

Point-by-point responses to the comments & suggestions from the editor and reviewers

Journal: Atmospheric Chemistry and Physics (ACP)

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Title: “Long-term evolution and effects of primary brown carbon aerosol in China”

We appreciate the comments from the editor and reviewers on this manuscript. We addressed the comments and suggestions carefully and made necessary changes as shown in the marked revision of manuscript. The detailed point-by-point responses are listed below in blue and modifications in the manuscript are in red.

Comments from the reviewers:

Reviewer #1:

This manuscript investigates the long-term sources, distributions, and impacts of primary brown carbon (PBrC) aerosol in China using a modeling framework with multiple PBrC sensitivity scenarios. The topic is important because brown carbon remains a major uncertainty in aerosol radiative effects, especially in regions with strong anthropogenic emissions. The study attempts to quantify contributions from different emission sources and evaluate uncertainties associated with optical properties, wildfire emissions, and aging processes. The manuscript has several strengths. The authors consider multiple PBrC scenarios, including a no-PBrC case, a baseline PBrC case, a no-wildfire case, a sensitivity case for the imaginary refractive index, and an OH-aging case. The authors also acknowledge several important uncertainties, including the exclusion of secondary brown carbon, fixed PBrC-to-OC ratios, and possible overestimation of wildfire emissions in FINN.

However, several aspects require clarification or revision before the conclusions can be fully supported. In particular, the scenario definitions and differencing methods need to be made clearer and more internally consistent. The model evaluation remains indirect for PBrC, relying mainly on OC and other aerosol-related constraints, and the

implications of this limitation should be discussed more explicitly. The uncertainty treatment should also be strengthened, especially for optical properties, emissions, aging, and the exclusion of secondary brown carbon. Therefore, I recommend “major revision” .

Major Comments

1: The manuscript defines five scenarios: No_PBrC, PBrC_baseline, PBrC_nowldf, PBrC_k_adj, and PBrC_age. This scenario design is useful, but the explanation of how differences between scenarios are used to quantify effects needs revision. For example, the manuscript states that anthropogenic contributions were obtained from “PBrC_baseline minus PBrC_nowldf”. Since PBrC_nowldf is described as the baseline case excluding wildfire emissions, this difference appears to represent the wildfire contribution, not the anthropogenic contribution. If the authors intend to quantify anthropogenic contributions, the no-wildfire case itself may be closer to the anthropogenic component, depending on the treatment of other natural sources. Please clarify and correct this wording or methodology. Similarly, the manuscript states that the impacts of k variability were estimated from “PBrC_k_adj minus PBrC_nowldf”. This comparison appears to conflate two differences: optical property changes and wildfire emission changes. To isolate the effect of k variability, PBrC_k_adj should presumably be compared with PBrC_baseline, unless there is a specific reason to compare it with the no-wildfire case. Please clarify the rationale and ensure that each scenario difference isolates only the intended factor.

Response: We greatly appreciate you for pointing out this important typographical error in our manuscript regarding the scenario descriptions. Your understanding is correct: we indeed evaluated the wildfire impact and the sensitivity of k exactly as your description. Specifically, to quantify the wildfire contribution, we used the differences of “PBrC_baseline minus PBrC_nowldf”, as illustrated in Figure 5. For the assessment of the k effect, we used the differences of “PBrC_k_adj minus PBrC_baseline”, since the k-adjusted scenario was directly derived from the baseline case. We sincerely apologize for this writing oversight. We have now corrected the descriptions in the

manuscript and re-examined all scenario-related statements throughout the text to prevent similar errors. Thank you again for your careful reading and valuable feedback.

Changes in manuscript: “Wildfire contributions were obtained from PBrC_baseline minus PBrC_nowldf, while the impacts of k variability and aging were estimated from PBrC_k_adj minus PBrC_baseline and PBrC_age minus PBrC_baseline, respectively.”
(Lines 128 – 130)

2: The tracer scaling method requires stronger physical justification. The authors scale PBrC mass by 10^{-5} and compensate by scaling the imaginary refractive index k by 10^5 apparently assuming that PBrC optical effects scale linearly with k . However, AAOD is determined by the vertically integrated absorption coefficient, where $\int k dz$, not simply by k . Although k influences MAC, MAC also depends on particle size, wavelength, density, mixing state, and the optical mixing rule. Since the model calculates aerosol optical properties using an internal-mixing core-shell assumption with shell properties determined by component volume fractions, reducing the PBrC mass by 10^{-5} and increasing k by 10^5 may not produce an optically equivalent aerosol. The authors should demonstrate that this scaling preserves absorption coefficient, AAOD, heating rate, and radiative forcing by comparing the scaled-tracer method against an unscaled PBrC simulation. Otherwise, the resulting optical and radiative effects may be affected by numerical or physical artifacts. In addition, multiplying by 10^5 could introduce physically unrealistic optical properties or numerical artifacts. The authors should justify why these specific scaling factors were selected and provide a validation test showing that the scaled-tracer method produces equivalent absorption, AAOD, heating rates, and radiative effects compared with an unscaled PBrC simulation

Response: Thank you for your comments. In this study, we adopted a tracer scaling approach to derive the mass concentration of PBrC and scaled its complex refractive index to compensate for its optical effects. This treatment is justified and does not introduce physical uncertainties about particle size, wavelength, or other properties. Through this approach, we are able to produce equivalent estimates of absorption, AAOD, and radiative effects. The added tracer mass itself is negligibly small and

therefore almost exerts no influence on the overall atmospheric physiochemical processes simulated by the model. The tracer is only restored at the post-processing stage to display the PBrC concentration in the final results. This methodology has been widely applied in previous source-oriented studies (Guo et al., 2018; Hu et al., 2016, 2017; Shi et al., 2017).

For aerosol optical effects, CMAQ calculates the AAOD by vertically integrating the layer-by-layer absorption, where the absorption for each layer is computed online based on the simulated aerosol properties.

$$AAOD = \sum_L b_{abs}(L) \cdot \Delta z_L$$

Where L denotes the vertical layer index, Δz_L is the thickness of layer L , and $b_{abs}(L)$ is the aerosol absorption coefficient for that layer. $b_{abs}(L)$ is derived from Mie scattering calculations, which integrate the computed refractive indices over the particle size distribution for each mode and wavelength band. The calculation of the refractive indices and the subsequent steps that incorporate particle size into the optical computation are decoupled in the workflow.

The absorption calculation employs a core-shell approach, in which the optical properties of the shell (core) are determined from the volume-weighted average of the refractive indices of the constituent species:

$$AE_NI_SHELL = \frac{\sum_{spc} \left[\left(1.0 \times 10^{-9} \cdot \frac{6\pi}{\rho_{spc}} \right) \cdot C_{spc} \cdot k_{spc} \right]}{\sum_{spc} \left(1.0 \times 10^{-9} \cdot \frac{6\pi}{\rho_{spc}} \right) \cdot C_{spc}}$$

Here, AE_NI_SHELL represents the imaginary part of refractive index for outer shell in a given mode, at a specific vertical layer, and for a particular wavelength band. The real part of the shell refractive index, as well as the refractive index for the inner core, are computed using an entirely analogous approach—the only differences lie in the species pools and the corresponding replacement of k (the imaginary part) with n (the real part). ρ_{spc} is the density of species spc , C_{spc} is the concentration of species spc , and k_{spc} is the imaginary part of the refractive index of species spc . This formulation is from CMAQ module (Beyer et al., 1996; Horvath, 1995).

In our modification, we introduced the optical properties of PBrC while setting the original OC absorption to zero to avoid double counting the brown carbon effect. We did not modify other OC properties, such as particle size. This is because PBrC is not a new species—it is inherently part of OC—and we did not wish to introduce additional uncertainties. The physical properties of PBrC are identical to those of OC, and retaining OC's physical attributes is essential for correctly representing properties such as particle size. Since our modification affects only the calculation of the refractive index of shell and does not alter the size distribution parameters, it does not influence subsequent physical calculation processes simulated in the model.

This approach is physically justified because PBrC is a subset of OC. We consider the existing model representation of OC size to be satisfactory for the purpose of this study; our objective is solely to modify the absorption of OC to emphasize the role of PBrC. Introducing additional changes to size distribution would introduce unnecessary uncertainty and shift the focus away from our central question—the impact of PBrC light absorption—toward a more general investigation of OC optical effects, which is beyond the scope of this work. Our aim is specifically to isolate and quantify the radiative effect of PBrC through its light-absorbing properties.

Changes in manuscript: “Since PBrC is intrinsically part of primary OC, we set the absorption of primary OC to zero and prescribed other bulk properties of PBrC to be identical to those of primary OC. This approach avoids double-counting of brown carbon absorption and prevents introducing additional uncertainties.” (Line 157)

3: The manuscript evaluates carbonaceous aerosol performance using observed OC concentrations from published studies and reports a correlation of $R = 0.61$ between modeled and observed OC. This is helpful because PBrC is a subset of OC and shares similar emission, transport, and deposition processes. However, OC is still an indirect proxy for PBrC. The authors should more explicitly acknowledge that agreement with OC does not necessarily validate PBrC absorption, source-specific PBrC fractions, or radiative effects. If available, the authors should compare model results with observations of aerosol absorption, absorption Ångström exponent, single scattering

albedo, organic aerosol absorption, BrC absorption, EC/OC ratios, or field-campaign measurements in China. If such observations are not available, the manuscript should clearly state this limitation and discuss how it affects confidence in the conclusions.

Response: Thank you for your comments. In the model evaluation, we included several indirect metrics, such as PM_{2.5} and OC concentrations as supplementary evaluation. Beyond these indirect metrics, we also compared lots of direct observational quantities that reflect PBrC, namely shortwave AOD (Fig. S3; R = 0.52), absorption AOD, and PBrC absorption). Specifically, we collected reliable ground-based measurements of aerosol optical properties from AERONET and selected shortwave absorption AOD, which better represents the absorption properties of brown carbon, for comparison with the model results (Fig. 1b; R = 0.43) (Li et al., 2025). We further compared PBrC absorption from field measurements in China with the simulated results (Fig. 1c; R = 0.54). Both direct comparisons indicate reasonable agreement between simulation and observation, which supports the credibility of our results. A description of these data has also been added to the revised manuscript.

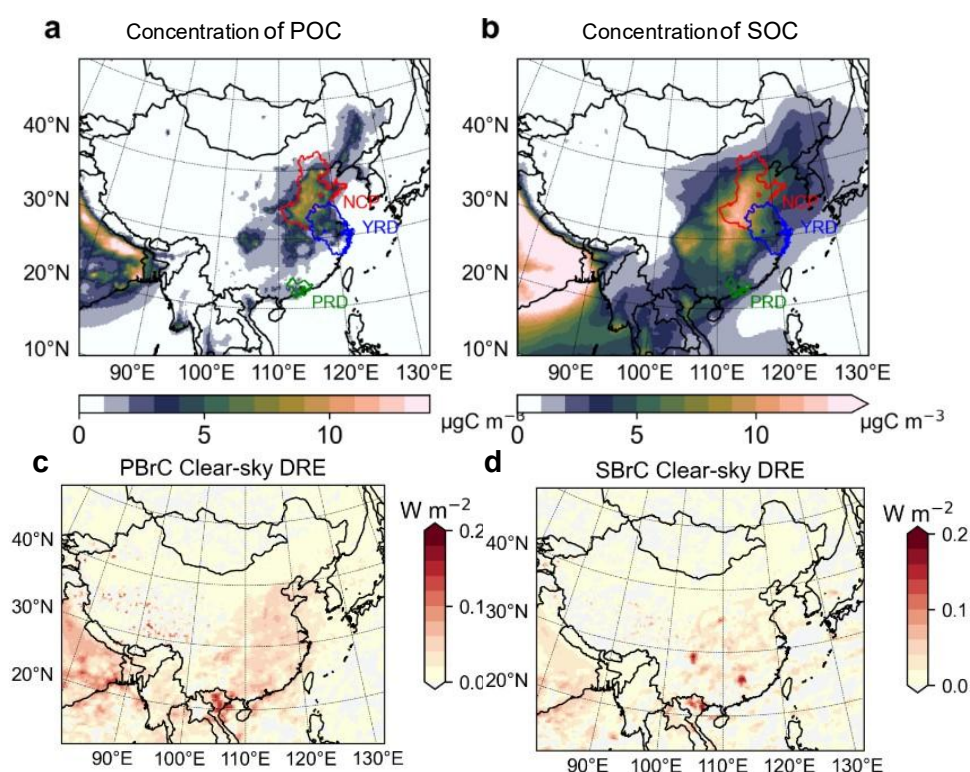
Changes in manuscript: “To assess whether our simulations realistically reproduce atmospheric conditions, both indirect (e.g., aerosol concentrations) and direct (e.g., AAOD and PBrC absorption) indicators were employed to evaluate the model’s ability to capture atmospheric dynamics and PBrC characteristics. The comparison indicates overall good model performance.” (Lines 185 – 187)

4: The manuscript acknowledges that SBrC is excluded and that this may lead to underestimation of BrC effects, especially in China. This is an important limitation. However, because the study discusses the impacts of brown carbon on atmospheric composition and radiative effects, the potential magnitude of this missing contribution should be discussed more quantitatively.

Response: Thank you for your comment. This study focuses on the evolution and role of PBrC over China, and the role of SBrC is therefore not addressed specifically. According to previous studies, the overall radiative effect of SBrC is approximately 15 %

of PBrC (Yang et al., 2025). While SBrC concentrations may be higher than those of PBrC, its contribution to radiation remains limited.

In view of the potential importance of SBrC, we have performed an additional experiment in which both PBrC and SBrC are simulated over the same domain for November 2022. Figure a and b compare the simulated concentrations of primary organic carbon and secondary organic carbon, and Figure c and d present the estimations of their radiative effects. The results show that the radiative effect of SBrC is generally weaker than that of PBrC. However, the analysis of SBrC is still ongoing, and these results remain subject to considerable uncertainty and are under further investigation. Therefore, we present these results for reviewer's information only and have not included them in the revised manuscript. We have added description of the influence of SBrC to the discussion section based on previous studies.



Changes in manuscript: “In addition, SBrC is not included in our simulations. Previous research suggested that SBrC may contribute DRE equivalents to roughly 15% of that of PBrC in China (Yang et al., 2025). While this does not affect our conclusions regarding PBrC, which is the focus of this study, further research on SBrC is warranted.” (Lines 379 – 381)

The authors should consider adding a discussion comparing their estimated PBrC effects with previous studies that include both primary and secondary brown carbon. If possible, provide an approximate range of how much total BrC absorption or radiative forcing could be underestimated due to the exclusion of SBrC.

1: The manuscript notes that PBrC emissions are estimated from OC emissions and fixed PBrC-to-OC ratios, and that this approach is constrained by limited chemical characterization and the lack of correction factors for technological improvements over time. This is a significant uncertainty, especially for a long-term study. The authors should provide more information on the PBrC-to-OC ratios used for different sectors and explain whether these ratios vary by fuel type, technology, region, or year. If fixed ratios are used over the entire period, the manuscript should discuss how changes in combustion technologies, emission controls, and fuel use may bias the temporal trends. A sensitivity analysis using alternative PBrC-to-OC ratios would be valuable.

Response: Thank you for your comments. The ratios used in this research are those compiled by Xiong et al. (2022), who synthesized measured BrC emission factors and laboratory studies to derive source specific PBrC-to-OC ratios. The ratios generally do not vary with combustion technology or emission controls, because the influence of technology is instead carried by the OC emission factors from which PBrC emissions are derived. For coal-fired power plants, for example, the OC emission factor differs by more than three orders of magnitude between unabated combustion and baghouse control, whereas the PBrC-to-OC ratio is held at 0.16. The ratios are also not varied by region or by year in our implementation, following Xiong et al. (2022), who applied them over 1960-2010 and found that the global anthropogenic PBrC-to-OC ratio changed only slightly over that period. Because the ratio is overall constant, the influence of its uncertainty on the spatial pattern or temporal trend is limited. Changes in combustion technology and emission controls, which are the dominant drivers of the temporal trend, affect through the OC emission inventory. We have modified the manuscript.

Changes in manuscript: “The current estimation of PBrC remains constrained by the use of fixed PBrC to OC ratios, owing to the lack of detailed chemical characterization of PBrC (Xiong et al., 2022).” (Lines 356 – 358)

2: The authors identify the imaginary part of the complex refractive index, k , as a major source of uncertainty and conduct a sensitivity test multiplying or dividing k by a factor of five. This is useful, but the results should be more clearly connected to the major conclusions. For example, do the main conclusions about trends, source contributions, or radiative impacts remain valid across the full range of k values? Please provide uncertainty ranges in the abstract and conclusions where appropriate.

Response: Thank you for your comments. In this study we examined how the estimated radiative effect responses to different k values. The results show that increasing or decreasing k has limited influence on the spatial pattern of the DRE, and the dominant effect is a shift in the absolute magnitude of the DRE. This is expected, because the trend and sources of PBrC are controlled primarily by changes in emissions. Varying k within its range of uncertainty does affect aerosol-driven atmospheric photochemistry, but it does not modify PBrC itself significantly. We therefore consider that the uncertainty in k does not affect our main conclusions: it changes the range of the absolute DRE, but not the trend, source attribution, and the trend in DRE. We have modified our manuscript.

Changes in manuscript: “The results show corresponding changes of -1.0% and +0.3% in surface O_3 , and +0.056 $W\ m^{-2}$ and -0.017 $W\ m^{-2}$ in PBrC DRE relative to the baseline, respectively. The estimated PBrC DRE in 2010 therefore ranges from +0.012 to +0.085 $W\ m^{-2}$ around a baseline value of +0.029 $W\ m^{-2}$, underscoring the need for accurate field measurements of k .” (Lines 372 – 374)

1: The manuscript includes an OH-induced bleaching scenario and reports that OH aging reduced nationwide PBrC by approximately 3.9% and O_3 by less than 0.1% in 2010. The authors interpret this as a relatively minor effect under anthropogenically dominated and heavily polluted conditions. This is reasonable, but additional

explanation is needed. Please clarify whether the OH-aging rate constant is applied uniformly to all PBrC sources and particle types. In reality, aging and bleaching may vary by source, chemical composition, coating state, and atmospheric conditions. The manuscript should discuss whether a single aging parameter could underestimate or overestimate aging effects for particular source categories.

Response: Thank you for comments. In the discussion section, we designed a scenario that accounts for OH aging to assess potential magnitude of this aging process on PBrC. OH bleaching is one of the main aging pathways, and we therefore included it to provide a reference estimation of the PBrC aging effect. BrC aging has become a focus of recent BrC research, but existing studies treat BrC as a single bulk component and BrC from different sources may differ in how it ages. However, evidence for source-dependent differences in aging is still lacking, and insufficient measurements are currently available to constrain them. We therefore did not distinguish between the aging processes of PBrC from different sources. We have added a corresponding modification to the revised manuscript.

Changes in manuscript: “Unlike most global regions where PBrC is dominated by biomass burning, our study region is characterized by infrequent wildfires, anthropogenic-dominated and heavily polluted background. The OH-induced aging of PBrC is therefore relatively less pronounced, consistent with previous findings of minimal DRE changes from OH aging in East Asia (Wang et al., 2018). Nevertheless, further investigation of the aging processes of PBrC from different sources is warranted.”
(Lines 365 – 368)

2: It is not clear how you used the core-shell model to calculate optical properties. Are you considering core-shell for all types of particles, including organics, inorganics, organic-inorganic mixture, soot, etc.? What is the coating, and what is the core? What is their RI? What is the mixing ratio between core and shell? All of this information is missing. Did you use Mie simulation or T-matrix?

Response: Thank you for your comments. In this study we used the core-shell treatment implemented in CMAQ (Bohren and Huffman, 2008; Bond et al., 2006), which

represents all particles, with black carbon (BC) as the core and all other species as the shell. The complex refractive index of the shell is obtained by volume-weighted averaging: for each species, its volume-weighted concentration is multiplied by the corresponding optical properties, and the products are summed to give the refractive index of the shell (see Equations *AE_NI_SHELL* in Question 2). The relative amounts of core and shell are derived automatically from the concentrations of BC and the other species. Mie scattering as implemented in CMAQ was used for optical calculations. All these calculations follow the default scheme provided in the current CMAQ version. No changes were made to the underlying mechanism, and the scheme is therefore well established. We have added the description in the revised manuscript.

Changes in manuscript: “In addition, no modifications were made to the optical calculation scheme in CMAQ, which represents particles as a BC core surrounded by a shell whose complex refractive index is a volume-weighted average of the other shell species, and obtains the optical properties from Mie theory.” (Line 157)

3: Please clarify which wavelengths were used for k , AAOD, AOD, and DRF.

Response: Thank you for comments. We focused the optical metrics on short and intermediate wavelengths because the optical properties of BrC are expressed primarily in the shortwave region. For the k values, we used the wavelengths 2094, 303, 310, 316, 333, 381, and 607 nm, which are the default wavelengths in CMAQ and are concentrated in the shortwave. For AOD and AAOD we used 381nm and 440nm, respectively, which are the original wavelengths reported by AERONET. Retaining the original wavelengths avoids the uncertainty associated with converting between wavelengths and these wavelengths are also widely used in BrC studies. In addition, we used light absorption data at 370nm and AAOD at 550nm, taken from the original observational datasets we collected (field measurements and satellites). In the radiative transfer calculation, all wavelengths were considered so that the DRE is estimated as completely as possible.

Minor Comments

1: The SI tables and figures should be labeled and numbered according to the order in which they are first cited in the main text.

Response: Thank you for reminding. The relevant labels have been corrected in the revised manuscript.

2: The manuscript refers to Table S4 when discussing the scaled refractive index used to restore PBrC optical effects. Please ensure that Table S4 is cited in the correct order and contains all necessary optical parameters, including the real and imaginary refractive index values, wavelength dependence, and source assumptions.

Response: Thank you for comments. The citation order has been corrected. Table S4 listed the source and the imaginary refractive index, together with its spectrum of wavelength. In addition, because OC is already included in the model and represents the scattering component of the total organic carbon (including BrC), no additional real part of refractive index (scattering) is assigned to PBrC, so that scattering is not double counted.

3: Please ensure that all acronyms are defined at first use.

Response: Thank you for reminding. We have checked and revised the manuscript.

4: Please be consistent with PBrC and PBrA.

Response: Thank you for comments. We have checked and revised the manuscript.

5: Could you clarify what you mean by carbon and non-carbon fractions/components inside PBrC? Are you referring to different elements? Or organic vs inorganic? But you mentioned “brown non-carbon organic matter” in line 145. What does that mean? How could organic be non-carbon?

Response: Thank you for your comments. Within the concept of brown carbon, the relationship between BrC and its non-carbon fraction is analogous to that between OC and OA in organic aerosol: the non-carbon fraction of brown carbon corresponds to the

hydrogen, oxygen, nitrogen, and other elements in brown organic aerosol, while the term brown carbon refers specifically to the carbon element of that aerosol. In this sense, BrC is to brown organic aerosol what OC is to OA.

1: Figure captions and axis labels should specify whether the reported quantity is PBrC carbon mass or total PBrA mass including non-carbon components.

Response: Thank you for your comments. In both the figures and the text, we use “ $\mu\text{gC m}^{-3}$ ” unit to denote the mass of the PBrC carbon component.

2: The manuscript notes that OC is underestimated under heavily polluted conditions and attributes part of this bias to SOA underestimation [1]. Please discuss how this bias may affect simulated PBrC concentrations and radiative effects in highly polluted regions.

Response: Thank you for your comments. We estimate PBrC emissions from the primary organic carbon emission inventory, so an underestimation of OC may indeed lead to an underestimation of both PBrC concentrations and associated DRE. Although part of the OC underestimation can be attributed to an underestimation of SOA, the influence of secondary organic aerosols on BrC is nevertheless limited (see response in Question 4). In the model, the underestimation of OC stems largely from uncertainties inherent in the emission inventory. We have discussed this in the Discussion section and have also modified the manuscript.

Changes in manuscript: “In addition, SBrC is not included in our simulations. Previous research suggested that SBrC may contribute DRE equivalents to roughly 15% of that of PBrC in China (Yang et al., 2025). While this does not affect our conclusions regarding PBrC, which is the focus of this study, further research on SBrC is warranted.” (Lines 379 – 381)

3: Should it be PBrC_k_adj minus PBrC_baseline in line 129?

Response: Thank you for pointing this out. The typo has been corrected and the wording in the manuscript has been revised accordingly (see response in Question 1).

Changes in manuscript: “Wildfire contributions were obtained from PBrC_{baseline} minus PBrC_{nowldf}, while the impacts of k variability and aging were estimated from PBrC_{k_adj} minus PBrC_{baseline} and PBrC_{age} minus PBrC_{baseline}, respectively.” (Lines 128 – 130)

4: Why estimate OA absorption using BC:OA ratio is only apply to biomass and biofuel combustion? Saleh et al (<https://pubs.acs.org/doi/full/10.1021/acs.estlett.8b00305>) and Cheng et al (<https://www.tandfonline.com/doi/full/10.1080/02786826.2019.1566593>) show this method can also be potentially applied to any type of combustion of organic fuels. Also, it is not clear to me what patterns and effects you refer to in line 143.

Response: Thank you for your comments. The BC:OA ratio method for estimating OA absorption is indeed widely used in studies of OA light absorption and can be applied to OA from a wide range of organic fuels. However, our study focuses on the evolution of PBrC pollutants and its effects on atmosphere and environment. Resolving both the concentration distribution and the spatiotemporal variation of PBrC requires a PBrC-specific emission inventory as model input, whereas the BC:OA ratio method, although it constrains the optical effect of OA well, cannot provide the concentration of PBrC itself. China has large anthropogenic emissions that have changed substantially over recent decades and is an important source of PBrC. We therefore used a PBrC emission inventory and prescribed the optical properties of PBrC (Xiong et al., 2022). We have revised the relevant statements in the manuscript, including the wording that was unclear.

Changes in manuscript: “However, this approach only provides the light absorption of OA and cannot resolve the concentration of PBrC, which is needed to quantify the evolution of this pollutant in China, where anthropogenic emissions dominate. Therefore, to simulate the spatiotemporal evolution of PBrC and its effects on the atmospheric environment, we adopted the method proposed by Xiong et al. for PBrC emission estimation (Xiong et al., 2022).” (Lines 141 – 144)

5: Can you make figures instead of a table for Table S1?

Response: Thank you for your suggestions. We chose a table rather than a figure because a table allows us to present more timescales (annual and monthly) and more metrics, and to report the values with greater precision. Because these indirect metrics are of secondary importance, the table is placed in the Supplementary Information.

6: There is no significant difference between with and without PBrC in Fig. 1 b. Also, please justify the improvement you mentioned in lines 211 and 212.

Response: Thank you for your comments. We acknowledge that the improvement in AAOD at 440 nm is not significant, because AAOD at 440 nm is influenced not only by BrC but also by other absorbing aerosols such as BC and mineral dust, whose contributions are also substantial. Changing the k of PBrC alone may therefore have a slight effect on the simulated AAOD at 440 nm. We have added a description of this to the revised manuscript.

Changes in manuscript: “The agreement between the simulated and observed AAOD improved slightly after PBrC was incorporated (Fig. 1b), with the regression slope increasing from 0.61 ($R=0.41$) without PBrC to 0.67 ($R=0.43$) with PBrC. This modest improvement may reflect the fact that AAOD at 440 nm is contributed jointly by several light-absorbing aerosol components, namely BC, BrC, and mineral dust (Li et al., 2025). Changing the complex refractive index of PBrC alone may therefore yield only a limited improvement in the total AAOD at 440 nm.” (Lines 210 – 212)

7: Please explain what the BC-tracer method is.

Response: Thank you for your comments. This method is the observational approach used in the studies we cite, and we have added a description of it to the revised manuscript.

Changes in manuscript: “In addition, field Aethalometer measurements of PBrC light absorption at 370 nm from previous studies were collected to further evaluation (Table S3) (Wang et al., 2019). In these studies, BrC absorption was isolated with the BC-tracer method, in which the absorption arising from primary combustion sources is

estimated as the measured BC mass concentration multiplied by a representative ratio of primary absorption to BC mass concentration, and the remaining absorption is attributed to SBrC.” (Line 174)

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