Author's response

Characteristics of particulate-bound *n*-alkanes indicating sources of PM2.5 in Beijing, China

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Referee Comment (from Anonymous Referee):

The authors quantify C13-C40 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 PM2.5 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/m3"

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 and our revisions/corrections in below.

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 for individual homolog of *n*-alkanes and added the analysis results in Section 3.3 (**L201-203:** Statistical tests on the differences in concentration of individual homolog of *n*-alkanes between day and night in different seasons showed that fewer *n*-alkane homologs with significant differences in winter (C_{16}, C_{17}) and spring (C_{21}) while more *n*-alkane homologs (C>21) with significant differences in summer and autumn.).

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 PM2.5 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 PM2.5 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 PM2.5 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 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.

Reply: Thank you for your comments. We reanalyzed the correlation between particulate-bound *n*-alkanes and PM2.5, 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 (**L182-183:** As shown in Figure 8, correlation analysis indicated that the n-alkane and PM2.5 concentrations significantly positively correlated (p<0.01, r = 0.618).; **L330-332:** As shown in Figure 8, a significant positive correlation was found between the PM2.5 and n-alkane concentrations (p<0.01), so n-alkanes could be used as indicators of the sources of PM2.5 in the atmosphere.), added the references (**L333:** Cass, 1998; Kavouras et al., 2001; Bi et al., 2003; Xu et al., 2013; Zhao et al., 2016; Han et al., 2018; **L340:** Lv et al., 2020; Qi et al., 2018) and draw the scatter plot as Figure 8 (**L599**).

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 (**L596**), revised the discussion about explaining the differences in factors (**L247-268:** The PMF model can quantify the contributions of specific sources of n-alkanes relatively accurately. The n-alkane homolog contributions to each factor identified by the PMF model were used to analyze and identify the corresponding source. As shown in factor 1 of Figure 6, the n-alkanes with carbon chain lengths of C_{13}–C_{18} were dominant, which similar to the n-alkane homolog (C<20) pattern for emissions during coal combustion found by Oros and Simoneit and Niu et al. (Oros et al., 2000; Niu et al., 2005). Therefore, we concluded that factor 1 indicated n-alkanes emitted through coal combustion. Vehicle emissions are important sources of n-alkanes in particulate matter in urban areas (Lyu et al., 2019). n-Alkanes emitted by vehicles mainly have carbon-chain lengths <30 (Wang et al., 2017). However, there are marked differences between the patterns of n-alkanes emitted in particulates in gasoline vehicle and diesel vehicle exhaust gases. Cmax for n-alkanes is lower and the proportion of low-carbon-chain length n-alkanes is higher for particulates in diesel vehicle exhaust gases than gasoline vehicle exhaust gases. This feature can be used to distinguish

between n-alkanes emitted by diesel and gasoline vehicles in fine particulate matter (Fujitani et al., 2012; Yuan et al., 2016). As shown in Figure 6, the homologs with a higher proportion of n-alkane species in factor 2 are concentrated around C_{20}, while in factor 3 are concentrated around C_{27}. According to studies of sachuer et al. for gasoline and diesel vehicle emissions (Schauer et al., 1999; Schauer et al., 2002), we determine that factor 2 and factor 3 indicated diesel and gasoline vehicle emission sources, respectively. C_{27}–C_{38} (i.e., high-carbon-chain-length) n-alkanes made large contributions and low-carbon-chain-length n-alkanes made small contributions to the pattern for factor 4. Studies have shown that C_{26}–C_{36} n-alkanes are mainly emitted from cuticular waxes in terrestrial plants (Alves et al., 2001; Lyu et al., 2016), so we inferred that factor 4 indicated n-alkanes emitted by terrestrial plants. n-Alkanes do not have an obvious regularity in composition and there was no clear n-alkane homolog pattern for factor 5, but long-chain n-alkanes with carbon chain lengths ≥ 34 were dominant. We found that road dust is one of the sources of particulate-bound n-alkanes (Anh et al., 2019), n-alkanes with $\geq C_{34}$ may come from road dust (Daher et al., 2013) and biogenic source (Liebezeit et al., 2009). Therefore, we concluded that factor 5 may be a mixed source of n-alkanes from road dust and biogenic emissions.).

Minor Comments:

1. page 1 line 16: Change "4.51-153 ng/m" to "4.51-153 ng/m3"

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:** The n-alkane concentrations were 4.51–153 ng/m³).

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:** Ma et al., 2012).

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:** Beijing Ecology and Environment Statement, 2016-2021).

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 (**L46-47:** The products of reactions involving short-chain *n*-alkanes (C≤16) in the environment strongly contribute to secondary organic aerosol formation (Michoud et al., 2012). Long-chain *n*-alkanes (C>16) are relatively stable in the environment and generally accumulate in particulate matter (Chrysikou et al., 2009).).

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:** Fine particulate matter samples were collected between November 2020 and October 2021).

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:** Beijing is a typical heavily populated and traffic-intensive Chinese city).

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 (**L122:** Temperature of the GC inlet was 290°C).

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 (**L154:** WNA% and PNA% (petrogenic *n*-alkane ratio) can be used to assess the relative contributions of biological and anthropogenic sources of *n*-alkanes in particulate matter (Simoneit, 1985)).

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 (**L160:** ("*n*" is an odd number)).

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 (**L316-318:** Particulate-bound *n*-alkanes from vehicular emissions usually of low molecular weight (Lyu et al., 2019), diesel emissions have higher concentrations of particulate-bound *n*-alkanes with carbon chain lengths less than 25 (Schauer et al., 1999).)

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 PM2.5 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 (**L179-180:** The *n*-alkane and PM2.5 concentrations in the different seasons are

shown in Table 1 and temporal variations in the average concentrations between day and night are shown in Figure 1.; **L580**).

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 (**L588-589**).

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:** The average concentration distributions of C_{13}–C_{40} n-alkanes in the different seasons are shown in Figure 4.; **L591**).

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 (**L590**; **L592**).

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 (**L592**).

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 (**L595**).

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.

Reference:

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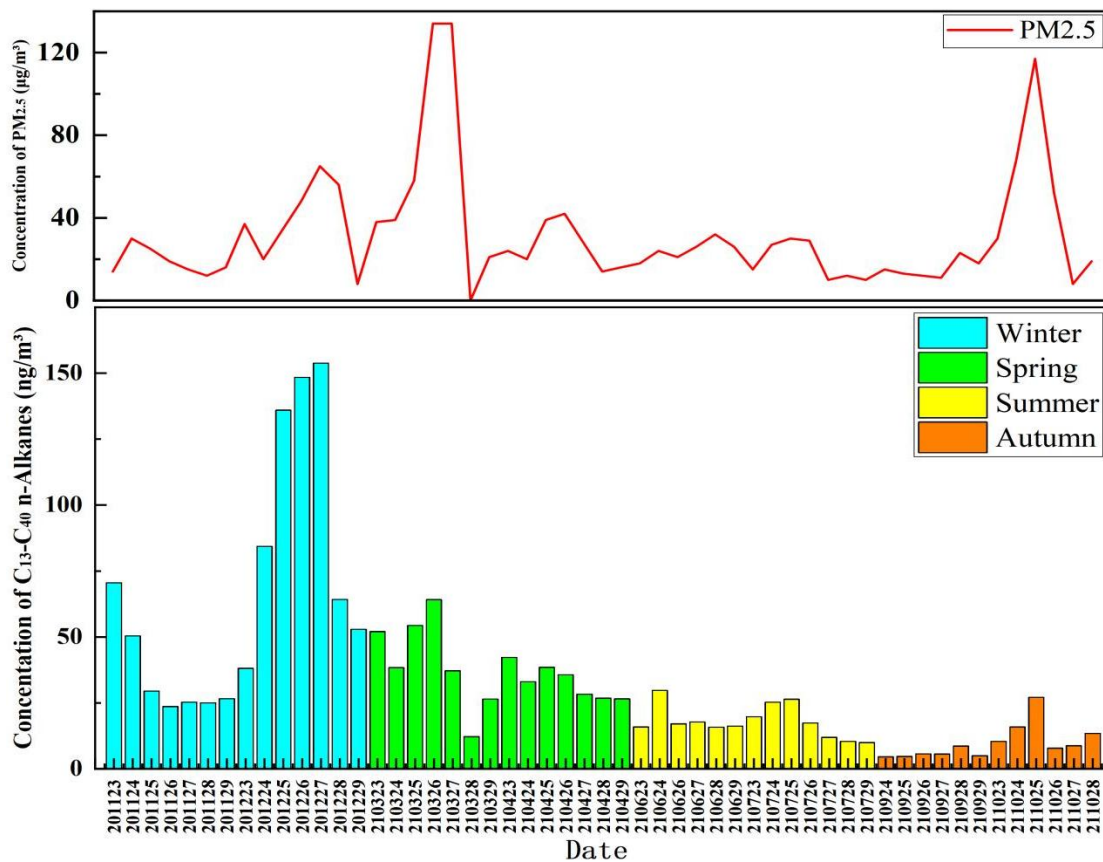
Figure 1. Temporal variations in PM2.5 and particulate-bound *n*-alkane concentrations during the sampling period in Beijing.

(The concentrations of C$_{13}$-C$_{40}$ *n*-Alkanes and PM2.5 are the average of the day and night).
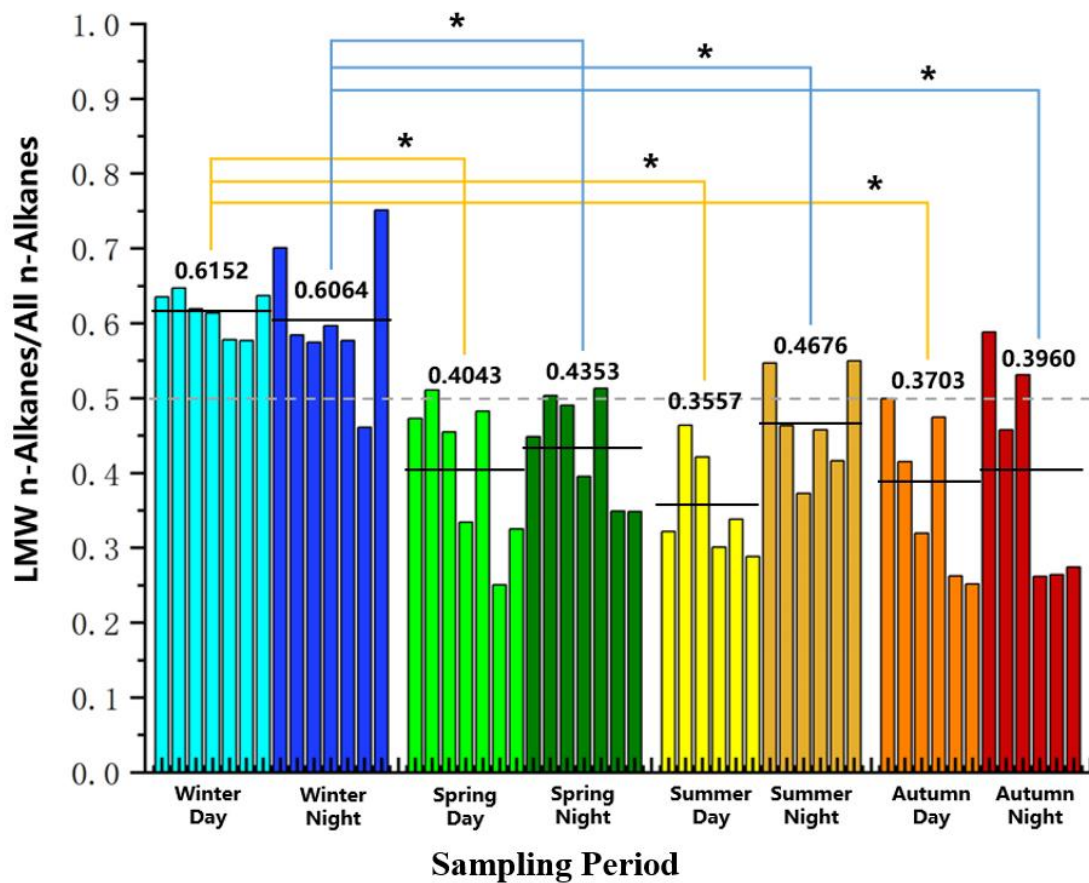
Figure 3. Contributions of low molecular weight *n*-alkanes in the day and night samples in the different seasons of Beijing.

(* indicates a significant difference, dashed line represents the 50% percentage, solid line shows the average proportion of LMW *n*-alkanes).
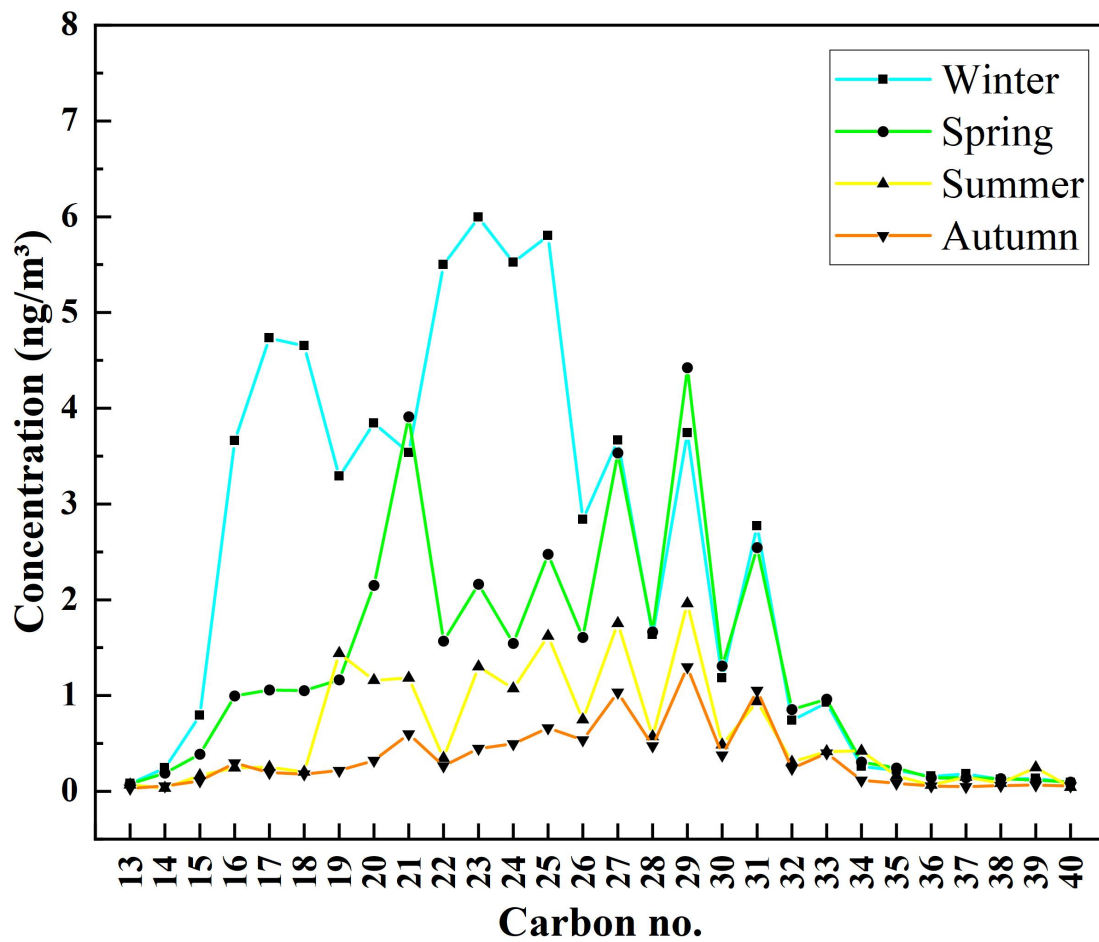
Figure 4. Average concentration distributions of the particulate-bound *n*-alkane homologs in the different seasons of Beijing.
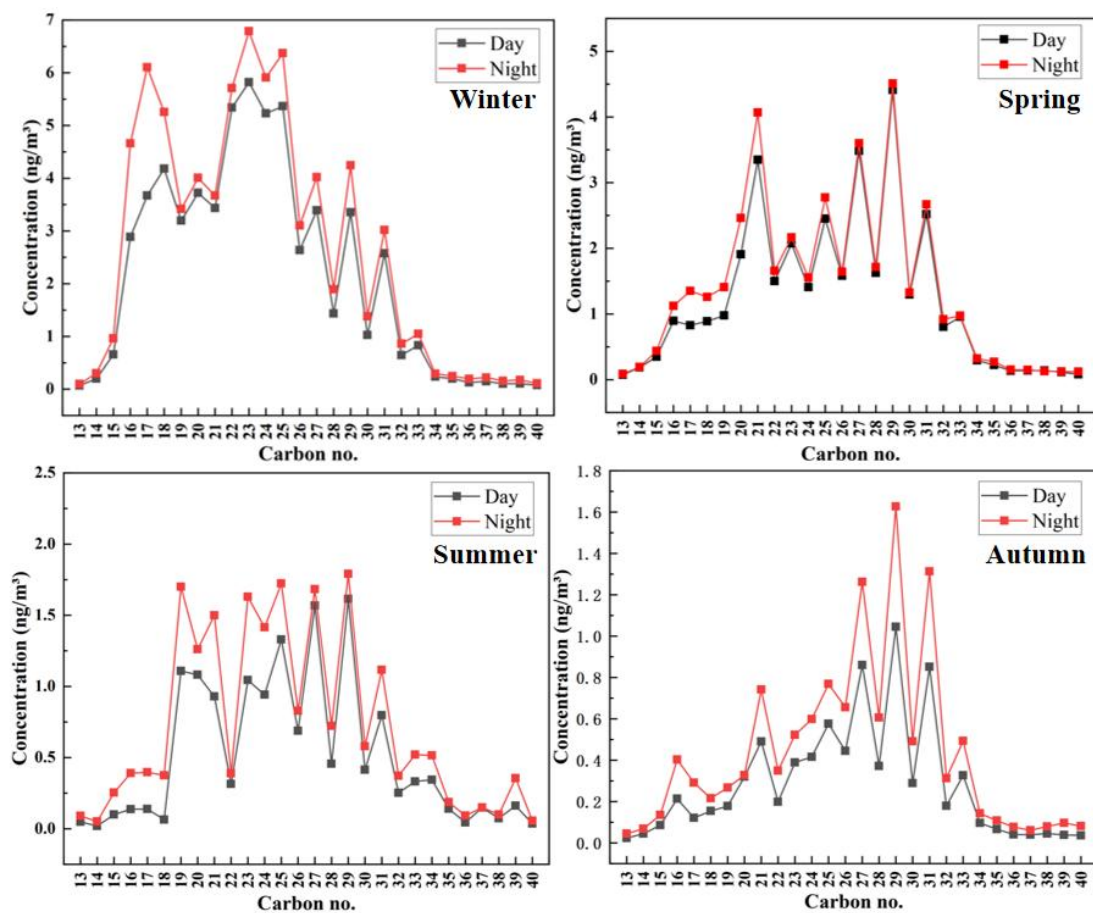
Figure 5. Concentration distributions of the particulate-bound *n*-alkane homologs in the day and night in the different seasons of Beijing.
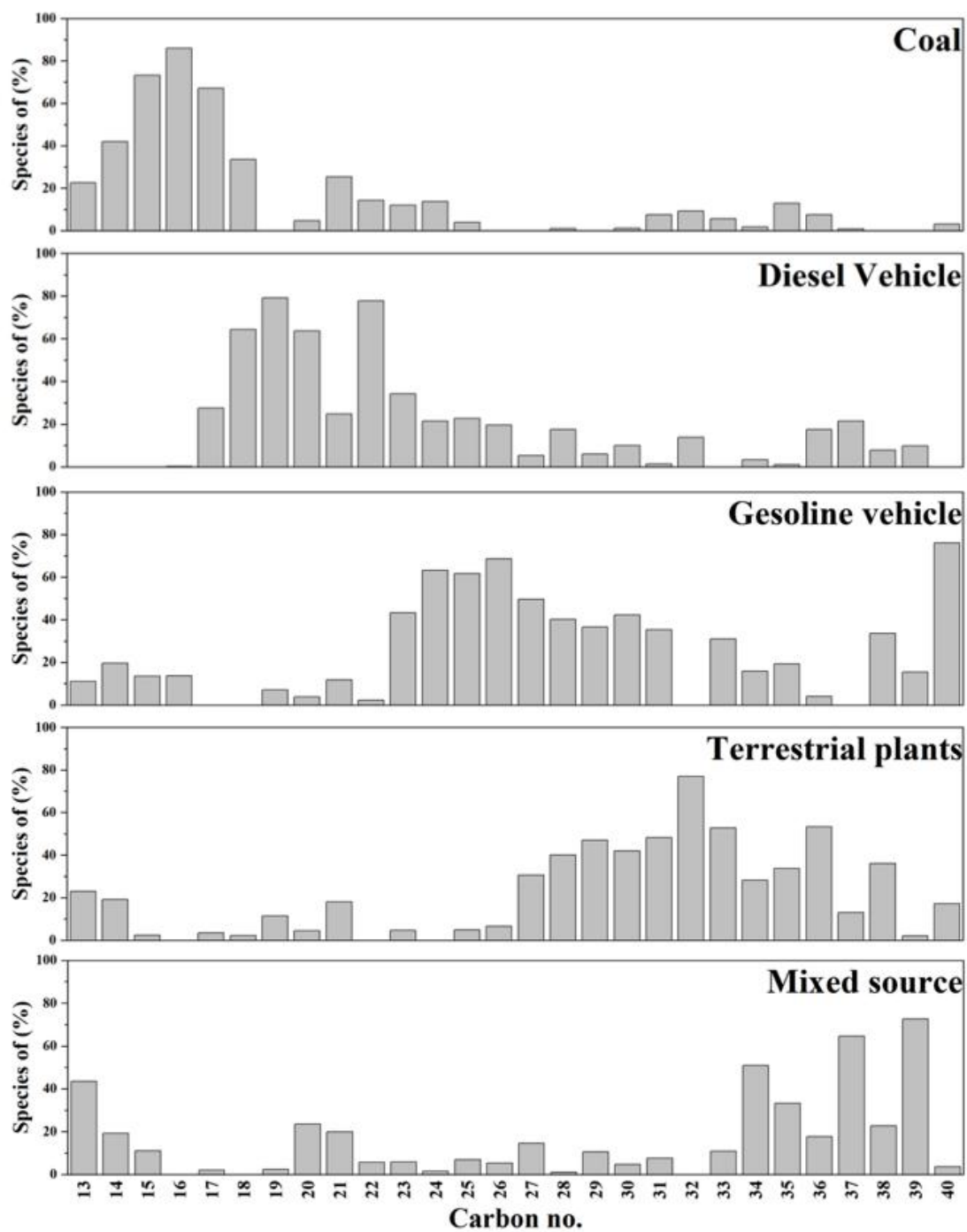
Figure 6. Proportions of the different *n*-alkane homologs in the factors identified by the positive matrix factorization model.
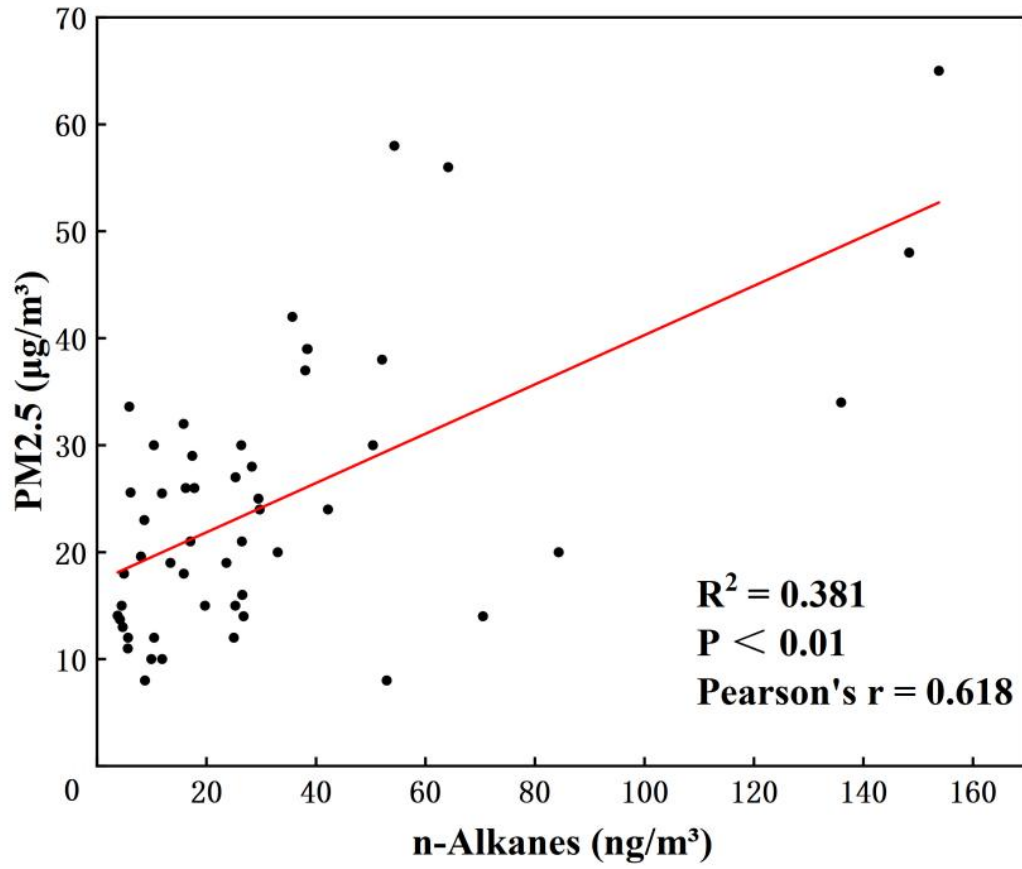
Figure 8. Association between particulate-bound *n*-alkanes and PM2.5 in Beijing.