Response to reviewers

Reviewer comments are in black ***italic*** type. Author responses are indented and in normal font labeled with [R]. Line numbers in the responses correspond to the revised manuscript with track-changes. Modifications to the manuscript are in *italics*.

Reviewer #1

Comments:

This manuscript describes mobile measurements of PM mass and composition, inorganic gases, and organic vapors on haze and non-haze days in Beijing. I like the study design, which focuses on quantifying the broad spatial patterns by repeatedly driving a ring road. This is in contrast to many previous mobile sampling studies that focused on obtaining neighborhood-level details at high spatial resolution.

However, I have some major criticisms that need to be addressed.

[R0] We thank the reviewer for the valuable feedback and constructive suggestions. Detailed responses are given below.

Specific comments:

(1) Amount and representativeness of data: The analyses (all figures except Fig 4) rely on only two days of data (November 14 and 18, 2018). Additionally, the authors primarily discuss midday concentrations on those days. For example, Fig 1 show data from the midday drives between 11:00 am and 12:30 pm local time. Since each drive takes around 70 minutes, this means that the majority of the analysis focuses on one or two drives on each of two days. The authors claim some large conclusions (they imply that their results are representative of all haze days and all clean days). They therefore need to show more than a small slice of data on two days. The results they present here are for two days, and therefore not necessarily representative of broader conditions in Beijing. A revised version of the manuscript should include analysis from multiple clean and haze days to get a better sense of how robust the results are.

[R1] As replied in [R4] of Reviewer #3, the day-to-day or diurnal variations of particle composition clearly present during the mobile campaign (Figure S3). Averaging the data for the whole measurement period or all clean days would smooth out the spatial variability. We therefore only presented the noon cycles to visualize the spatial variabilities of pollutants as examples. The spatial-distribution graphs are only for two days but the conclusions are made based on all the data measured during the campaign (and even data from another study in 2021).

To enrich the discussion, we have added a new graph as Figure 2. This graph shows the CV (i.e., spatial variability) distributions of all clean-day cycles vs. the haze-day cycles for the mass fractions of major particle components. Despite of the day-to-day variations, the clean-day CV values are significantly greater than the haze-day for all time periods, supporting greater spatial variabilities of aerosol composition during the clean days. We only had one haze-day data during the 2018 campaign. To support the haze-day results, we have added the haze-day results from another campaign in 2021 in Beijing in Figure 2 and Figure S10. The data also show homogeneous distributions of particle composition and

featured correlation heatmaps of VOCs and OVOCs during the haze event that are similar to the results herein. Related discussion is added in Line 188-195 and Line 307-309. The spatial-temporal distributions of VOCs are also presented in Figure 4 (now Figure 5), and we have already discussed it in the main text.

While writing this review I looked up a 2018 calendar. November 14 was a Wednesday, and November 18 was a Sunday. I am unfamiliar with the typical Chinese workweek or people's activity patterns in Beijing, but it seems like there is a good chance that most of this paper's analysis compares a single working day to a single non-working day.

[R2] We thank the reviewer for the good suggestion. But industrial or work activities do not vary much in China over the week compared with those in western countries. As indicated by the satellite observations of the tropospheric vertical column density of NO_2 and from the near-surface observations of NO_x, the weekend effect is insignificant in China (Hayn et al., 2009; Wang et al., 2014). Previous measurements for air pollutants in Beijing show some weekend-weekday differences but within the measurement uncertainties (Sun et al., 2015; Liu et al., 2020). Haze conditions are often associated with meteorological conditions that favor the accumulation of pollution (e.g., stagnant and humid conditions).

(2) Interpretation of spatial homogeneity on the haze day: The authors need to provide readers with a better sense of meteorological conditions on the clean versus haze days, and how those conditions relate to their interpretation of the mobile measurements. My assumption is that the haze days have low wind speed and perhaps a low mixing height, whereas the non-haze days are windier and better mixed. That seems to be the case from the data shown in Figure S3, but the authors need to include some of that context in the manuscript. Since the haze day has lower wind speed and presumably poorer mixing, I would expect significant spatial variability, especially for primary emissions. I might even expect larger spatial gradients on haze than non-haze days because of poor dispersion. Instead, the authors explain the more homogeneous conditions on the haze day as a result of "regional transport." That doesn't make sense to me as an explanation, since the haze day seems to be a case of stagnant air where local emissions are trapped.

[R3] We agree with the reviewer that the haze days usually have low wind speed and perhaps a low mixing height and thus the local emissions are likely accumulated more locally. However, this does not mean greater spatial heterogeneity for the mobile measurements because the on-road measurements sample air from both urban background and instantaneous plumes. The haze pollution in NCP usually develops regionally, transports to Beijing from the south, and linger in urban Beijing for days before the northwesterly/northeasterly wind with high speed blows away the pollution (An et al., 2019). Studies show that regional transport could contribute 60-70% of $PM_{2.5}$ during severe haze events in Beijing. When background air makes a major contribution to the on-road concentration of the pollutants, the impacts of accumulated local emissions on spatial distributions are perhaps reduced and spatial homogeneity presents for those pollutants. Similar to our study in Beijing, a study in Zurich shows that more than half of PM_1 measured in Zurich during winter are not from local emissions due to thermal inversions, resulting in a lower local/measured ratio and a rather uniform distribution of pollutant concentrations and particle composition throughout the whole Swiss plateau region. To

clarify, we have revised the discussion in Line 178-187 as follows: "*Although stagnant conditions facilitate the accumulation of local emissions (e.g., vehicle emissions on the road), over 60% of the $PM_{2.5}$ mass in Beijing can be contributed by regional transport during the winter haze episodes (Sun et al., 2014; Wu et al., 2021). The predominant contribution of regional transport suggests similar sources of $PM_{2.5}$ in Beijing. Similar particle composition suggests a spatial chemical homogeneity at least on the megacity scale in terms of gas-to-particle equilibrium or partitioning as well as the heterogeneous or particle-phase production. The north-south difference in mass concentration is perhaps driven by the differences in atmospheric dilution on the intracity scale (Sun et al., 2016; Chen et al., 2020). The uniform spatial distributions of PM composition under haze conditions are similar to the observations in the metropolitan area of Zurich when thermal inversions occur over the Swiss plateau and secondary pollution is built up regionally (Mohr et al., 2011), highlighting stagnant metrological conditions as one of the key drivers of the city-scale chemical homogeneity*."

The local emissions seem to be significant. Figure 4 shows that there are strong enough local emissions on the clean day to replenish pollutant concentrations after the boundary layer rises in the morning (e.g., hydrocarbon concentrations are higher from 12-14 and 14-16 than from 10-12). Thus, if emissions were similar on the two days, one would expect a larger daytime increase in concentrations, not a flat profile. If the haze day was a non-working day (Sunday, see comment above), emissions would be very different, and would have a major impact on the temporal patterns.

[R4] As replied in [R6] for Reviewer #3's comments, the on-road measurements of hydrocarbons are largely affected by instantaneous vehicle plumes. Therefore, the hydrocarbon measurements herein do not represent urban background conditions. The greater concentrations of hydrocarbon in the afternoon than from 10-12 suggest less vehicle plumes that the mobile measurements captured form 10-12. This may be explained by the less traffic volume on the road. During the haze day, the stagnant condition may favor the mobile measurements to capture the high emitting plumes from 10-12 and therefore shows a rather flat profile. By contrast, for VOC and OVOCs that vehicles are not a significant source, their concentrations are affected by urban background concentrations. To clarify, we have revised the text in Line 279-294 as follows: "*The on-road measurements of hydrocarbons are largely affected by instantaneous vehicle plumes. The greater concentrations of hydrocarbon in the afternoon (2:00 p.m. to 4:00 p.m.) than in the earlier period (11:00 a.m. to 2:00 p.m.) suggest that the mobile measurements captured less vehicle plumes, which is consistent with the less traffic volume on the road. Their concentrations decrease first as the boundary layer develops, and then increase in the afternoon as the pollution accumulates in the boundary layer under non-haze conditions. Under non-haze conditions, the spatial variabilities of hydrocarbons vary significantly during the day. Their CV values are high in the morning and low in the afternoon. It is likely that the photochemistry and the better mixing conditions in the afternoon smooth out some of the spatial variabilities caused by on-road vehicle emissions (Mellouki et al., 2015; Karl et al., 2018). By contrast, their concentrations keep decreasing during the day under haze conditions, and the greater day-time concentrations of $\sum$ hydrocarbons than during the clean days are plausibly driven by the greater contribution of regional transport to on-road air and stagnant meteorological conditions that favour the accumulation of on-road*

vehicle plumes. Under non-haze conditions, the spatial variabilities of hydrocarbons vary significantly during the day. Their CV values are high in the morning and low in the afternoon. It is likely that the photochemistry and the better mixing conditions in the afternoon smooth out some of the spatial variabilities caused by on-road vehicle emissions. Under haze conditions, the spatial variability of hydrocarbons is slightly greater in the afternoon, which is probably because of the change of regional transport direction in the afternoon."

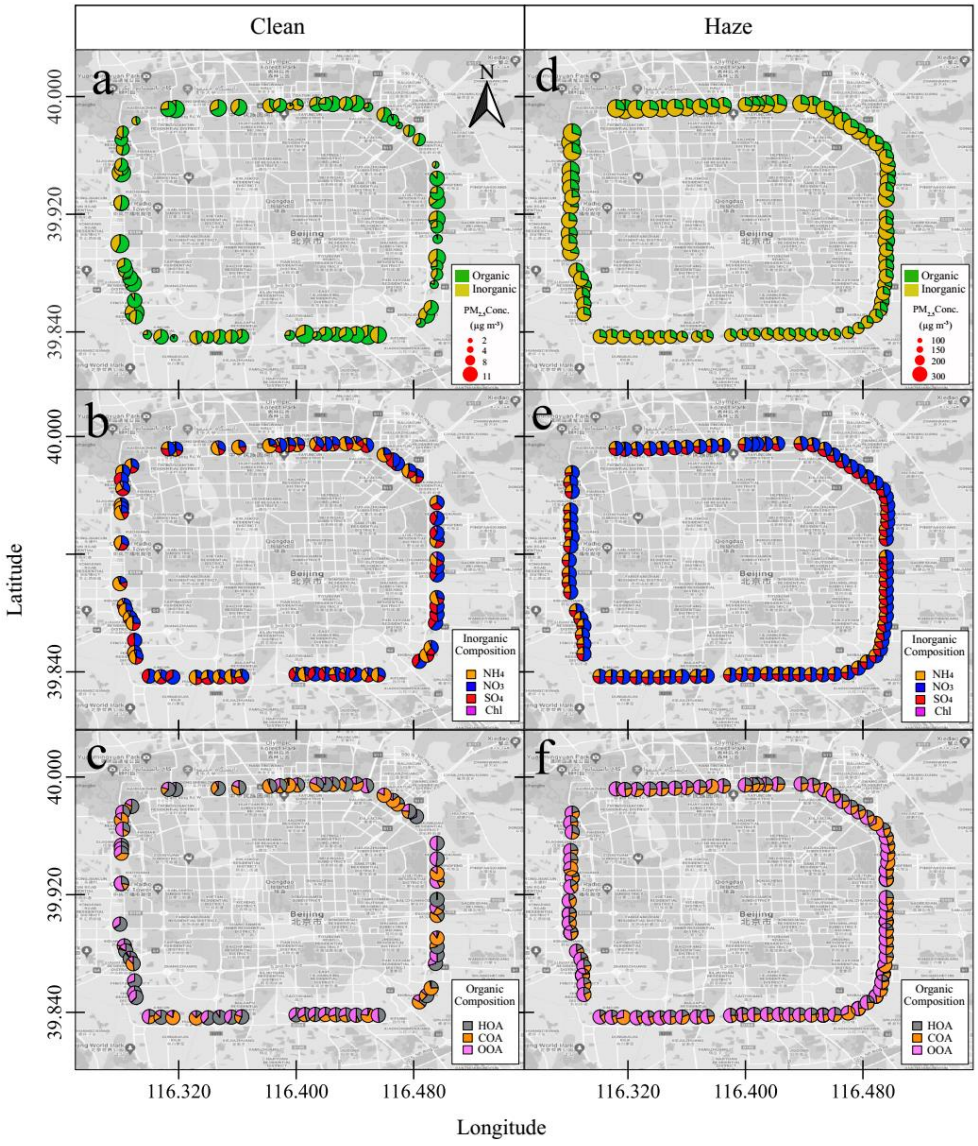
(3) With the exception of Figure 4, the authors do not show any temporal variations. I would expect that there is a lot to learn from comparing spatial patterns at different times of day (e.g., morning rush versus midday). Not showing this data in more detail seems like a major missed opportunity.

[R5] The temporal variations of the particle composition and the concentrations of gas pollutants and VOCs in Beijing have been investigated in tremendous studies and their sources have been extensively studied. We focus here only the spatial variabilities of these pollutants and their broad implications. With the addition of the new Figure 2 and its related discussions, the spatial patterns at different times are discussed in more detail in the revised version.

Additional comments:

(1) Figure 1a and 1d show the spatial variation of PM1 concentrations on two days. This figure is supposed to show that there is more variability on the clean day, however that is not obvious given the scaling of the symbols. The two days both look homogenous to me.

[R6] We have adjusted the lay out of the composition pies in the revised Figure 1 (below) to visualize the heterogeneity better. The pies are different along the 4th Ring Road in Figure 1a,b,c but rather uniform in Figure 1 d,e,f. Table 1 lists the CV values of the mass concentrations and the mass fractions of NR-$PM_{2.5}$ components, providing quantitative information for their spatial variabilities.



(2) Lines 129-130 note that most of the OA spatial variability on the clean day is due to variations in POA. However, the CV for OOA mass concentration (0.76) is similar to the CV for HOA (.79). This suggests that OOA is also variable. Though, as the authors note,

I would expect OOA to be more spatially homogeneous. Perhaps this high CV for OOA points to some misapportionment of other OA types as OOA.

[R7] We agree with the reviewer that OOA also shows a great spatial variability. But we don't think this is because of the misapportionment of POA as OOA. As described in Line 106-108, BBOA or CCOA were not resolved in this data set and were perhaps mixed with OOAs. Their contributions to OA are however expected to be small because of the emission control actions according to the previous results (Zheng et al., 2020; Duan et al., 2020). OOA can be contributed by many precursors and processes. It is not surprised to see a great spatial variability. We have clarified this part in Line 143-150 as follows: "*The spatial variations of the OA mass are attributed to both of POA and OOA. As shown in Fig. 1c, the mass fractions of POA factors such as HOA and COA show a large spatial heterogeneity with hot spots (mass fraction > 60%) in various segments of the 4th Ring Road. These hot spots are plausibly contributed by exhaust plumes from on-road vehicles and nearby restaurants that have not yet well mixed with urban background air. Similarly, the measurements in Pittsburgh show a significant spatial heterogeneity of primary carbonaceous components such as HOA, COA, and BC (Gu et al., 2018). The Pittsburgh study show less spatial variabilities of OOAs, whereas the CV value for the OOA concentration are high in Beijing during the clean day. This is perhaps because the precursors and formation pathways of OOAs are more complicated in Beijing than in Pittsburgh (Li et al., 2021; Yang et al., 2019)".*

(3) Fig 4 - Make it clean which panels are haze versus clear days. I assume that grey shading is for the haze days.

[R8] We have revised Figure 4 (now Figure 5) accordingly.

(4) Fig 4 - how many days are in each plot? Please be clear about how much data is being shown.

[R9] There are 7 non-haze days and 1 haze day of mobile measurements in winter in 2018. We have added Section A in the supplement and revised Figure S3 to show the mobile measurement periods.

Reviewer #3

Comments:

The paper reported the on-road mobile measurement results in megacity in China. It is interesting that homogeneous and heterogeneous spatial distributions were observed respectively for haze and clean days. The fine spatial resolution measurement provided a lot of information on localized sources, which is potentially useful for the development of future pollution control strategies. Overall, the paper is well written and logically organized. High-spatial resolution measurements is important yet scarce in China. As one of the pioneering studies in China, I recommend the paper be published subject to minor revision.

[R0] We thank the reviewer for the valuable feedback and constructive suggestions. Detailed responses are given below.

Specific comments:

1. Line 85, both mass resolution and time resolution of the ToF-ACSM sampling should be provided.

[R1] We have added those information in Line 93-97 as follows: *"Gas pollutants were detected by gas analyzers including NO_2 (Teledyne, T500U), NO-NO_x (Ecotech, EC9841A), SO_2 (Ecotech, EC9850A), CO (Ecotech, EC9830A), and O_3 (Ecotech, EC9810A) with a time resolution of 2 s. The chemical composition of NR-$PM_{2.5}$ was measured by an Aerodyne time-of-flight aerosol chemical speciation monitor (TOF-ACSM) with $PM_{2.5}$ lens and a capture vaporizer with a time resolution of 40 s and a mass resolution of about 400 (Zheng et al., 2020)"*.

2. Line 94 and Line 100, I don't think this is a good way to describe how the PMF results were derived and how the instruments were run during the campaign. Although experimental details had been published in the papers from the same group, readers may not have read the other ones and it is not their duty to do so. As an independent submission, at least all the necessary experimental details should be provided in SI to aid understanding of the whole manuscript.

[R2] We have added Section A as well as Figures S11 and S12 in the Supplement for the experimental details and the PMF analysis. The text in Line 104-106 has also been revised to guide the readers for the supplementary material.

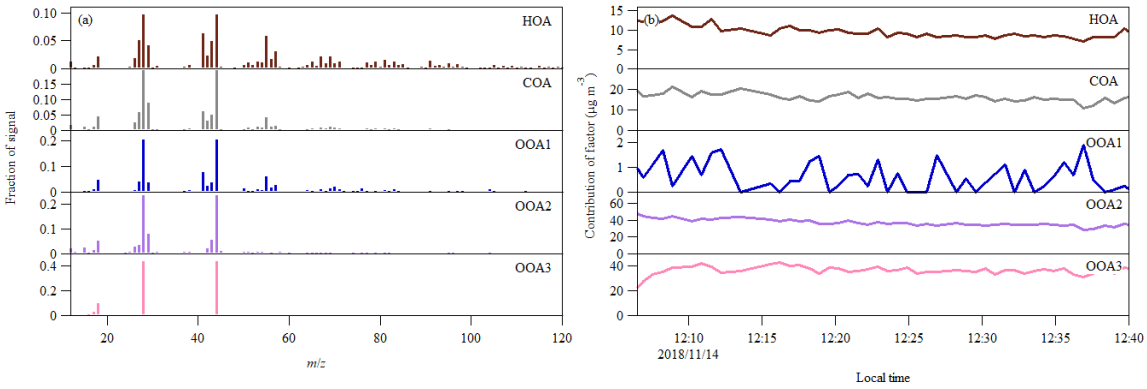


Figure S11. (a) Mass spectra and (b) time series of the OA factors identified by PMF (5-factor solution).

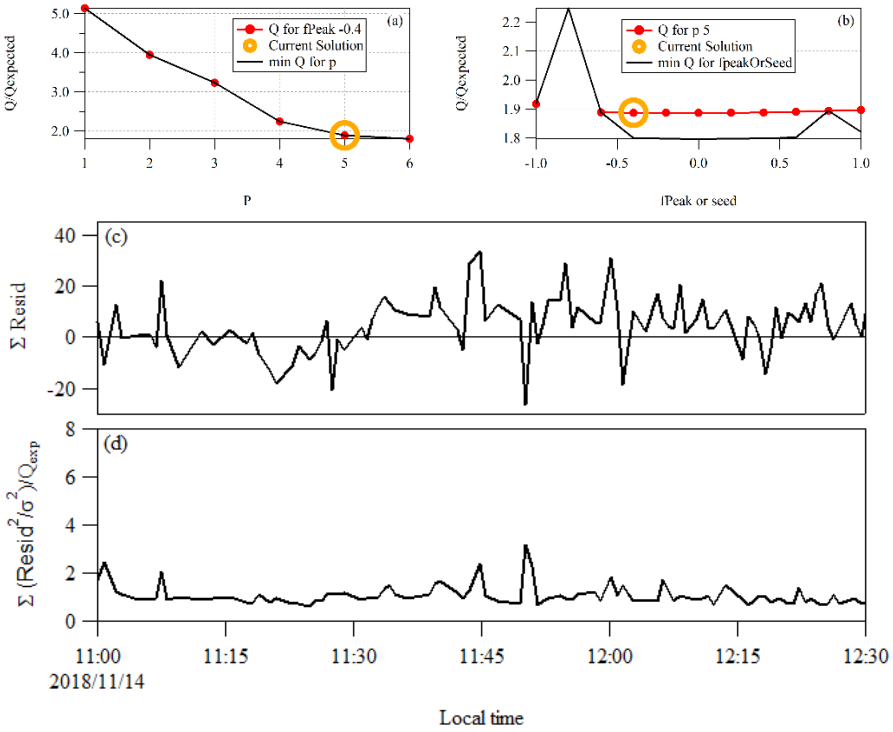


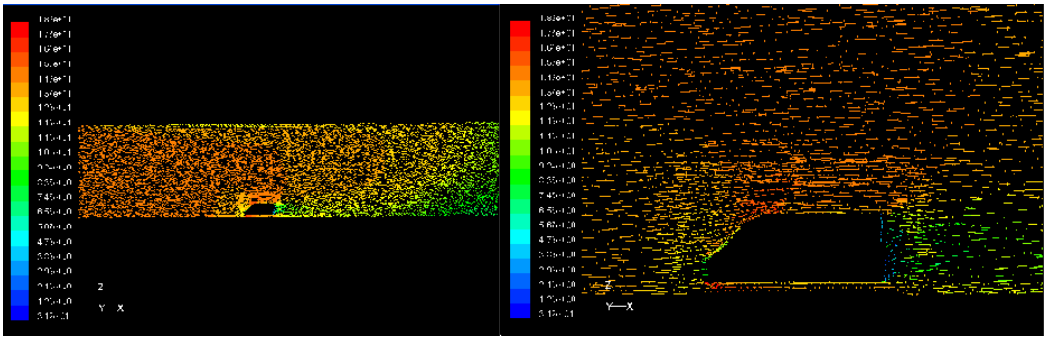
Figure S12. PMF diagnostics for Q/Q_{exp}, variance, and residuals. Residuals are shown for the example noon cycle during the haze day.

3. Line 125: The authors run the mobile lab on the highway, which is largely affected by the on-road vehicle emissions. Although self-contamination from the exhaust of the mobile lab could be eliminated, I'm not sure whether the data could represent the characteristic the specific area as shown on each pie in Figure 1. In another word, if the mobile lab was run on the road several meters away from the highway, would similar composition distributions be derived?

[R3] The sampling inlets were installed at the top front of the vehicle, 3.4 m above the ground (Figure X1). The wind speed was 0.5-2 m s^{-1} and sometimes 4-6 m s^{-1} during the

measurement period (Fig. S3). When the mobile lab ran for cycles on the 4th Ring Road, the $PM_{2.5}$ measurements by TOF-ACSM (40 s) may roughly represent a maximum area of 0.16 km^2 upwind (e.g., the wind is persistently perpendicular to the mobile path at a speed of 6 m s^{-1}). This means a bigger footprint of our measurements than the stationary measurements on the roadside (e.g., several meters away from the highway). By contrast, the measurements of gas pollutants (2 s) represent a rather small area.

Similar composition distribution between road-side and on-road measurements may be derived for pollutants that are well mixed in the urban background and not affected by vehicle emissions (Gentner et al., 2017). For example, the particle compositions measured on the 4th Ring Road were similar to those measured by a long time-of-flight aerosol mass spectrometer (LTOF-AMS) at the PKU roof station (Figure S3). To clarify, we have added some discussion in Line 171-177 as follows: "*The sampling inlets of the PKU mobile lab are located at 3.4 m above the ground, which may sample air from both of urban background and instantaneous plumes. The 40-s $PM_{2.5}$ measurements by TOF-ACSM may roughly represent a maximum area of 0.16 km^2 upwind when the mobile laboratory was run on the 4th Ring Road by cycles. The similar chemical composition along the road suggests relatively homogeneous spatial distributions of the mass concentration and composition of NR-$PM_{2.5}$ across the city under haze conditions. This is supported by the fact that the particle composition observed at the PKU roof site was similar to our mobile measurements (Figure S3)*". Figure S3 is also revised with the LTOF-AMS results.



Z（m）	0.16	0.27	0.39	0.47	0.51	0.89	1.09
V（m/s）	6.71	10.71	15.59	15.57	15.55	15.48	15.50

Figure X1. The wind field in front of the PKU mobile laboratory at a speed of 50-60 km h^{-1} modeled by FLUENT. The sampling height of Z refers to the height above the vehicle. The sampling inlet of PKU mobile laboratory was located at Z=0.4 m.

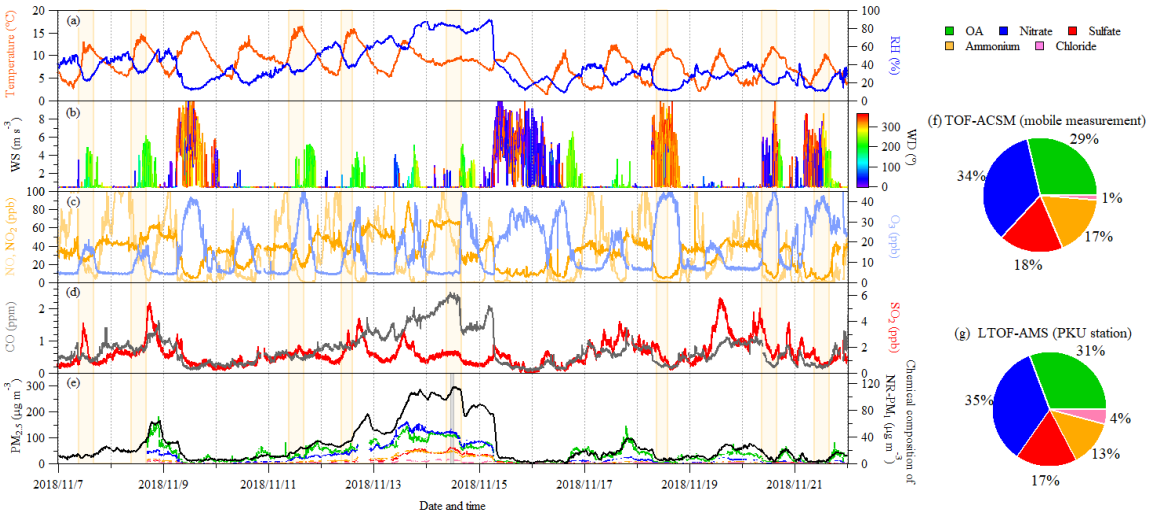


Figure S3. Time series of (a) temperature and relative humidity (RH), (b) wind speed (WS) and wind direction (WD), (c) NO, NO_2, and O_3, (d) CO and SO_2, (e) $PM_{2.5}$ mass concentration and chemical composition of NR-PM_1 measured by a long time-of-flight aerosol mass spectrometer (LTOF-AMS) at the PKU campus roof site during the entire mobile campaign in the winter of 2018. (f) and (g) particle composition of NR-$PM_{2.5}$ measured by TOF-ACSM in the mobile laboratory and particle composition of NR-PM_1 measured by LTOF-AMS at the PKU campus roof station during the time period of a 4th Ring Road cycle on November 14 (marked in grey in (e)), respectively. The yellow-shaded periods represent the periods having the mobile measurements.

4. Lines 125-145, it is interesting that on clean days great spatial variability of aerosol components was observed. What about the daily variation? I'm curious whether the observed spatial variation can well represent the local emission. Also, why the authors specifically present the results of the noon cycles instead of the average of the whole cycles for one day or during all clean days' sampling since the campaign lasted for around 2 weeks.

[R4] We have added the results of the chemical composition of non-refractory submicron particles measured by the LTOF-AMS at the PKU roof station to Figure S3. As shown in Figure S3, the day-to-day or diurnal variations of particle composition clearly present. Averaging the data for the whole measurement period or all clean days would smooth out the spatial variability. We therefore only presented the example noon cycles in Figure 1. To support our conclusion that the spatial variability of aerosol composition is greater during the clean days than during the haze day, we have added a new graph as Figure 2. This graph shows the CV (i.e., spatial variability) distributions of all clean-day cycles vs. the haze-day cycles for the mass fractions of major particle components. During the 2018 mobile campaign, we only had one haze-day data. In the revised manuscript, we have added another haze-day data that were collected on 21 January 2021 in Beijing. Despite of the day-to-day variations, the clean-day CV values are significantly greater than the haze-day values for all time periods.

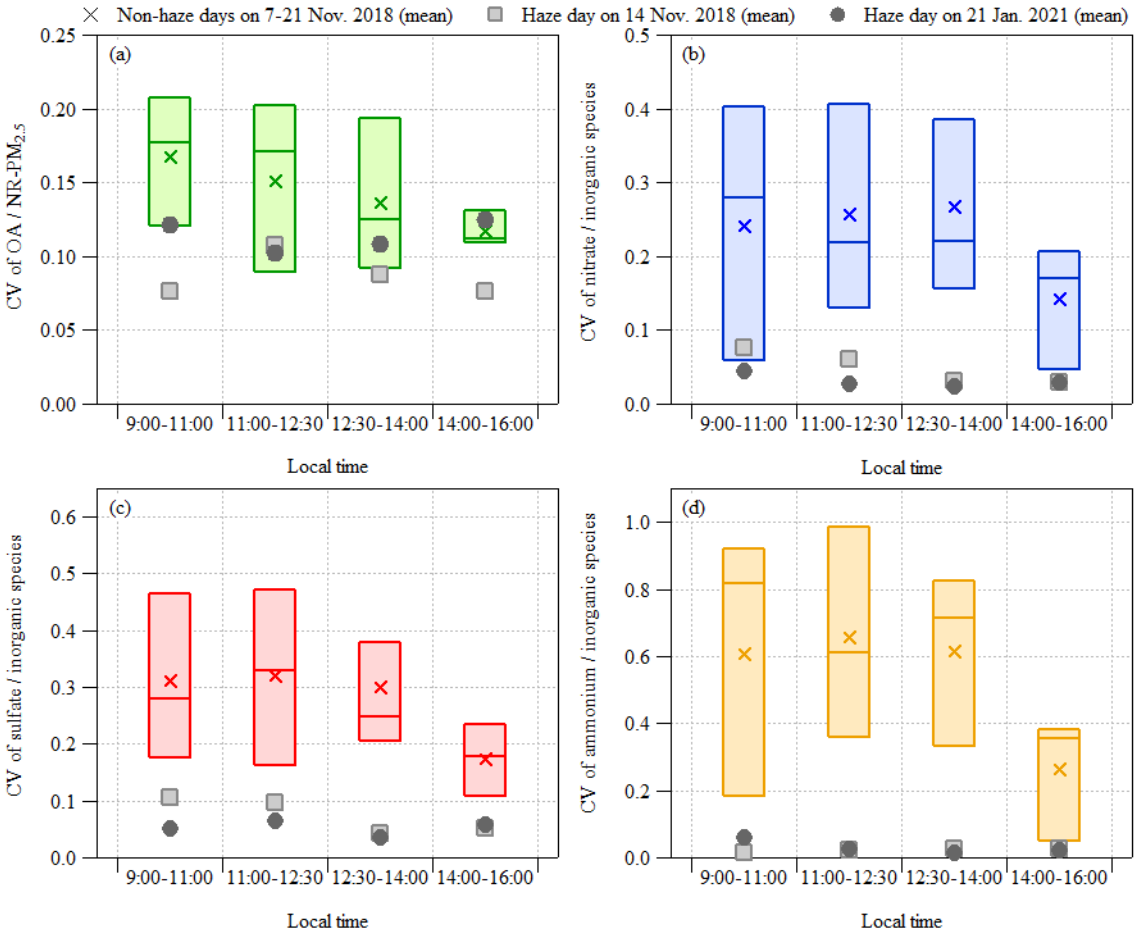


Figure 2. The CV values for the inorganic and organic mass fraction in NR-$PM_{2.5}$ for all cycles during the mobile campaign. The box plots show the 75th, median, and 25th percentiles.

5. Line 164: megacity scale? Or the authors meant the regional scale?

[R5] Yes, we meant megacity scale. The severe winter haze is typically a regional event. But we have only measured in Beijing and have no data to tell whether the particle composition were similar outside urban Beijing. To be clear, we have revised the text as follows: "*The similar particle composition may suggest a chemical homogeneity at least on the megacity scale*" in Line 171.

6. Line 248: Why hydrocarbons accumulated in the afternoon (12:00pm-14:00pm)? Hydrocarbons should decrease during the noon time because of photochemical consumption as observed from on-site measurements in literature.

[R6] We agree with the reviewer that photochemical consumption may lead to a noontime valley of hydrocarbon concentrations as observed in urban background site. The on-road measurements of hydrocarbons are however largely affected by instantaneous vehicle plumes. Therefore, the measurements herein do not represent urban background conditions for hydrocarbons. As shown in Figure 4, the median concentrations during 12:00-14:00 are lower than the morning concentrations but the data span in a wide range.

To clarify, we have revised the text in Line 279-290 as follows: "*The on-road measurements of hydrocarbons are largely affected by instantaneous vehicle plumes. The greater concentrations of hydrocarbon in the afternoon (2:00 p.m. to 4:00 p.m.) than in the earlier period (11:00 a.m. to 2:00 p.m.) suggest that the mobile measurements captured less vehicle plumes, which is consistent with the less traffic volume on the road. Under non-haze conditions, the spatial variabilities of hydrocarbons vary significantly during the day. Their CV values are high in the morning and low in the afternoon. It is likely that photochemistry and better mixing conditions in the afternoon smooth out some of the spatial variabilities caused by on-road vehicle emissions (Mellouki et al., 2015; Karl et al., 2018). By contrast, their concentrations keep decreasing during the day under haze conditions, and the greater concentrations of* $\sum$ *hydrocarbons than during the clean days are plausibly driven by the greater contribution of regional transport to on-road air and stagnant meteorological conditions that favour the accumulation of on-road vehicle plumes*"

7. From the discussion in Section 3.3, it seems variations of VOCs and OVOCs species are predominantly driven by on-road vehicles or high-emitting plumes. The running cycles on the 4th Ring Road cover different regions characterized by different functions, such as industrial area, residential area, etc., yet the VOC characteristics in different regions were not discussed in detail except vehicle emission. Could more information on local sources for different regions be derived from the measurements? After all, mobile emission is not the only emission source.

[R7] Yes, we agree with the reviewer that local sources can affect the on-road mobile measurements. For example, as we mentioned in Line 246 that the high T/B ratios in the south region of the 4th ring road may be explained by industrial plumes (e.g., from chemical plants, painting processes, or constructions involving evaporation emissions). Cooking exhaust plumes present as well as indicated by the COA hotspot in Figure 1c. We have clarified in Line 250-251 that mobile emissions are not the only source that influence the on-road air.

8. Line 540: Legend, non-haze and haze days should be denoted in Figure 4.

[R8] We have revised this figure (now Figure 5) to clarify the non-haze and haze-day results.

Reference

An, Z. S., Huang, R. J., Zhang, R. Y., Tie, X. X., Li, G. H., Cao, J. J., Zhou, W. J., Shi, Z. G., Han, Y. M., Gu, Z. L., and Ji, Y. M.: Severe haze in northern China: A synergy of anthropogenic emissions and atmospheric processes, Proc. Natl. Acad. Sci. U. S. A., 116, 8657-8666, https://doi.org/10.1073/pnas.1900125116, 2019.

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