

## Responses to reviewer #1

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

The manuscript addresses a topic of scientific interest during the last 2 years, such as the variation in air pollution during the COVID-19 lockdown. The study is interested in analyzing the variation of the chemical composition of PM<sub>2.5</sub> (not only its concentration) obtained in an area with particular geographical conditions such as a semi-arid city of northern China. Several studies have reported changes in the concentrations of atmospheric pollutants such as PM, O<sub>3</sub> and NO<sub>2</sub> during the lockdown measures, but few studies have delved into the variation in the chemical composition of PM. This approach allows carrying out more detailed analyzes of atmospheric chemistry by relating the fluctuation of emission sources and the implications on the chemical composition of PM.

I appreciate if the authors can offer a response/discussion to each of the following comments:

1. The authors define as objective of the study "identify the long-term chemical characteristics of PM<sub>2.5</sub> in a semi-arid city". However, can one year of study be considered a long-term study?

**Response:** Thank you for pointing it out. We revised the inappropriate sentence. (L92)

2. The manuscript suggests that the results obtained "can provide a new insight for the formulation of effective policies to improve aerosol pollution in semi-arid regions". The authors should go beyond the generality and could suggest concrete measures to improve public policies based on the results achieved.

**Response:** Thank you for your suggestion. We add some practical suggest on improving air quality in the study area, which also can be applied in semi-arid region.

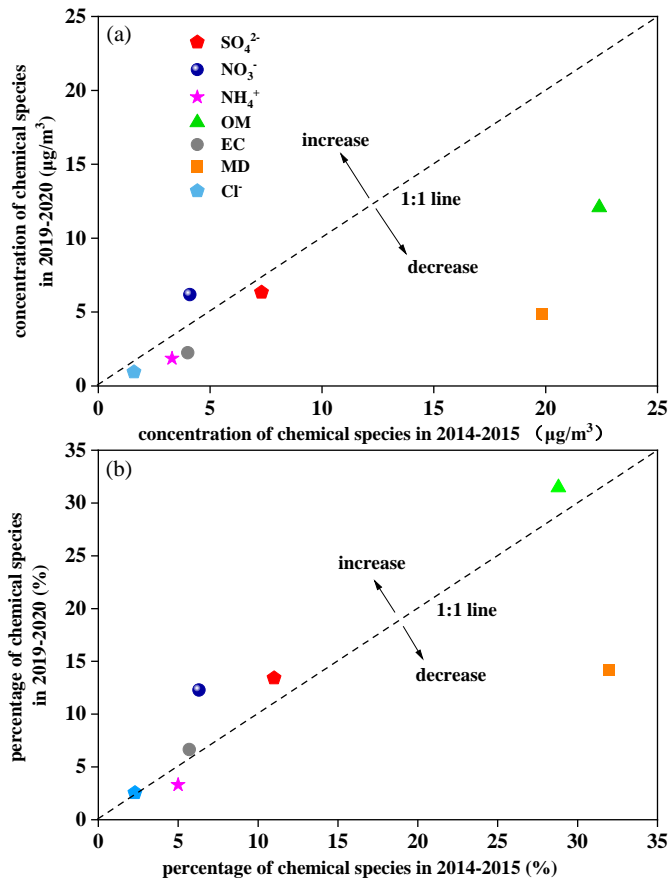
**L422-433:** "A relatively high contribution of primary sources, such as coal combustion and dust source, was observed in Hohhot. Therefore, the control of primary sources, *such as*

*increasing the proportion of clean energy to reduce coal consumption, could be an effective way to reduce the concentration of PM<sub>2.5</sub> in Hohhot. The unfavorable meteorological conditions played an integral role during winter and promoted SNA formation and accumulation, causing frequent heavy pollution events. The reduction in anthropogenic activities and the important role of meteorology in the formation of air pollutants should be considered in aerosol quality and policy measures. The emission reduction of gaseous precursors (SO<sub>2</sub> and NO<sub>x</sub>) under adverse meteorological conditions, can prevent heavy pollution events driven by SNA. The control of coal combustion sources and accurate ambient air quality forecasting techniques will do much good to reduce annual concentrations of PM<sub>2.5</sub> and the occurrence of heavy pollution days, respectively. This study can provide a new insight for the formulation of effective policies to improve aerosol pollution in semi-arid regions. ”*

3. It would be interesting to present a comparative analysis of the variation in the composition of PM<sub>2.5</sub> (not only concentrations) between the year of study and an average of previous years (to be possible). This is a good way to identify PM<sub>2.5</sub> chemical composition anomalies during the COVID-19 lockdown measures.

**Response:** We add a comparison of chemical composition of PM<sub>2.5</sub> between previous study (in 2014-2015) (Wang et al., 2019) and our result (in 2019-2020).

**L223-239:** *“The annual mean concentration of OM, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, MD, EC, NH<sub>4</sub><sup>+</sup>, and Cl<sup>-</sup> were 12.1, 6.6, 6.4, 4.9, 2.2, 2.0, and 1.1 μg/m<sup>3</sup> (Figure 4a), accounting for 31.5%, 13.4%, 12.3%, 14.2%, 6.6%, 3.3%, and 2.5% of PM<sub>2.5</sub>, respectively (Figure 4b). Compared with the result of Hohhot in 2014–2015(Wang et al., 2019), the annual mean concentration of NO<sub>3</sub><sup>-</sup> increased, whereas the concentration of the other species decreased (Figure S10a). Due to the implemented measures, a sharp decrease in OM and MD was observed, resulting in a considerable decrease in PM<sub>2.5</sub> (decreased from 66 μg/m<sup>3</sup> in 2014–2015 to 42.6 μg/m<sup>3</sup> in 2019–2020). The proportion of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and OM increased considerably, whereas the proportion of MD showed a substantial decrease (Figure S10b). The result indicates that the contribution of chemical composition related to secondary formation has increased in recent years. 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.”*



**Figure S3.** Comparison of chemical composition in (a) 2019-2020 and (b) 2014-2015 in Hohhot. The data of chemical composition in 2014-2015 were obtained from Wang (Wang et al., 2019). The organic matter (OM) and mineral dust (MD) were calculated by the following equations with the composition data,  $OM=1.6 \times [OC]$  and  $MD=2.14 \times [Si]+1.89 \times [Al]+1.40 \times [Ca]+1.43 \times [Fe]+1.58[Mn]+1.21 \times [K]+1.67 \times [Ti]$ .

- The authors calculated and reported two indicators related to secondary aerosols, the sulfur oxidation ratio (SOR) and the nitrogen oxidation ratio (NOR). What is the usefulness of these indicators and how are the results interpreted? What additional information do the indicators provide regarding the concentrations of SO<sub>2</sub> and SO<sub>4</sub>?

**Response:** The sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) are important indicators to estimate the secondary formation of inorganic aerosols. The increasing SOR and NOR were observed during heavy pollution process, indicating that the secondary formation of sulfate and nitrate is the main driving factor for the rapid increase of fine particulate matter on haze days in Hohhot. We revised section 2.3 and the related text in our manuscript.

L142-145: “The organic matter (OM) and mineral dust (MD) were calculated using the following equations (1 - 2). To estimate the secondary formation of inorganic and organic aerosols, the sulfur oxidation ratio (SOR), nitrogen oxidation ratio (NOR), and secondary organic carbon (SOC) were calculated using the following equations (3 - 5) (Xie et al., 2019; Liu et al., 2021)”

L186-187: “High RH is conducive to the secondary formation of sulfates and nitrates, presenting higher SOR and NOR in these pollution periods (Figure 3a, 3b).”

L192-200: “Higher SOR was observed in winter and summer in Hohhot (Figure 3a). High SOR in winter is mainly caused by heterogeneous processes under high RH conditions, while that in summer is caused by homogeneous gas-phase oxidation reactions under high temperatures and O<sub>3</sub> concentrations (Zhang et al., 2018; Li et al., 2017a). NOR was higher in winter, whereas it was lower in summer (Figure 3b). The higher NOR in winter can be ascribed to the rapid formation of nitrate under high RH. The lower NOR in summer may be related to the high temperature, which is favorable for nitrate volatilization (Daher et al., 2012). The higher SOR and NOR in winter indicate the higher secondary formation of sulfate and nitrate that resulted the heavy pollution episodes in pre-LD and LD periods.”

L305-314: “From CP to HP, the percentages of SNA increased (from 26.1% to 52.4%), whereas the percentages of OM and MD decreased (from 33.5% to 23.8%, and from 16.7 to 2.4, respectively). This response is related to the adverse meteorological conditions characterized by high RH and low WS, leading to the enhanced formation of SNA (higher SOR and NOR). The values of SOR increased from 0.18 during CP to 0.48 during HP. The values of NOR increased from 0.07 during CP to 0.25 during HP. The results suggest enhanced SNA formation during heavy pollution episodes. The coupled effects of high RH and low WS promoted the rapid increase of fine particulate matter on haze days in Hohhot. The high WS is beneficial for the elimination of atmospheric pollutants, resulting in low concentrations of SO<sub>2</sub> and NO<sub>2</sub> on dust storm days. Furthermore, the low RH is detrimental to the secondary formation of SNA (lower SOR and NOR), resulting in a lower SNA content in dust storm days.”

L334-338: “NOR was negatively correlated with T at  $p < 0.001$ , which may be related to the volatility of NH<sub>4</sub>NO<sub>3</sub>. The higher T is favorable for nitrate volatilization, resulting in lower NOR (He et al., 2012). SOR and NOR was positively correlated with RH at  $p < 0.001$  and  $p < 0.05$ , respectively, suggesting that both SOR and NOR were influenced by RH.”

5. Please check in the title "3.2 Factors influencing PM<sub>2.5</sub>" the word "metrological" since it should be "meteorological".

**Response:** We checked through our manuscript and corrected the typos. (L321 , L330, and L425)

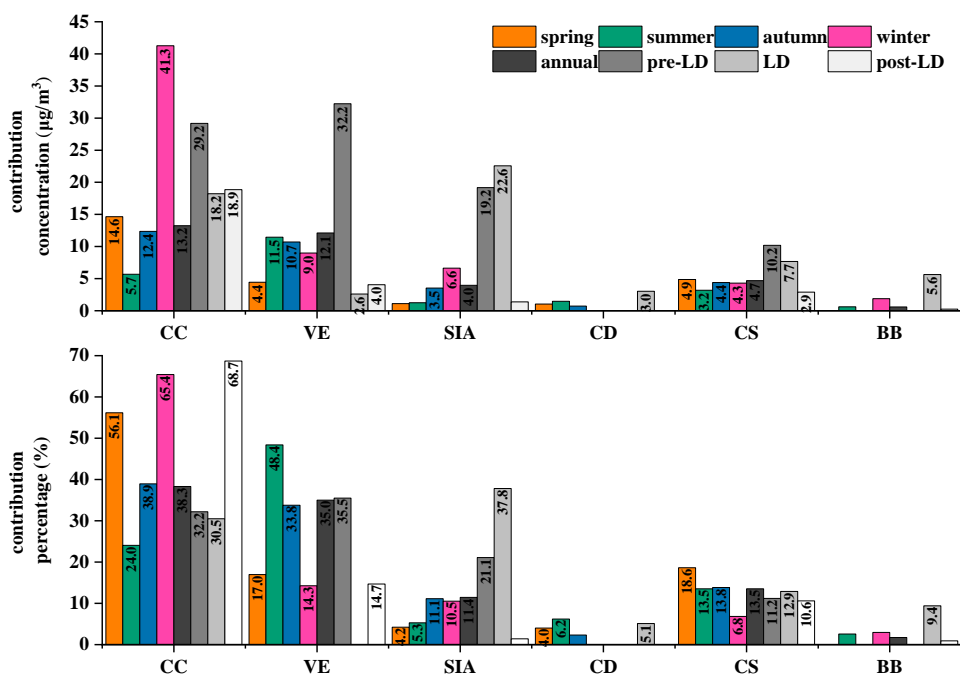
6. The source apportionment of PM<sub>2.5</sub> was carried out for each of the four seasons. But how did the COVID-19 lockdown measures impact on sources of PM<sub>2.5</sub>?

**Response:** Thanks for your constructive comment. We conducted a study on the impact of COVID-19 lockdown measures on PM<sub>2.5</sub> sources. We added some discussions to the section 3.3 and made a revision on the 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<sub>2.5</sub>, respectively, accounting for 37.8%, 30.5%, 12.9%, 9.4%, 5.1%, and 4.4% of the total PM<sub>2.5</sub> 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<sub>2.5</sub> 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.*

## References

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