

Responses to reviewer #2

We appreciate the reviewer for the constructive comments on our manuscript. We have studied the comments carefully and revised our manuscript accordingly, which can be found in the attached file (Track Changes). Our point-by-point replies to the comments are provided below. Referee comments are given in black, and our replies are given in blue. Additionally, we checked our figures using the Coblis – Color Blindness Simulator and revised the color schemes accordingly.

Comments

This manuscript analyses the composition and sources of ambient PM_{2.5} in the Hohhot region in China before and during the COVID-19 lockdown. The information presented in the study is relevant because, unlike other existing studies, data were collected well before beginning and after the lockdown period which allowed capturing business-as-usual and lockdown PM_{2.5} samples. Results are well presented and structured, and the discussion goes straight to the relevant findings. By applying the PMF model, it can be proposed those sources that contribute significantly to the ambient levels observed with statistical confidence. Nevertheless, some details must be addressed before it is accepted for publication at ACP.

1. Some sentences are repetitive in the abstract, e.g. L25-27, and within the entire document.

Response: Thank you for pointing it out. We conducted a statistical test and revised the related descriptions.

L26-29: We deleted the description of not-significant variation.

L279-283: We rephrased the sentence.

“Compared with the pre-LD period, the concentration of sulfate ($p < 0.01$), nitrate ($p < 0.01$), ammonium ($p < 0.01$), OM ($p < 0.001$), and EC ($p < 0.001$) decreased due to the decline in the emission intensity under the strict control measures during the LD period (Figure 4a, Table S10). The percentage of sulfate (not significant for LD and $p < 0.01$ for post-LD), nitrate (not significant for LD and $p < 0.05$ for post-LD), and ammonium ($p < 0.05$ for LD and $p < 0.01$ for post-LD) decreased continuously during LD and post-LD, while the MD ($p < 0.01$ for LD and $p < 0.001$ for post-LD), OM (not significant for both two periods), and EC (not significant for LD and $p < 0.01$ for post-LD) increased (Table S10).”

L413-415: We rephrased the sentence.

“Compared with the pre-LD period, the concentration of SNA, OM, and EC decreased substantially during LD and post-LD periods due to the lockdown measures.”

2. One aim is to identify the long-term characteristics of PM_{2.5} in the studied region, however, analysing one year is not sufficient to understand long-term variations unless their results are discussed and compared with those in existing studies, which are not reported.

Response: Thank you for your suggestion. We revised the inappropriate sentence. (L92)

3. Introduction includes studies from most regions of the world, but Latin America was not included where also interesting studies have been made. I recommend you revise and include the following studies where appropriate:

-Mendez-Espinosa, J. F., Rojas, N. Y., Vargas, J., Pachón, J. E., Belalcazar, L. C., & Ramírez, O. (2020). Air quality variations in Northern South America during the COVID-19 lockdown. *Science of the Total Environment*, 749, 141621.

-Hernández-Paniagua, I. Y., Valdez, S. I., Almanza, V., Rivera-Cárdenas, C., Grutter, M., Stremme, W., García-Reynoso, A. & Ruiz-Suárez, L. G. (2021). Impact of the COVID-19 lockdown on air quality and resulting public health benefits in the Mexico City Metropolitan Area. *Frontiers in public health*, 9, 642630.

-Nakada, L. Y. K., & Urban, R. C. (2020). COVID-19 pandemic: Impacts on the air quality during the partial lockdown in São Paulo state, Brazil. *Science of the Total Environment*, 730, 139087.

Response: Thanks for your comment. The references have been added to the manuscript. (L72-73)

4. In line 129: the authors did not define what a strict analytical procedure is.

Response: The detailed analytical procedures were reported in previous studies, and we conducted our analysis according to the referred methods strictly. We added some quality assurance descriptions to make our data reliable.

L136-139: *“Field blank and replicate analyses were carried out once per 10 samples. The concentrations of field blanks were all lower than the method detection limits, and the relative deviations of replicate analyses were < ~ 5%. All the analytical procedures were strictly controlled according to the referred methods to reduce artificial interference.”*

5. In several sections, calculations and results are reported for a year time-scale, but it is not defined if this refer to a calendar or sampling year.

Response: All the years mentioned in the manuscript refer to the sampling year. We revised the ambiguous sentences. (L268, L352, and L402)

6. It would be convenient if the authors propose a hypothesis in introduction and then discuss their findings in light of it, e.g. L196-199.

Response: We revised it accordingly.

L77-80: *“An increase ($p < 0.01$) in $PM_{2.5}$ was found in Hohhot during the LD period, whereas a considerable improvement was reported in most of the cities globally. The response of chemical composition and sources of $PM_{2.5}$ in Hohhot to lockdown measures and the driving factors behind the abnormal increase in $PM_{2.5}$ are still unclear.”*

7. In most sections, $PM_{2.5}$ composition is claimed to be different from other Chinese regions but the reason behind this is not discussed. This issue is critical and must be addressed.

Response: Thanks for the comment. We added some discussions about the composition and source differences between Hohhot and other cities.

L232-237: *“However, the proportion of MD was still substantially higher than those of other cities in South China (Huang et al., 2013), southwest China (Feng et al., 2021), southeast China (Li et al., 2017b), and the Central Plains Urban Agglomeration (Liu et al., 2019), which is close to the cities in northern China (Liu et al., 2021; Xie et al., 2019) and northwest China (Zhou et al., 2021). The lower relative humidity, higher wind speed, and larger area of uncovered surface soil lead to frequent dust storms in semi-arid regions, resulting in a higher contribution of MD than in the humid area. The result indicates that the cities in arid or semi-arid regions (such as in northern China and northwest China) are more susceptible to mineral dust sources.”*

L254-265: *“During this period, the SNA contributed 55.3% by to the total $PM_{2.5}$, slightly higher than those of the cities in northern China such as Xi’an (50.0%) (Tian et al., 2021) and Beijing (48.5%) (Ren et al., 2021), and lower than the cities in southern China such as Guangzhou (78.7%) (Wang et al., 2021), Nanjing (68.2%) (Ren et al., 2021), and Shanghai (75.4%) (Chen et al., 2020). Sulfate was the predominant component of SNA in Hohhot during this period, whereas nitrite was the main contributor to SNA in Guangzhou, Nanjing, and Shanghai. The result indicated that higher SNA contributions in megacities of southern China are mainly related to vehicular emission. The higher contribution of sulfate in Hohhot is mainly related to coal combustion for winter heating. OM contributed by 27.8% to the total $PM_{2.5}$, lower than that of Xi’an (42.0%) (Tian et al., 2021), and higher than that of the other cities listed in Table S9. The contribution of EC is higher than all of the cities listed in Table S9. The higher contribution of sulfate, OM and EC in Hohhot indicated that coal combustion may have been a predominant source of $PM_{2.5}$ during the pre-LD period.”*

L353-358: *“The contribution of primary sources such as CC, VE, and dust source (refer to the sum of construction dust and crustal sources in this study) in Hohhot was higher than the megacities such as Beijing (Zková et al., 2016), Tianjin (Tian et al., 2021), and Shanghai (Feng et al., 2022), whereas the SIA and BB contributions were lower than in these cities*

(Table S11). The result indicates that the contribution of secondary aerosols is predominant in megacities, while the primary source is predominant in semi-arid regions.”

8. L230-235: Statistical tests must be conducted to identify if the changes observed for each component between periods were significant.

Response: We conducted a statistical test for the changes of each component between different periods and marked the significance level to the text and figures. We added a table (Table S10) to the supplement.

L186-188: *“In the heating period, in addition to the contribution of SNA to $PM_{2.5}$, the primary pollutants such as Cl^- ($p < 0.001$) and EC ($p < 0.001$) were higher than those in the non-heating period (Figure 3d, 3e)”*

L213-216: *“The comparison of atmospheric pollutants ($PM_{2.5}$, PM_{10} , SO_2 , NO_2 , O_3 , and CO) between the LD period and the same period in 2017–2019 are shown in Figure S1. The average concentrations of $PM_{2.5}$, PM_{10} , O_3 , and CO increased by 77.8% ($p < 0.01$), 34.6% ($p < 0.05$), 14.5% ($p < 0.001$), and 5.9% ($p < 0.001$), respectively, whereas the average concentrations of SO_2 and NO_2 decreased by 43.2% ($p < 0.05$) and 8.6% ($p < 0.001$), respectively.”*

L274-283: *“Compared with the pre-LD period, the concentration of sulfate ($p < 0.01$), nitrate ($p < 0.01$), ammonium ($p < 0.01$), OM ($p < 0.001$), and EC ($p < 0.001$) decreased substantially due to the decline in the emission intensity under the strict control measures during the LD period (Figure 4a, Table S10). The percentage of sulfate (not significant for LD and $p < 0.01$ for post-LD), nitrate (not significant for LD and $p < 0.05$ for post-LD), and ammonium ($p < 0.05$ for LD and $p < 0.01$ for post-LD) decreased continuously during LD and post-LD, while the MD ($p < 0.01$ for LD and $p < 0.001$ for post-LD), OM (not significant for both two periods), and EC (not significant for LD and $p < 0.01$ for post-LD) increased (Table S10).”*

Table S10 The changes of chemical composition of PM_{2.5} in Hohhot during pre-LD, LD, post-LD

species	period	Concentration		Percentage	
		change (µg/m ³)	p	Change (%)	p
SO ₄ ²⁻	LD	-12.42	0.004	-7.02	0.055
	post-LD	-17.46	0.000	-11.36	0.003
NO ₃ ⁻	LD	-10.38	0.007	-4.75	0.210
	post-LD	-13.41	0.001	-8.23	0.036
NH ₄ ⁺	LD	-4.27	0.005	-3.04	0.032
	post-LD	-5.41	0.001	-4.44	0.003
Cl ⁻	LD	-0.81	0.129	+0.96	0.154
	post-LD	-1.71	0.002	-0.14	0.841
OM	LD	-9.51	0.000	+2.35	0.423
	post-LD	-17.55	0.000	+2.97	0.326
EC	LD	-1.53	0.000	+0.79	0.276
	post-LD	-2.27	0.000	+2.07	0.006
MD	LD	+1.89	0.187	+6.96	0.003
	post-LD	+0.51	0.726	+11.55	0.000

Pre-LD, LD, and post-LD represent pre-lockdown, lockdown, and post-lockdown period, respectively. “-” and “+” represent “decrease” and “increase”, respectively.

9. L236-238: The percentage of SNA decreased during and post lockdown, but the reason behind this behaviour is not discussed.

Response: We revised it accordingly.

L274-287: *“Compared with the pre-LD period, the concentration of sulfate ($p < 0.01$), nitrate ($p < 0.01$), ammonium ($p < 0.01$), OM ($p < 0.001$), and EC ($p < 0.001$) decreased substantially due to the decline in the emission intensity under the strict control measures during the LD period (Figure 4a, Table S10). The percentage of sulfate (not significant for LD and $p < 0.01$ for post-LD), nitrate (not significant for LD and $p < 0.05$ for post-LD), and ammonium ($p < 0.05$ for LD and $p < 0.01$ for post-LD) decreased continuously during LD and post-LD, while the MD ($p < 0.01$ for LD and $p < 0.001$ for post-LD), OM (not significant for both two periods), and EC (not significant for LD and $p < 0.01$ for post-LD) increased (Table S10). The mean value of RH declined continuously from pre-LD to LD and post-LD, while the mean value of WS showed an opposite trend (Figure S11). The lower RH and higher WS were not conducive to the secondary formation and accumulation of SNA. Therefore, due to the emission reduction and improved atmospheric conditions, the proportion of SNA decreased sufficiently (from 55.3% in pre-LD to 40.1% in LD and 21.6% in post-LD).”*

10. Since the main objective of the study was identifying changes in emissions during the lockdown, why PMF was not applied to conduct an additional analysis of sources prior and during lockdown?

Response: We conducted a study on the impact of COVID-19 lockdown measures on PM_{2.5} sources, and revised the abstract, discussion, and conclusion part accordingly.

11. How did the apportionment to PM_{2.5} change during the lockdown? This is not reported.

Response: We added some discussions to section 3.3 and revised the abstract and conclusion section.

L383-399: *“During the LD period, the contribution of SIA, CC, CS, BB, CD, and VE was 22.6, 18.2, 7.7, 5.6, 3.0, and 2.6 $\mu\text{g}/\text{m}^3$ to PM_{2.5}, respectively, accounting for 37.8%, 30.5%, 12.9%, 9.4%, 5.1%, and 4.4% of the total PM_{2.5} mass (Figure 7). The contribution of CC and dust source (the sum of CS and CD) during the LD period in Hohhot was much higher than those of Tangshan (Wang et al., 2021), Taiyuan (Wang et al., 2022), and Xiamen (Hong et al., 2021) (Table S11). The contribution of SIA was lower than Tangshan and Taiyuan, while higher than Xiamen. Hohhot, Tangshan, and Taiyuan are located in northern China, and consume large amount of coal for winter heating. The high intensity of gaseous precursors emitted from coal combustion is reasonable for a high contribution of SIA. The contribution of VE in Hohhot was lower than Xiamen and Taiyuan. The contribution of VE decreased from 35.5% to 4.4%, whereas the SIA increased from 21.1 % to 37.8 %. The substantial reduction*

in VE was associated with the strict traffic restrictions during the LD period, which is consistent with the findings in Taiyuan (Wang et al., 2022). Compared with the LD period, the contribution of VE increased from 4.4% to 14.7% during the post-LD period, which can be ascribed to the canceled traffic restrictions. The contribution of CC increased from 30.5% during the LD period to 68.7% during the post-LD period, while the concentration decreased from 29.2 to 18.2 $\mu\text{g}/\text{m}^3$. The contribution of SIA decreased from 37.8% during the LD period to 5.0% during the post-LD period, which can be attributed to the improved atmospheric conditions.”

L29-30: “The contribution of secondary inorganic aerosols increased (from 21.1 to 37.8%), whereas the contribution of vehicular emissions was reduced (35.5% to 4.4%) due to lockdown measures.”

L417-421: “The source contribution of secondary inorganic aerosols and vehicular emission decreased during the lockdown period, whereas coal combustion increased. The substantial reduction in the contribution of vehicular emissions was associated with the strict traffic restrictions during the lockdown period, the increase in vehicular emission contributions during the post-lockdown period can be attributed to the canceled traffic restrictions.”

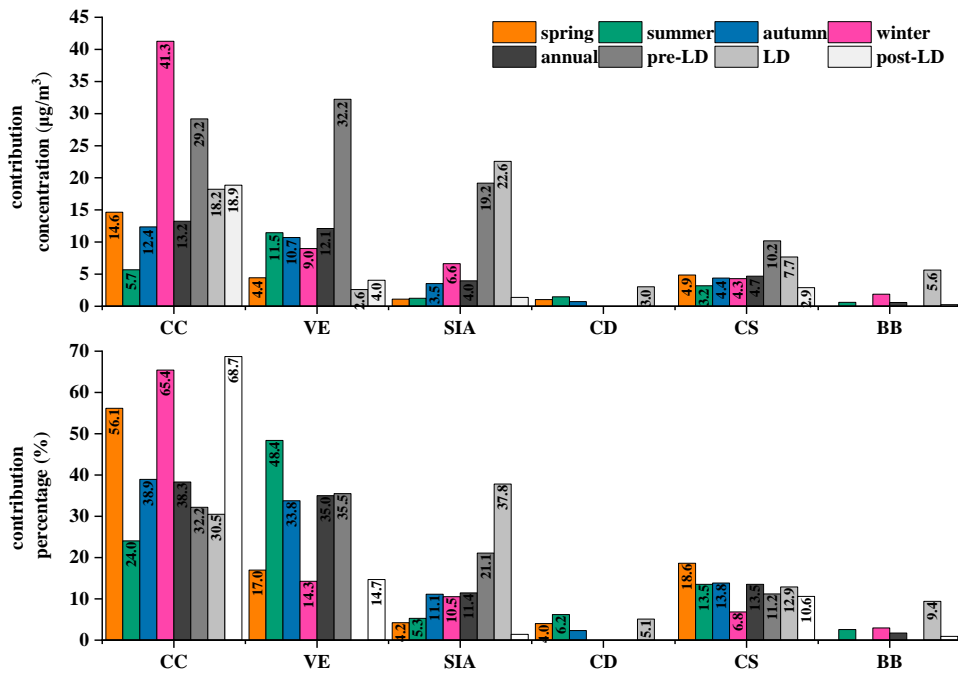


Figure 7. (a) Concentration and (b) percentage of source contribution to $PM_{2.5}$ in Hohhot in spring, summer, autumn, winter, over the sampling year, pre-lockdown, lockdown, and post-lockdown. CC, VE, SIA, CD, CS, and BB represent coal combustion, vehicular emission, secondary inorganic aerosol, construction dust, crustal sources, and biomass burning, respectively.

12. Overall, the text is clear and understandable but there are some sentences that require re-writing and re-wording (L95-inhabitants?, L114-analysis, L181, L222-contributed by X % to total PM_{2.5}..., 278-benefical?, L283-easier, you meant faster?...).

Response: We checked through our manuscript and corrected the typos accordingly. (L101-102, L121, L254, L262, L267, L327, and L335-336)