Responses to the Reviewer

"Measurement Report: Four-year Variability and Influence of Winter Olympics and other Special Events on Air Quality in Urban Beijing during Wintertime" by Guo et al.

Referee #1

This study focuses on relating air pollutants concentrations in wintertime in Beijing to meteorological conditions and to temporary modifications in the anthropogenic emissions caused by "special events" like the Chinese New Year days, the COVID lockdown period or the 2021 Olympic games. The aim of this study is to infer information useful to guide future air pollution mitigation actions in Beijing based on an analysis of the effects of the restrictions during these special events. In this reviewer's opinion, the real value of this study is in contributing to understanding the atmospheric processes driving air pollution in this area of the world as a function of both meteorological factors and changes in the emissions. Besides, I consider comments like "the concentrations of $PM_{2.5}$ […] showed similar year-to-year variabilities, decreasing from 2019 to 2021 and then increasing in 2022" (Abstract, lines 25 - 27) somewhat misleading, because the year-to-year variability is interpreted as part of long-term trend triggered by the changes in the emissions, while here just four years are contrasted and the effects of meteo anomalies is big. In some parts of the paper, the simple Authors' judgement not the data is used to explain phenomena, like the rise in concentrations in 2022 shown as an effect of the resumed social activities. I encourage the Authors to first present the observed changes in atmospheric composition as a consequence of the variability in the meteorology, then show the effects of restrictions net of the variability caused by the changing meteo conditions. Although all these effects are analysed in detail in the main part of the text, the Abstract and the conclusions fail in presenting a fair description of such complexity.

We appreciate the valuable comments from the reviewer, which help to improve our manuscript. As suggested, we changed the "year-to-year variability" to "four-year variability" or "four-year variation" throughout the text (including the title) and clarified it in the main text (see the response to Specific Comment 4). We also modified the Abstract and Conclusion parts, making them more organized and cover the main points of our study. The changes of "Abstract" can be found in the response to Comment 2, and the revisions of "Summary and conclusions" can be found in the response to Comment 13. Besides, following the reviewer's suggestions, when discussing the variations of parameters, we first considered the influence of meteorological conditions, and then move to the effects of emissions and other sources, which should make our analysis easier to follow. The point-to-point responses to the comments are given below. The comments, our replies, and corresponding changes in this response document, revised manuscript and supplementary information are marked in black, blue, and green texts, respectively.

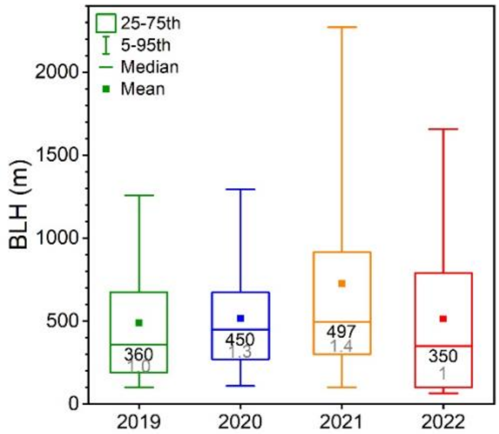
Specific comments:

1. The presentation of the results is well organized and supported by clear figures. It remains, however, a bit qualitative. The inter-period variability of the single aerosol and gas parameters is well monitored in the text, but it is often unclear whether when two variables share qualitatively the same pattern they also do quantitatively (see e.g., CO and NO_2 in Fig. 3 vs total OOMs in Fig. 4). Maybe, by normalizing the data, the magnitude of the changes will show up more clearly.

Response: Thanks a lot. Your suggestion is really helpful.

To evaluate to what extent those variables have changed, we normalized the median values to the lowest median value within each boxplot. For example, as shown in the modified **Fig. 2 e**, the median BLH in 2022 was the lowest, and it was chosen as the base value of 1, then median BLHs in 2019, 2020 and 2021 were ~

1.0, 1.3 and 1.4 times of that in 2022. Those figures are marked in gray adjacent to the median values in each box.



Modified Figure 2. (e) Boundary layer height (BLH) for different years (1st to 22nd January). The black value inside each box is the median value of corresponding parameter. The gray values are the normalized median values to the lowest median value in each subplot, which is used for quantitative variation comparison between different parameters.

Same normalizations have also been accomplished for almost all parameters except for temperature, fraction weighted oxygen number of OOMs (nO) and fraction weighted nitrogen number of OOMs (nN). Corresponding figures and captions, including **Fig. 2 – 4**, **Fig. 6 – 7**, **Fig. 9 – 10** and **Fig. 13 – 19**, have all been updated in the revised manuscript. Below are the caption changes:

"The black value inside each box is the median value of corresponding parameter. The gray values are the normalized median values to the lowest median value in each subplot, which is used for quantitative variation comparison between different parameters." (Line 954 – 956 for Fig. 2, Line 959 – 961 for Fig. 3, Line 966 – 968 for Fig. 4, Line 976 – 978 for Fig. 6, Line 982 – 984 for Fig. 7, Line 993 – 995 for Fig. 9, Line 999 – 1001 for Fig. 10, Line 1015 – 1017 for Fig. 13, Line 1022 – 1024 for Fig. 14, Line 1029 – 1031 for Fig. 15, Line 1034 – 1036 for Fig. 16, Line 1040 – 1042 for Fig. 17, Line 1045 – 1047 for Fig. 18, and Line 1051 – 1053 for Fig. 19)

Based on that, quantitative change comparison among different parameters can be made and some discussions in the manuscript have been revised, which will be shown in later responses.

2. Finally, the presentation of air ions and condensable vapors monitoring clearly provides new information on the processes governing SOA and $PM_{2.5}$ formations in and out periods of restrictions. Such results should be better emphasized in the Abstract.

Response: Thank you for your suggestions. As suggested, we have emphasized the results of atmospheric particles, ions and condensable vapors, and added a few sentences of pollutants during the special-event periods in the revised Abstract:

"Meanwhile, both the oxygen and nitrogen contents of oxygenated organic molecules increased year by year, implying that not only the oxidation state of those compounds increased, but also NO_x was involved more efficiently in their formation processes. With higher sulfuric acid concentrations and NPF frequencies in 2021 than in 2022, and with the lowest concentrations of background aerosols and the lowest ambient temperatures in 2021, $N_{1.3\text{-}3}$ was still the lowest in 2021. Unlike $N_{1.3\text{-}3}$, the ion concentrations in both 0.8-2 and 2-4 nm size ranges were higher in 2021 than in the other years." (Line 29 – 34)

"Therefore, CO, NO_x, SO_2, total OOMs, accumulation mode particles ($N_{100\text{-}1000}$), total $PM_{2.5}$ and its compositions were much lower compared with the Reference period." (Line 36 – 37)

"Then, the days after 4th February were chosen to explore the influence of special events, the non-event days within this date range in 2019 and 2021 was chosen as the "Reference period". Due to the favorable meteorological conditions together with reductions in anthropogenic emissions, there were basically no haze events during Beijing Winter Olympics. Therefore, CO, NO_x, SO_2, total OOMs, accumulation mode particles ($N_{100\text{-}1000}$), total $PM_{2.5}$ and its compositions had the lowest concentrations, while ions showed the highest concentration during the Olympics. Influenced by SO_2, condensation sink and sunlight, the sulfuric acid concentration was found to be comparable between Olympics and Reference period. Although there was also emission reduction during the COVID, especially for NO_x, the enhancement of secondary inorganic aerosol formation, together with unfavorable meteorological conditions, caused severe haze events during this period. Hence, CO, total OOMs and all $PM_{2.5}$ compositions during the COVID increased dramatically compared with the Reference period. Additionally, $N_{1.3\text{-}3}$ was almost at the same level during different periods, indicating that the special events had only little impacts on the new particle formation processes. These results provide useful information to the development of more targeted pollution control plans." (Line 39 – 45)

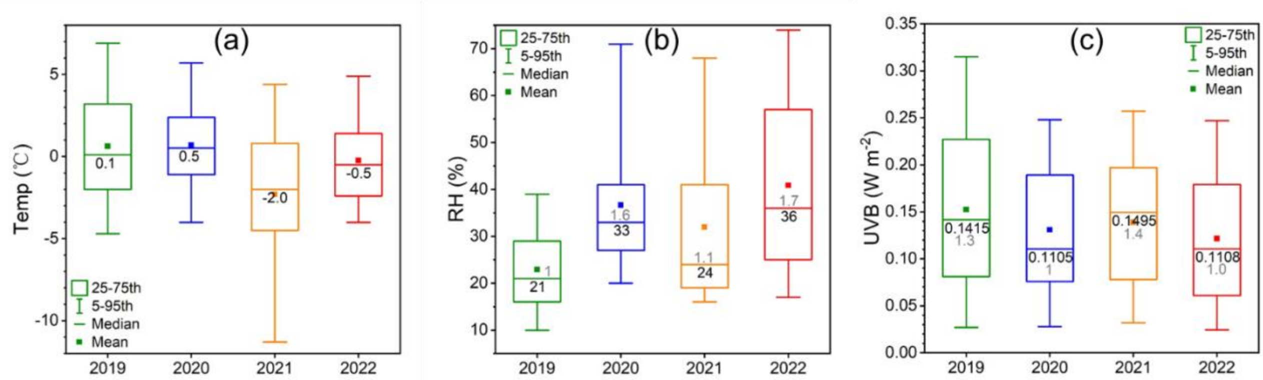
3. Section 3.1.1. This section does not discuss the difference in wind direction of year 2022 with respect to the previous years (Fig. 1). In addition, does the higher RH and lower UVB in 2020 and 2022 with respect to 2019 and 2021 (Fig. 2) has something to do with an anomaly in cloudiness/fog?

Response: Thanks a lot. Firstly, we added more discussion about the wind in the revised manuscript following the reviewer's suggestion:

"In 2022, the wind blew mostly from the southeast from midnight (00:00) to noon (12:00), but came from northeast from afternoon (13:00) to midnight (00:00)." (Line 182 – 183)

Secondly, the wintertime is typically very dry in Beijing with clear sky. Based on our observations, the median RH was under 40% in every year and also the 95th percentile was just over 70%. Therefore, there has not been any (at least substantial and long lasting) fog events. Even if the wintertime in Beijing is typically rather cloudless, differences in cloudiness could have had a minor effect on RH and UVB together with varying aerosol concentrations and meteorological conditions. The effect of wind direction to RH has been discussed in the text (Line 184 – 188). We have added additional descriptions of these in the text (Line 194 –196) as follows:

"… whereas the RH was higher in 2020 and 2022, which could be partly attribute to more southern and eastern winds in these two years. In addition, differences in cloud conditions could have a minor effect on UVB together with varying pollution conditions."



Modified Figure 2. (a) Temperature (Temp), (b) relatively humidity (RH) and (c) UVB for different years (1st to 22nd January). The black value inside each box is the median value of corresponding parameter. The gray values are the normalized median values to the lowest median value in each subplot, which is used for quantitative variation comparison between different parameters. Please note that for UVB, only daytime (08:00 – 16:00) dataset was used.

4. Line 165 (Section 3.1.1): clarify that the "year-to-year variability" refers to the 1st – 22nd Jan period.

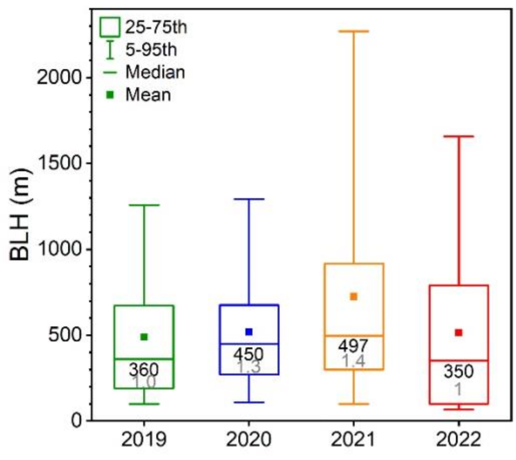
Response: Thanks. As suggested, we clarify the year-to-year variability (now the four-year variability) period in the revised manuscript as follows:

"Therefore, the period of four-year variability covered days from 1st to 22nd January of each year, lasting for 22 days." (Line 167 – 168)

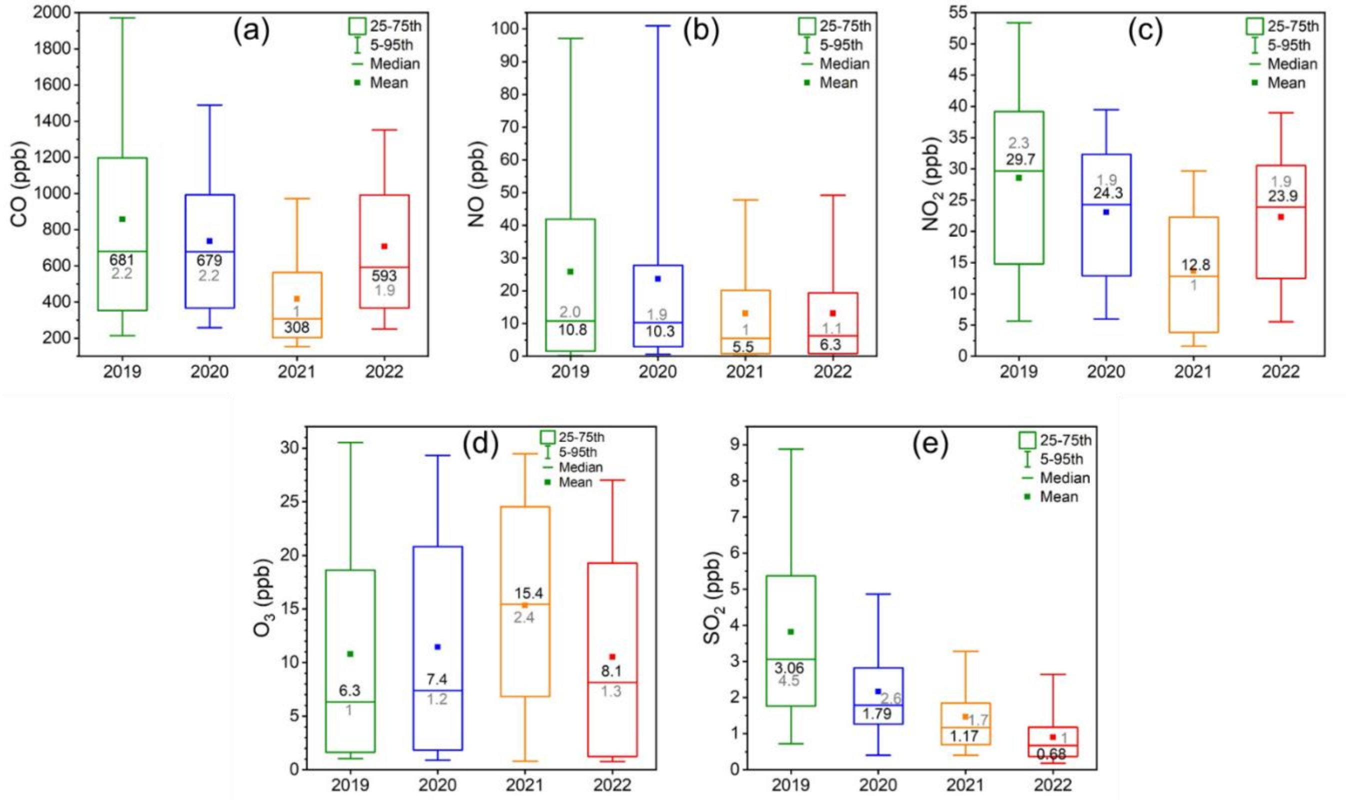
5. Section 3.1.2. The year-to-year variations in trace gas levels are interpreted only in terms of variations in the emissions (decline, "rebound", "recovery" etc) and completely neglect the meteo anomalies discussed in the previous section; why? Clearly the drop in concentrations of year 2021 must be influenced by the windy conditions favouring pollutant dispersion. In this reviewer's opinion, the drop of the mean NO_x and CO concentrations from 2019 to 2020-2022 can be attributed to changes in the emissions only in light of the different meteo conditions: in 2019, the winds were less frequently flowing from polluted sectors and still concentrations of NO_x and CO were higher than in 2020 and in 2021, hence this can footprint an actual change in traffic emissions.

Response: Thanks. Your suggestions really make sense. In the revised manuscript, we discuss the influence of meteorological conditions first. We modified our expression for the variations of CO, NO, NO_x and SO_2 in the revised manuscript as follows:

"The four-year variations of CO, NO and NO_2 were generally opposite to that of BLH, which decreased from 2019 to 2021 and rebounded in 2022, resulting in the lowest concentrations in 2021 (**Fig. 3 a, c**, median values are 308 and 12.8 ppb for CO and NO_2, respectively). Although the relative changes of them were larger than BLH, the variations of them should be partially attributed to the change in BLH, or the change in atmospheric diffusion capacity. Besides, the winds in 2021 came less frequently from polluted sectors, which might also play a role. We then compared the four-year variations of CO and NO_2 under $PM_{2.5}$ < 35 μg cm^{-3}, and found that their concentrations in 2022 increased substantially back to similar level as before COVID in 2020 (Fig. S6). This could indicate that the social activities have been recovered during the post COVID-19 period. Despite the influence of different wind direction and speed, SO_2 monotonically decreased from 2019 (median 3.06 ppb) to 2022 (median 0.68 ppb). Besides, this trend is quite different from that of BLH, indicating that the drop of SO_2 is not caused by variations of meteorology conditions, but by the decline in SO_2 emissions. Our result is in line with the long-term variation of SO_2 in China, where SO_2 decreased from 24.8 ppb in 2013 to 8.0 ppb in 2017 for the Beijing-Tianjin-Hebei area after the clean air action in 2013 (Wang et al., 2020)." (Line 204 – 216)



Modified Fig. 2. (e) Boundary layer height (BLH) for different years (1st to 22nd January). The black value inside each box is the median value of corresponding parameter. The gray values are the normalized median values to the lowest median value in each subplot, which is used for quantitative variation comparison between different parameters. Please note that for UVB, only daytime (08:00 – 16:00) dataset was used.



Modified Figure 3. Mixing ratios of (a) CO, (b) NO, (c) NO_2, (d) O_3 and (e) SO_2 for different years (1st to 22nd January). The black value inside each box is the median value of corresponding parameter. The gray values are the normalized median values to the lowest median value in each subplot, which is used for quantitative variation comparison between different parameters.

6. Lines 187-188: "The variations of CO, NO and NO_2 were generally opposite to that of the BLH, suggesting that atmospheric diffusion capacity could play some role". It is well established that the concentrations of primary pollutants are affected by the BLH, so why using the conditional tense?

Response: Thanks. We revised it accordingly.:

7. Lines 191-192: "This is likely associated with the recovery of social activities during the Post COVID-19 Period". This interpretation is actually what the paper is supposed to demonstrate, while here this statement is provided unsupported of meaningful data.

Response: Thanks, we have rephrased this sentence in the revised manuscript (see response to comment 5).

8. Lines 194-195: "Consequently, our results show that Beijing was successful in the reduction of SO_2, while some challenges still exist in the restriction of NO". The paper does not discuss the air quality targets in China, therefore this statement remains somewhat subjective and out of focus.

Response: Thanks. We have deleted this sentence in the revised manuscript as suggested.

9. Section 3.1.3. The sharp year-to-year change in OOMs concentration, their progressive increase in oxygen and nitrogen stoichiometric ratios and especially the gradual decrease with respect to $PM_{2.5}$ (Fig. S7) deserves further discussion. Apparently such changes can be useful to unmask the nature of SOA formation in this environment. Since $PM_{2.5}$ levels can be thought as an overlap between a background contribution and a locally-formed fraction, the decrease in OOM/$PM_{2.5}$ with time seems to be related to a sharp decrease in the local sources: such sources are apparently characterized by OOMs rich of carbon.

Response: Thanks for your suggestions. The changes of OOM concentration and its oxygen and nitrogen contents really need detailed discussion. As suggested, we modified and added the following text in the revised manuscript:

"Specifically, the median OOM concentration in 2019 (2.4×10^7 cm^{-3}) was ~ 8.6 times of that in 2021 (2.8×10^6 cm^{-3}) (**Fig. 4c**), while median and peak BLH values in 2019 (360 m and ~940 m for median and peak respectively) were only ~0.7 – 0.8 times of that in 2021 (497 m and ~1200 m for median and peak respectively) (**Fig. 2e and 2f**). Thus, the reasons behind this drastic yearly change of the OOM concentration cannot be solely attributed to changes in the BLH. Under polluted conditions, regional transportation of lower-volatility OOMs could result in higher concentration of OOMs (Guo et al., 2022). Besides, accumulation of precursor VOCs, and possibly an enhancement of heterogeneous reactions (Riedel et al., 2015), may promote the chemical formation of OOMs. However, due to the lack of VOC measurements and inadequate understanding of heterogeneous reactions, it is hard to conclude which one is the determining factor for the observed OOM concentration changes. Additionally, the total OOM concentrations at same $PM_{2.5}$ levels generally decreased from 2019 to 2022 (**Fig. S7**), which implies that the OOM concentrations have reduced in recent years. In terms of the OOM composition, **Figs. 4d and 4e** show that both oxygen and nitrogen contents of OOMs increased year by year. None of the meteorology parameters showed such a behavior, so changes in the chemical pathways is likely the main cause. The increase in the oxygen content indicates an enhancement of the overall oxidation state of OOMs, and the increase in the nitrogen content is related to an enhancement of the NO_x involvement in OOM formation. Although there are no direct VOC measurements, studies have shown that the VOC levels are continuously decreasing in the region (Yao et al., 2022). Therefore, by assuming that the level of oxidants stays the same, the oxidants to VOCs ratio increases, thus likely leading to more oxidation of each VOC molecule. Besides, long-term observations have shown that the concentration decrease of NO_x is slower than that of VOCs (Yao et al., 2022;Li et al., 2020). As a result, the NO_x to VOCs ratio should have increased with time, which may partially explain the increased OOM nitrogen content. Furthermore, the oxygen content decreases as $PM_{2.5}$ is increasing. This is not surprising, as OOMs with lower oxygen contents possess higher volatilities, and thus are more easily to be re-evaporated back to the atmosphere and transported along with $PM_{2.5}$. The nitrogen content first increases with an increasing $PM_{2.5}$ levels and then decreases when $PM_{2.5}$ exceeds 150 μg cm^{-3}. Higher $PM_{2.5}$ levels seem to be associated with higher NO_x concentrations (**Fig. S6**), which promotes the formation of organonitrates, leading to a higher nitrogen content. But the reason why the nitrogen content decreases under serve haze remains a puzzle, deserving further studies." (Line 231 – 254)

Besides, to roughly evaluate the OOM contribution to SOA formation through condensation, the parameter [ELVOCs+LVOCs]×CS was used as a simple surrogate for OOM condensation flux. The following discussion and **Fig. 4f** have be added in the revised manuscript:

"In order to evaluate the OOM contribution to SOA formation through condensation, the parameter [ELVOCs + LVOCs] × CS was used as a simple surrogate for OOM condensation flux (**Fig. 4f**). [ELVOCs + LVOCs] × CS was the highest in 2019 and the lowest in 2021, with 2020 and 2022 lying between and being comparable. This suggests that the SOA formation potential of OOMs was highest in 2021 and lowest in 2021, which is in consistent with the organic aerosol concentration in **Fig. 10**." (Line 254 – 258)

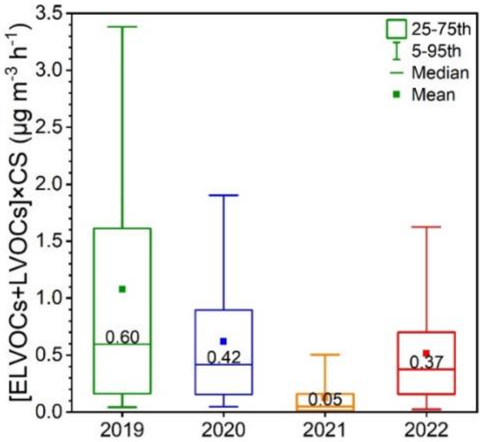


Figure 4. (f) [ELVOCs+LVOCs]×CS for different years (1st to 22nd January). The value inside each box is the median value.

For $PM_{2.5}$, apart from the contribution of background aerosols and local formation, it also has regional transport sources (Han and Zhang, 2018;Wen et al., 2018;Wang et al., 2015). Therefore, the decrease in $PM_{2.5}$ is not solely caused by the decrease of local sources. OOMs rich of carbons should have low volatility, so they tend to partition into the particle-phase compared with stay in the gas-phase. As a consequence, measured gaseous OOMs with higher nC have low concentration and take small fraction in total OOMs (Nie et al., 2022;Guo et al., 2022), and it is challenging to capture the OOMs rich of carbons based on current measurement technique.

10. Line 313 (Section 3.2.1) "Meteorological conditions were not substantially different among different special events, with a few notable exceptions" this statement seems unsupported by the data shown in Fig. 12 and 13 where several variations can be found.

Response: Thanks, we revised the manuscript accordingly:

"There are three notable meteorological characteristics for the special event periods." (Line 347)

11. Section 3.2.2. The discussion in this section is sometimes difficult to follow. It would need a sum-up too. The current conclusions "As for the CNY period, its restriction effect on NO_x was comparable with Olympics, but not for SO_2 due to fireworks. Overall, our results suggest that the restrictions during Olympics were the most effective in controlling primary gaseous pollutants." are ambiguous: can this "restriction effect" be identified based on atmospheric composition data are just derived from general information about the emissions (e.g. the fireworks)? The simple conclusion about "the restrictions during Olympics were the most effective in controlling primary gaseous pollutants" is somewhat too little in summarizing the information provided by **Fig. 14 and S10**.

Response: Thanks for your suggestion. The "restriction effect" was identified based on measured datasets in this study as well as sources of trace gases from previous studies. As suggested, we restructured most of the discussions in the revised manuscript as follows:

"During Olympics, the southerly wind was not that frequent so that this period was the least influenced by polluted air masses. Consequently, CO, NO, NO_2 and SO_2 were the lowest, and O_3 was the highest during Olympics compared with other three periods. During COVID, however, the southerly wind appeared with the highest frequency, leading to the most severe pollution transportation. Thus, the CO concentration was the highest (849 ppb). It can also be found that, despite the most polluted air masses during COVID, the concentrations of NO, NO_2 and SO_2 were lower than the Reference period, suggesting that the restrictions on NO, NO_2 and SO_2 during that period were quite efficient. Besides, compared with NO_2, the restriction on NO and SO_2 were much more obvious. NO_2 has large sources from secondary formation and is able to transport over long distances (Zhu et al., 2021;Tan et al., 2022), hence, the transportation of NO_2 likely partly compensates the effect of the NO_2 emission control. Under the synergistic influences of pollution transport and COVID control measures, the O_3 concentration was only slightly higher during COVID than during the Reference period. As mentioned before, during CNY, the wind direction had a pattern similar to that during the Reference period, and the wind speed as well as BLH were only slightly higher than those during the Reference period. Therefore, meteorological conditions were similar during these two periods. Consequently, CO was only a bit lower during CNY than during the Reference period. For NO and NO_2, however, their concentrations were much lower during CNY than during the Reference period. This is likely due to the reduction of NO_x emissions from vehicles, as a large fraction of people went hometown from Beijing. Lower NO_x levels also led to higher O_3 levels. The SO_2 concentration was only slightly lower during CNY than during the Reference period. During the winter heating-period in urban Beijing, SO_2 should mainly come from the combustion of fossil fuel (Xu et al., 2016;Meng et al., 2016), so that the reduced traffic emissions had minor influences on SO_2 concentrations. Besides, from New Year's Eve to the next following days, SO_2 can also be emitted from fireworks (Foreback et al., 2022)." (Line 364 – 382)

To better clarity the influences of different special events, we also gave a brief summary of this section:

In summary, in urban Beijing, concentrations of gaseous pollutants are controlled by their emission sources and the meteorological conditions. Thus, during Olympics, favored by the cleaner air mass, CO, NO, NO_2 and SO_2 were dramatically reduced. During COVID, under the influence of extremely polluted air masses, the strict restrictions on people's movement which affects traffic emissions, and probably other production activities, CO increased a lot, while NO and SO_2 reduced, and NO_2 reduced slightly. During CNY, under the influence of unfavorable meteorology conditions, reduced traffic emission and probably other production activities, CO and SO_2 did not decrease, while NO_x reduced." (Line 392 – 399)

12. Lines 370 – 371 "OOM concentration also showed a strong association with the pollution level (Fig. S11).". This is clear from Fig. S11, but why in the period of no restriction the same dependence looks so much variable between different observation periods (Fig. S7)?

Response: There might be a misunderstanding since these two figures have different y-axis scales (Fig. S7 up to 7*10^7, Fig. S11 up to 1.4*10^8). OOMs could be formed from gaseous oxidation of VOCs or oxygenated VOCs (OVOCs), generated from heterogeneous reactions at the surface of aerosols, or be transported along with pollution. Due to the lack of VOC measurement and limited knowledge about heterogeneous reactions, now it is hard for us to determine which factor has the most significant impact. In the revised manuscript, we have modified the discussions about the four-year variation of OOM concentration, which can also be found in the response to Comment 9.

13. Section 4 "Summary and conclusions" not fully acknowledges the complexity of the results presented in the previous sections. The information provided here is often uncomplete: for instance, line 453 "The control measures during COVID and Olympics were effective in reducing NO_x and SO_2" makes no mention to the effect that the COVID period restrictions had no effect on CO instead. I suggest to expand the summary making a more systematic survey of the changes observed in the single special events (COVID, Olympics and CNY) with respect to the reference, highlighting what can be explained by meteo anomalies, what can be actually attributed to changes in the emissions and what is yet to be explained. The results about NPF events, air ions, SA and OOM should be summarized in a dedicated paragraph, because these are unconventional parameters providing originality to the present study.

Response: Thanks for your suggestions. We have restructured the "Summary and conclusions" part and added some key points. In the revised manuscript, the first and the last paragraphs stay as before, giving an overview and pointing out the possible significance of this study, respectively.

From the second to the fourth paragraphs, the findings related to four-year changes are discussed. We added the second paragraph to summarize the variations of meteorological conditions:

"Generally, the meteorological conditions for 2019 and 2020 were similar, where temperature, wind speed and BLH are comparable, and the wind mainly blew from the northwest from night to noon and turned to the south from the afternoon. In 2022, the wind mostly blew from the southeast from midnight to noon and turned to the northeast from afternoon. The wind speed in this year was a little higher than 2019 and 2020, and the temperature and BLH were slightly lower than 2019 and 2020. In 2021, the wind was mostly from the west or northwest. Thus, the air masses in this year were much cleaner than other three years due to the least influence of polluted south air masses. This clean condition in 2021 was also associated with the highest wind speed and the maximum BLH, leading to the strongest atmospheric diffusion capacity. Besides, RH showed an increasing pattern from 2019 to 2022, which may facilitate the formation of secondary aerosols through heterogeneous reactions (Sun et al., 2018)." (Line 478 – 486)

The third paragraph summarizes the variations of conventional gaseous and particulate pollutants. The main content stays as before. Below are some revisions:

"For conventional gaseous and particulate pollutants, the four-year changes of CO, NO_2, total $PM_{2.5}$, organic aerosol, chloride and black carbon were generally opposite to that of BLH that they decreased from 2019 to 2021 and increased in 2022. The atmospheric conditions of 2021 were also the cleanest. Thus, the atmospheric diffusion capacity and the overall cleanness of the air masses may control the variations of the above pollutants." (Line 487 – 490)

As suggested, we separated the discussions about NPF-related parameters, including sulfuric acid, OOMs and particles, in another paragraph. The main content stays as before. Below are some revisions:

"For unconventional parameters …" (Line 498)

"For total OOMs, however, its concentration decreased from 2019 to 2021 and increased in 2022. This variation was generally opposite to BLH but with larger extent, suggesting that OOM concentration was controlled both by meteoroidal conditions as well as local chemical production." (Line 500 – 502)

From the fifth to the seventh paragraphs, the results related to special events are summarized. As suggested by the reviewer, all the discussions are made event by event. Most of the content has been modified:

"The wind direction distribution during the Reference, COVID and CNY periods were similar that the wind mostly blew from northwest or northeast from midnight to noon and turned to the south or southeast from the afternoon. During the Olympics, however, the dominant wind was from the southwest from midnight to noon, while there was no prevailing wind from the afternoon. Thus, the air masses during Olympics was the cleanest due to less frequent southerly wind. Besides, there was not too much different in wind speed and BLH for four periods. One thing needs to be mentioned is that the RH during COVID was much higher than other three periods, which should promote the growth of secondary aerosols." (Line 510 – 516)

"During Olympics, favored by the clean conditions, most trace gases (CO, NO, NO_2 and SO_2) and particulate matters (total $PM_{2.5}$, organic aerosol, nitrate, ammonium, chloride and black carbon) were dramatically reduced compared with Reference period. During COVID, influenced by the extremely polluted conditions, CO and all particulate matters (total $PM_{2.5}$, organic aerosol, sulfate, nitrate, ammonium, chloride and black carbon) increased a lot. Despite the serve haze, NO and SO_2 reduced, and NO_2 reduced slightly, suggesting that the restriction on traffic emission and probably other production activities were effective in reducing NO_x and SO_2." (Line 517 – 522)

"During CNY, affected by unfavorable meteorological conditions, reduced traffic emission and probably other production activities, CO, SO_2, total $PM_{2.5}$, nitrate and black carbon were comparable with the Reference period, while NO_x reduced, organic aerosol, ammonium and chloride increased, and sulfate increased to a large extent." (Line 524 – 526)

"SA and OOMs are the secondary products so that their variations were complex and were not directly linked to the changes of meteorology or restriction. The level of SA1 was mainly influenced by CS, SO_2 and

UVB (Yang et al., 2021;Petäjä et al., 2009). Compared with Reference period, SA1 was comparable during Olympics, lower during CNY and much lower during COVID. The concentration of SA2 was mainly controlled by SA1 and CS. Compared with Reference period, SA2 during Olympics and CNY were comparable, while much lower during COVID. As COVID was the most polluted, the above results suggest that the formation of SA was suppressed under polluted condition. For total OOMs, however, it had the lowest concentration in Olympics while the highest concentration in COVID, indicating that the level of total OOMs was higher under polluted conditions. Meanwhile, the oxygen content was the highest during Olympics while the lowest during COVID, which implies the oxidation state of OOMs likely decreased with increasing pollution level." (Line 530 – 538)

Referee #2

This study presents results of a comprehensive measurement campaign for gaseous and particulate atmospheric compositions during wintertime in Beijing, China. Data and information presented in this study would be useful for the reader. I appreciate the authors for providing such an important data to be available to open public. Since the manuscript type of this paper is Measurement Reports, I respect the interpretation made by authors as much as possible. I recommend this manuscript for publication after the reasonable revision.

We thank the reviewer for the constructive comments and suggestions. We have carefully revised our manuscript and supplement accordingly. The point-to-point responses to the comments are given below. And the comments, our replies, and corresponding changes in this response document, revised manuscript and supplementary information are marked in black, blue, and green texts, respectively.

1. Please mention whether any precipitation/rain events occurred or not during the observation periods, since such events have significant impact on the compositions of gaseous and particulate atmospheric compositions.

Response: Thank you for your suggestion. This study focuses on the winter periods so that there should be seldom rain events but probably snow events. It is a pity that we don't have the reliable precipitation measurement, so we used the precipitation data from NOAA. It should be noted that the longitude and latitude of the NOAA measurement spot (40.08° N, 116.59° E) is different from our site (39.95° N, 116.31° E), so the precipitations may exist large difference between those two sites. As shown in Fig. R1, the precipitation in 2019 and 2021 was quite low, while in 2020 was much higher. Hence, we first used the 2020 datasets to explore the influence of precipitation on gaseous and particulate compositions.

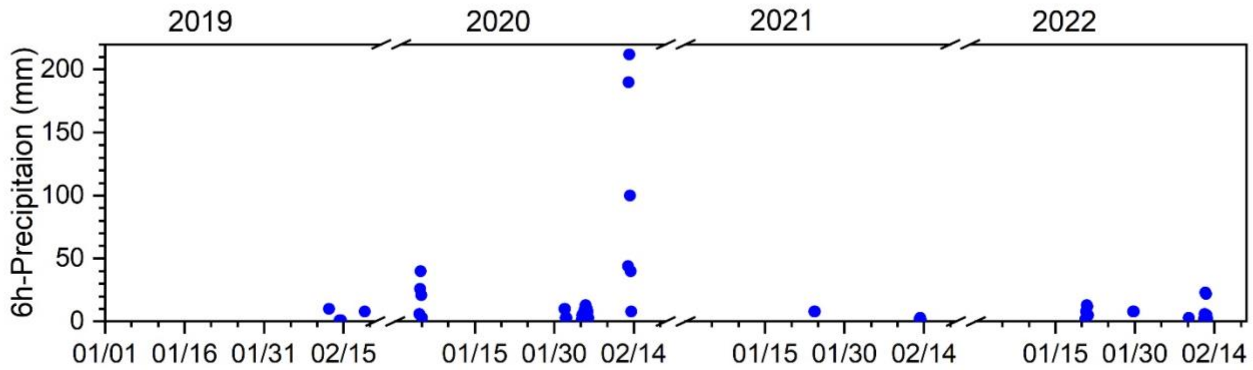


Figure R1. 6h-precipitation from 1st January to 28th Feburary in 2019 – 2022. The datasets were from NOAA, and the location of the measurement spot is 40.08° N, 116.59° E.

As shown in Fig. R2, the precipitation events mostly lowered the concentration of gaseous and particulate species by wet deposition, but its influence on the fractions of different particulate compositions was not clearly seen. To confirm this, we then plotted the datasets of 2022. As shown in Fig. R3, the concentrations of gaseous and particulate species were not always decreased during precipitation events, which may be due to the precipitation being too small to have an influence or that the precipitation from NOAA cannot represent the one at our site. Consequently, due to the lack of reliable precipitation measurement at our site, the small intensity of precipitation in urban Beijing as well as the unclear influence of precipitation, we prefer not to discuss the precipitation in detail in the manuscript. Nevertheless, we added a few sentences in the revised manuscript to clarify the concern about precipitation as suggested:

"Furthermore, precipitation can increase the wet deposition of almost all gaseous and particulate species, influencing their concentrations in the atmosphere. However, since there was no reliable measurements of precipitation at our site, detailed conclusions on this matter cannot be drawn. It should be mentioned that the intensity of snow precipitation in winter Beijing is generally small, except during special case(s), so that the overall effects of precipitation in winter Beijing is not as larger as that in summer Beijing or in other humid areas." (Line 198 – 202)

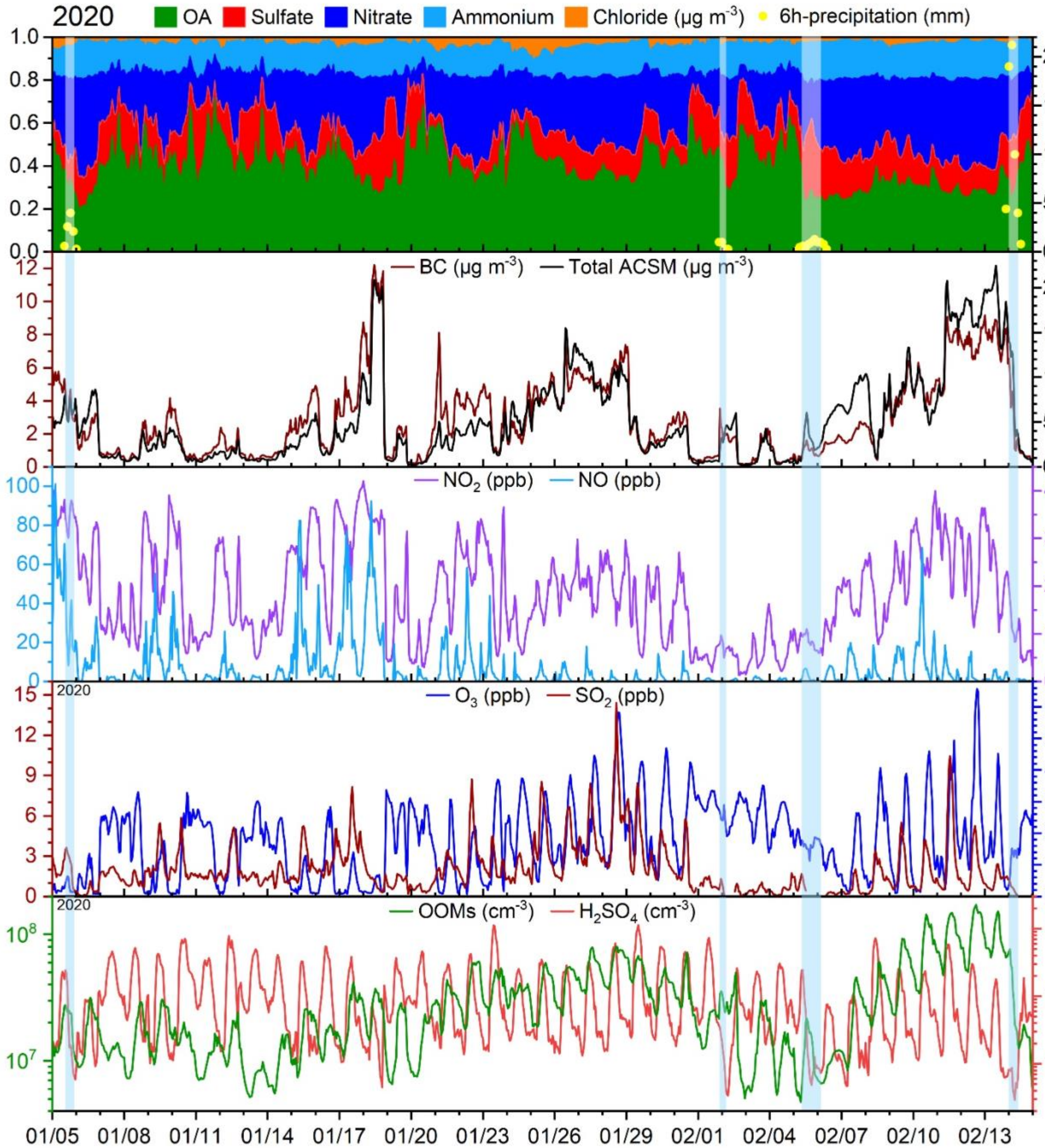


Figure R2. Time variation of fractions of organic aerosol (OA), sulfate, nitrate, ammonium and chloride, 6h-precipitation from NOAA of Beijing spot (40.08° N, 116.59° E), black carbon (BC), particulate mass concentration measured by ACSM (total ACSM), NO_2, NO, O_3, SO_2, total OOMs and sulfuric acid (H_2SO_4) from 5th January to 14th Feburary in 2020. The Blue regions denote precipitation events.

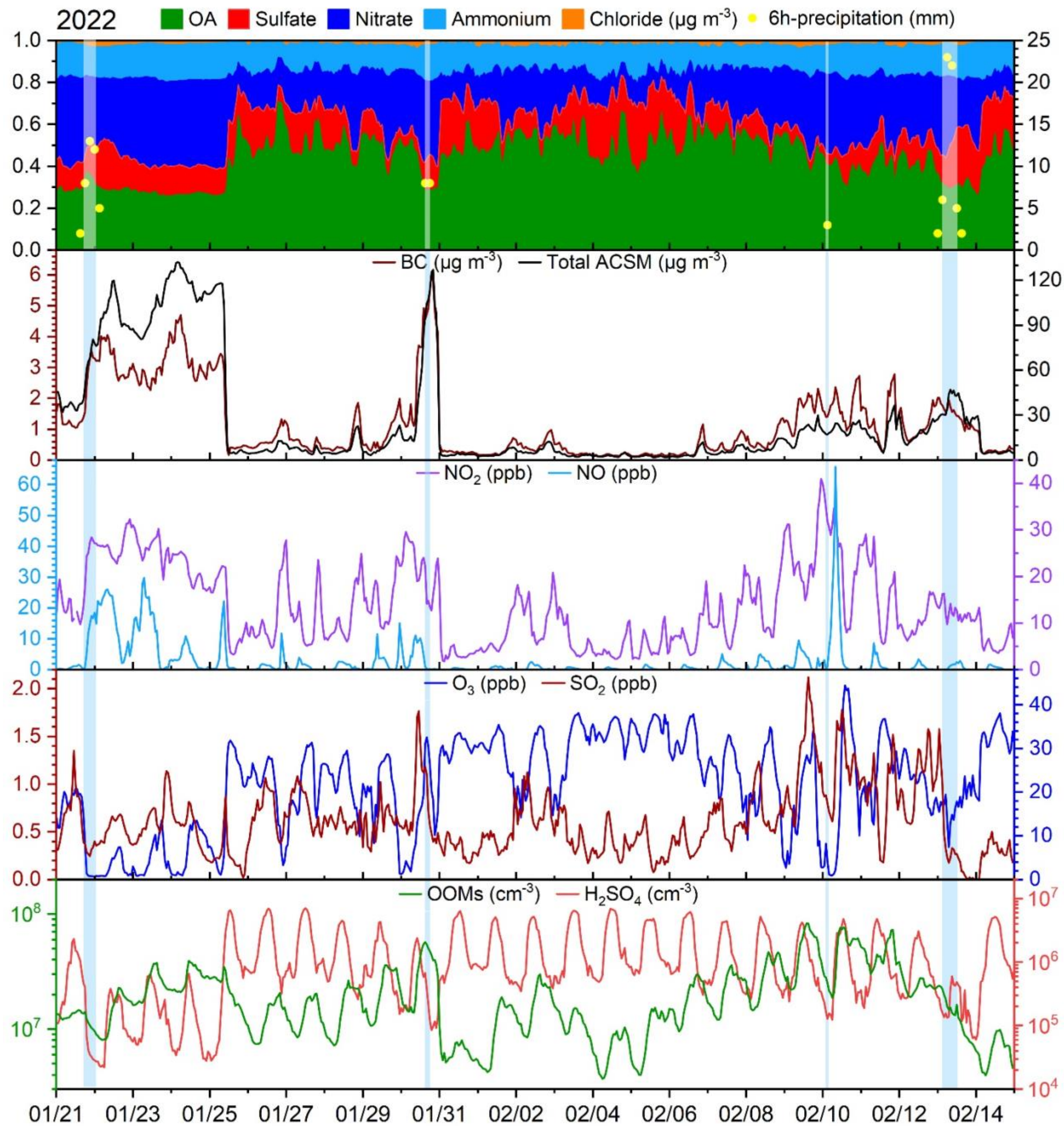


Figure R3. Time variation of fractions of organic aerosol (OA), sulfate, nitrate, ammonium and chloride, 6h-precipitation from NOAA of Beijing spot (40.08° N, 116.59° E), black carbon (BC), particulate mass concentration measured by ACSM (total ACSM), NO_2, NO, O_3, SO_2, total OOMs and sulfuric acid (H_2SO_4) from 21st January to 14th Feburary in 2022. The Blue regions denote precipitation events.

2. Line 79-80: Needed appropriate citations for this sentence. I guess that annual Chinese New Year celebrations did not always result in reductions in air pollution.

Response: Thank you for your suggestion. We agree with the reviewer that the pollution level during Chinese New Year is not always lower compared with other periods, as it is synergistically influenced by emissions, air mass transportation as well as the atmospheric diffusion capacity. But here we are discussing about anthropogenic emissions rather than air pollution. During the Chinese New Year, plenty of migrant population return their hometown from Beijing. And less people in Beijing should cause the reduction in anthropogenic emissions. As suggested, we have added two citations for the sentence in the revised manuscript:

"Additionally, annual Chinese New Year celebrations are typically associated with reductions in anthropogenic emissions during the 7-day holiday (Tan et al., 2009;Lin and McElroy, 2011)." (Line 92)

3. Line 81-87: In context of the impact of special event such as the Olympic Games on the air quality in Beijing, I think several previous studies regarding the effect of the 2008 Beijing Olympic Games should be cited.

Fan, S.-B. et al. (2009) Road fugitive dust emission characteristics in Beijing during Olympics Game 2008 in Beijing, China, Atmos. Environ., 43, 6003-6010, doi:10.1016/j.atmosenv.2009.08.028

Okuda, T. et al. (2011) The impact of the pollution control measures for the 2008 Beijing Olympic Games on the chemical composition of aerosols, Atmos. Environ., 45, 2789-2794, doi:10.1016/j.atmosenv.2011.01.053

Schleicher, N. et al. (2012) Efficiency of mitigation measures to reduce particulate air pollution-A case study during the Olympic Summer Games 2008 in Beijing, China, Sci. Total Environ. 427-428, 146-158, doi:10.1016/j.scitotenv.2012.04.004

Response: Thank you very much for your suggestion and providing the citations. The 2008 Beijing Olympic Games was mentioned in the last paragraph. Therefore, we added the above three citations to where the 2008 Olympics lies in the revised manuscript:

"Beijing has imposed strict short-term emission reductions during highly visible international events, such as Beijing Summer Olympics in 2008 (Wang et al., 2010;Shou-bin et al., 2009;Okuda et al., 2011;Schleicher et al., 2012), …" (Line 85 – 86)

4. Line 250-256: I'm curious that the N_{1-3} decreased with the increasing $PM_{2.5}$ levels while sub-2 nm ions showed relatively constant concentrations. The diffusion coefficient of sub-2 nm ions is larger than that of N_{1-3}; therefore, sub-2 nm ions be possibly scavenged by surrounding aerosol particles, but reality was not. Please add some comments on this.

Response: We agree with the reviewer that sub-2 nm ions should be more vulnerable to be scavenged by background particles than sub-3 nm particles. We are also surprised by the phenomenon that sub-2 nm ions showed relatively constant concentrations with the increasing $PM_{2.5}$ level. We think that it might be that the production rate increased as $PM_{2.5}$ concentration increased, but we do not have any proof to prove it. As suggested, we added some comments in the revised the manuscript:

"Both sub-3 nm particles and sub-2 nm ions are easily to be scavenged by larger particles, and ions are more vulnerable considering their higher diffusivity. The relatively constant concentrations of sub-2 nm ions under different pollution levels might result from that the ion source strengths increased as $PM_{2.5}$ concentration increased, which needs further exploration." (Line 287 – 290)

5. Line 280-283: This is just a comment, but I think that this sentence is only valid in wintertime. In summer, the intensity of solar radiation would have significant impact on sulfate concentration.

Response: Thanks. We agree with the reviewer and revised the sentence accordingly.

"… reflecting the significant reduction of SO_2 emissions that China has made during the recent years, especially in winter." (Line 316)

6. Line 349-351, As mentioned in L409-410, it is possible that particle concentration would increase during CNY due to fireworks, but I'm suspicious that it is applicable to SO_2 concentration.

Response: Thanks for your suggestions. As you mentioned, according to previous studies, the particle concentration always increases during firework periods (Tian et al., 2014;Jiang et al., 2015;Kong et al., 2015;Foreback et al., 2022). While there was study showing that SO_2 concentration also increased during the firework event in 2018 (Foreback et al., 2022). This study was also conducted in our sampling site. Here, we used data from the four surrounding national stations to demonstrate the behaviors of SO_2 due to fireworks during CNY from 2015 to 2018. It can be found that there were sharp increases for SO_2 during the CNY's

Eves for all four years. Although SO_2 can be transported along with polluted air masses, such explosion of SO_2 was likely caused by local emission(s) rather than transportation. Thus, the fireworks during CNY were the most possible causes. It should also be mentioned that at the end of 2017, the Beijing Municipal Government passed a regulation, saying that fireworks were forbidden within the Fifth Road (http://www.bjrd.gov.cn/zyfb/zdgz/jyjd/202012/t20201222_2179517.html). Our measurement site and four surrounding National Stations are all inside the Fifth Road so that the strength of fireworks after 2018 for these sites should be weaker.

Due to the above uncertainty in SO_2 from fireworks, we modified our expression with more soft tone in the revised manuscript:

"Besides, from New Year's Eve to the next following days, SO_2 can also be emitted from fireworks (Foreback et al., 2022)." (Line 381 – 382)

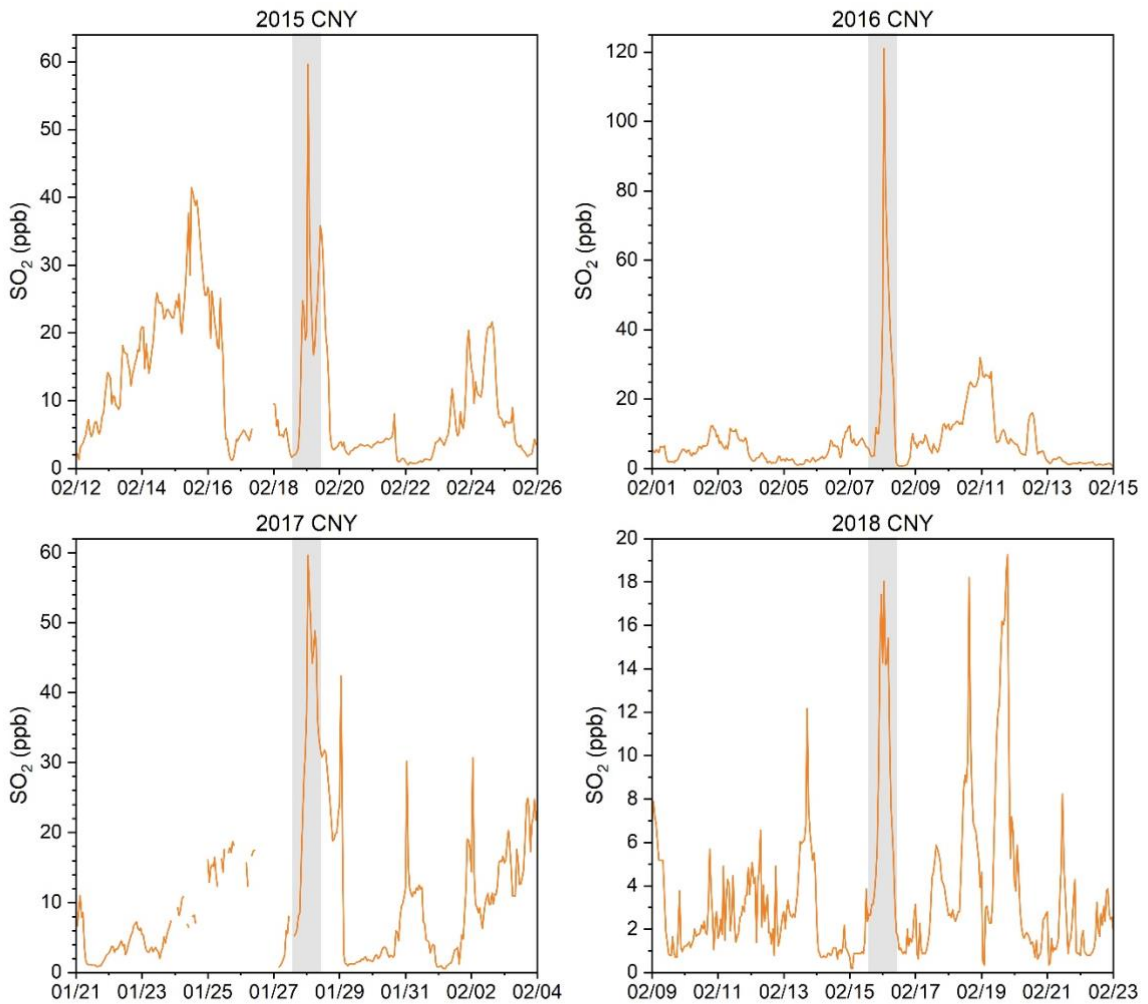


Figure R4. SO_2 variation before and after several days of Chinese New Year (CNY) for the years from 2015 to 2018. As there was no SO_2 measurement for our site before 2018, the SO_2 concentration shown here is the averaged value of four surrounding National Stations.

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