Author's response

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

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Referee Comment (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 PM2.5. 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 PM2.5 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 C4 to C10 range while diesel fuel consists of C8 to C25 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 >C34?

8. Lines 257-258 and 267-270. The authors assume that temporal distribution of LMW and HMW alkanes is explained by the behaviour of Cmax 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/m3
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 and revisions/corrections in below.

1. The authors should further justify the interest in studying alkanes in PM2.5. 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≤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 PM2.5. The study of *n*-alkanes can help to better explain the source of PM2.5 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 PM2.5 in the introduction (**L50-51:** Particulate-bound *n*-alkanes play an important role in studying organic aerosols and the sources of the PM2.5) and added some references (**L44:** Chen et al., 2019; **L46:** Aumont et al., 2013; **L47:** Michoud et al., 2012; **L48:** Chrysikou et al., 2009).

2. Line 44. What are the human health effects of alkanes in PM2.5 mentioned by the authors? Provide the references.

Reply: Thank you for your questions. The *n*-alkanes in PM2.5 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_8-C_{16} *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 PM2.5 (**L44:** Chen et al., 2019).

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_8-C_{40} *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 2.2 (**L111-117:** Spiked recovery experiment was used to evaluate the recovery efficiency of particulate-bound *n*-alkanes. Mixed standard solution of C_8-C_{40} n-alkanes (20 μL, 1 ppm) was added to the blank samples, then the blank samples was pre-treat according to the same methods and the concentrations of *n*-alkanes was detected by GC-MS. The recovery was calculated based on the theoretical concentrations of *n*-alkanes standard solution and the measured concentrations of *n*-alkanes in the blank spiked samples. The blank spiked recovery experiments were repeated three times and the final recovery was averaged over the three experiments, the extraction recovery for *n*-alkanes range from 43.6% to 128%, the RSD for the concentrations of n-alkanes is 3.51%.).

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_8-C_{40} *n*-alkanes with concentration gradients of 10 ppm, 1 ppm, 500 ppb, 100 ppb, 50 ppb and 10 ppb were prepared. The peak area of each homolog of *n*-alkanes in standard solutions measured by GC-MS was used as the horizontal coordinate and the corresponding concentration was used as the vertical coordinate to draw the calibration curve, 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 (**L129-135:** Particulate-bound *n*-alkanes were quantified by external standard method. We prepared standard solutions of C_8-C_{40} *n*-alkanes with concentration gradients of 10 ppm, 1 ppm, 500 ppb, 100 ppb, 50 ppb and 10 ppb. The calibration curves is plotted with the concentrations of the standard solution as the vertical coordinate and the corresponding peak areas obtained by GC-MS as the horizontal coordinate, the correlation coefficient of each individual calibration curve is greater than 0.99. The concentrations of particulate-bound *n*-alkanes were finally quantified by the peak areas of the samples and the calibration curves.).

6. Lines 237-242, 283-285. It is well known that gasoline is mostly comprised of hydrocarbons in the C4 to C10 range while diesel fuel consists of C8 to C25 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 PM2.5, 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_{18}-C_{29} and C_{15}-C_{29}, 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 >C34?

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 ≥C34 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:** *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 ≥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.).

8. Lines 257-258 and 267-270. The authors assume that temporal distribution of LMW and HMW alkanes is explained by the behaviour of Cmax 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 PM2.5 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 (**L279-287:** 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 PM2.5 and *n*-alkanes in winter were influenced by the mixing layer height. In addition, 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).; **L297-300:** In addition, the seasonal distribution of *n*-alkanes is influenced by the temperature. The temperature in Beijing is high in summer and low in winter, 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 proportion in winter also affected by temperature.)

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:** 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).).

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 PM2.5. As shown in Figure 8, we found a positive correlation between particulate-bound *n*-alkanes and PM2.5. 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 PM2.5.

Modification: We have added the scatter plot as Figure 8 to further illustrate the relationship between particulate *n*-alkanes and PM2.5 (**L599**).

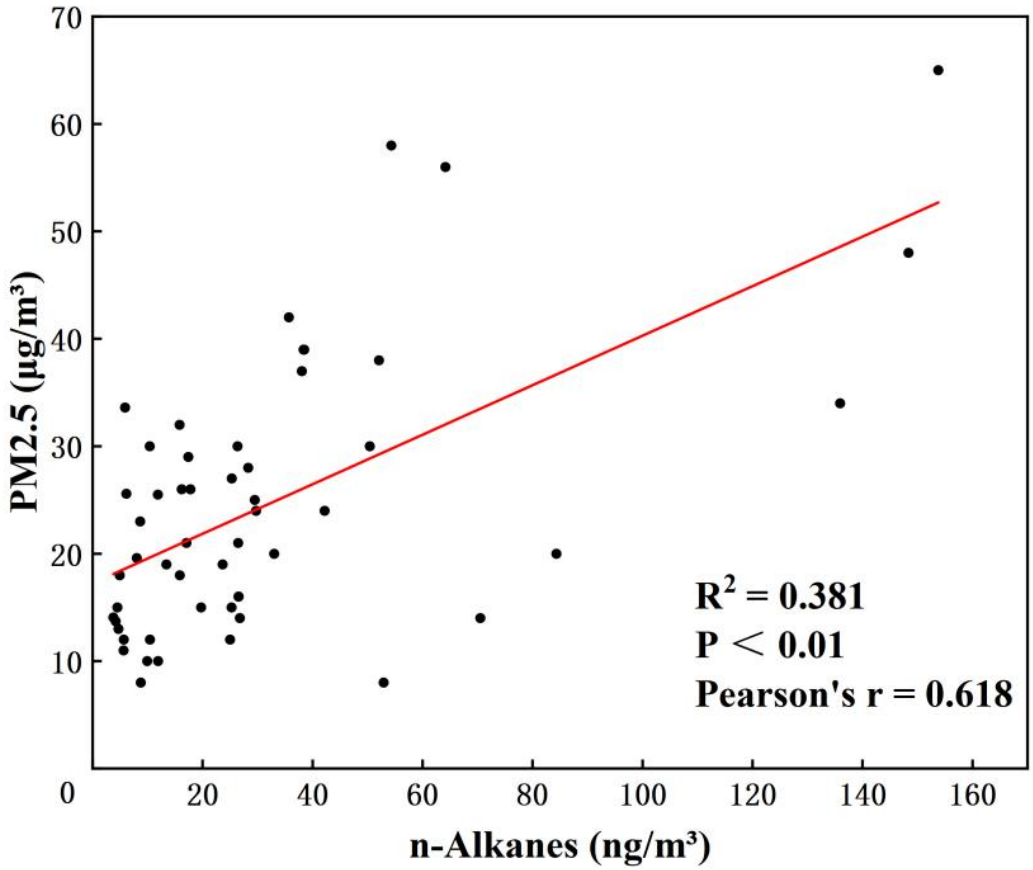

Figure 8. Association between particulate-bound *n*-alkanes and PM2.5 in Beijing.

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 their 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, PM10, and PM2.5, 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 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 (**L279-287:** 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 PM2.5 and *n*-alkanes in winter were influenced by the mixing layer height. In addition, 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).; **L297-300:** In addition, the seasonal distribution of *n*-alkanes is influenced by the temperature. The temperature in Beijing is high in summer and low in winter, 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 proportion in winter also affected by temperature.).

13. Line 16. It should say 153 ng/m3

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:** The *n*-alkane concentrations were 4.51–153 ng/m^3).

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:

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