the measurements (shown in Figure 1). To this end, we have replaced the 720 hPa layer with the model layer corresponding to 550 hPa. The vertical range from the surface to 550 hPa encompasses much of the altitude ranges from the POLAR6 flights. Given that the model does not overlap the observations over this range, we do not feel an average level is helpful in this instance. Further, this may mislead readers into thinking we sampled the model along the flight track which is not the case.

Revisions to manuscript:
In Figures 5 and 6, we replace the 720 hPa level with 550 hPa (see revised submission). We make minor changes to the text, replacing references to 720 hPa with 550 hPa.