

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

Journal: Atmospheric Chemistry and Physics (ACP)

Manuscript ID: EGUSPHERE-2026-1196

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 #2:

This manuscript investigates the long-term (2005–2020) evolution and atmospheric impacts of primary brown carbon (PBrC) aerosols in China – a region marked by intensive human activity and substantial emission shifts over the study period. Using an internal mixing model, the authors comprehensively simulate PBrC surface and vertical concentrations, source contributions, direct radiative effects (DRE), and impacts on formation across the entire country. A key strength of this work is its nationwide scope rather than a narrow regional focus. While using the PBrC/OC ratio method leads to a recognized underestimation of the PBrC DRE, the authors adequately justify this approach. Overall, despite this limitation, I recommend accepting the manuscript for publication after minor revisions.

Major Comments

1: Page 12, Line 300-305: The discussion in this section and in other relevant parts (Lines 345-350) of the manuscript (e.g., the graphical abstract) seems counterintuitive. The manuscript reported that as PBrC decreases, its suppression of weakens – meaning PBrC's "braking effect" on formation is removed. This should actually contribute to the

increase in, but that is not the case it seems (look at graphical abstract). Please explain/clarify?

Response : Thank you for your comments. O_3 formation is influenced by photochemistry: a larger JNO_2 favors O_3 production, whereas the light absorption by PBrC reduces JNO_2 and therefore suppresses O_3 formation. As PBrC concentrations decrease, its reduction of JNO_2 is weakened, which in turn weakens the suppression of O_3 by PBrC. It should be noted, however, that PBrC is still present and therefore still suppresses O_3 formation and it does not increase O_3 . This is what the figures show for the years with low PBrC concentrations (2017-2020): the reduction in JNO_2 is weaker (the green dots become lighter in color), and the suppression of O_3 by PBrC is weakened (the absolute value of the negative blue line becomes smaller). Note that the negative values are plotted on the right side of the y-axis, so a higher line indicates a weaker PBrC effect. We have revised the figure caption to make this clearer.

Changes in manuscript: “Green circles represent the relative reduction in NO_2 photolysis rates caused by PBrC, with darker colors indicating a larger reduction. The right side of the y-axis shows the PBrC-induced change in O_3 concentration, with more negative values indicating a stronger suppression of O_3 .” (Lines 309 – 310)

2: Please explain why and levels declined at different percentage rates between 2005 and 2020. While the authors briefly note that levels were dominated by the power and industrial sectors whereas was influenced mainly by agriculture, it remains unclear how these distinct sources caused the differing decline rates. Government clean air actions typically target regulated and organized sectors like industry rather than unorganized sectors like agriculture or residential heating; thus, further clarification on how these policy dynamics affected the respective emission trends would strengthen the discussion.

Response: Thank you for your comments. The different decline rates reflect both government regulation and continuing advances in technology and management. Power generation and industry have been the primary targets of China's clean air actions since 2013, and their emissions consequently declined markedly between 2013 and 2017

(Wang et al., 2025; Zheng et al., 2018). Agriculture and residential combustion, by contrast, are more closely tied to the continuous improvement of policy management and technology and to the rapid urbanization of China over recent decades. The decline in agricultural emissions is related mainly to the phase-out of open crop-residue burning under the straw-burning bans, whereas residential emissions declined as coal was progressively replaced by gas and electricity for household heating, particularly after 2017. The penetration of these measures is partial and spatially uneven, and the associated decline has been gradual and is still ongoing (Wang et al., 2025; Zheng et al., 2018). In addition, PBrC originates mainly from residential sources, therefore their reduction contributes to the decline in total PBrC significantly. We have added a description to the Discussion in the manuscript.

Changes in manuscript: “The phase-out of open crop-residue burning under the straw-burning bans, the replacement of coal by gas and electricity for household heating, and the rapid urbanization of China over recent decades declined their emissions significantly (Zheng et al., 2018).” (Lines 252 – 256 in the revision)

Minor Comments

1: Page 1, Lines 14 and 16: comma missing after “globally” and “underexplored in China”.

Response: We greatly appreciate you for pointing this out. We have corrected the punctuation errors and re-examined the manuscript to prevent similar errors.

Changes in manuscript: “Although biomass burning is recognized as the major source of primary BrC (PBrC) globally, anthropogenic sources can contribute comparably or more to PBrC in regions with intensive human activities, yet variations in concentrations and effects of PBrC remain underexplored in China, where dramatic emission changes occurred in last two decades.” (Lines 14 and 16)

2: Page 2, Line 40: “Although the chemical consist of BrC remains some debates,” Confusing, rewrite it.

Response: Thank you for your suggestions. We have revised the manuscript to make it clearer.

Changes in manuscript: “Although the chemical composition of BrC has not yet been fully resolved, it is generally categorized into primary brown carbon (PBrC) and secondary brown carbon (SBrC) according to their sources” (Lines 40 – 41)

3: Page 3, Line 53: “in regional scale” is redundant.

Response: Thank you for your suggestions. We have deleted this redundant word.

Changes in manuscript: “Consequently, SBrC contributes less to the global radiative balance (Li et al., 2025), while in certain regions with abundant precursors, it can exceed PBrC due to the enhanced production of secondary organic aerosols (SOA) (Tsigaridis and Kanakidou, 2018).” (Lines 51 – 54)

4: Page 3, Line 56: “absorptions” should be “absorption”.

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

Changes in manuscript: “Its effective absorption in the UV range can suppress the photolysis rates of photochemically active gas.” (Line 56)

5: Page 3, Line 61: Year information is missing for the citation.

Response: Thank you for reminding us. We have corrected the citation errors and checked the manuscript again.

6: Page 7, Line 184: should be “remains poorly constrained...”

Response: Thank you for your correction. We have modified the text, and the grammar of the manuscript has been checked throughout.

Changes in manuscript: “The chemical composition of PBrC remains constrained,” (Line 184)

7: Page 7: Line 188-189: The sentence is confusing: “model’s performance in PM_{2.5} and gaseous O₃ concentrations”.

Response: Thank you for your comments. We have modified the manuscript.

Changes in manuscript: “the model’s performance in reproducing observed PM_{2.5} and O₃ concentrations can serve as an indicator of its ability to represent the physical and chemical processes governing PBrC in the atmosphere.” (Lines 188 – 189)

8: Page 7: Line 188-189: should be “physical and chemical processes”

Response: Thank you for your comments. We have modified the manuscript.

9: Fig. S4: AOD wavelength should be same (380 nm) for both simulated and observed values. Pls recheck.’

Response: Thank you for your comments. We have modified the figures in the Supplement.

10: Fig S8: Type mistake. In think figure ‘b’ is for ‘TRAN’ not for ‘WSTE’.

Response: Thank you for your correction. The errors in the figures in the Supplement have been corrected.

11: Line 377: Typo: “stud3y”?

Response: Thank you for your correction. The typographical errors in the manuscript have been corrected.

12: Line 378: Should be Figure S11. Please check.

Response: Thank you for comments. The citation order has been corrected

13: Line 396: “insensitive”?

Response: Thank you for your careful reading. We have corrected the typographical errors.

Changes in manuscript: “across East Aasia, South Asia, and Southeast Asia with intensive human activity.” (Line 396)

References

Wang, Y., Chen, G., Yang, Y., Zhang, Z., Zhang, R., Wang, P., and Zhang, H.: Anthropogenic emissions shape long-term changes in PM_{2.5} concentrations and health risks in China, *Eco-Environment & Health*, 4, 100198, <https://doi.org/10.1016/j.eehl.2025.100198>, 2025.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, *Atmospheric Chemistry and Physics*, 18, 14095–14111, <https://doi.org/10.5194/acp-18-14095-2018>, 2018.