

## Author's response

### Characteristics of particulate-bound *n*-alkanes indicating sources of PM<sub>2.5</sub> in Beijing, China

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#### Referee Comment 1 (from Omar Amador-Munoz):

The article describes the temporal behaviour of *n*-alkanes in Beijing China on some days during 1 year of measurements. The authors propose emission sources using some ratios as well as a multivariate test. The study is a longitudinal descriptive analysis. Methodological details are lacking, some comments are not properly supported, information on atmospheric criteria pollutants associated with the air quality is missing, as well as the meteorology. The article cannot be published in its current state.

1. The authors should further justify the interest in studying alkanes in PM<sub>2.5</sub>. The alkanes are irrelevant from the point of view of air quality and human health, since alkanes are not toxic to humans.
2. Line 44. What are the human health effects of alkanes in PM<sub>2.5</sub> mentioned by the authors? Provide the references.
3. Lines 102-103. In the gravimetric procedure carried out at 20°C for 24 h to obtain the particulate mass, the mass of some organic compound is lost due to evaporation during this time. How was the alkanes mass lost considered to calculate their concentrations in the air?
4. Lines 109-110. Explain what was the procedure to evaluate the recovery efficiency of the alkanes by the analytical method used? What were the concentrations spiked? and how many repetitions were made?.
5. It is not clear how the alkanes were quantified. Was a calibration curve used, external standards, internal standards, isotopic dilution, etc.? Explain the details of the quantitative analysis.
6. Lines 237-242, 283-285. It is well known that gasoline is mostly comprised of hydrocarbons in the C<sub>4</sub> to C<sub>10</sub> range while diesel fuel consists of C<sub>8</sub> to C<sub>25</sub> hydrocarbons (Han et al., 2008; Schauer et al., 1999; Lough et al., 2005; Gentner et al., 2012; Wang et al., 2005). Explain the discrepancy with the results obtained by the authors of the actual study.
7. Line 245. Why can road dust be a source of alkanes with >C<sub>34</sub>?

8. Lines 257-258 and 267-270. The authors assume that temporal distribution of LMW and HMW alkanes is explained by the behaviour of  $C_{max}$  and WNA%, however, they do not consider the effect of temperature or the mixing layer height. How do these variables affect the temporal behaviour of the alkanes?
9. Lines 282-283. The reference by Simonet et al (2004). doi:10.1029/2004JD004565, does not mention alkanes in diesel.
10. Lines 294-296. Show scatter plot corroborating this association.
11. Authors should incorporate atmospheric criteria pollutants and the association with alkanes.
12. Meteorology was not included. It is extremely important to describe the meteorological variables, for example to observe differences in temperature between the four seasons, as well as wind speed and wind direction in order to propose emission sources.
13. Line 16. It should say 153 ng/m<sup>3</sup>
14. The PMF 5.0 user guide can not be a supplemental material. Authors should only cite it.

### **Author's response:**

#### **Dear referee:**

Thank you for your constructive comments on our manuscript. We have carefully considered the suggestions and revised the manuscript accordingly. We added the methodological details involved in the experimental procedure and additional literature support was provided for some of the ideas in the manuscript. In addition, we have consider the effect of meteorological factors on particulate-bound *n*-alkanes and have added the corresponding discussion. Finally, some of the statements and diagrams in the article have been revised. We have tried our best to improve this manuscript, please find our itemized responses in below and our revisions/corrections in the re-submitted files.

1. The authors should further justify the interest in studying alkanes in PM<sub>2.5</sub>. The alkanes are irrelevant from the point of view of air quality and human health, since alkanes are not toxic to humans.

**Reply:** Thank you for your questions. *n*-Alkanes are important environmental pollutants, their health effects are cytotoxicity resulting from interaction with other organic matter in particulate matter (Chen et al., 2019). Compared to the health effects, the research significance of *n*-alkanes is more focused on the environmental impacts. Short-chain *n*-alkanes ( $C \leq 16$ ) are one of the precursors of secondary organic pollutants in the atmosphere and have good reactivity, they are easily involved in the formation of other pollutants (Michoud et al., 2012). Medium-chain and long-chain *n*-alkanes ( $C > 16$ ) are relatively stable in the environment and can be used as indicators to reflect the source of atmospheric particulate matter through source resolution (Chrysikou et al., 2009; Han et al., 2018). We are more interested in the role of *n*-alkanes for indicating the sources of organic aerosol and PM<sub>2.5</sub>. The study

of *n*-alkanes can help to better explain the source of PM<sub>2.5</sub> in order to reduce emissions at the source to control particulate pollution and improve air quality.

**Modification:** We have modified the statement on the interest in studying *n*-alkanes in PM<sub>2.5</sub> in the introduction (L50-51) and added some references (L44, L46, L48).

2. Line 44. What are the human health effects of alkanes in PM<sub>2.5</sub> mentioned by the authors? Provide the references.

**Reply:** Thank you for your questions. The *n*-alkanes in PM<sub>2.5</sub> can be cytotoxic together with PAHs, the concentration of *n*-alkanes can affect cytotoxicity (Chen et al., 2019). In addition, we found that *n*-alkanes have narcotic toxicity and the extent of harm to humans depends on the length of their carbon chains. C<sub>8</sub>-C<sub>16</sub> *n*-alkanes can cause neurological disorders and strong irritation of the respiratory system. When the carbon chains continues to increase, *n*-alkanes can lead to skin damage and even skin cancer (Yang, R. M., 2001; Horiguchi, H., 1978). Therefore, particulate-bound *n*-alkanes can have health effects on humans.

**Modification:** We added the references for the human health effects of *n*-alkanes in PM<sub>2.5</sub> (L44).

3. Lines 102-103. In the gravimetric procedure carried out at 20°C for 24 h to obtain the particulate mass, the mass of some organic compound is lost due to evaporation during this time. How was the alkanes mass lost considered to calculate their concentrations in the air?

**Reply:** Thank you for pointing this out and your questions. We minimized this mass loss by wrapping and sealing the filter with aluminum foil in the gravimetric procedure. We calculated the recoveries of these substances by the recovery experiment, which includes the effect of mass loss due to the experimental process. The overall analysis of concentration variations and sources is not affected.

4. Lines 109-110. Explain what was the procedure to evaluate the recovery efficiency of the alkanes by the analytical method used? What were the concentrations spiked? and how many repetitions were made?.

**Reply:** Thank you for pointing this out. We used blank spiked recovery experiment to evaluate the recovery efficiency of particulate-bound *n*-alkanes. We added a mixed standard solution of C<sub>8</sub>-C<sub>40</sub> *n*-alkanes (20 μL, 1 ppm) to the blank samples and process the samples according to the method in section 2.2. After pre-treatment, we detected the concentrations of *n*-alkanes in blank spiked samples by GC-MS, according to the formula: recovery efficiency = measured concentrations / theoretical concentrations \* 100% to calculate the recovery of blank spiked samples. The blank spiked recovery experiments were repeated three times and the final recovery was

averaged over the three experiments.

**Modification:** We have added the procedure and details of the spiked recovery experiment in Section 2.5 (L133-139).

5. It is not clear how the alkanes were quantified. Was a calibration curve used, external standards, internal standards, isotopic dilution, etc.? Explain the details of the quantitative analysis.

**Reply:** Thank you for your questions. We quantified the concentration of *n*-alkanes using the external standard method. Standard solutions of C<sub>8</sub>-C<sub>40</sub> *n*-alkanes with concentration gradients of 10 ppm, 1 ppm, 500 ppb, 100 ppb, 50 ppb and 10 ppb were prepared. The calibration curves is plotted with the concentrations of the standard solution as the abscissa axis and the corresponding chromatographic response obtained by GC-MS as the ordinate axis, the correlation coefficient of each individual calibration curve is greater than 0.99. Finally, we quantified the concentration of *n*-alkanes from the peak area and the calibration curve.

**Modification:** We have added the methods and details of the quantitative analysis of *n*-alkanes in Section 2.4 (L122-127).

6. Lines 237-242, 283-285. It is well known that gasoline is mostly comprised of hydrocarbons in the C<sub>4</sub> to C<sub>10</sub> range while diesel fuel consists of C<sub>8</sub> to C<sub>25</sub> hydrocarbons (Han et al., 2008; Schauer et al., 1999; Lough et al., 2005; Gentner et al., 2012; Wang et al., 2005). Explain the discrepancy with the results obtained by the authors of the actual study.

**Reply:** Thank you for pointing this out. Our study focused on *n*-alkanes in PM<sub>2.5</sub>, the exhaust from gasoline and diesel vehicles is one of the sources of such *n*-alkanes (Wang et al., 2017). In fact, the composition of hydrocarbons in gasoline and diesel is quite different from the composition and distribution of particulate-bound *n*-alkanes in vehicle exhaust. Schauer et al. found that the composition of *n*-alkanes in particle phase from the exhaust of gasoline and diesel vehicles ranges from C<sub>18</sub>-C<sub>29</sub> and C<sub>15</sub>-C<sub>29</sub>, respectively (Schauer et al., 1999; Schauer et al., 2002). Therefore, we believe that the composition of particulate-bound *n*-alkanes in vehicle exhaust differs from the composition of hydrocarbons in gasoline and diesel.

7. Line 245. Why can road dust be a source of alkanes with >C<sub>34</sub>?

**Reply:** Thank you for your questions. We reconsidered the possible sources represented by factor 5 obtained from the PMF model analysis. As the *n*-alkanes from this source do not have an obvious regularity in composition, the *n*-alkanes have homologs with a high proportion of species in both the low and high carbon chains. Based on the results of previous studies, we found that *n*-alkanes with ≥C<sub>34</sub> may come

from road dust (Daher et al., 2013) and biogenic source (Liebezeit et al., 2009), road dust is one of the sources of particulate-bound *n*-alkanes (Anh et al., 2019). Therefore, we infer that factor 5 may be a mixed source of road dust and biogenic emissions.

**Modification:** We have modified the analysis and extrapolation of the corresponding sources for factor 5 (L264-268).

8. Lines 257-258 and 267-270. The authors assume that temporal distribution of LMW and HMW alkanes is explained by the behaviour of  $C_{max}$  and WNA%, however, they do not consider the effect of temperature or the mixing layer height. How do these variables affect the temporal behaviour of the alkanes?

**Reply:** Thank you for pointing this out. We reconsidered and added the effect of temperature and atmospheric mixing layer height on the temporal behavior of *n*-alkanes. Temperature affects the concentration of *n*-alkanes by influencing gas-particle partitioning, the gas-particle partitioning varies with the change of temperature. When the temperature is lower in winter, gaseous *n*-alkanes are more likely to partition into particles with the higher partition coefficient of gas-particle partitioning (Lyu et al., 2016; Wick et al., 2002). Therefore, the increase of LWM *n*-alkanes concentration in winter also affected by temperature. The mixing layer height influences the concentration of *n*-alkanes by affecting the particulate matter, it's shown that the mixing layer height is correlated with the concentration of particulate matter and the peak concentration of particulate matter increases as the mixing layer height decreases (Wagner et al., 2017). The atmospheric mixing layer height in Beijing has obvious seasonal characteristics, showing low in winter and high in summer (Wang et al., 2020; Tang et al., 2016). Therefore, the increased concentrations of PM<sub>2.5</sub> and *n*-alkanes in winter were influenced by the mixing layer height.

**Modification:** We have added the the effect of temperature and atmospheric mixing layer height on the temporal behavior of *n*-alkanes in Section 4.2 (L299-304, L307-310).

9. Lines 282-283. The reference by Simonet et al (2004). doi:10.1029/2004JD004565, does not mention alkanes in diesel.

**Reply:** Thank you for your corrections. Simonet et al. were not mention *n*-alkanes in diesel in this paper. We have cited the wrong reference in this part of the discussion.

**Modification:** We have revised the discussion in this part and corrected the incorrect references (L316-318).

10. Lines 294-296. Show scatter plot corroborating this association.

**Reply:** Thank you for your suggestions, we have added scatter plot to corroborate the association between particulate-bound *n*-alkanes and PM<sub>2.5</sub>. As shown in Figure 8, excluding the sharp increase in the concentration of PM<sub>2.5</sub> during the haze pollution, we found a positive correlation between particulate-bound *n*-alkanes and PM<sub>2.5</sub>. Therefore, we believe that they have similar trends and that particulate-bound *n*-alkanes can be used as an indicator to reveal the source of PM<sub>2.5</sub>.

**Modification:** We have added the scatter plot as Figure 8 to further illustrate the relationship between particulate *n*-alkanes and PM<sub>2.5</sub> (L596).

11. Authors should incorporate atmospheric criteria pollutants and the association with alkanes.

**Reply:** Thank you for your proposals. We found that the association between particulate-bound *n*-alkanes and atmospheric criteria pollutants such as nitrogen oxides, sulfur dioxide, carbon monoxide and ozone has not been considered in the studies of *n*-alkanes in particulate matter by other researchers. Because there is no correlation between particulate-bound *n*-alkanes and these pollutants, the concentration, distribution and sources of particulate-bound *n*-alkanes are not directly influenced by these pollutants. *n*-Alkanes are associated with particulate matter such as TSP, PM<sub>10</sub>, and PM<sub>2.5</sub>, widely distributed in different particle sizes and mainly concentrated in fine particulate matter (Mirante et al., 2013; Wang et al., 2017). Therefore, only the association between the two was considered in our study.

12. Meteorology was not included. It is extremely important to describe the meteorological variables, for example to observe differences in temperature between the four seasons, as well as wind speed and wind direction in order to propose emission sources.

**Reply:** Thank you for your suggestions. We considered the influence of meteorology on the particulate-bound *n*-alkanes. Temperature and atmospheric mixing layer height affect the ambient concentrations of particulate-bound *n*-alkanes by affecting the gas-particle partitioning and the concentration of particulate matter, respectively, but do not affect the emission sources of particulate-bound *n*-alkanes. Studies have shown that wind speed and humidity do not have a direct effect on the source of particulate-bound *n*-alkanes (Owoade et al., 2012), but the difference of average wind speed between day and night would affect the concentration of *n*-alkanes by influencing the atmospheric diffusion conditions (Yao et al., 2009; Wehner et al., 2008). Wind direction is one of the factors affecting the seasonal differences in particulate matter and *n*-alkanes, the northwest wind in winter brought the polluted air masses from inland to Beijing, while the southeast wind in summer transported cleaner aerosols from oceans to here (Wei et al., 2020). We will add the analysis and discussion of meteorological impacts to the manuscript.

**Modification:** We have supplemented our discussion of seasonal differences in particulate-bound *n*-alkanes with the analysis of meteorological effects in Section 4.2 (L299-310).

13. Line 16. It should say 153 ng/m<sup>3</sup>

**Reply:** Thank you for pointing out this mistake, we will correct it.

**Modification:** We have modified the incorrect units for the concentration of particulate-bound *n*-alkanes (L16).

14. The PMF 5.0 user guide can not be a supplemental material. Authors should only cite it.

**Reply:** Thank you for pointing this out, we will cite the PMF 5.0 user guide rather than take it as a supplemental material.

**Modification:** We have corrected the citation to the PMF 5.0 user guide.

We would like to thank you again for taking the time to review our manuscript.

## Reference:

Anh, H. Q., Tran, T. M., Thuy, N. T. T., Minh, T. B., and Takahashi, S.: Screening analysis of organic micro-pollutants in road dusts from some areas in northern Vietnam: A preliminary investigation on contamination status, potential sources, human exposure, and ecological risk, *Chemosphere*, 224, 428-436, doi: 10.1016/j.chemosphere.2019.02.177, 2019.

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Yao, L., Li, X. R., Guo, X. Q., Liu, X. R., and Wang, Y. S.: Pollution Characteristics of n-alkanes in Atmospheric Fine Particles During Spring Festival of 2007 in Beijing, *Environ. Sci.* (in Chinese), 30, 589-593, doi: 10.13227/j.hjkk.2009.02.042, 2009.

## **Referee Comment 2 (from Anonymous Referee):**

The authors quantify C<sub>13</sub>-C<sub>40</sub> n-alkanes in Beijing from 2020-2021 on some days. The authors use a multivariate test to identify sources of these n-alkanes. The overall presentation of the data needs to be improved, and there is further justification needed to make air quality strategy statements based on n-alkane measurements. The following concerns need to be responded to before the article is suitable for publication.

### **Major Comments:**

Meteorological seasons are defined as December-February for winter, March-May for spring, June-August for summer and September-November for autumn, respectively. Why is November defined as winter in this study? Why are several months omitted? For a year-long study, I think these omissions and changes in definitions need to be explained, as the conclusions of this paper rely on the collection dates in the paper.

Page 5 Line 185: Were the statistical tests done for individual carbon numbers between day and night? For example, the concentrations during winter for carbon numbers > 22 look almost identical between day and night. There needs to be additional explanation for the statistical tests, and where they were significantly different or not.

Page 7 Lines 257-266 lines 273-275: To what extent is the LMW increase in winter due to a) the larger amount of PM<sub>2.5</sub> allowing for a greater condensable surface area for intermediate volatility compounds b) the lower temperature resulting in a reduced vapor pressure of all compounds, resulting in sufficiently low vapor pressure for LMW n-alkanes to partition to the condensed phase?

Page 8 lines 299-300: Figure 7 indicated emissions in vehicle exhaust gases and

through coal combustion contributed up to 72.4% of particulate n-alkanes, not PM2.5. Given that the correlation between particulate n-alkane and PM2.5 is quite low (Pearson's  $r = 0.313$ ), the conclusions regarding controlling PM2.5 concentrations are quite speculative. The concentrations of the sum of particulate n-alkanes are ~1 part per thousand of the total PM2.5. One cannot state that all of the PM2.5 scales based on a particular n-alkane source. In order to demonstrate vehicle exhaust as the key source of PM2.5, a greater survey of the literature will be required to make this claim. It would be clearer in Figure 6 to label the profiles as the interpretations (i.e. gasoline) rather than "Factor 1, factor 2, etc." The discussion to interpret each factor should be re-written accordingly to account for this difference.

**Minor Comments:**

page 1 line 16: Change "4.51-153 ng/m" to "4.51-153 ng/m<sup>3</sup>"

Page 1 line 24: Remove sentence "Air quality in Beijing needs to be improved." Or place in the beginning to provide motivation, or include context for this sentence.

Page 1 Line 27: Include citation, likely from a review of air quality in China.

Page 1 lines 35-36: Include citation for the PM2.5 concentrations listed.

Page 2 Lines 45-48: Define short-chain and long-chain by carbon number, as these descriptions are vague.

Page 2 line 47: Why are long-chain n-alkanes relatively stable in the environment and generally accumulate in particulate matter compared to short-chain n-alkanes?

Page 3 Line 85: Abstract and introduction say particulate matter was collected between 2020 and 2021, materials and methods state 2021 and 2022. Please correct.

Page 3 line 87: Change "population" to "populated".

Page 3 line 116: What was the temperature of the GC inlet?

Page 4 Line 139: Define PNA% here, not in line 142.

Page 4 line 145: The WNA% needs a clearer mathematical definition to make it clear that n are odd numbers only. This is stated in the text, but should also be stated mathematically.

Page 8 Lines 279-281: Suggest explanations, rather than state that these are the causes. Without meteorological data, you cannot state these explanations as fact.

Figure 1: Specify whether the data shown is during the day, night or an average of the two.

Figure 3: Define the dashed and solid lines in the figure.

Figure 4: State whether the concentration distributions are for the day, night, or an average of the two.

Figure 4 and 5: Remove minor ticks in x-axis, and "C"s in the tick labels. Since these are describing natural numbers, minor ticks are meaningless, and the x-axis label "Carbon number" makes the "C"s redundant.

Figure 4 and 5: Figure 5 seasons are mislabeled compared to Figure 4. Switch "spring" and "winter" labels in figure 5.

Figures 4 & 5: Error bars representing standard deviations of methods should be included for Figure 4 and 5.

Figure 6: Remove "C"s in the tick labels. The y-axis is confusing, as the sum of the bars will add to more than 100%. It seems that the percentages for each carbon

number equal 100% between all 4 seasons, but the y-axis is not intuitive to understand.

Page 7 Lines 243: The term “higher plants” is not defined in the paper. Please use clearer terminology, or define higher plants.

### **Author's response:**

#### **Dear referee:**

Thank you for your constructive comments and detailed revisions on our manuscript. We have carefully considered the suggestions and made some changes on the details of the manuscript accordingly. We have added to the analysis and discussion in the manuscript based on your suggestions, and provided more references support to our proposed views. We have provided additional explanations in the manuscript based on your major comments and revised some of the statements and diagrams according to your minor comments. We have tried our best to improve this manuscript in order to it can be published successfully, please find our itemized responses in below and our revisions/corrections in the re-submitted files.

### **Major Comments:**

1. Meteorological seasons are defined as December-February for winter, March-May for spring, June-August for summer and September-November for autumn, respectively. Why is November defined as winter in this study? Why are several months omitted? For a year-long study, I think these omissions and changes in definitions need to be explained, as the conclusions of this paper rely on the collection dates in the paper.

**Reply:** Thank you for your questions. In our manuscript, the definition of each season was influenced by the sample collection process. In the Chinese Lunar Calendar, November 7, 2020 is the Start of Winter and November is defined as winter. In addition, Beijing started centralized heating in November, and considered the possible impact of the heating process on air quality, we included this period in the sampling process and defined it as winter. Due to the impact of the epidemic control of COVID-19 in China during the sample collection process, samples were not collected in some months as planned so several mouths were omitted. Therefore, only samples from November and December 2020, March and April 2021, June and July 2021 and September and October 2021 were finally selected as the four seasons of winter, spring, summer and autumn respectively to encompass all seasons of the year.

2. Page 5 Line 185: Were the statistical tests done for individual carbon numbers between day and night? For example, the concentrations during winter for carbon numbers > 22 look almost identical between day and night. There needs to be additional explanation for the statistical tests, and where they were significantly different or not.

**Reply:** Thank you for your questions and suggestions. In our study, the statistical tests on the differences of particulate-bound *n*-alkanes between day and night were conducted by examining the mean concentrations of all homologs of *n*-alkanes in different seasons, without considering statistical tests on individual *n*-alkanes. We attempted a statistical test for individual homolog of *n*-alkanes between day and night in different seasons. Combined with the results of previous statistical tests, we found that there are significantly different in whole *n*-alkanes while the difference of individual homolog has no obvious pattern. As shown in Figure 5, fewer *n*-alkane homologs ( $C < 25$ ) with significant differences between day and night in winter and spring, more *n*-alkane homologs ( $C > 21$ ) have significant differences between day and night in summer and autumn.

**Modification:** We have done the statistical tests for individual homolog of *n*-alkanes and added the analysis results in Section 3.3 (L201-203).

3. Page 7 Lines 257-266 lines 273-275: To what extent is the LMW increase in winter due to a) the larger amount of PM<sub>2.5</sub> allowing for a greater condensable surface area for intermediate volatility compounds b) the lower temperature resulting in a reduced vapor pressure of all compounds, resulting in sufficiently low vapor pressure for LMW *n*-alkanes to partition to the condensed phase?

**Reply:** Thank you for pointing this out. We have tried to further analyze the reasons for the increase of LMW *n*-alkanes in winter and explain the extent. In fact, we think that the LWM *n*-alkanes increase in winter is influenced by the combination of factors such as increased emissions from sources, lower temperature affected the gas-particle partitioning lead to LMW *n*-alkanes entered the particle phase, and the higher PM<sub>2.5</sub> concentration. However, it's hard to quantify the extent of the contribution of these factors, so we only used these factors as the possible explanations for the increase of LMW *n*-alkanes in winter like other research (Lyu et al., 2016). Since we observed no significant increase in LMW *n*-alkanes when the PM<sub>2.5</sub> concentration increased significantly in hazy pollution days, we believe that the low temperature affects the gas-particle partitioning of *n*-alkanes leads to a greater increase in LWM *n*-alkanes.

4. Page 8 lines 299-300: Figure 7 indicated emissions in vehicle exhaust gases and through coal combustion contributed up to 72.4% of particulate *n*-alkanes, not PM<sub>2.5</sub>. Given that the correlation between particulate *n*-alkane and PM<sub>2.5</sub> is quite low (Pearson's  $r = 0.313$ ), the conclusions regarding controlling PM<sub>2.5</sub> concentrations are quite speculative. The concentrations of the sum of particulate *n*-alkanes are ~1 part per thousand of the total PM<sub>2.5</sub>. One cannot state that all of the PM<sub>2.5</sub> scales based on a particular *n*-alkane source. In order to demonstrate vehicle exhaust as the key source of PM<sub>2.5</sub>, a greater survey of the literature will be required to make this claim.

**Reply:** Thank you for your comments. We reanalyzed the correlation between particulate-bound *n*-alkanes and PM<sub>2.5</sub>, and used the scatter plot to reflect the

association between them after excluding the effect of the sharp increase in PM2.5 concentration during the haze pollution period. As shown in Figure 8, there is a positive correlation between particulate-bound *n*-alkanes and PM2.5 (Pearson's  $r = 0.618$ ,  $P < 0.01$ ). Although the total concentration of particulate-bound *n*-alkanes represents only accounts for one thousandth of the mass of PM2.5, many studies have shown that *n*-alkanes can be used as indicators to apportion the sources of PM2.5 (Cass, 1998; Xu et al., 2013; Zhao et al., 2016; Han et al., 2018). In addition, studies on PM2.5 sources analysis also proved that vehicle exhaust is a main source of particulate matter (Cheng et al., 2010; Andrade et al., 2012; Qi et al., 2018).

**Modification:** We have revised the analysis of the correlation between *n*-alkanes and PM2.5 in the manuscript (L181-183, L330-332), added the references and draw the scatter plot as Figure 8 (L596).

5. It would be clearer in Figure 6 to label the profiles as the interpretations (i.e. gasoline) rather than “Factor 1, factor 2, etc.” The discussion to interpret each factor should be re-written accordingly to account for this difference.

**Reply:** Thank you for your suggestions, We will revise the labeling of Figure 6 in the manuscript as you suggested and revise the discussion explaining each factor in order to explain the differences between the factors more clearly.

**Modification:** We modified the Figure 6 by labeling the profiles as the interpretations (L592), revised the discussion about explaining the differences in factors (L247-268).

#### **Minor Comments:**

1. page 1 line 16: Change “4.51-153 ng/m” to “4.51-153 ng/m<sup>3</sup>”

**Reply:** Thank you for pointing this out, we will correct it.

**Modification:** We have modified the incorrect units for the concentration of particulate-bound *n*-alkanes (L16).

2. Page 1 line 24: Remove sentence “Air quality in Beijing needs to be improved.” Or place in the beginning to provide motivation, or include context for this sentence.

**Reply:** Thank you for your suggestions, we decide to remove this sentence.

**Modification:** We have removed this sentence (L24).

3. Page 1 Line 27: Include citation, likely from a review of air quality in China.

**Reply:** Thank you for your suggestions, we will add the citation.

**Modification:** We have added the citation (L27).

4. Page 1 lines 35-36: Include citation for the PM2.5 concentrations listed.

**Reply:** Thank you for your suggestions, we will add the citation.

**Modification:** We have added the citation for the PM2.5 concentrations listed (L37).

5. Page 2 Lines 45-48: Define short-chain and long-chain by carbon number, as these descriptions are vague.

**Reply:** Thank you for pointing this out, we will add the definition.

**Modification:** We have added the definition of carbon number for short-chain and long-chain *n*-alkanes, and added description of the length of the carbon chain (L46-48).

6. Page 2 line 47: Why are long-chain *n*-alkanes relatively stable in the environment and generally accumulate in particulate matter compared to short-chain *n*-alkanes?

**Reply:** Thank you for your questions. *n*-Alkanes can participate in atmospheric chemical reactions, but the volatility and reactivity of the *n*-alkanes decrease as the carbon chain length increases (Aumont et al., 2013). Long-chain *n*-alkanes are more stable than short-chain *n*-alkanes because they are less likely to react in the environment (Chrysikou et al., 2009), so long-chain *n*-alkanes generally accumulate in particulate matter.

7. Page 3 Line 85: Abstract and introduction say particulate matter was collected between 2020 and 2021, materials and methods state 2021 and 2022. Please correct.

**Reply:** Thank you for pointing this out, we will correct it.

**Modification:** We have corrected the wrong state of particulate matter sample collection period in Section 2.1 (L86).

8. Page 3 line 87: Change “population” to “populated”.

**Reply:** Thank you for your suggestions, we will change this word.

**Modification:** We have changed the “population” to “populated” (L88).

9. Page 3 line 116: What was the temperature of the GC inlet?

**Reply:** Thank you for your questions, the temperature of the GC inlet was 290°C.

**Modification:** We have added the temperature of the GC inlet in Section 2.3 (L115).

10. Page 4 Line 139: Define PNA% here, not in line 142.

**Reply:** Thank you for pointing this out, we will modify it.

**Modification:** We have moved the definition of PNA% to line 154 (L153-154).

11. Page 4 line 145: The WNA% needs a clearer mathematical definition to make it clear that  $n$  are odd numbers only. This is stated in the text, but should also be stated mathematically.

**Reply:** Thank you for your suggestions, we will add the mathematical definition.

**Modification:** We have added a mathematical definition of “ $n$ ” as an odd number to the formula (L159).

12. Page 8 Lines 279-281: Suggest explanations, rather than state that these are the causes. Without meteorological data, you cannot state these explanations as fact.

**Reply:** Thank you for pointing this out, we decided to revise the explanation of this part to make it more reasonable.

**Modification:** We have rewritten the explanation of this part (L316-319)

13. Figure 1: Specify whether the data shown is during the day, night or an average of the two.

**Reply:** Thank you for your suggestions, the  $n$ -alkanes and PM<sub>2.5</sub> data shown in Figure 1 is the average of the day and night. We will add an explanation in the text and Figure 1.

**Modification:** We have added the explanation of the date in the text and Figure 1 (L178-179, L577).

14. Figure 3: Define the dashed and solid lines in the figure.

**Reply:** Thank you for pointing this out. The dashed line in Figure 3 represents the 50% percentage, which is for the convenience of comparing the proportion of LMW  $n$ -alkanes in different periods. The solid lines in Figure 3 shows the average proportion of LMW  $n$ -alkanes in day and night in different seasons. We will supplement the definition and explanation of dashed and solid lines in Figure 3.

**Modification:** We have added the explanation of the dashed and solid lines in Figure 3 (L585-586).

15. Figure 4: State whether the concentration distributions are for the day, night, or an average of the two.

**Reply:** Thank you for your suggestions, the concentration distributions of *n*-alkanes are the average of the day and night in different seasons, we will add an explanation in the text and Figure 4.

**Modification:** We have added the explanation of the concentration distributions in the text and change the title of Figure 4 (L193, L587).

16. Figure 4 and 5: Remove minor ticks in x-axis, and “C”s in the tick labels. Since these are describing natural numbers, minor ticks are meaningless, and the x-axis label “Carbon number” makes the “C”s redundant.

**Reply:** Thank you for your proposals, we will modify Figure 4 and Figure 5 according to your comments.

**Modification:** We have modified the x-axis in Figure 4 and Figure 5 (L587, L589).

17. Figure 4 and 5: Figure 5 seasons are mislabeled compared to Figure 4. Switch “spring” and “winter” labels in figure 5.

**Reply:** Thank you for pointing out this mistake, we will change the labels in Figure 5.

**Modification:** We have switched the “spring” and “winter” labels in Figure 5 (L589).

18. Figures 4 & 5: Error bars representing standard deviations of methods should be included for Figure 4 and 5.

**Reply:** Thank you for your suggestions. Figure 4 and Figure 5 aim to show the difference in the concentration of each homolog of *n*-alkanes in different seasons and between day and night, the concentration of each homolog of *n*-alkanes is the average concentration of the homolog in all samples during the corresponding time. Due to the variation in the concentration of each homolog of *n*-alkanes in different samples, the addition of error bars would affect the visual effects of Figure 4 and Figure 5. Error bars were not added to such figures in similar studies (Li et al., 2013; Wang et al., 2017), so we decide not to add the error bars for now.

19. Figure 6: Remove “C”s in the tick labels. The y-axis is confusing, as the sum of the bars will add to more than 100%. It seems that the percentages for each carbon



number equal 100% between all 4 seasons, but the y-axis is not intuitive to understand.

**Reply:** Thank you for your suggestions, we will change the tick labels on the x-axis and removed “C”s. Figure 6 represents the proportions of individual *n*-alkane homologs in the factors identified in the positive matrix, so the sum of the proportions of the five factors for each *n*-alkanes homolog is 100%.

**Modification:** We have modified the tick labels on the x-axis in Figure 6 (L593).

20. Page 7 Lines 243: The term “higher plants” is not defined in the paper. Please use clearer terminology, or define higher plants.

**Reply:** Thank you for pointing this out. We decided to use a more clearer terminology to describe this source of particulate-bound *n*-alkanes, replace “higher plants” with “terrestrial plants”.

**Modification:** We have changed the term “higher plants” to “terrestrial plants” .

We would like to thank you again for taking the time to review our manuscript.

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