Response to the Reviewers' comments

Dear Editor:

First, we would like to thank you and the anonymous reviewers for the constructive comments and suggestions. We have carefully addressed all comments and revised our manuscript accordingly. We think these opinions and suggestions can help to improve the quality of the manuscript. We have submitted three documents, including:

1. A clean copy of the revised manuscript, named "Clean Manuscript."
2. A copy of the revised manuscript with all changes highlighted, named "Revised Manuscript."
3. A detailed, point-by-point list of our replies to the comments of the reviewers, named "Response to comments."

Please find below our detailed response to the comments point-by-point. In addition, the manuscript has been improved according to the manuscript formatting request. Hope these will make the manuscript more acceptable for publication in Atmospheric Chemistry and Physics.

Yours sincerely,

Yingjun Chen

Reviewer(s)' Comments to Author:

Our point-to-point responses are presented in the following. We hope that the revision would satisfactorily address the comments and concerns of the editors and reviews.

Reviewer #1:

This manuscript focused on carbonyl compounds emitted by four types of combustion sources, including biomass burning, residential coal combustion, on-road vehicles and non-road mobile machineries. The emission factors were carefully determined and their influencing factors were comprehensively discussed. I think this manuscript could be considered for publication in ACP, after minor revisions.

Response: Thanks for your positive comments. We have carefully revised this manuscript based on the reviewer's comments. We hope that the revisions and improvements would satisfactorily address the reviewer's concerns. Our point-to-point responses are as follows.

1. Major point: Section 2.1, softwood and hardwood should be clearly distinguished. Section 3.1, please clarify whether the emission characteristics were comparable between softwood and hardwood. If not, please give possible explanations.

Response: Thanks for your valuable comments. We sincerely apologize for the oversight in not clearly distinguishing between softwood and hardwood in the original manuscript. In the revised version, we have explicitly differentiated these two fuel types in Section 2.1. Additionally, in Section 3.1, we have added a detailed comparison of the emission characteristics between softwood and hardwood. Our analysis reveals that softwood (e.g., pine) tends to emit higher concentrations of carbonyl compounds, particularly small-molecule carbonyl compounds such as formaldehyde and acetaldehyde, which is likely attributed to differences in lignin content and combustion efficiency. This finding has been discussed in the revised manuscript, along with potential explanations for the observed variations.

A detailed comparative analysis of carbonyl compound emission profiles across different fuel types has been incorporated, with new findings presented in Section 3.1 (Lines 213-234).

2. Abstract, re-write the first sentence as "Fuel combustions are important primary sources of carbonyl compounds (CCs)……"

Response: Thanks for your valuable comments. As suggested, we have rephrased the first sentence of the Abstract to: "Fuel combustions are important primary sources of carbonyl compounds (CCs) ……" This revision improves the clarity and precision of the statement.

See the revised version in the Abstract section (Line 18).

3. Abstract, line 30, suggest removing the "tend".

Response: Thanks for your valuable comments. The word "tend" has been removed as recommended. The revised sentence now reads: "High-temperature promotes small molecules like F+A to cyclize, supplying ample precursors for the formation of acetone and aromatic aldehydes."

See the revised version in the Abstract section (Line 30).

4. I noticed that "EFccs" appeared only once. Remove this abbreviation or use it to replace "EFs of CCs" throughout the manuscript.

Response: Thanks for your valuable comments. We have carefully reviewed the manuscript and replaced all instances of "EFs of CCs" with "EF_{CCs}" to ensure consistency. This change has been applied throughout the text.

All modifications concerning EF_{CCs} have been highlighted in red throughout the annotated manuscript. Given the extensive nature of these revisions, a comprehensive enumeration has been omitted for conciseness.

5. Some abbreviations were repeatedly defined. For example, CCs was defined twice in Page 2; EFs was defined again in Page 5.

Response: Thanks for your valuable comments. We apologize for the redundancy in defining abbreviations. In the revised manuscript, we have ensured that each abbreviation (e.g., CCs, EFs) is defined only once upon its first appearance.

The definition of carbonyl compounds (CCs) appears exclusively in two locations in the revised manuscript: Line 18 of the Abstract section and Line 36 of the Introduction.

6. Grammar mistakes like "Fuel types is key factors…(Page 1)" and "Fuel types determines the composition of…(Page 15)" should be avoided.

Response: Thanks for your valuable comments. We sincerely apologize for the grammatical errors in the original manuscript. The following corrections have been made:

"Fuel types is key factors…" has been corrected to "Fuel type is a key factor…"

“Fuel types determines the composition of…” has been corrected to “Fuel type determines the composition of…”

All other instances of subject-verb disagreement have been carefully reviewed and corrected.

The revised content appears in two locations: (1) Line 18 of the Abstract section, and (2) Line 450 of the Conclusions and Future Perspectives section.

7. Equations in Page 5, suggest using hyphens, which could be clearly distinguished from the minus signs, for the subscripts (e.g., c-fuel).

Response: Thanks for your valuable comments. Thank you for pointing out the inconsistency in the use of hyphens and minus signs in the equations. We have revised the equations to use hyphens for subscripts (e.g., c-fuel) and minus signs for mathematical operations, ensuring clarity and consistency.

The revised content appears in Section 2.3, page 6.

8. Page 7. Suggest briefly explaining the “bell-shaped distribution theory”.

Response: Thanks for your valuable comments. We have added a brief explanation of the “bell-shaped distribution theory” in the revised manuscript. Specifically, we describe it as a theoretical framework used to explain the relationship between the volatile content of residential coal and its emission characteristics, where emissions peak at intermediate volatile content and decrease at both very high and very low volatile content levels. This addition provides better context for readers unfamiliar with the concept.

Reviewer #2:

This manuscript presents a comprehensive and in-depth study on the emission characteristics and influencing factors of carbonyl compounds (CCs) from various typical combustion sources. It provides the latest data for emission inventories and offers a scientific basis for targeted emission reduction strategies. This manuscript will make a significant contribution to the control and management of air pollution.

Response: We sincerely appreciate your thorough and constructive review of our manuscript titled "Emission Characteristics and Influencing Factors of Carbonyl Compounds from Various Typical Combustion Sources." We are grateful for your recognition of the manuscript's potential contribution to air pollution control and management, as well as for your detailed suggestions, which have significantly improved the quality of our work. Below, we provide a point-by-point response to your comments and outline the revisions we have made accordingly.

1. The abstract should be revised to better articulate the scientific innovation and applied value of this study, specifically by explicitly stating its contributions to optimizing combustion systems, guiding emission control policies, and enhancing atmospheric chemistry modeling frameworks.

Response: Thanks for your valuable comments. We have revised the Abstract to better highlight the scientific innovation and applied value of this study. Specifically, we have explicitly stated its contributions to optimizing combustion systems, guiding emission control policies, and enhancing atmospheric chemistry modeling frameworks. The revised Abstract now reads:

"This study provides the latest data for emission inventories and offers a scientific basis for targeted emission reduction strategies. Its findings contribute to optimizing combustion systems, guiding emission control policies, and enhancing atmospheric chemistry modeling frameworks."

The newly added content appears in the Abstract section (Lines 32-34).

2. The conclusion section should include specific estimates of the emission reduction effects of the proposed measures on atmospheric oxidizing capacity. For example, the impact of increasing combustion temperature and upgrading emission standards on ozone (O_3) formation should be quantified.

Response: Thanks for your valuable comments. As suggested, we have added specific estimates of the emission reduction effects of the proposed measures on atmospheric oxidizing capacity. For example, we have quantified the impact of increasing combustion temperature and upgrading emission standards on ozone (O_3) formation. The revised Conclusion now includes:

"At a high combustion temperature of 800°C, the ozone formation potentials (OFP) of BB and RCC are 0.8 (g O_3/kg-fuel) and 0.6 (g O_3/kg-fuel), respectively, indicating that increasing the combustion temperature can reduce ozone formation by 91.0% and 53.8%, respectively. Similarly, for both on-road and non-road sources, upgrading vehicle emission standards can significantly reduce ozone formation (46.8%~65.0%), with the most notable reduction effects observed for DV (63.6% reduction) and AM (65.0% reduction), which are the sources with higher emission levels. In conclusion, the proposed measures in this study demonstrate significant emission reduction effects on atmospheric oxidizing capacity. ”

The additional content can be found in the 3.4.2 section (Lines 437 to 443, Page 19).

3. Spelling errors. In line 32 of the abstract, "allevite" should be corrected to "alleviate".

Response: Thanks for your valuable comments. We sincerely apologize for the spelling error in line 32 of the Abstract. The word “allevite” has been corrected to “alleviate.”

The revised content appears in line 32.

4. Terms such as “EF_{CCs}” should be defined upon their first appearance, and consistent usage should be maintained throughout the manuscript. For example, “EF_{CCs}” and “EFs of CCs” should be standardized. Additionally, some abbreviations need to be redefined to avoid confusion with commonly used abbreviations, such as AA and OA.

Response: Thanks for your valuable comments. We have ensured that “EF_{CCs}” is defined upon its first appearance and used consistently throughout the manuscript. Additionally, In the fields of chemistry and environmental science, “AA” is a commonly used abbreviation for aromatic aldehydes. However, to avoid confusion with other terms, its definition has been explicitly stated upon its first use in this manuscript. Therefore, “AA” has been retained as the abbreviation for aromatic aldehydes in this article. As for “OA”, which represents "other aldehydes and ketones" in this study, it is indeed prone to confusion with "organic aerosols" (OA). To address this, the term "other aldehydes and ketones" has been replaced with “Other CCs” throughout the manuscript.

All modifications concerning EF_{CCs} have been highlighted in red throughout the annotated manuscript. Given the extensive nature of these revisions, a comprehensive enumeration has been omitted for conciseness. Furthermore, all instances of 'OA' have been systematically replaced with 'Other CCs' throughout the manuscript, with corresponding revisions made to all figures and tables to maintain consistency.

5. Some sentences are lengthy and complex. Simplifying sentence structures can improve readability. For instance, the second paragraph of the introduction (line 49) could be broken into multiple sentences, and it's necessary to add the following reference in revised manuscript.

Emission of Intermediate Volatile Organic Compounds from Animal Dung and Coal Combustion and Its Contribution to Secondary Organic Aerosol Formation in Qinghai-Tibet Plateau, China. *Environmental Science & Technology* 2024 58 (25), 11118-11127.

Response: Thanks for your valuable comments. We have simplified lengthy and complex sentences to improve readability. For instance, the second paragraph of the Introduction (line 49) has been broken into multiple sentences. The revised Abstract now reads:

"Cheng et al. (2022) investigated the emission characteristics of CCs from BB, such as emission factors (EFs) and chemical composition, using a tube-furnace. They found that combustion conditions, including temperature and oxygen concentration, as well as fuel characteristics like composition, significantly affect the EFs and composition of CCs."

We have also added the suggested reference in the second paragraph of the introduction:

"Emission of Intermediate Volatile Organic Compounds from Animal Dung and Coal Combustion and Its Contribution to Secondary Organic Aerosol Formation in Qinghai-Tibet Plateau, China. *Environmental Science & Technology* 2024 58 (25), 11118-11127."

The revised content appears on page 2, line 51. The newly added references are presented on page 2, lines 56-58.

6. Line 105: For biomass samples, the selection of 2g is difficult. Besides, the combustion process is very fast. How can the author ensure the accuracy of sampling throughout the entire combustion process? the following are some references related to experiment process that can be cited:

Examination of long-time aging process on volatile organic compounds emitted from solid fuel combustion in a rural area of China. *Chemosphere* 333 (2023) 138957.

Response: Thanks for your valuable comments. We acknowledge the concern regarding the selection of 2g biomass samples and the rapid combustion process. To ensure sampling accuracy, we have implemented a continuous sampling protocol and validated it through repeated experiments. We have also cited the following reference to support our methodology:

"Examination of long-time aging process on volatile organic compounds emitted from solid fuel combustion in a rural area of China. *Chemosphere* 333 (2023) 138957."

The newly added references are presented on page 4, lines 126.

7. Ensure that the font size in all figures and tables is consistent to present a more organized and visually appealing layout. For example, the legend font size in Figure 1a is slightly larger than in the other three subfigures.

Response: Thanks for your valuable comments. We have ensured that the font size in all figures and tables is consistent. For example, the legend font size in Figure 1a has been adjusted to match the other subfigures, presenting a more organized and visually appealing layout.

8. The manuscript should include (1) uncertainty quantification analyses of ozone formation potential across diverse CCS emission sources, and (2) a schematic diagram of the sampling system in Section 2.1 to enhance methodological transparency.

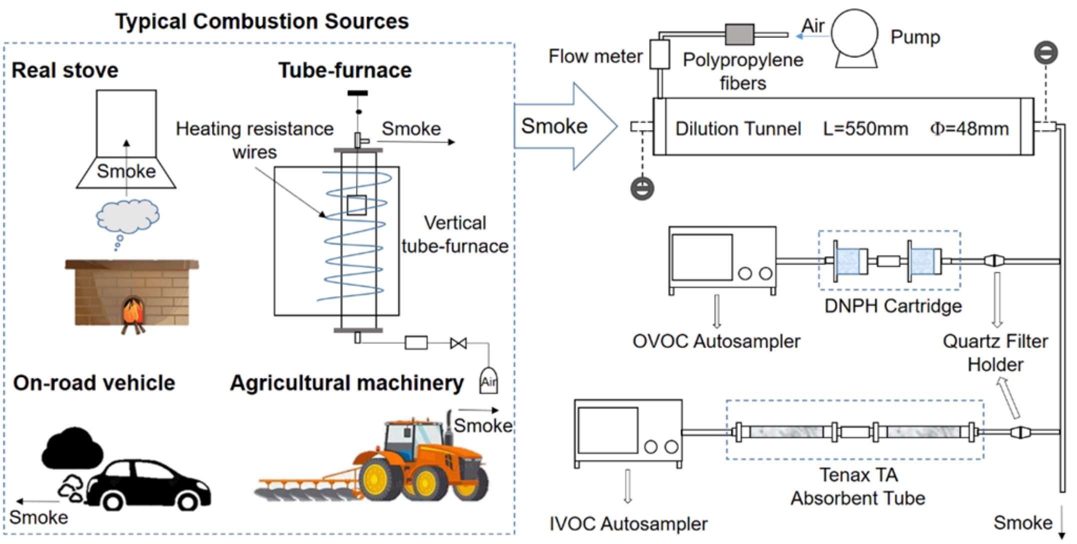
Response: Thanks for your valuable comments. We have added (1) uncertainty quantification analyses of ozone formation potential across diverse CCs emission sources, and (2) a schematic diagram of the sampling system in Section 2.1 to enhance methodological transparency. The schematic diagram illustrates the sampling setup and process, providing readers with a clear understanding of the experimental design. Here are some of the new additions:

(1) Uncertainty quantification analyse:

Combustion sources	Max	Min
BB	21.5%	-3.1%
RCC	13.8%	-10.9%
E-GV	3.66%	-2.68%
GV	3.84%	-2.31%
DV	8.38%	-6.63%
AM	10.27%	-4.52%

The table presents the uncertainty ranges of ozone formation potential estimates for different combustion sources, expressed as maximum (Max) and minimum (Min) deviation percentages. It can be observed that the deviations are generally positively correlated with the emission factors of carbonyl compounds, meaning that the higher the emission factor, the greater the uncertainty in the calculated ozone formation potential for the combustion source. Among these, BB, RCC, and RCC are identified as high-uncertainty combustion sources, indicating that their emission characteristics are complex and may significantly contribute to ozone formation. It is recommended to conduct more precise measurements and modeling of the emission factors for these combustion sources to reduce uncertainty.

(2) Schematic diagram of the sampling system:



The newly added schematic diagram of the sampling system appears in Section 2.2 (page 5), while the additional uncertainty analysis is presented in Section 2.5 (page 7).

9. The figures should be replaced with higher resolution versions to ensure graphical clarity essential for proper interpretation of the experimental data.

Response: Thanks for your valuable comments. All figures have been replaced with higher resolution versions to ensure graphical clarity essential for proper interpretation of the experimental data.

Reviewer #3:

This manuscript conducts an in-depth investigation into the emission characteristics and influencing factors of carbonyl compounds (CCs) from four typical combustion sources, based on laboratory simulations and road measurements. The findings fill the data gap regarding the emission characteristics of CCs from different combustion sources, providing significant scientific and innovative value for atmospheric pollution control and policy formulation.

Response: Thanks for your valuable comments.We sincerely thank the reviewer for their thorough and constructive feedback on our manuscript. We greatly appreciate the time and effort dedicated to evaluating our work and providing valuable suggestions for improvement.

1. The experimental design mentions a more detailed classification of fuel types, but the results and discussion section does not address these finer distinctions, such as whether there are differences in CCs emissions between southern and northern straw burning, or between softwood and hardwood.

Response: Thanks for your valuable comments. We appreciate the reviewer's observation regarding the finer distinctions in fuel types, such as differences in carbonyl compounds (CCs) emissions between southern and northern straw burning, as well as between softwood and hardwood. In response, we have expanded the Results and Discussion section to include a more detailed analysis of these distinctions. Specifically, we have added a subsection comparing the emission characteristics of CCs from southern and northern straw burning, as well as softwood versus hardwood combustion. These additions are supported by additional data and references to relevant studies. Below is a brief description of the newly added content:

The carbonyl compounds generated from the combustion of southern straw (rice straw) are higher than those from northern straw. The emission factor of CC from rice straw combustion (3865.6 ±

558 mg/kg) is significantly higher than that of corn (2829.8 ± 1771.8 mg/kg) and wheat (1772.6 ± 847.2 mg/kg), indicating that the carbonyl compounds generated from southern straw combustion are 1.4-2.2 times higher than those from northern straw combustion. This phenomenon may be attributed to differences in the content of biomass components (cellulose, hemicellulose, and lignin) and combustion efficiency. Cheng et al. (2022) found that among the three biomass components, cellulose combustion generates the highest amount of CCs, followed by hemicellulose, with lignin producing the least. Meanwhile, Zhao et al. (2019) discovered that the holocellulose content (cellulose + hemicellulose: ~56.3%) of rice straw is higher than that of corn and wheat. Additionally, in this study, the combustion efficiency of rice straw (90.9%) is slightly lower than that of corn and wheat (92.0%-92.8%).

The carbonyl compounds generated from the combustion of softwood (pine) are higher than those from hardwood. The emission factor of CCs from pine combustion (1415.3 ± 431.8 mg/kg) is significantly higher than that of poplar (1020.3 ± 249.1 mg/kg) and willow (905.5 ± 109.6 mg/kg), indicating that the carbonyl compounds generated from softwood combustion are 1.4-1.6 times higher than those from hardwood. This phenomenon may be due to differences in combustion efficiency. In this study, the combustion efficiency of pine (94.0%) is significantly lower than that of poplar and willow (95.6%-96.0%). Studies have shown that incomplete combustion of fuels is more likely to generate CCs.

A detailed comparative analysis of carbonyl compound emission profiles across different fuel types has been incorporated, with new findings presented in Section 3.1 (Lines 213-234).

2. While the discussion on residential solid fuel combustion is thorough, the analysis of the formation mechanisms of CCs from on-road sources, particularly for ethanol-blended gasoline and biodiesel, is relatively brief. It is recommended to include additional discussion on the formation mechanisms of CCs from ethanol-blended gasoline and biodiesel, along with relevant references.

Response: Thanks for your valuable comments. We agree with the reviewer that the discussion on the formation mechanisms of CCs from on-road sources, particularly for ethanol-blended gasoline and biodiesel, was relatively brief. To address this, we have expanded the discussion in Section 4.2 to include a more detailed analysis of the formation mechanisms of CCs from ethanol-blended gasoline and biodiesel. We have also added relevant references to support this discussion, ensuring a more comprehensive understanding of the underlying processes.

For the ethanol gasoline section, an in-depth discussion on different emission standards has been added. It was found that the "acetaldehyde/formaldehyde" ratio in carbonyl compounds emitted by China VI emission standard ethanol gasoline vehicles is approximately 0.38, which is about 2.5 times that of gasoline vehicles under the same emission standard. This indicates that the use of ethanol gasoline significantly increases acetaldehyde emissions. In contrast, this ratio for China V ethanol gasoline vehicles is 1.80, suggesting that higher emission standards can effectively reduce carbonyl compound emissions while also decreasing acetaldehyde generation. The reason for this phenomenon may be that, as emission standards are continuously upgraded, exhaust treatment technologies are improved, and emission requirements become stricter, leading to further oxidation of harmful substances such as acetaldehyde into formaldehyde.

For the biodiesel section, two additional references have been included to enhance the understanding of the formation mechanisms:

Lin, Y.C., Hsu, K.H., and Chen, C.B.: Experimental investigation of the performance and emissions of a heavy-duty diesel engine fueled with waste cooking oil biodiesel/ultra-low sulfur diesel blends, *Energy*, 36, 241–248, https://doi.org/10.1016/j.
energy.2010.10.045, 2011.

Chien, S.M., Huang, Y.J., Chuang, S.C., and Yang, H.H.: Effects of biodiesel blending on particulate and polycyclic aromatic hydrocarbon emissions in nano/ultrafine/fine/ coarse ranges from diesel engine, *Aerosol Air Qual Res*., 9, 18–31, https://doi.org /10.4209/aaqr.2008.
09.0040, 2009.

The expanded discussion on ethanol gasoline appears on page 12 (lines 285-291), while the additional references for biodiesel are presented on page 14 (line 343).

3. Grammatical errors. There are several instances of subject-verb disagreement in the text, for example, in line 24 of the abstract.

Response: Thanks for your valuable comments. We sincerely apologize for the grammatical errors in the manuscript, particularly the subject-verb disagreement in line 24 of the abstract. The revised

Abstract now reads:

"Fuel type is a key factor affecting the CCs components."

We have conducted a thorough proofreading of the entire manuscript to correct grammatical errors, improve sentence structure, and ensure consistency in language usage. All instances of subject-verb disagreement have been resolved.

The revised content appears in Line 24 of the Abstract section.

4. The terms EF_{CCs} and EFs of CCs convey the same meaning. It is recommended to use EF_{CCs} throughout the manuscript to replace "EFs of CCs".

Response: Thanks for your valuable comments. We thank the reviewer for pointing out the inconsistency in the use of EF_{CCs} and EFs of CCs. As suggested, we have replaced all instances of "EFs of CCs" with "EF_{CCs}" throughout the manuscript to ensure consistency and clarity.

All modifications concerning EF_{CCs} have been highlighted in red throughout the annotated manuscript. Given the extensive nature of these revisions, a comprehensive enumeration has been omitted for conciseness.

5. The first occurrence of an abbreviation in the main text should be clearly annotated with its full name. For example, "ES" appears for the first time in line 74 of the abstract without its full name being provided.

Response: Thanks for your valuable comments. We apologize for the oversight in not providing the full name for the abbreviation "ES" upon its first occurrence in line 74 of the abstract. We have now clearly annotated all abbreviations with their full names upon their first appearance in the main text. "ES" is now defined as "emission standard" in line 74.

6. The font format in the formulas is inconsistent. It is recommended to unify the font format throughout.

Response: Thanks for your valuable comments. We acknowledge the inconsistency in the font format used in the formulas. To address this, we have standardized the font format for all formulas throughout the manuscript, ensuring a consistent and professional presentation.