We thank Cenlin He for suggesting additional references and suggesting areas requiring further discussion. The suggested references are quite relevant and helpful to the paper. We have responded to each of the points below and have copied our added text (in underlined font).

The authors used the SP2 and UHSAS measurements of BC mixing states to provide important observational constraints for model estimates of DRE over the Arctic. This work provides valuable information to advance the current understanding. I have a few comments/suggestions.

1. Previous observations (China et al., 2015) have shown a large variety of non-core-shell BC coating/mixing structures/morphology. Recent modeling studies (e.g., He et al., 2015, 2016) further indicated a substantial variation (up to a factor of 2) in BC optical properties by using those different coating/mixing structures/morphology. The authors used a core-shell assumption for internally mixed BC, which may lead to some uncertainty. I suggest including these recent studies and add some discussions on this coating/mixing structure issue.

References:

China, S., et al.: Morphology and mixing state of aged soot particles at a remote marine free troposphere site: Implications for optical properties, Geophys. Res. Lett., 42, 1243–1250, doi:10.1002/2014gl062404, 2015.

He, C., et al.: Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison, Atmos. Chem. Phys., 15, 11967-11980, doi:10.5194/acp-15-11967-2015, 2015.

He, C., et al.: Intercomparison of the GOS approach, superposition T-matrix method, and laboratory measurements for black carbon optical properties during aging, J. Quant. Spectrosc. Radiat. Transf., 184, 287–296, doi:10.1016/j.jqsrt.2016.08.004, 2016.

Thank you for suggesting these citations. We agree it is important to be quite clear on our assumptions regarding BC morphology and have added several statements discussing this assumption. In this study, we seek to constrain only the population mixing state and leave particle morphology to future work. In the introduction of the paper, we attempt to distinguish two types of mixing state: chemical mixing state and morphological mixing state. The former we constrain through observations and the latter we retain the core-shell assumption. We have added the following sentences to our introductory paragraph on morphology to discuss the observed variability in structure and impacts on absorption:

<u>At a remote observation site, China et al. (2015) found substantial variability in the fractal dimension and structure of mixed (i.e., fully encapsulated versus partly encapsulated) BC</u>

particles. Despite this variability, a common assumption for the morphological mixing state of BC is…

However, the degree of absorption enhancement is a strong function of the structure and geometry of the mixed particle as well as the core diameter and shell thickness (He et al., 2015; He et al., 2016).

We have added the following statements to state our measurements only constrain population mixing state:
We use these measurements as constraints on the population mixing state to estimate the direct radiative effect (DRE) in the springtime Arctic, and compare these estimates to the DRE calculated using bounding cases of completely external or internal mixing state-assumptions. Note, these measurements do not allow us to constrain the morphological mixing state.

In Section 2.5.3, where we discuss mixing states, we have added the following sentence explicitly stating that we constrain only the population mixing state:
These measurements constrain only the population mixing state. For cases of mixed BC, we assume an ideal core-shell mixture, but note morphological mixing state is an important uncertainty (China et al., 2015).

In Section 3.4, where we discuss study limitations, we state morphology as a limiting assumption. We have added the citation to China et al. (2015) here as well:
First, the measurements described here constrain only population mixing state. With regard to particle morphology, we assume a core-shell configuration with BC at the exact center of the particle. Several studies have suggested this may not always be representative of atmospheric aerosol (e.g., Cappa et al., 2012; China et al., 2015).

2. Page 3, Lines 13-15: For the authors' information, a recent study (Qi et al., 2017a) also used the GEOS-Chem adjoint model to investigate the BC source contributions to the Arctic, which included most recent BC emissions (e.g., gas flaring, small-fire biomass burning, anthropogenic). This recent study is a useful reference that can be cited here.

References:

Qi, L., et al.: Sources of Springtime Surface Black Carbon in the Arctic: An Adjoint Analysis for April 2008, Atmos. Chem. Phys., 17, 9697-9716, doi:10.5194/acp-17- 9697-2017, 2017a.

Yes, thank you, this is a very helpful citation for this paragraph. We have added a citation to Qi et al. (2017) to the list of citations in Line 16. We have also added the following sentence to the same paragraph:

Similarly, Qi et al. (2017) found BC concentrations in the Arctic in April 2008 to be largely from anthropogenic sources in Asia and biomass burning sources in Siberia.

3. Section 2.5.1: recent studies (Qi et al., 2017b,c) have improved BC dry and wet deposition schemes in GEOS-Chem model, which leads to better BC simulations over the Arctic. The authors did not describe the BC dry and wet deposition scheme used in the present study, which are key factors influencing BC simulations. I suggest adding some descriptions on BC deposition scheme used in the model and also including some discussions associated with the aforementioned recent improvements.

References:

Qi, L., et al.: Factors controlling black carbon distribution in the Arctic, Atmos. Chem. Phys., 17, 1037-1059, doi:10.5194/acp-17-1037-2017, 2017b.

Qi, L., et al.: Effects of the Wegener-Bergeron-Findeisen Process on Global Black Carbon Distribution, Atmos. Chem. Phys., 17, 7459-7479, doi:10.5194/acp-17-7459- 2017, 2017c.

We suspect that the dry and wet deposition schemes are likely the cause of the lower BC mass concentrations in GEOS-Chem-TOMAS relative the observations. The deposition scheme for GEOS-Chem-TOMAS is different from that in the standard GEOS-Chem model. As a measurement-model comparison is not the focus of this paper, we leave detailed descriptions of the aerosol microphysical routines in TOMAS to other publications (and we cite them in the relevant section). We have, however, added a sentence to Section 3.2 discussing Qi et al. (2017b,c) and the importance of wet and dry deposition:
The lower BC mass concentrations may be due to the representation of dry and wet scavenging in TOMAS, which does not include recent updates described in Qi et al. (2017b,c).