

**Reviewer #3:**

**Major Comments:**

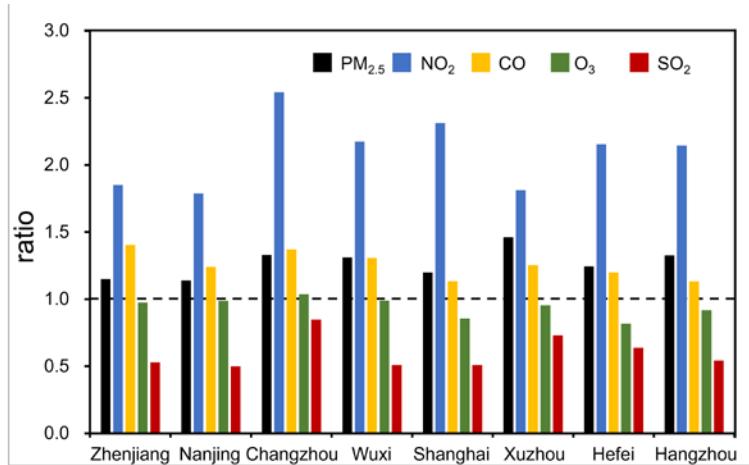
1. The authors claimed that “the number fraction of freshly emitted BCc particles decreased to 28% during the lockdown (LD), with that from vehicle emissions experiencing the most substantial reduction”, and also expressed that “the uncontrolled reductions of nitrogen oxides (NO<sub>x</sub>) and volatile organic compounds (VOCs) likely contributed to increased ozone (O<sub>3</sub>) concentrations, increased the oxidizing capacity”. My question is what are the emission sources of NO<sub>x</sub> and VOCs under the scenario of “the vehicle emissions experiencing the most substantial reduction”? Therefore, I think the authors analysis is paradoxical. Therefore, the mechanism of increase of more aged BCs particles is unreasonable.

Thanks for the insightful comment. The emissions of NO<sub>x</sub> and VOCs originate from various sources, including vehicles, industrial processes, power plants, and agricultural activities. Despite notable reductions in vehicle emissions, these other sources may still play a significant role in influencing atmospheric composition.

Furthermore, as depicted in **Figure S4**, the levels of NO<sub>2</sub> in the major cities of the YRD was significantly higher than that in Yangzhou during the LD period. While NO<sub>x</sub> emissions are primarily a local concern, they can also be transported over considerable distances via air currents, as documented in previous studies (Hertel et al., 2012).

Additionally, the elevated ozone levels observed in Yangzhou relative to other major cities in the YRD (**Figure S4**) underscore the regional variability in atmospheric oxidation processes during the LD period.

Overall, the intricate interplay between diverse emission sources and atmospheric processes presents a valuable opportunity to investigate the characteristics of BCc particles and assess the impacts of lockdown measures on regional air quality. Such analyses are essential for developing effective strategies to mitigate air pollution and protect public health.



**Figure S4.** The ratios of gaseous and particulate levels in the major cities of the YRD compared to those in Yangzhou during the LD period. The black dashed line represents the pollution levels of Yangzhou.

**2. During the observation period, precipitation occurred intermittently (Figure 2b), and the author only mentioned “the data collected during the precipitation were excluded from the analysis” in Section 3.1. This method is obviously not enough to eliminate the impact of precipitation. Additionally, the maximum daily precipitation at the observation site does not exceed 10 mm (Figure 2b), however, the author has repeatedly mentioned heavy precipitation. How is the degradation of precipitation defined?**

Thank you for your comment. The precipitation data depicted in Figure 2b were obtained from the ERA5 reanalysis dataset. According to the definition provided by the China Meteorological Administration, a rainstorm is typified by substantial precipitation, usually falling within the range of 50 to 100 mm per day. On July 28, the daily precipitation amounted to 90 mm, indicating that it reached the threshold for a rainstorm event. Therefore, the heavy precipitation significantly influenced the atmospheric environment during the early stage of the LD period.

Regarding the exclusion of precipitation data from the analysis in Section 3.1 of the study, we chose to exclude data collected during precipitation events to eliminate the potential impact of precipitation on the observed variables. This approach allowed for

a more precise comparison between different lockdown periods.

In response to your question about the definition of precipitation degradation, we acknowledge that precipitation can have complex and varied effects on atmospheric conditions. These effects may include changes in air quality, alterations in aerosol composition, shifts in atmospheric chemistry, scavenging of atmospheric pollutants, and interactions with atmospheric aerosols.

**3. In Section 3.5, the authors propose a method to roughly estimate the local and non-local proportions for each type of BCc particles in Yangzhou only during the ALD period. And the distribution of BCc particles during LD is unclear and there is no comparison before and after. The manuscript focuses on the COVID-19 lockdown, how do the authors provide valuable insights for future air pollution control measures? As the authors pointed out, “Since there was a heavy precipitation on July 28th (the day before lockdown) which removed most atmospheric pollutants, the air pollutants might be influenced mostly by regional transport as local emissions were significantly cut down during the LD period”. Therefore, is it necessary to analyze the distribution of BCc particles during LD period?**

Thank you for your thoughtful comment. We agree with your suggestion, and upon consideration, we find that estimating the local and non-local proportions for each type of BCc particles in Yangzhou is indeed ambitious and may introduce unnecessary complexity. Therefore, we have decided to delete Section 3.5 from the manuscript.

**Minor Comments:**

**1. A detailed description of the shading in Figure 1 should be given.**

We appreciate the suggestion. We have enhanced the annotation of Figure 1 with additional details, as follows:

Line 193~196: “**Figure 1. Geographical overview of the Yangtze River Delta (YRD)**

*Region in China, depicting the major cities within the YRD and the sampling site located in Yangzhou. The color gradient from green to white indicates varying altitudes across the region (Maps were generated by using Arcgis Pro)."*

**2. (Line 177-181) NO<sub>2</sub> (NRTI/L3 NO<sub>2</sub>) obtained from the TROPOspheric Monitoring Instrument (TROPOMI) with a spatial resolution of 3.5×7 km<sup>2</sup>; Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2 SO2SMASS) with a spatial resolution of 69×55 km<sup>2</sup>. My question is, how do the authors think about the impacts of resolution difference between two datasets on the results?**

Thanks for the comment. We recognize the disparities between the spatial resolutions and units of the bands in TROPOMI and MERRA-2, which complicates the comparison of remote sensing results from these two different sensors. Consequently, we have transitioned to using the Copernicus Atmosphere Monitoring Service (CAMS) Global Near-Real-Time dataset for analyzing the distribution of NO<sub>2</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub>, ensuring consistency in resolution. Section 2.2.1 has been updated accordingly.

**3. (Line 228) "Dramatically", the authors should objectively describe and carefully consider the modifiers.**

Thanks for the comment. The word "dramatically" has been replaced with "significantly" as requested in Line 260.

**4. (Line 244-246) "Surface O<sub>3</sub> concentration showed an increase of 12.6 μg m<sup>-3</sup> (19%) during the LD period compared to the BLD period, which may attribute to the reduction of fresh NO emissions that alleviates O<sub>3</sub> titration". In fact, the O<sub>3</sub> concentration showed sustained higher values during the ALD period compare with those in BLD and LD periods (Figure 3). What is the reason?**

Thank you for the comment. We have carefully considered the factors contributing to the higher O<sub>3</sub> concentration during the ALD period. Firstly, the significantly lower duration of precipitation and rainfall during the ALD (54 hours, 40 mm) compared to the BLD (377 hours, 302 mm) and LD (202 hours, 228 mm) periods suggests a reduced removal of pollutants, including ozone, from the atmosphere through wet deposition during the ALD period. Additionally, the lower frequency of precipitation during the ALD period implies increased opportunities for atmospheric photochemical reactions, potentially leading to heightened ozone production compared to the BLD and LD periods. Furthermore, the lower average relative humidity (RH) during the ALD period (77%) compared to the BLD (84%) and LD (86%) periods contributed to decreased net ozone production, as supported by previous studies (Kavassalis and Murphy, 2017; Li et al., 2021). In conclusion, these three factors collectively contribute to the observed higher O<sub>3</sub> concentration during the ALD period.

**5. (Line 246-248) “However, the average concentrations of PM<sub>2.5</sub> (19.9 vs. 21.2  $\mu\text{g m}^{-3}$ ) and SO<sub>2</sub> (9.4 vs. 9.5  $\mu\text{g m}^{-3}$ ) were very close between BLD and LD stages (Figure 3).” According to this logic, TVOC was also very close between BLD and LD stages (Figure 3). Why was it not mentioned?**

Thank you for bringing this to our attention. We have incorporated the TVOC concentration into the text as requested. The revised section now reads as follows:

Line 281~283: “*However, the average concentrations of PM<sub>2.5</sub> (19.9 vs. 21.2  $\mu\text{g m}^{-3}$ ), SO<sub>2</sub> (9.4 vs. 9.5  $\mu\text{g m}^{-3}$ ), CO (0.61 vs. 0.64 mg  $\text{m}^{-3}$ ) and TVOC (58 vs. 55  $\mu\text{g m}^{-3}$ ) were very close between BLD and LD stages (Figure 3).*”

**6. (Line 254) There is a discrepancy between the segmentation of the chart and the textual expression. Additionally, are all observation elements conducted at a rooftop laboratory 20 m above ground? If yes, is it appropriate to use “surface”?**

According to the "Automated Methods for Ambient Air Quality Monitoring Standard of China (HJ 664-2013)," the height of the air automatic monitoring sampling port from the ground should be within the range of 3~20 meters. Our sampling site, located in a rooftop laboratory at the fourth floor, falls within this standard requirement, with a height between 18~20 meters. Observing atmospheric pollutants at a height of 20 meters above the ground is generally considered representative of near-surface concentrations. While it may not be directly at ground level, it still provides valuable insights into pollutant levels close to the surface, where people and ecosystems are most affected.

**7. (Line 270-273) “Such results highlight the short-term, limited-scale, and human-induced reduction in air pollutants as a result of the lockdown measures in Yangzhou, and demonstrate the effectiveness of regional stringent emission control in reducing local atmospheric pollutant concentrations”. From the analysis of the results, it is obvious that the result cannot be obtained. Furthermore, this conclusion is completely opposite to that in Section “Abstract” (Line 46-50).**

We apologize for the confusion caused by the original manuscript. The conclusions presented in the "Abstract" (Lines 46-50) pertain specifically to PM<sub>2.5</sub>, suggesting that short-term, stringent local emission controls may not effectively reduce PM pollution. However, the conclusions in section 3.1 (Lines 270-273) were found lack precision and have been removed. In order to provide clarity, the text has been revised accordingly.

Line 44~47: *“Our research highlights that short-term, strict local emission controls may not effectively reduce PM pollution due to the complex generation and transmission characteristics of BCc and the non-linear responses of PM<sub>2.5</sub> to its precursors.”*

**8. (Line275-280) Calculations were only conducted for the regions with  $PM_{2.5} > 10 \mu g m^{-3}$ ,  $NO_2 > 0.2$  Dobson units (DU), and  $SO_2 > 0.2$  DU in the BLD period. What is the reason for defining these thresholds? Please provide an explanation.**

Thanks for the comment. In response, we utilized the formula  $(LD - BLD)/BLD$  to calculate fractional changes. The initial use of thresholds in our original manuscript aimed to mitigate abnormal values of fraction change, particularly caused by pollutants with low levels during BLD (especially noticeable with TROPOMI  $NO_2$  due to its higher resolution and presence of low-value pixels). Similar thresholds were also employed by Le et al. in their study (Le et al., 2020). However, in the revised manuscript, we transitioned to using the Copernicus Atmosphere Monitoring Service (CAMS) Global Near-Real-Time dataset to analyze the distribution of  $NO_2$ ,  $SO_2$ , and  $PM_{2.5}$ . Throughout the calculation of fraction changes, we didn't encounter such issues, so we decided to remove these thresholds.

**9. (Line 297) Figure 5, please add a horizontal axis identifier.**

We are grateful for the suggestion. We have supplemented the labels for the x-axis in Figure 5.

**10. (Line 354) As shown in Figure 1, it can be seen that significant precipitation occurs during the LD period. The authors' expression here is “the day before lockdown”, please unify it.**

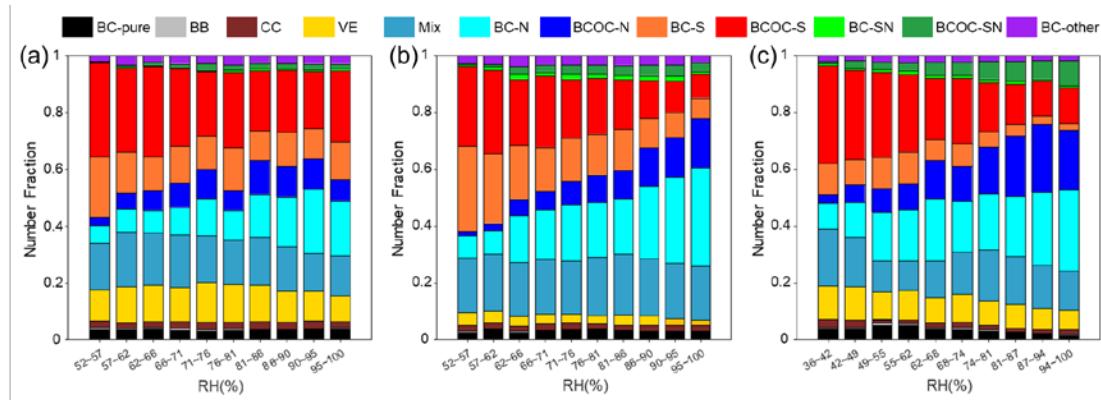
Thank you for bringing this to our attention. We have rechecked the monitoring and meteorological data, and indeed, heavy precipitation occurred on the evening of July 27<sup>th</sup> and early morning of July 28<sup>th</sup>, the eve of the lockdown. The text has been updated accordingly.

Line 346~348: “*During the transition of BLD to LD, heavy precipitation occurred from*

*the evening of July 27<sup>th</sup> to early morning of July 28<sup>th</sup> (the eve of lockdown), resulting in the removal of a majority of the pollutants (PM<sub>2.5</sub>: 4  $\mu\text{g m}^{-3}$ , O<sub>3</sub>: 35  $\mu\text{g m}^{-3}$ , NO<sub>x</sub>: 8  $\mu\text{g m}^{-3}$ )."*

**11. (Line 462, 477) RH is a key element which is responsible for the formation of BCOC and BC-SNA particles. Is there a simple positive or negative correlation between them?**

Thank you for the comment. **Figure R1** illustrates the variation in number fractions of different types of BCc particles under varying RH conditions during different lockdown periods. The data show that the majority of BC<sub>aged</sub> types exhibit a strong correlation with RH, while the number fractions of BC<sub>fresh</sub> types decline with increasing RH, indicating that higher RH environments are conducive to BC aging (Zhang et al., 2021). Specifically, sulfur-containing BCc particles (BCOC-S and BC-S) exhibit a decrease in fraction with increasing RH, whereas nitrogen-containing BCc particles (BCOC-N and BC-N) demonstrate a gradual increase in fraction with higher RH levels, eventually dominating BCc particles at elevated RH levels. However, it's important to note that the variation in number fractions of different BCc types during the BLD period significantly differs from that during the LD and ALD periods. This discrepancy may be attributed to frequent precipitation events, during which sulfate is likely removed less efficiently particularly in warmer seasons (Isokäätä et al., 2022). Consequently, the relationship between RH and the formation of BCOC and BC-SNA particles is complex and influenced by various factors. As such, it cannot be simplified to a simple positive or negative correlation.



**Figure R1.** Variations of number fractions of BCc particle types with relative humidity (RH) during (a) the BLD period, (b) the LD period, and (c) the ALD period.

**12. (Line 474-477) “As shown in Figure 9, BCc particles with ~400 nm Dva exhibited significant diurnal fluctuations in the OC/Cn and SNA/Cn ratios, during the LD period. Moreover, there was a noticeable increase in the proportion of BC-SNA particles during nighttime when RH was relatively high”. Compared with the LD period, BCc particles exhibited more significant diurnal fluctuations in the OC/Cn and SNA/Cn ratios during the ALD period. What is the reason?**

Thank you for your insightful comment. The diurnal fluctuations observed in the OC/Cn and SNA/Cn ratios of BCc particles during the ALD period can be attributed to several factors:

- Changes in atmospheric conditions:** Variations in temperature, humidity, and solar radiation during the ALD period could influence the formation and transformation of OC and SNA, contributing to their diurnal fluctuations.
- Atmospheric photochemical processes:** Enhanced solar radiation during the ALD period could accelerate photochemical reactions involving OC and SNA, leading to their rapid formation or removal throughout the day.
- Changes in pollutant transport and dispersion:** Differences in atmospheric stability and boundary layer dynamics during the ALD period may result in variations in the transport and dispersion of pollutants, affecting the observed diurnal fluctuations in BCc particle composition.

**13. (Line 490-491) Please confirm the relationship between “BCc particles” and “RH” again to prevent conflicting results.**

Thank you for your comment. The diurnal variations of the OC/C<sub>n</sub> and SNA/C<sub>n</sub> ratios (Figure 9) reveal that higher relative humidity (RH) during the night facilitates the formation of a thicker coating on the surfaces of BC cores. Additionally, there is a higher fraction of BC<sub>age</sub> during the LD period with higher RH compared to the BLD and ALD periods (Figure 6). Furthermore, the majority of BC<sub>aged</sub> types exhibit a strong correlation with RH throughout the entire observation period (Figure R1). These findings demonstrate that higher RH environments promote the aging process of BCc particles.

**References:**

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