Response to Dr. Bahreini's comments

We really appreciate the editor Roya Bahreini's comments on our manuscript which help improve the robustness of our conclusions. Following the suggestions, we have revised the manuscript accordingly. Listed below is our point-to-point response in blue to each comment.

1. With the MRS approach, the inherent assumption is that the sources of BC and BrC are different. However, in the real world, many of the processes that emit BC directly also emit BrC or its precursors, so in reality there is a correlation between BC and BrC (even if you assume BrC is only secondary). In this case, how valid is the interpretation of the MRS approach?

We agree with the editor that many processes emit both BC and BrC, for instance, biomass burning. The directly emitted BrC is defined as primary BrC in our study. The MRS approach is mainly used to differentiate the secondary BrC from primary BrC which has been reported in previous studies (Wang et al., 2019b; Zhu et al., 2021). Based on the investigation of MRS method in a previous study (Wu and Yu, 2016), the key of this method is to determine an appropriate $b_{abs,BrC}$/eBC ratio that represents primary combustion emission sources, i.e., $(b_{abs,BrC}/eBC)_{pri}$ at the observation site. The hypothetical $(b_{abs,BrC}/eBC)_{pri}$ that generates a minimum R^2 (secondary BrC vs. eBC) represents the actual $(b_{abs,BrC}/eBC)_{pri}$ ratio if the variations of eBC and secondary BrC are independent and $(b_{abs,BrC}/eBC)_{pri}$ is relatively constant during the study period. Wu and Yu (2016) used numerically simulated data to evaluate the accuracy of estimated secondary components by the MRS method and also compared with the results from two commonly used methods. The bias of MRS result is <23% when the measurement uncertainty is within 20%. In addition, the MRS method can reduce the uncertainty caused by the different arbitrary selection criterion of $(b_{abs,BrC}/eBC)_{pri}$. Therefore, previous studies suggested that the MRS method is a useful method to distinguish primary and secondary components.

One assumption of the MRS method is that the sources of BC and secondary BrC are different although the sources of BC and primary BrC can be the same. This assumption might have some uncertainties in remote environment where BC and secondary aerosol are both from long-range transport. However, in urban atmosphere, secondary organic aerosol (SOA) that is mainly from the oxidation of volatile organic compounds (VOCs) (Hallquist et al., 2009) often exhibits largely different variations from primary organic aerosols, such as biomass burning OA, traffic-related OA, and coal combustion OA (Cappa et al., 2019; Xu et al., 2021). As part of the organic carbon, secondary BrC in this study is also expected to show different variations with primary components. To verify the results of secondary BrC absorption, we further calculated the secondary BrC absorption at 470 nm, 520 nm, 590 nm and 660 nm with the MRS approach, and then determined the AAE of secondary BrC. As shown in Figure R1, within the 10th to 90th percentiles, the AAE distribution of secondary BrC in our study ranged from 2 to 8, which is consistent with the AAE values of secondary organic carbon reported in

previous studies (Cheng et al., 2016; Qin et al., 2018; Kasthuriarachchi et al., 2020; Soleimanian et al., 2020; Kaskaoutis et al., 2021). Based on the above analysis, we concluded that the MRS approach is reasonable to determine the secondary BrC in urban area although there may have some uncertainties we cannot quantify now.

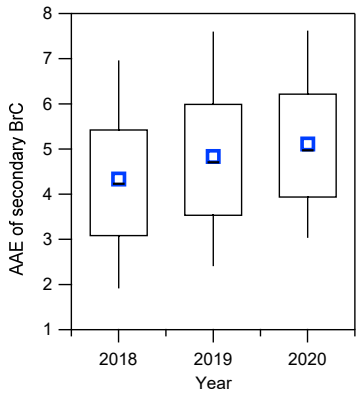

Figure R1. Annual variations of AAE of secondary BrC. The median (horizontal line), mean (markers), 25th and 75th percentiles (lower and upper box), and 10th and 90th percentiles (lower and upper whiskers) are also shown.

2. In the calculation of the radiative forcing values, constant back scatter fractions were assumed. However, the backscatter fraction is aerosol size and shape dependent. Please justify use of the constants here.

According to previous studies in Beijing, we found that BC exhibited an approximately lognormal size distribution with the median diameter about 170~210 nm, and the size distribution also showed some differences in different seasons (Liu et al., 2019; Yang et al., 2019; Liu et al., 2020b). Therefore, we did not analyze the variation of ΔF_R in different seasons in our study, instead, we focus on the changes of ΔF_R in the same season when the changes in size distribution of BC from 2013 to 2019 was small (Figure R2) (Wu et al., 2021), and the difference in annual median radius difference was less than 20 nm. Such a small difference affected the backscatter fraction of BC less than 0.04 (Chylek and Wong, 1995). Liu et al. (2017) found the difference of asymmetry factor (g) below 0.01 when the range of BC particle diameter was at 150~250 nm. Considering g as a function of backscatter fraction (Sviridenkov, 2015; Horvath et al., 2016), we further calculated the difference of BC particle backscattering fraction at 170 ~ 210 nm, and it was below 0.005. Therefore, the uncertainty of backscatter fraction caused by same change in size distribution of BC could be negligible in our study.

Besides, as the editor mentioned, the backscatter fraction also depends on BC particle shape. However, the effect of BC shape on the radiative effect is likely much lower than that caused by the large decrease (71%) in mass concentration. In previous studies, the

key parameter to represent the shape of BC particles is fractal dimension (D_f) (China et al., 2013). Previous studies found that most D_f of BC particles from biomass burning and fossil fuel combustion was at 1.7~2.2 (Wei et al., 2020). According to the relationship between D_f, BC diameter and g in previous study (Liu et al., 2017), the maximum g difference of BC particles was at ~180 nm in the range of BC particle diameters of 150 ~ 250 nm (median mass diameter in recent years). At the same time, the range of g was only 0.47~0.45, corresponding to a D_f of 1.8~2.8, respectively. We then calculated the backscattering fraction according to the relationship with g (Sviridenkov, 2015; Horvath et al., 2016) and found that the difference in the backscattering fraction was about 0.012.

Therefore, we considered these uncertainties to be acceptable and chose an appropriate backscatter fraction (Charlson et al., 1992; Chylek and Wong, 1995) which could fit the usual size distribution of BC in Beijing (Schwarz et al., 2008; Liu et al., 2019; Liu et al., 2020a; Liu et al., 2020b; Wu et al., 2021). To clarify this, we added the description about the backscatter fraction (β) as follows:

"Based on the small differences in mass size distribution of BC in recent years (Wu et al., 2021) and little effect of BC particle shape on backscatter fraction (Liu et al., 2017), we used a constant value of β (0.29) (Charlson et al., 1992; Chylek and Wong, 1995) which could be adapted to the usual size distribution of BC in Beijing (Liu et al., 2019; Liu et al., 2020a; Liu et al., 2020b; Wu et al., 2021)."

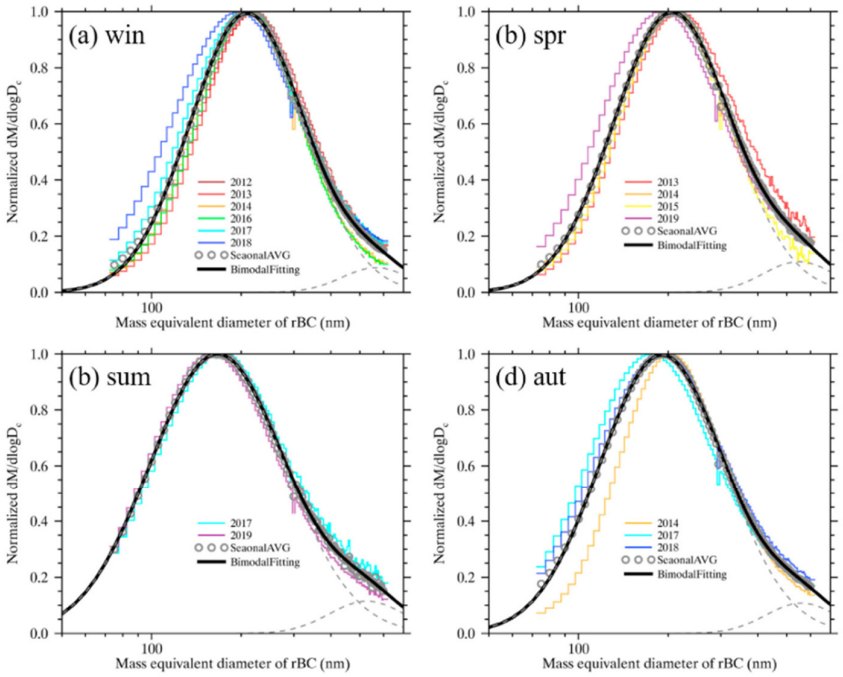

Fig. 1. Mass size distributions of rBC ($dM/d\log D_c$) in urban Beijing in (a) winter, (b) spring, (c) summer, and (d) autumn normalized by the maximum $dM/d\log D_c$ values of respective distributions. The dashed gray lines represent the two modes combining the bimodal fitting.

Figure R2 (Wu et al., 2021).

3. Lastly, the SSA is also a function of aerosol size. With aerosol growth but under fixed composition, SSA increases. Therefore, a change in SSA alone cannot be taken as an indicator of a relative change between the contribution of the absorbing and non-absorbing species. Are there any measurements of aerosol size distributions that you can use to examine changes in SSA as a function of size parameter to be able to conclude any changes in the composition? If not, please elaborate on this caveat in the discussion.

We agree with the editor that aerosol optical properties are also related to particle size distributions in addition to chemical composition. As shown in Figure R3, the differences in aerosol volume size distribution from 2013 to 2019 in Beijing (available from the Aerosol Robotic Network data archive) are relatively small. Generally, the aerosol volume concentration exhibits a bimodal distribution and the first peak often occurs at ~200 nm except 2014 and 2019. These results indicate that the annual variation of aerosol optical properties (SSA and MEE) in recent years is more likely caused by the change in aerosol chemical components when the changes in aerosol particle size is relatively small.

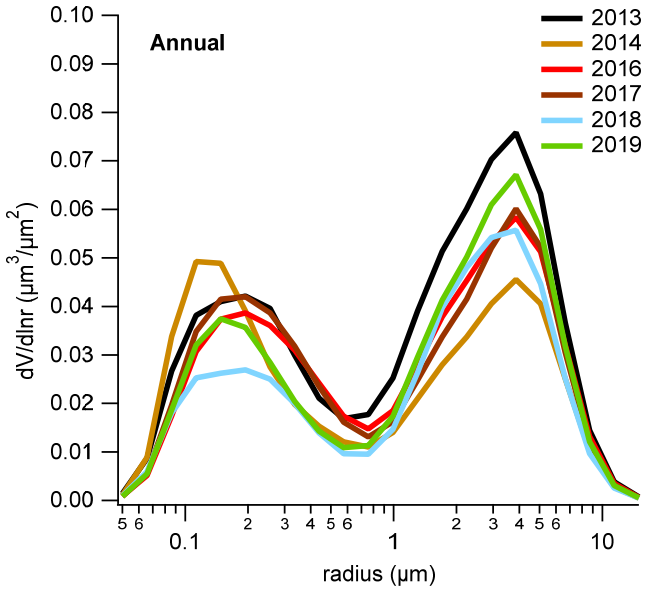

Figure R3. Annual variations of aerosol volume size distribution in Beijing.

Previous studies also found that the aerosol volume size distributions showed seasonal variations in Beijing (Zheng et al., 2020). Following the editor's suggestion, we added an analysis of aerosol volume size distribution variation in different seasons (available from the Aerosol Robotic Network data archive). According to Figure R4, we found that the particle radius corresponding to the first volume concentration peak was the same at 200 nm in the fall of 2013 and 2018, while it became smaller in 2019. These results indicate that the increase in SSA in fall before 2019 was mainly due to the changes in aerosol chemical composition. Differently, the particle size in summer showed a yearly increasing trend from 2013 to 2018, and decreased in 2019 and

increased again in 2020. In spring in 2014 and 2015, higher volume concentrations occurred at larger radius (~180 nm) than those in other years. However, in winter the particle radius corresponding to the first volume concentration peak was larger in 2012 and 2018 (~200 nm) than other years. Because the measurements of particle size distributions were not available in this study, those derived from the Aerosol Robotic Network were mainly used to indicate the uncertainness in analyzing aerosol optical properties.

Following the editor's suggestion, we expanded the discussions of aerosol optical properties in the revised manuscript. In addition, we added the Figures R4 and R5 in supplementary to let readers know the uncertainties in this study.

The revised part in section 3.2 are as follows:
"In particular, the annual mean MEE and SSA increased despite the decreases in eBC and b_{ext} and the relatively stable variations in particle size distribution (Fig. S7) in the past decade, which indicated that scattering aerosol species played more important roles than absorbing aerosol species in radiative forcing."

"While the increase of SSA in fall was mainly due to the changes in aerosol chemical composition such as the decrease in BC, the increase of SSA in summer was also likely affected by the increase in aerosol particle sizes. These results were supported by the similar particle size (~ 200 nm) in fall, while a clear increase in summer from 2013 to 2018 (Fig.S8). Note that the particle size in summer 2019 was smaller than that in 2018, yet the seasonal mean SSA was higher in 2019, suggesting that the changes in aerosol chemical components contributed significantly to the increased SSA."

"Considering the increased SSA and particle size (Fig. S8) yet relatively constant mass concentrations of eBC during 2017 ~2019, we inferred that the increased light extinction in winter was mainly caused by scattering aerosols that can vary substantially in different years due to the changes in meteorological conditions (Zhou et al., 2019)."

"The diurnal cycles of b_{ext} and SSA in four seasons are shown in Fig. 4. Because the diurnal variations of particle size distributions are not available in this study, our discussions are mainly focused on the influences of chemical components."

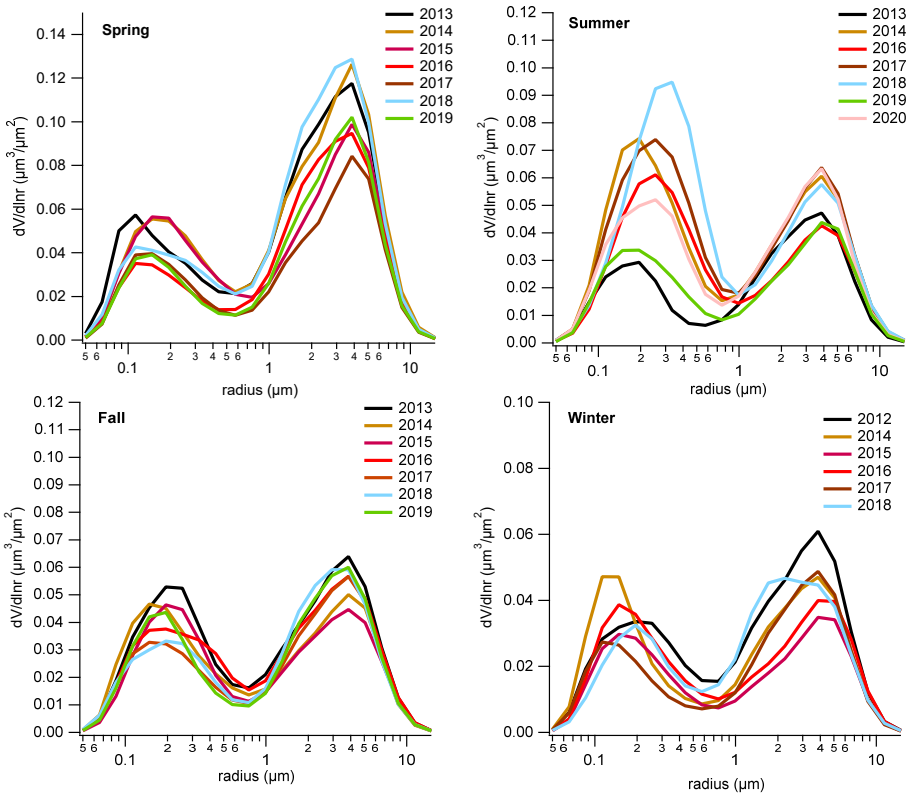

Figure R4. Seasonal variations of aerosol volume size distribution in Beijing.

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