Reviewers' Comments, Author's Replies, and Revisions in the Text

(Second Round)

Manuscript Number: egusphere-2023-598

Title: Source differences in the components and cytotoxicity of $PM_{2.5}$ from automobile exhaust, coal combustion, and biomass burning contributing to urban aerosol toxicity

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MS type: Research article

Dear editor Dr. Prof. Su and dear reviewers,

We are very grateful for your helpful comments again. Based on these kind advices and suggestions, we have made further careful modifications and detailed improvements in depth to the previous manuscript. The changes are shown in the track-changes. The point-to-point replies, explanations, and clarifications for all of the revisions are listed below for easy reference. We have also polished the overall manuscript again.

We hope the revised manuscript this time could be published in ACP after these important modifications and significant improvements.

Responses to Reviewers' Comments:

Reviewer #1:

Thanks to the authors for the replies. I just have one more comment. Regarding the statistical analysis in Figure 5 & 6, it is not clear in terms of the letter “a”, “b” and “ab”. I was wondering about the differences among the different types of samples. For example, was there any significant difference between the ROS production of group automobile exhaust and coal combustion (Figure 6-2)? It would be better if the authors could put a horizontal line above the two compared groups, and label the statistical comparison result with asterisk (statistically significant) or NS (no statistically significant difference). BTW, it is not common to use number “1” to label each panel (Figure 6)

Reply and revision:

We appreciate your kindly evaluations again very much for our manuscript. Thanks for the reminding about the marking style of statistical differences. It is possible that the unclear statements of the descriptions about Figure 5 and 6 in our previous manuscript caused the confusion. These figures used letters to indicate the statistical difference between groups, which was significant if they do not contain the same letter, and was not significant if they contain the same letter. The horizontal lines with asterisk or NS above the compared groups is indeed good idea, but if we use this method to indicate the statistical differences between groups in figures of current study, then each set of data would need 5 lines to indicate the significances between different groups, and it might be a bit too crowded in the graphs, so we finally prefer to choose using letters to indicate the difference significances between groups. In addition, according to your nice comments, numbers labeling sub-graph have been replaced in the updated Figure 6.

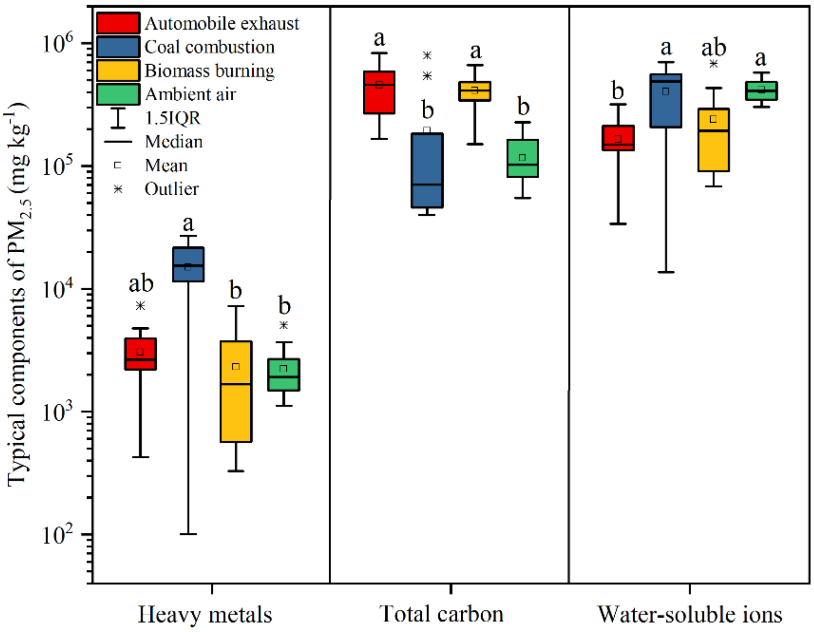

Figure 5. Cumulated typical measured components (mg kg^{-1}) in $PM_{2.5}$ from various specific sources (n=10 for each combustion source and n=16 for urban ambient air). Statistically significant differences between the groups are indicated by different letters (Kruskal-Wallis test, $p < 0.05$).

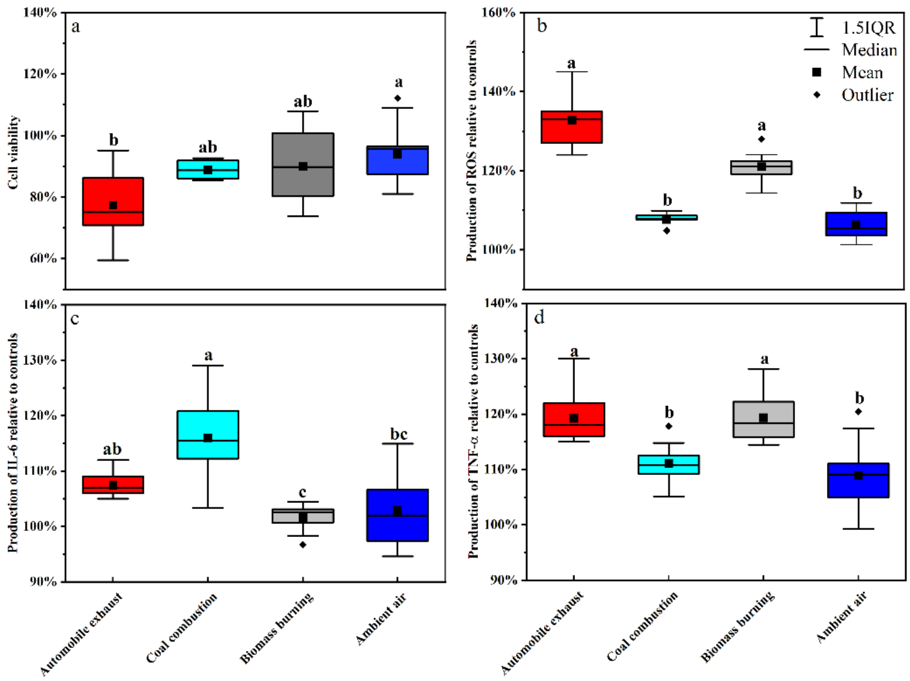

Figure 6. Cell viability, oxidative stress and inflammation levels of human alveolar epithelial cell lines (A549) exposed to $PM_{2.5}$ suspension (80 mg L^{-1}) from various specific sources (n=10 for each combustion source and n=16 for urban ambient air). Statistically significant differences between the groups are indicated by different letters (Kruskal-Wallis test, $p < 0.05$).

Reviewer #2:

The manuscript is an exhausting work considering both the emission tests and toxicity tests on particles emitted from vehicle emission, coal combustion, biomass burning. Also the ambient air was also investigated. The most interesting thing is the toxicity identification of particles emitted from various sources. While the other research points (the toxicity of ambient $PM_{2.5}$ and its source apportionment) are not so impressive as they have been done for a long period. The research contents are a bit of redundant. There are also obvious shortages for the sample collection and PMF modeling. Therefore, I suggested the authors to focus on the toxicity of particles emitted from various sources. It can be accepted after a thorough correction and polishing.

Reply and revision:

Thanks very much for your critical evaluation on our manuscript, which is an important guidance for us to improve the overall quality of this paper. It's really an exhausting $PM_{2.5}$ work with abundant both chemical and toxicological results for three major types of combustion sources, and also supported by their final sink - - the ambient air samples. So yes, our aim is focusing on the toxicity of particles emitted from various sources, and many supporting information beneficial to understand the findings are listed in the supplemental materials.

Because last manuscript was the revision after a round of reviewing, we revised and added many contents in accordance with two reviewers' comments during the first round revision. Some comments of the previous round reviewing might be conflicting with this round, but we will integrate them, and have revised the manuscript thoroughly according to your following advices on those substantial issues.

The detailed comments:

1. The authors should not overemphasize the significance of this study, especially in optimizing air quality standards and prioritizing $PM_{2.5}$ control strategies. For optimizing air quality standards and prioritizing $PM_{2.5}$ control strategies, this simple study is quite not enough. Corresponding descriptions should all be corrected, including

the description in Line 15-19, Line 34-36, Line 39-42, Line 48-49, Line 92-93, Line 101, Line 456-457.

Reply and revision:

Thanks for your comments. This study should be valuable for optimizing air quality standards and prioritizing $PM_{2.5}$ control strategies, but we really still can't overemphasize these significances just by results of our study. We have made the necessary updates to these corresponding statements according to your suggestion, including rewriting and reorganization.

2. For conducting the $PM_{2.5}$ source apportionment, the 16 samples are so limited that I do not believe the authors can obtain a reasonable result or the result can be accepted. 19 chemical components were input into the models, while there are only 16 samples. I suggest the author to use the CMB model, but not the PMF model, as the authors have the source profiles.

Reply and revision:

Thank you for the reminding and nice suggestion.

The three universally specific combustion sources are the most key objects of this study, while the ambient air is their final sink in the environment, therefore, as the actual mixture of various source particles in real environment, totally 16 typical urban $PM_{2.5}$ samples covering a year monthly were collected to represent the ambient air samples. Although the sample size of ambient $PM_{2.5}$ are limited, as real examples, their chemical and toxicological results could be compared with the source samples, and also imply and explained by the contributions of source samples, indicating the necessity of investigating specific sources samples.

Of course, as you suggested, besides the PMF model applied previously, we further use the CMB model to identify the ambient $PM_{2.5}$ source. We found that the results from the CMB and PMF models are very similar (Figure 1). Therefore, we decided to include a comparison of the results from these two models in the revised manuscript.

The following is a description of the CMB model analysis and results:

Due to the high concentration of sulfate and nitrate in ambient $PM_{2.5}$, and being lack of specific actual source to emit sulfate and nitrate, we added the virtual source profiles of secondary sources in CMB model. The virtual source profiles of secondary sources are represented by the proportion of sulfate, nitrate and ammonium in pure ammonium sulfate and ammonium nitrate. The source profiles of coal combustion, plant biomass burning, automobile exhaust, and secondary sources are shown in the table below.

The parameters of CMB are set as follows: the mass percentage range of $PM_{2.5}$ is 80%-120%, 0< Chi^2 (χ^2)<4, and 0.8<R^2<1. CMB model identified four major sources of the ambient $PM_{2.5}$, including primary particles of coal combustion, plant biomass burning, automobile exhaust, and secondary aerosols, which account for 20.19%, 8.31%, 25.88%, and 26.07%, respectively. The mass percentage range of $PM_{2.5}$ is 80.45%, Chi^2 (χ^2)=1.08, R^2=0.82. All parameters show that the CMB model results are well. As the mass percentage range of $PM_{2.5}$ is 80.45%, we further normalized the above four source classes: coal combustion (25.10%); plant biomass burning (10.32%); automobile exhaust (32.17%); secondary aerosols (32.40%).

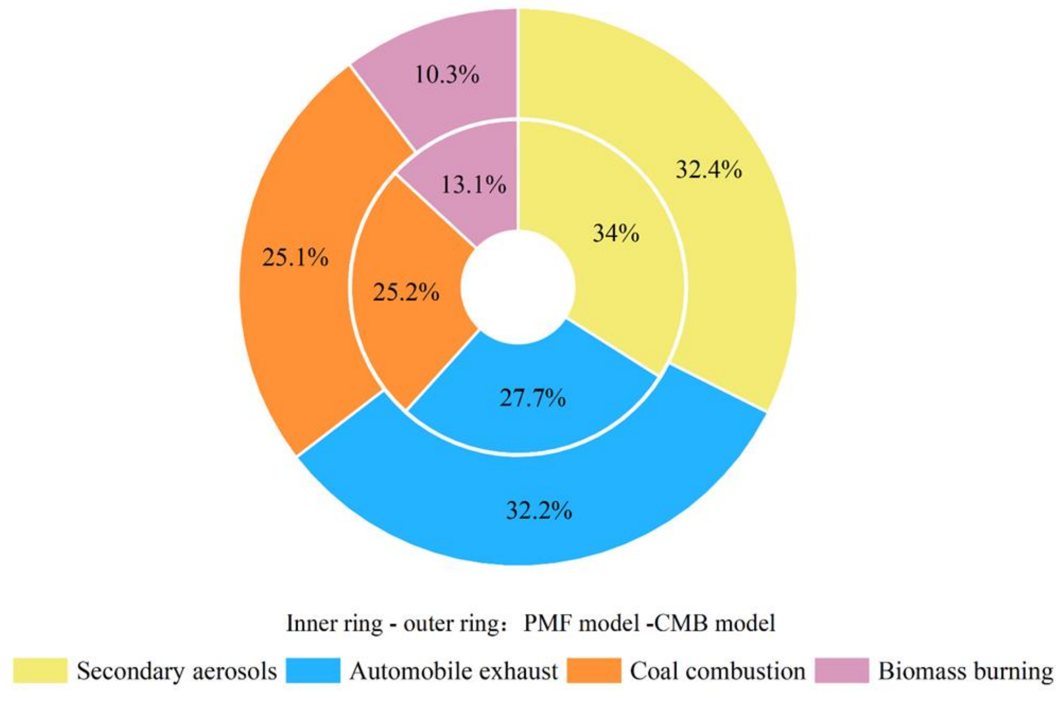

Figure 1. Source contributions (%) to the urban ambient air $PM_{2.5}$ (PMF model vs CMB model).

Table S4. The CMB source profiles of coal combustion, plant biomass burning, automobile exhaust, and secondary sources (g/g).

Source	**Coal combustion**		**Biomass burning**		**Automobile exhaust**		**Sulfate**		**Nitrate**	
	Mean	Standard deviation	Mean	Standard deviation	Mean	Standard deviation	Mean	Standard deviation	Mean	Standard deviation
OC	0.1548	0.2046	0.2828	0.1075	0.2159	0.0945	0.0000	0.0000	0.0000	0.0000
EC	0.0386	0.0609	0.1280	0.1119	0.2420	0.1589	0.0000	0.0000	0.0000	0.0000
V	0.0002	0.0001	0.0001	0.0001	0.0007	0.0011	0.0000	0.0000	0.0000	0.0000
Cr	0.0001	0.0002	0.0012	0.0011	0.0004	0.0002	0.0000	0.0000	0.0000	0.0000
Mn	0.0001	0.0001	0.0001	0.0001	0.0004	0.0003	0.0000	0.0000	0.0000	0.0000
Co	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Ni	0.0001	0.0000	0.0006	0.0005	0.0004	0.0002	0.0000	0.0000	0.0000	0.0000
Cu	0.0007	0.0006	0.0002	0.0002	0.0006	0.0002	0.0000	0.0000	0.0000	0.0000
As	0.0006	0.0008	0.0000	0.0001	0.0003	0.0005	0.0000	0.0000	0.0000	0.0000
Pb	0.0132	0.0083	0.0001	0.0001	0.0002	0.0001	0.0000	0.0000	0.0000	0.0000
Na^+	0.0166	0.0243	0.0071	0.0064	0.0312	0.0166	0.0000	0.0000	0.0000	0.0000
NH_4^+	0.0860	0.0546	0.0073	0.0046	0.0074	0.0047	0.2730	0.0273	0.2250	0.0225
K^+	0.0055	0.0056	0.0929	0.0930	0.0039	0.0016	0.0000	0.0000	0.0000	0.0000
Mg^{2+}	0.0005	0.0003	0.0008	0.0006	0.0036	0.0015	0.0000	0.0000	0.0000	0.0000
Ca^{2+}	0.0043	0.0027	0.0055	0.0054	0.0322	0.0123	0.0000	0.0000	0.0000	0.0000
F^-	0.0018	0.0012	0.0031	0.0025	0.0108	0.0091	0.0000	0.0000	0.0000	0.0000
Cl^-	0.0273	0.0347	0.0928	0.0841	0.0148	0.0059	0.0000	0.0000	0.0000	0.0000
SO_4^{2-}	0.2504	0.1529	0.0185	0.0111	0.0191	0.0125	0.7270	0.0727	0.0000	0.0000
NO_3^-	0.0125	0.0081	0.0119	0.0065	0.0421	0.0283	0.0000	0.0000	0.7750	0.0775

3. In the abstract, the authors indicated that the contributions of $PM_{2.5}$ from combustion sources to the health risks are not unclear. I think it is not suitable, as former studies have conducted works on evaluate the health risks of $PM_{2.5}$ from various sources, though they combined the source apportionment results and carcinogenic and non-carcer risks calculated by the USEPA equations. I believe the toxicity of chemical components in $PM_{2.5}$ from some sources has also been conducted, but not so comprehensive like this study. Corresponding studies should be thoroughly summarized.

Reply and revision:

Sorry for the inaccurate statements leading misunderstanding. Indeed, former studies have conducted works on evaluate the health risks of $PM_{2.5}$ from some sources, mainly calculated by the human health risk assessment equations, but lack of considering the important different toxic roles of various chemical components in $PM_{2.5}$ related to pollution sources, which is one major trigger and objective of our study. According to your suggestion, we reorganized the corresponding descriptions. Corresponding literature were summarized and added in Introduction.

4. Line 34-36, the conclusion is not so meaningful. Without this study, the emission control measures including strengthening the emission standards, coal to gas and coal to electricity, cutting off the crop straw burning have been proposed. The authors should give more accurate conclusions through the key findings of this study, but not repeat formers.

Reply and revision:

Thanks for your comments. We have rewritten these sentences showing key findings of this study based on the specific source toxicity comparisons.

5. I do not agree with the authors that the world air quality guidelines treated the $PM_{2.5}$ as equally toxic. The establishment of air quality standards relied on the former toxicity and epidemiology researches, and they are not established by imaging. The WHO and abundant scientists know the toxicity of various chemical components. To establish a standard, lots of experiments should be done, and the performability should also be considered. I suggest the authors to read some papers to learn how to establish an air quality standard for air pollutants.

Reply and revision:

Thanks very much for your kindly reminding. We corrected and revised all corresponding statements to be more accurate by avoiding misunderstanding.

6. Line 51-56, we all know these knowledges and the sources of aerosol are not the key research points of this study. One sentence is enough. The toxicity of aerosols from various sources should be more summarized.

Reply and revision:

We have polished and condensed these contents according to your comments in the revision. Owing to the background information required for some readers, necessary sentences were kept to make the logic of this study clear.

7. Line 73-74, I don't agree with this description. The heavy metals and PAHs in $PM_{2.5}$ have been listed as the toxic components by the main countries, which means that their toxicity have been tested. Additionally, there are so many studies on the health risk assessment of heavy metals and PAHs (by USEPA methods) and the source apportionment of health risks published, the reviewers even do not want to read such kinds of papers.

Reply and revision:

We corrected and revised the inappropriate statements according to your comments. Yes, we know that the pure chemicals of heavy metals and PAHs are well known toxic pollutants with corresponding tested toxicity and have been widely monitored in environmental managements including air pollution, and there are really abundant routine papers reporting their health risks just assessed by the USEPA calculation methods, but the exposure to human lung cells of these mixed chemicals bound in $PM_{2.5}$ inducing toxicity by inhalation are still not clear. Moreover, air particles are so complicated mixture of many chemicals, such as the sulfate and nitrate salts, not only the general EPA listed pollutants available in various environmental media, their toxicity effects and mechanisms is really a significant question.

8. Line 79-80, how the source profiles be applied in elucidate the aerosol pollution and control strategies? The source profiles of different emission inventories indicated what? The emission inventory was established by activity data and emission factors. Source profiles do not belong to emission inventories.

Reply and revision:

Thank you for the reminding. We revised these statements. Due to variations in particle composition, sources, and toxicity in different urban environments, it is necessary to establish aerosol source emission inventories for different regions to elucidate local aerosol pollution characteristics and facilitate control strategies.

9. Line 81-89, the logic is confusing. For example, the author indicated that straw burning contributed to air pollution in many regions, then the author indicated that the aerosol from biomass burning in Amazon had an ability to induce ROS. Do the author want to say the aerosol from straw burning could induce ROS, or the aerosol from biomass burning in Amazon contributed to air pollution in many regions? Meanwhile, the many regions including where? The authors should give more accurate descriptions. Such kinds of descriptions are ubiquitous in this study.

Reply and revision:

Thanks for your reminding. We checked and revised the statements overall by more accurate descriptions to avoid confusions.

10. Line 85-86, the authors listed the sulfate is mainly from coal combustion, but not mention its toxicity. This example did not contain the similar meanings with other sentences.

Reply and revision:

Because a reviewer of last round has previously mentioned that there was not direct evidence suggesting sulfate itself is toxic or associated with oxidative stress, toxicity is not mentioned in this context. Our aim was twofold: firstly, to illustrate the detrimental effects of anthropogenic sources such as vehicle exhaust, coal combustion, and biomass burning on human health through these examples; and secondly, to emphasize that researchers often focused exclusively on one specific source or component, rarely comparing multiple sources with detailed components together as done in this work.

11. Line 94, the source samples are not abundant in the field of source profiles and emission factors studies.

Reply and revision:

Thanks for your reminding. We have replaced the word "abundant" with the specific number of samples.

12. Line 106, line 109-110, Line 120-123, Line 145, Line 165-167 are redundant. The authors should describe the sampling and analysis methods directly in this section, do not list such inessential information.

Reply and revision:

Because some detailed sampling information in this part were requested by the reviewers of last round reviewing, we polished and simplified these contents according to your comments.

13. Line 118, characteristic to physical-chemcial

Reply and revision:

Revised.

14. Line 127-130, there are four channels, the 160 L min-1 indicated each channel or the four channels together? What is the dilution ratio for the burning test? For each type of fuels, how many times were repeated? How many fuels were burned in each test? The detailed sampling information should be given. Same information for other combustion tests should also be given.

Reply and revision:

We clarified these statements. Some information has been showed in Tables of the Supplemental Materials, and we included more detailed information in Table S1-S3 of the revised manuscript.

Table S1. Characteristics and collection process of the investigated typical vehicles.

No.	Abbreviations	Vehicle types	Manufacture year	Emission standards	Fuel type	Collection time (min)	Weight (kg)
#1	SDGCs-1	Small duty gasoline coach	2015	CN.V	CN.92 #	120	1970
#2	SDGCs-2	Small duty gasoline coach	2019	CN.VI	CN.92 #	120	2110
#3	SDDCs	Small duty diesel coach	lost	CN.IV	CN.5#	20	1790
#4	MDDCs	Middle duty diesel coach	2009	CN.IV	CN.5#	20	3600

#5	HDDCs	Heavy duty diesel coach	2015	CN.V	CN.5#	20	15800
#6	LDDVs-1	Light duty diesel van	2009	CN.III	CN.5#	20	3970
#7	LDDVs-2	Light duty diesel van	2015	CN.IV	CN.5#	20	4500
#8	MDDVs	Middle duty diesel van	2014	CN.IV	CN.5#	20	7320
#9	HDDVs-1	Heavy duty diesel van	2015	CN.IV	CN.5#	20	29080
#10	HDDVs-2	Heavy duty diesel van	2019	CN.V	CN.5#	20	40000

Table S2. Characteristic analysis and collection process of typical coal samples.

Coal types	M_{ad} (%)	A_{ad} (%)	V_{ad} (%)	FC_{ad} (%)	Fuel consumption (g)	Burning duration (min)	Origin
HC-1	1.87	46.2	9.87	42.1	1169	158	Nanjing city
HC-2	2.15	49.3	9.63	38.9	1138	144	Nanjing city
AC-1	1.26	10.2	10.6	78	739	222	Ningxia province
AC-2	1.19	12.5	10.8	75.5	1024	180	Anhui province
AC-3	1.76	6.78	8.99	82.5	537	170	Shanxi province
BC-1	5.23	1.84	41.5	51.5	8117	102	Inner Mongolia province
BC-2	7.06	5.07	29.8	58	669	85	Henan province
IC-1	0.43	13	1.63	85	559	115	Nanjing Iron & Steel Co.
IC-2	1.74	11.1	30.3	56.9	601	90	China Resources Jiangsu Nanre Power Generation Co.
IC-3	4.37	8.17	30.9	56.5	652	95	Huaneng Nanjing Jinling Power Generation Co.

Note: M_{ad} is the moisture mass fraction of the sample on an air-dried basis; A_{ad} is the ash mass fraction of the sample on an air-dried basis; Vd_{ad} is volatile matter mass fraction of sample on dry air-dried basis; FC_{ad} is fixed carbon fraction of the sample on an air-dried basis; $FC_{ad} = 1 - M_{ad} - A_{ad} - Vd_{ad}$.

Table S3. Characteristic analysis and collection process of typical plant biomass fuel samples.

Biomass types	M_{ad} (%)	A_{ad} (%)	V_{ad} (%)	FC_{ad} (%)	Fuel consumption (g)	Burning duration
Rice straw	10.8	14.6	59.8	14.9	83	4′24″
Wheat straw	12.1	5.65	65.5	16.8	328	9′14″
Corn straw	11.6	4.22	66.1	18.1	108	4′39″
Soybean straw	11	4.62	68.4	16	360	11′24″
Peanut straw	15	10.8	61.4	12.8	49	1′20″
Rape straw	11.1	2.95	68.8	17.1	39	1′05″
Sesame straw	13.1	7.64	63.7	15.5	154	2′42″
Corncob	9.21	0.66	73.5	16.7	131	11′35″
Pine branches	13.4	0.33	66.6	19.7	148	12′20″
Peach branches	9.94	0.65	73.4	16	244	16′45″

Note: M_{ad} is the moisture mass fraction of the sample on an air-dried basis; A_{ad} is the ash mass fraction of the sample on an air-dried basis; Vd_{ad} is volatile matter mass fraction of sample on dry air-dried basis; FC_{ad} is fixed carbon fraction of the sample on an air-dried basis; $FC_{ad} = 1 - M_{ad} - A_{ad} - Vd_{ad}$.

15. Line 135-136, what is the diameter of the filter? How many filters were cut for

carbon, ion and toxicity tests, respectively?

Reply and revision:

The diameter of all filters is 47 mm. Carbon, ion, heavy metal, and toxicity testing each require one parallel sample filter, that uses a total of four filters across the four parallel channels.

16. Line 137, why the author said that 16 samples were representative? Representative of what?

Reply and revision:

The three universally typical combustion sources are the most key objects of this study, while the 16 ambient $PM_{2.5}$ samples investigated just provide representative real urban air samples in environment. In last round revision, we revised "typical" as "representative". As the actual mixture of various source particles in real environment, totally 16 example ambient air $PM_{2.5}$ samples (each time lasting a day) spanning all months and different seasons of a year were collected in an urban site surrounded by traffic, residential and commercial quarters of a typical megacity, which site is also very common in China and even globally. Although the ambient $PM_{2.5}$ sample number is limited, these environmental samples can still provide insights into the temporal variations in urban air quality related to the investigated sources and compositions. Undoubtedly, a larger sample size of ambient $PM_{2.5}$ would be much better for the study focusing on the ambient aerosols.

17. Line 141-143, the sentence is not essential.

Reply and revision:

Revised.

18. Line 148, some elements indicated which elements?

Reply and revision:

Revised with details.

19. Line 157, 1/2 or 1/4 or other fraction of filter was cut into pieces?

Reply and revision:

Details were added. Due to the small diameter (47 mm) of filters, each chemical analysis and toxicity testing require one whole parallel sample filter.

20. Line 163, medium (DMEM) medium repeat

Reply and revision:

Revised.

21. Line 193, why these species were selected for PMF modeling?

Reply and revision:

These components represent all the measured chemical data we obtained, and are also commonly used as source tracers in source apportionment studies.

22. Line 199, how the author got the daily $PM_{2.5}$ concentrations? You should give clearly information. Did the author just used the 16 filter samples to calculate its mass concentration? Exceeded the healthy guidelines obviously indicated exceeded how many times?

Reply and revision:

The word "daily" in this paper is not "every day" but "a day". As a routine standard method, we calculated the daily ambient $PM_{2.5}$ concentration through gravimetric measurement of each filter and sampled air volume lasting 23h for a day. Moreover, specific comparison results with healthy guidelines in Fig S3 were showed in the revision.

23. Plant biomass burning can be changed into domestic biofuel burning

Reply and revision:

Thanks for your nice suggestion. Yes, biomass burning can include biofuel burning (both solid, liquid, or gas), agricultural crop burning (both open and domestic), and wildfires of plants mainly trees. In this study, we typically burned 8 types of crop (straws of rice, wheat, corn, soybean, peanut, rape, and sesame, corncob), and also 2 types of firewood (branches of peach and pine), so “domestic biofuel burning” may not match all biomass investigated, and “plant biomass burning” or “solid biomass burning” might be more accurate. Because we focus on domestic solid biofuel burning in this study, we replaced "plant biomass burning" with "plant biomass (domestic biofuel) burning" or " domestic plant biomass burning" for corresponding statements.

24. Line 207-209 can be deleted directly. The authors should give the results directly. Please do not give repeat or meaningless information. This question should be corrected for the whole manuscript.

Reply and revision:

Thanks for your kind reminding. We simplified the manuscript overall to keep streamlined.

25. Line 213, why it indicated that the OC in ambient $PM_{2.5}$ was lower. It may also indicate that the OC in the ambient air may be aged or cleaned. The authors should read more papers on the atmospheric chemistry of OC.

Reply and revision:

Thanks for the comments. We added more discussions according to your suggestions.

26. Line255-257, Line 291, Line 292, Line 293, ….much higher, higher…. The authors should give quantitative description. Such problem should be corrected for the whole paper.

Reply and revision:

Thanks for your kind reminding. We checked the whole paper again and used quantitative descriptions in the revised manuscript.

27. Line 264-265, of course the anthropogenic combustion sources should be controlled. without this study, we all know this.

Reply and revision:

According to your comment, we simplified general knowledge and summarized new findings to make the revised manuscript more concise and succinct.

28. Line 276, they are not new markers.

Reply and revision:

We revised the description.

29. Line 286-288, the sentence is meaningless.

Reply and revision:

This sentence was revised, which was a remark explaining the limitations of PMF source models applied in current study owing to the absence of natural sources.

30. Line 296-297, the volatile fraction is composed of organic matter. Of course, it is composed of organic matter. The description is right, but so boring. I suggest the author read more papers to give the formation mechanisms of OC during combustion sources and to support the higher TC contents from coal burning.

Reply and revision:

Thanks for your suggestion. We simplified the general knowledge. For the formation mechanisms of OC, we also added more discussion and reference on coal combustion in the revised manuscript. Below are the additional references added:

He K, Shen Z, Zhang B, et al. Emission profiles of volatile organic compounds from various geological maturity coal and its clean coal briquetting in China. Atmospheric Research, 2022, 274: 106200.

Zhou W, Jiang J, Duan L, et al. Evolution of submicrometer organic aerosols during a complete residential coal combustion process. Environmental Science & Technology, 2016, 50(14): 7861-7869.

De la Puente G, Iglesias M J, Fuente E, et al. Changes in the structure of coals of different rank due to oxidation effects on pyrolysis behaviour. Journal of Analytical and Applied Pyrolysis, 1998, 47(1): 33-42.

31. Line 302, there were only 16 samples, one for one month. Of course, they varied seasonally. Another thing is not reasonable is that one sample can not be represent for one month and three samples can not be represent for one season. I can not agree with this. Other similar description and conclusion could also not be accepted.

Reply and revision:

Thank you for your suggestion, we have modified the statements about ambient samples in the revised manuscript by providing a more comprehensive perspective to avoid arbitrary expressions. Totally 16 ambient $PM_{2.5}$ samples were indeed limited in representing a year, but in current exhausting study focusing on the various combustion source samples, we can only choose representative real air samples covering each moth and season to support the main source research meaningfully as the mixture sink of their contributions. In fact, for ambient air $PM_{2.5}$, we have collected massive samples frequently in four different sites of this megacity lasting 8 years, but the corresponding research for these abundant ambient samples with spatial-temporal characteristics would be another exhausting valuable work in future.

32. Line 306-307, the author drew the conclusion subjectively. My question is that how to control EC from diesel vehicles? In line 330-332 and in line 347-348, how to control these elements and ions in the particles emitted from these sources selectively. Can you tell us the method?

Reply and revision:

Thanks for your critical comments, we made revision in accordance. The exact effective methods to control these specific key toxic components from the emissions of various combustion sources indeed a challenge, but need to be explored. The chemical findings of our toxicological research point a specific direction for better air pollution control, that should be helpful for the environmental technology with potential methods, targeting source materials, combustion processes, or final emissions. Both the basic findings of this study and the corresponding technological solutions not investigated in this manuscript would be valuable for the clean air and public health. Moreover, environmental management policies might also be beneficial to such aims, such as the choice of fuel types.

33. Line 397, are you sure, the metals and ions can be controlled by strengthening the emission standards? For biomass burning, the particles may all hold high concentrations of OC, Cl^- and K^+, how to control them?

Reply and revision:

Thanks for your reminding about the accurate descriptions. We made corresponding revisions. The measures mentioned have an overall impact on source emissions. Besides the environmental technological methods of controlling toxic components targeting source materials, combustion processes, and final emissions, the environmental management policies are also beneficial to this aim, such as the choice of fuel types, especially for the management of domestic biomass fuel burning. For examples, potential measures include promoting new green energy vehicles and low-ash clean coals, depressing the diesel exhaust and rural crop straw burning emissions.

34. Line 438, I think this sentence say nothing, and we all know these knowledges.

Reply and revision:

According to your comment, we simplified the general knowledges and summarized new findings.

35. Line 442, the $PM_{2.5}$ can not reflect the air situation of eastern China. The site is just a site in a megacity. There are totally 16 samples and the source apportionment results are quite inconvincible.

Reply and revision:

We have updated the relevant descriptions. Yes, we know the limitations of these 16 ambient samples and just use them for comparisons with specific source samples and representation of mixed multi-source samples in real environment. The source apportionment results were cross-validated by models PMF and CMB, supporting the main objectives of this study focusing on various combustion source samples.

36. Line 456-457, the authors just compared the compositions and toxicity of $PM_{2.5}$ from some types of sources, and the manuscript can not provide supports for establishing economical composition-source-based strategies for aerosol pollution control.

Reply and revision:

Thanks for your kind reminding. We restricted the description accordingly.

37. References: the format should be corrected, such as the subscript and low level mistakes

Reply and revision:

We have checked and updated all references accordingly.

38. Figure S1, what is the dwell room? Size selector should be corrected into $PM_{2.5}$ cut inlets

Reply and revision:

We employ the term "residence chamber" as a substitute for the phrase "dwell room". The residence chamber constitutes a crucial element of the dilution tunnel sampler, serving the purpose of allowing the diluted and mixed exhaust to dwell for a specific duration. This facilitates the exhaust cooling and mixture of condensable particles at proper concentrations.

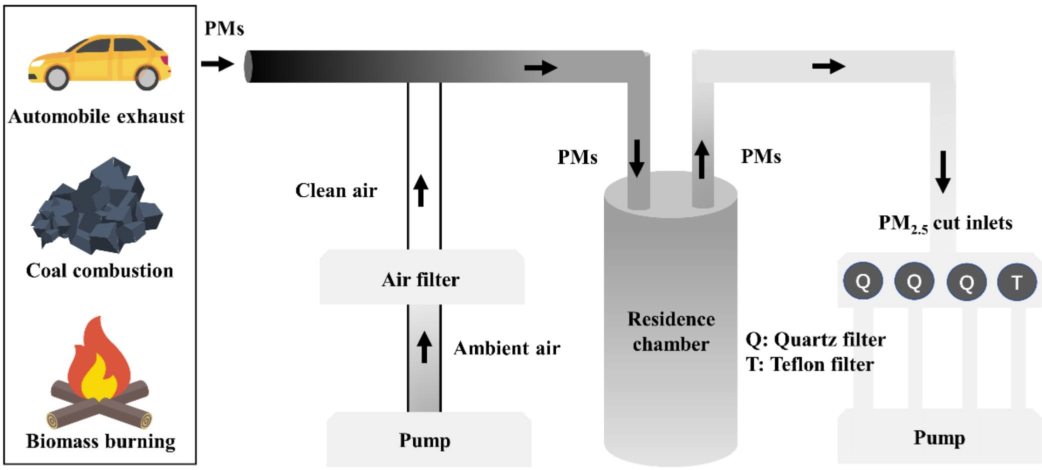

Figure S1. Schematic of a dilution 4-channel sampler used for collecting $PM_{2.5}$ directly from various combustion source emissions.

39. Figure S3, I suggest the authors use the air quality monitoring dataset from local environmental monitoring station for the seasonal comparison. The dot-line figure should be corrected into column figure as the data is not continuous.

Reply and revision:

Revised. The dot-line figure has been replaced with column figure. Considering the main objective of current study, we analyze the monitored data by the self-collected samples with constrained statements.

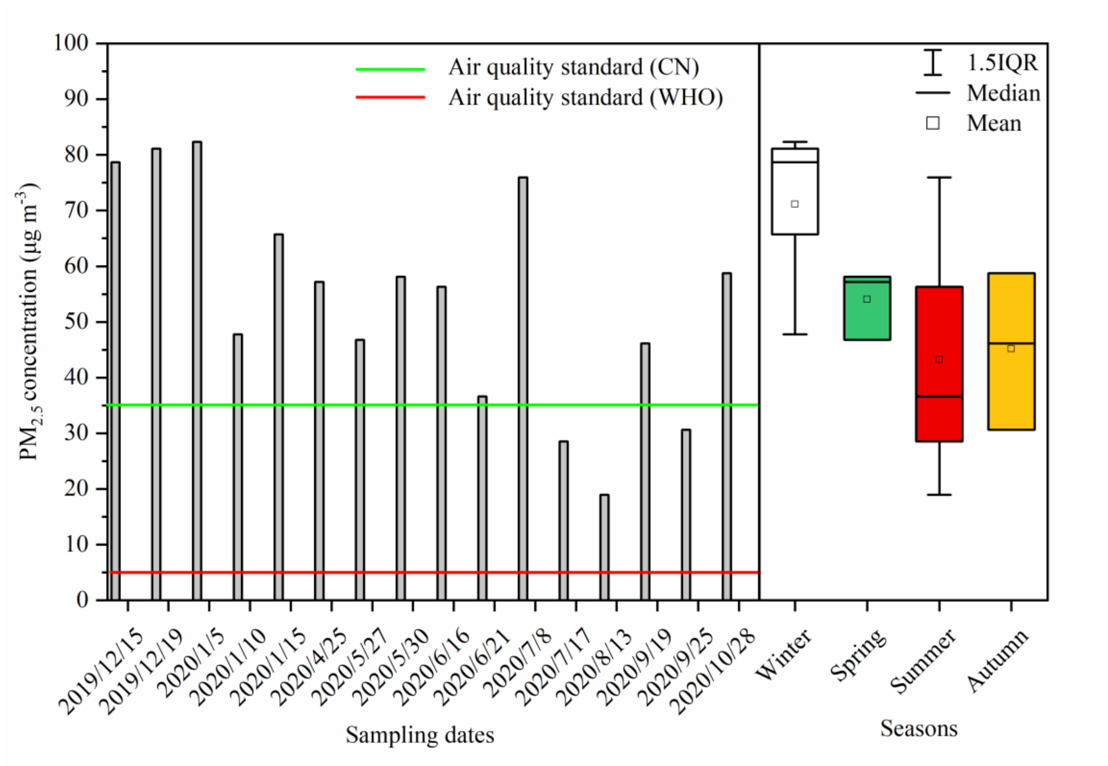

Figure S3. Example of daily urban air $PM_{2.5}$ concentrations (μg m^{-3}) monitored in Nanjing city, eastern China.

40. Figure S4-S10, Figure S12-S15, did the authors do parallel samples? Can the standard errors be given?

Reply and revision:

Thanks for the reminding. Owing to the huge cost of this exhausting work including several chemical and toxicological parameters for abundant samples, we did not analyze parallel samples for every samples. For quality control measures, reference materials were adopted and analytical experiments were performed only after recovery was achieved.

41. Figure S11, it is not monthly $PM_{2.5}$, and it is just $PM_{2.5}$ sample for the selected days.

Reply and revision:

We revised the description about samples from each month according to your comment.