Anonymous Referee #1

Wei et al. estimated long-term daily seamless different ground-level gaseous pollutants with high accuracy using machine learning and big data by combing monitors, satellites, and models. The public dataset are important to study air quality in China and also have been widely adopted in public health-related studies. The study is well organized and the results are well presented. However, the manuscript still suffers from some flaws. I recommend the manuscript for publication after the following comments are well addressed.

Major comments:

The authors have constructed many air quality dataset (e.g., PM5, PM10) across China. Please introduce the novelty of this study compared with previous studies. I think it is essential to add these contents in the introduction.

Response: Yes, we have constructed a virtually a complete set of major air quality parameters concerning both gaseous and particulate pollutants over a long period of time across China. While we have published a few studies on different parameters, this study adds to the list with the following unique aspects. First, it adds two new species of SO_2 and CO for the first time. Instead of devoting to a single pollutant, this paper deals with all gaseous pollutants of compatible quality over the same period with the same spatial coverage and resolution. This is highly valuable for the sake of studying their variations, relative proportions and attribution of emission sources. Per your suggestion, they are clarified in the following revised texts:

"To date, we have combined the advantages of artificial intelligence and big data to construct a virtually complete set of major air quality parameters concerning both particulate and gaseous pollutants over a long period of time across China, including PM_1 (2000–Present, Wei et al., 2019), $PM_{2.5}$ (2000–Present, Wei et al., 2020; Wei et al., 2021a), PM_{10} (2000–Present, Wei et al., 2021b), O_3 (1979–Present, Wei et al., 2022a; He et al., 2022), and NO_2 (2019–Present, Wei et al., 2022b), serving environmental, public health, economy, and other related research. This study is the continuation of our previous studies, which adds two new species of SO_2 and CO for the first time and also dates the data records of NO_2 back to 2013. Instead of devoting itself to a single pollutant, this paper deals with all gaseous pollutants of compatible quality over the same period with the same spatial coverage and resolution. In particular, considering that there are few public datasets of these three gaseous pollutants with such spatiotemporal coverages focusing on the whole of China, this is highly valuable for the sake of studying their variations, relative proportions, and attribution of emission sources, as well as their diverse and joint effects of different pollutant species on public health."

The authors should discuss the limitations of this paper and prospects for future work in the conclusion. The development of high-resolution dataset might not be the final aim.

Response: We have discussed the limitations of our study and prospects for future work in the revised conclusion as follows:

"Although a lot of new and/or useful data and analyses are presented in this study, they still suffer from some limitations. For example, input variables related to the emission inventory, modeled simulations, and assimilations still have considerable uncertainties. More influential factors stemming from regional economic and development differences need to be considered in more powerful artificial intelligence models to improve the prediction accuracy of air pollutants. The spatiotemporal resolutions of gaseous pollutants will be further improved by integrating information from polar-orbiting and geostationary satellites to investigate diurnal variations. In a future study, we will also reconstruct data records over the last two decades and investigate their long-term spatiotemporal variations, filling the gap of missing observations. This will help us understand their formation mechanisms and impacts on fine particulate matter and ozone pollution in China."

Specific comments:
Line 41-43: Please spell out these abbreviations, e.g., NOx, VOCs, et al. Also, please double-check and correct such issues throughout the paper.
Response: We have corrected and spelled out all abbreviations throughout the paper.

Lines 48 and 54: Should be MEE and WHO.
Response: Corrected.

Lines 64-69: The authors are suggested to highlight the main purpose and provide more descriptions of the main work here to enrich the Introduction.
Response: We have clarified the main purpose and added more descriptions of our study in the revised Introduction as follows:

"In view of the above problems, the purpose of this paper is to reconstruct daily concentrations of three ambient gaseous pollutants (i.e., NO_2, SO_2, and CO) in China. To this end, relying on the dense national ground-based observation network and big data, including satellite remote sensing products, meteorological reanalysis, chemical model simulations, and emission inventories, we are capable of mapping three pollutant gases seamlessly (100% spatial coverage) on a daily basis at a uniform spatial resolution of 10 km since 2013 in China. Estimates were made using an extended and powerful machine-learning model incorporating spatiotemporal information, i.e., space-time extra-trees. Natural and anthropogenic effects on air pollution, including their physical mechanisms and chemical reactions, were accounted for in the modeling. Using this dataset, spatiotemporal variations of the gaseous pollutants, the impacts of environmental protection policies and the COVID-19 epidemic, and population risk exposure to gaseous pollution are investigated."

Lines 83-88: A long sentence suggests splitting.
Response: Done per your suggestion.

Line 97: $0.1° \times 0.1°$?
Response: Corrected.

Figures 2 and 3: Please clarify which cross-validated method was used.
Response: We have clarified this in the captions of these figures.

Section 3.2.3: Besides annual variations, it is also interesting to see how three gaseous pollutants changed in different seasons on both the national and regional scales during the study period.
Response: Thanks for your suggestion. We have discussed the changes in gaseous pollutants at the seasonal level in China and three key regions in the revised Section 3.2.2 as follows:

"Large seasonal differences were observed in the amplitude of gaseous pollutant (Figure 6), e.g., surface NO_2 decreased the most in winter, especially in the three urban agglomerations (↓24–31%), changing the least in autumn (especially in the YRD). Surface SO_2 showed much larger decreases in all seasons, especially during the cold seasons (↓55–81%), due to the implementation of stricter "ultra-low" emission standards (Q. Zhang et al., 2019; Li et al., 2022a). Surface CO had similar seasonal changes as SO_2 but 1.5–3.3 times smaller in amplitude."

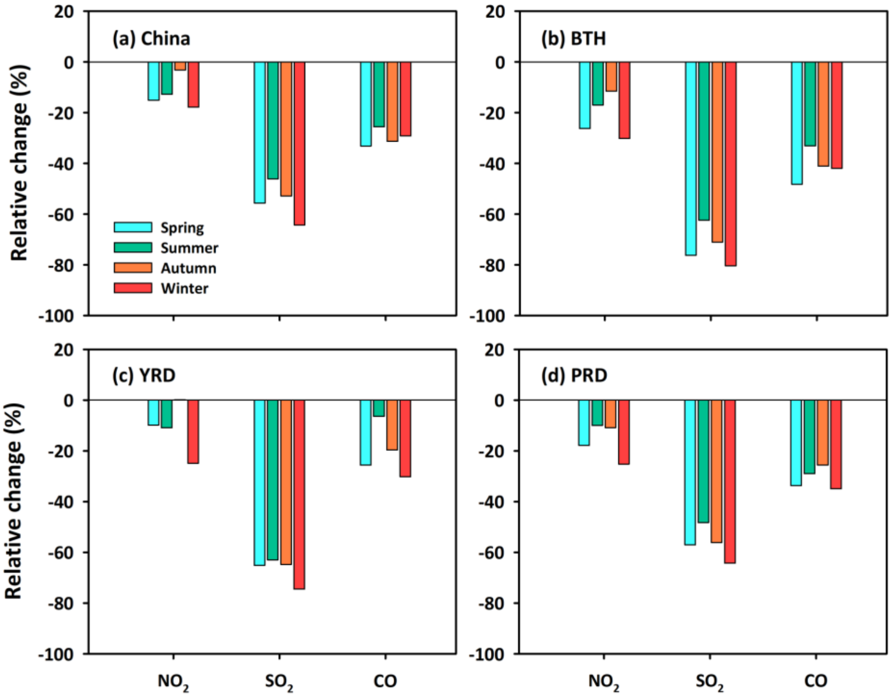

Figure 6. Relative changes (%) in seasonal mean surface NO_2, SO_2, and CO concentrations between 2013 and 2020 over (a) China, (b) the Beijing-Tianjin-Hebei (BTH) region, (c) the Yangtze River Delta (YRD), and (d) the Pearl River Delta (PRD).

Lines 286 and 294: References are needed to support the evidence here.
Response: Done per your suggestion.

Figures 9 and 10: Since the air quality guidelines have been newly updated in 2021, it is suggested to show the spatial distributions and variations of the percentage of polluted days exceeding both the WHO recommended long-term and short-term AQG levels and interim targets.

Response: Thanks for your suggestions. We have updated this section by discussing the spatiotemporal variations of national and regional polluted days according to the new WHO recommended AQG levels and interim targets in the revision as follows:

"With the daily seamless datasets, we can evaluate the spatial and temporal variations of short-term population-risk exposure to the three gaseous pollutants by calculating the number of days in a given year exceeding the new recommended short-term minimum interim target (IT1) and desired air quality guidelines (AQG) level defined by the WHO in 2021 (WHO, 2021). The area exceeding the recommended levels (i.e., daily NO_2 > 120 μg/m^3, SO_2 > 125 μg/m^3, and CO > 7 mg/m^3) was generally small in eastern China (Figure S7). High NO_2-exposure risks were mainly found in Beijing and Hebei Province and a handful of big cities (e.g., Jinan, Wuhan, Shanghai, and Guangzhou), while high SO_2-exposure risks were mainly observed in Hebei, Shandong, and Shaanxi Provinces. The risk of high CO pollution was small, only found in some scattered areas in the NCP. In general, both the area and the possibility of occurrence exposure to high pollution has gradually decreased over time, almost disappearing since 2018.

By contrast, most areas of eastern China had a surface NO_2 exposure exceeding the AQG level (Figure 8), especially in the north and economically developed areas in the south (proportion > 80%). Both the extent and intensity are decreasing over time, but it is still a problem, suggesting that stronger NO_x controls are needed in the future. Most of the main air pollution transmission belt in China (i.e., the "2 + 26" cities, Figure 1) had surface SO_2 levels exceeding the AQG level at the beginning of the study period. Thanks to strict control measures, these polluted areas sharply decreased after 2015, almost disappearing in 2020. Controlling CO was much more successful in China, with less than 10% of the days in the BTH exceeding the acceptable standard in the early part of the study period. Most areas have reached the CO AQG level since 2018.

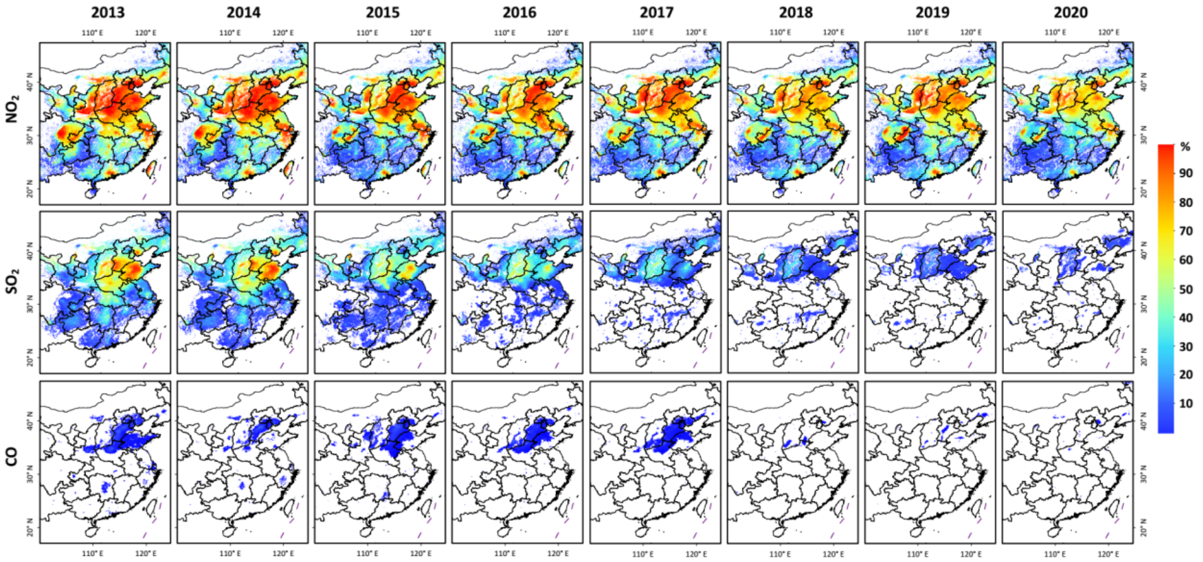

Figure 8. Spatial distributions of the percentage of days exceeding the WHO recommended short-term desired air quality guidelines level for surface NO_2 (daily mean > 25 μg/m^3), SO_2

(daily mean > 40 μg/m^3), and CO (daily mean > 4 mg/m^3) for each year from 2013 to 2020 in populated areas in eastern China.

Figure 9 shows the percentage of days with pollution levels exceeding WHO air quality standards in three key regions. BTH was the only region experiencing high NO_2 and SO_2 exposure risks (i.e., daily mean > IT1), dropping to zero since 2017 and 2016, while YRD and PRD had no high risks of exposure to the three gaseous pollutants (Figure 9a-b). There was also no regional high CO-pollution risk (Figure 9c). However, although declining continuously, regional surface NO_2 levels failed to meet the short-term AQG level in 2020, with 61–73% of the days exceeding the AQG level. More efforts toward mitigating NO_2 levels in these key regions are thus needed. Continual decreases in the number of days above the AQG level were also observed in surface SO_2, reducing to near zero in 2014, 2016, and 2018 in the PRD, YRD, and BTH, respectively. Less than 3% of the days in the BTH and YRD had surface CO levels exceeding the AQG level. Surface CO levels were always below the AQG level in the PRD."

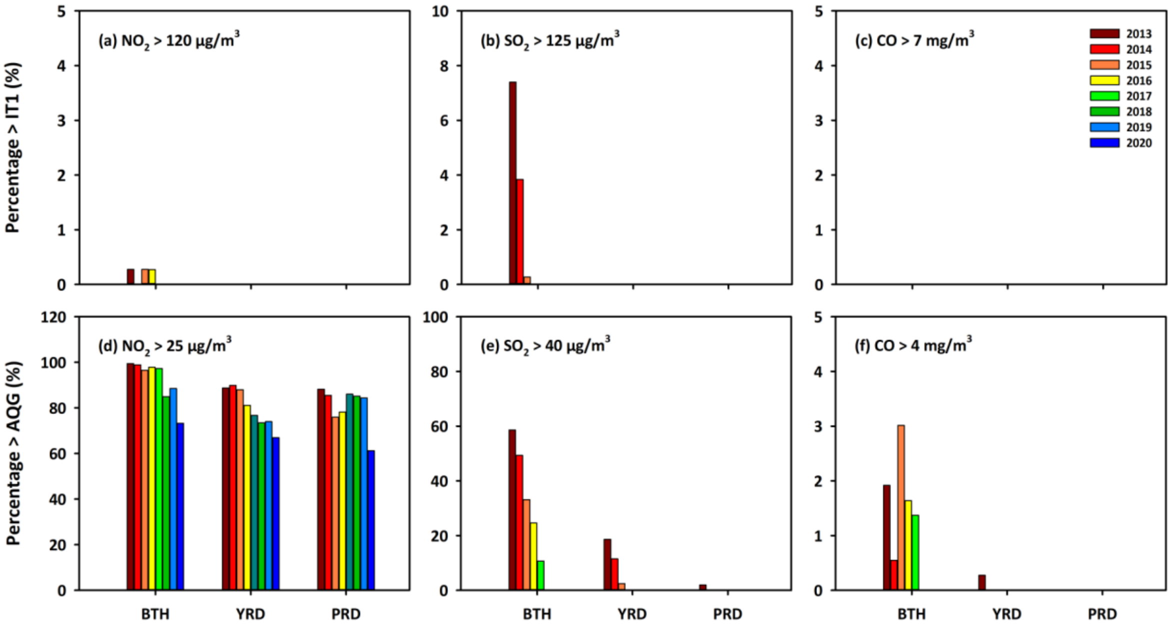

Figure 9. Percentage of days (%) exceeding the WHO recommended short-term (a-c) minimum interim target (IT1) and (d-f) desired air quality guidelines (AQG) level for surface NO_2, SO_2, and CO for each year from 2013 to 2020 in three typical urban agglomerations: the Beijing-Tianjin-Hebei (BTH) region, the Yangtze River Delta (YRD), and the Pearl River Delta (PRD)."