Dear Anonymous Referee #1,

We are submitting the point-by-point responses to your comments. We thank you for comments and suggestions, and hope you are satisfied with our responses.

The major changes made in the revised version include:

1. Add hourly in-situ NO_2 measurements analysis in the supplement.
2. Add discussions about the tropospheric O_3 transport and synoptic pattern effects on surface O_3 in the revised manuscript.
3. Add explanation about the preponderant role of NO titration in surface ozone increase in the revised manuscript.
4. Add the average diurnal profiles of pollutants in the revised supplement.

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Corresponding author: Dr. Janping Huang

Point-by-point responses to the Comments/Suggestions

Overview

The paper deals with the rapid increase in summer surface ozone over the North China Plain during 2013–2019 and the hypothesis that this decrease would be a side effect of reduction control of atmospheric particulate matter is examined. I would suggest publication of the paper, after the issues raised below are addressed.

General comments

1. I think that there are some key measurements missing in order to undertake a proper review of this manuscript. The most important is the lack of hourly in-situ NO2 measurements from the same stations providing the hourly ozone measurements, so that to be able to check if the ozone increase is directly related to the corresponding NO_2 decrease of surface concentrations The reason is that it is already known that for most urban stations the sum $NO_2+O_3=O_X$, called also potential ozone is constant (e.g. Kley et al., 1994; Kalabokas et al., 2000), so that any NO_2 decrease is directly related with an exactly equivalent increase in ozone (in ppb) through reaction with ozone (NO titration), which is very rapid. The presented data of total NO_2 column might provide some indication on that, but it is expected to be less efficient than in-situ measurements.

Thank the reviewer for the suggestion. The in-situ NO_2 measurements are now included for a comparison between year 2013 and year 2019 (Fig.T1). The in-situ measured NO_2 showed a similar decreasing trend to the total column NO_2 (Fig. T2).

To better understand the relationship of increase in surface O_3 with the decrease in NO_2, the change in monthly mean Ox (a sum of O_3 and NO_2) is plotted in Fig. T3. It is clear that Ox showed an increasing trend over the past 7 years during daytime and nighttime in both Beijing and the NCP region. The decrease in NO_x emissions could be part of the main reasons causing the rapid increase in O_3 over the NCP region where O_3 formation is dominated by the VOC-limited regime, but we do not see that "any NO_2 decrease is directly related with an exactly equivalent increase in ozone (in ppb) through reaction with ozone (NO titration)" as the reviewer expected. In fact, this isn't inconsistent with the statement that Ox is a conservative quantity over short time scales (Kley et al., 1994) since we are looking at the change over a long-time period (i.e., 7 years). Will add our response to address the comment in the revised version.

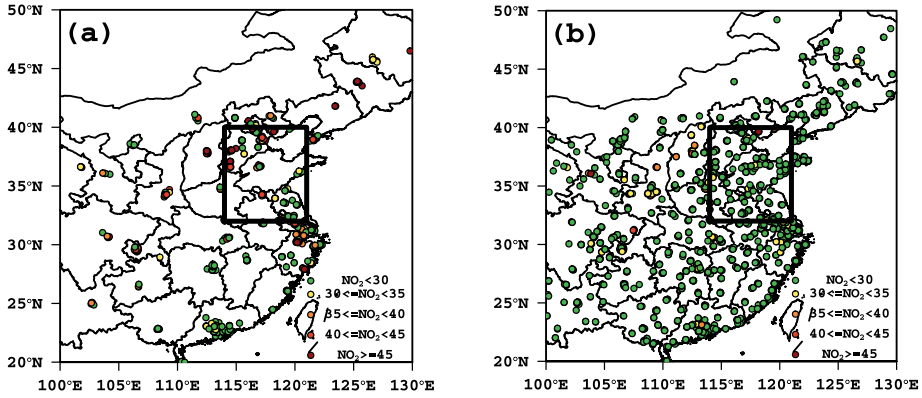

Figure T1. A comparison of spatial distributions of monthly mean of NO_2 (μg m^{-3}) monitored by China National Environmental Monitoring Center between (a) 2013 and (b) 2019 in eastern China (NCP indicated by the box).

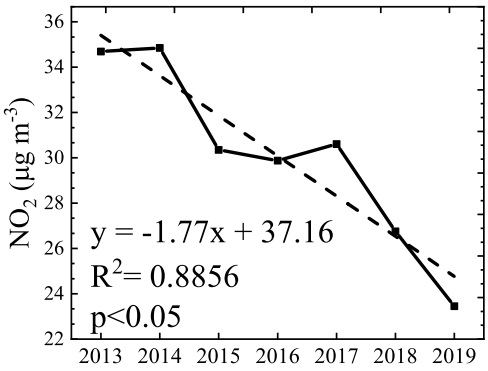

Figure T2. Long-term changes in monthly mean of observed NO_2 averaged over the North China Plain in June over the period of 2013–2019.

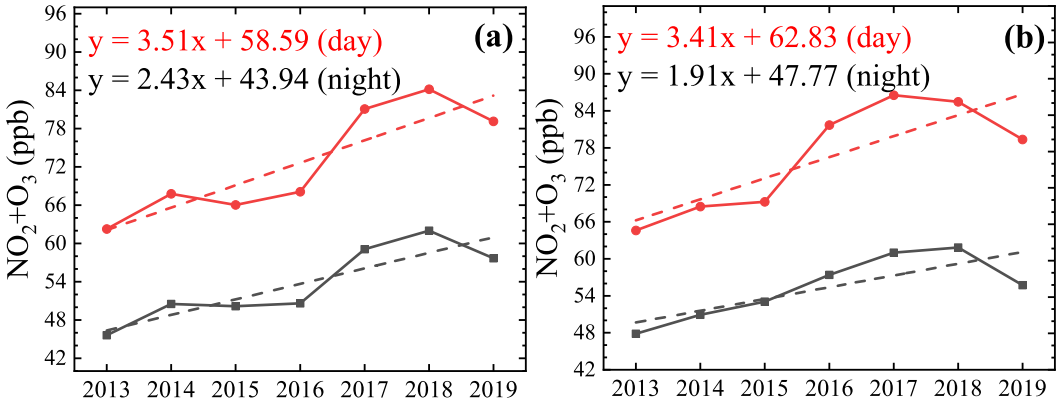

Figure T3. Long-term changes in monthly mean of observed NO_2 averaged over the North China Plain (a) and urban areas Beijing in daytime (redline) and nighttime (blackline) in June over the period of 2013–2019.

2. In addition, the issue of tropospheric ozone and its subsequent influence to the boundary layer and surface background ozone concentrations should be also taken into account. In relation to that, in my opinion, a weak point of the paper is that the levels of measured surface ozone are mainly related to the photochemical ozone production over the examined region of China. On the other hand, the issue of background ozone levels and their variability within the boundary layer and the free troposphere are not (or very little) discussed. For this purpose, I think that it would be quite helpful to take into account a relatively recent extended review paper on tropospheric ozone on global scale, including SE Asia which is one of the most important global tropospheric ozone hotspots (Gaudel et al, 2018, Elem Sci Anth, 6: 39. DOI: https://doi.org/10.1525/elementa.291 and also references therein). From my perspective and based on my expertise of analyzing ozone episodes in the Mediterranean region, I would just point out that the possibility of vertical ozone transport in the troposphere influencing the boundary layer and surface ozone values (a major factor in the Mediterranean, especially in

its eastern part during summer but also in its western part during spring) is not mentioned in the manuscript and so all measured ozone is considered to be produced by local photochemistry from precursor pollutant emissions emitted in China only. This might not be always the case, especially during the May-September period when the tropospheric influence to the boundary layer gets its maximum height while at the same time the tropospheric ozone maxima are observed during the same period of the year, with subsequent influence to the boundary layer and surface ozone values depending on the prevailing synoptic meteorological conditions.

We agree with the reviewer that the change in tropospheric O_3 could exert an important impact on ozone in the atmospheric boundary layer (ABL) or near surface. For instance, Jiang et al. (2015) presents an ozone episode in the southeast costal of China, and found that the downward transport of O_3 from the UTLS region driven by a typhoon is the key factor causing a large increase in surface O_3 by 21-42ppb. However, East Asia including the NCP region is considered as a net export region of O_3 from the ABL rather than an import (e.g., Cooper et al., 2010; Lin et al., 2012). On the other hand, the long-range transport of O_3 from Africa may exert an important impact on O_3 peaks in Asia around 25^0N, i.e., the south of our study area, NCP (32^0N-40^0N) with the largest impact in boreal winter and early spring (>10 ppb) and the lowest in boreal summer (<6ppb) (Han et al., 2018).

The impacts of downward transport of tropospheric O_3 and regional transport are not included in our current study mainly due to two reasons. First, the MM model does not have a capability of simulating vertical transport. Second, no vertical observational data (like ozone sounding or Lidar observational data in this region over the study period) are available. We have included the first point in the Discussion and will add a statement of "the impact of tropospheric O_3 should be taken into account when the observational data available in this region" in the revised version to address the reviewer's comment.

In relation to the above, tropospheric vertical ozone measurements over China (e.g. Ding et al., 2008; Zhang et al., 2020) would be needed for a thorough assessment together with tropospheric satellite ozone data. In addition, synoptic weather patterns might influence greatly the tropospheric as well as the surface ozone concentrations (e.g. Kalabokas et al., 2013; Kalabokas et al., 2017) and this issue is not discussed.

Assessment of the change in tropospheric O_3 is an attractive topic, and we like to pursue it in another study separately. To our knowledge, tropospheric O_3 retrieved from satellite measurements remains a large uncertainty given 90% of total O_3 is located in the stratosphere. In addition, no ozone sounding data (e.g., Zhang et al., 2020) and other observations such as Measurement of Ozone by Airbus In-service airCraft' (MOZAIC) program data (Ding et al., 2008) are available for performing further analysis in this region during the study period. We admit that the MM model used in this study does not have a capability of quantifying the impact of vertical transport on surface O_3, and we have included this in our discussion.

We agree with the reviewer that synoptic pattern may exert an important impact on tropospheric and surface O_3 concentrations. For instance, high concentrations of surface O_3 or O_3 episodes

occurred in western Mediterranean and central Europe were usually linked with anticyclone synoptic pattern which leads to a large-scale subsidence, clear sky, and high temperature (e.g., Kalabokas et al., 2013; Kalabokas et al., 2017). Yin et al. (2019) conclude that synoptic patterns play a critical role in summer ozone pollution in eastern China. Under the control of zonally enhanced East Asian deep trough, the local hot, dry air and intense solar radiation enhance the photochemical reactions and produce more O_3. However, the rapid increase of surface ozone in NCP during the past seven years was mainly due to anthropogenic emissions based on recent publications (Li et al., 2019; Liu and Wang, 2020a, 2020b). The inter-annual magnitude variations of the domain synoptic patterns may have an important impact on surface O_3 episodes, and its impact on the long-term changes needs a further investigation. Thus, we include discussion on impact of individual meteorological factors such as air temperature, the atmospheric boundary layer height, etc. rather than synoptic patten on long-term surface O_3 in our study.

To address reviewer's comment, we will add additional discussions on this comment in the revised version.

Overall, I think that the submitted paper presents some interesting data and ideas regarding the recent increasing trend of surface ozone in China but I think that the above described missing information is essential for a proper review of this manuscript.

Thanks for positive comments and we tried our best to include all the available observational data in this study. In-site NO_2 measurements are included in the revised version as suggested.

Specific comments

Page 2, lines 216-220: This is reasonable as higher NO/NO2 levels increase the ozone destruction in urban and semi urban stations, through NO titration.

Thanks for comments. We agree that higher NO/NO_2 levels contribute to more ozone through NO titration.

Page 9, lines 235-236: This applies to stations with low NO emissions in their surroundings. As mentioned before in most urban and semi urban stations, the NO titration is the controlling factor.

Thanks for comments. We will add the discussion about NO titration in the manuscript.

Page 14, lines 379-386: This in fact reflects the preponderant role of NO titration. Lower NO emissions destroy less ozone, which in most stations is originated from the tropospheric/boundary layer background.

Thanks for your comments. We will add explanation about the preponderant role of NO titration in surface ozone increase in the revised manuscript.

Supplement: I would suggest plotting also the average diurnal profiles of pollutants (O3, PM2.5) per season, at least for spring and summer.

Thanks for suggestion. We will add the average diurnal profiles of pollutants in the revised supplement.

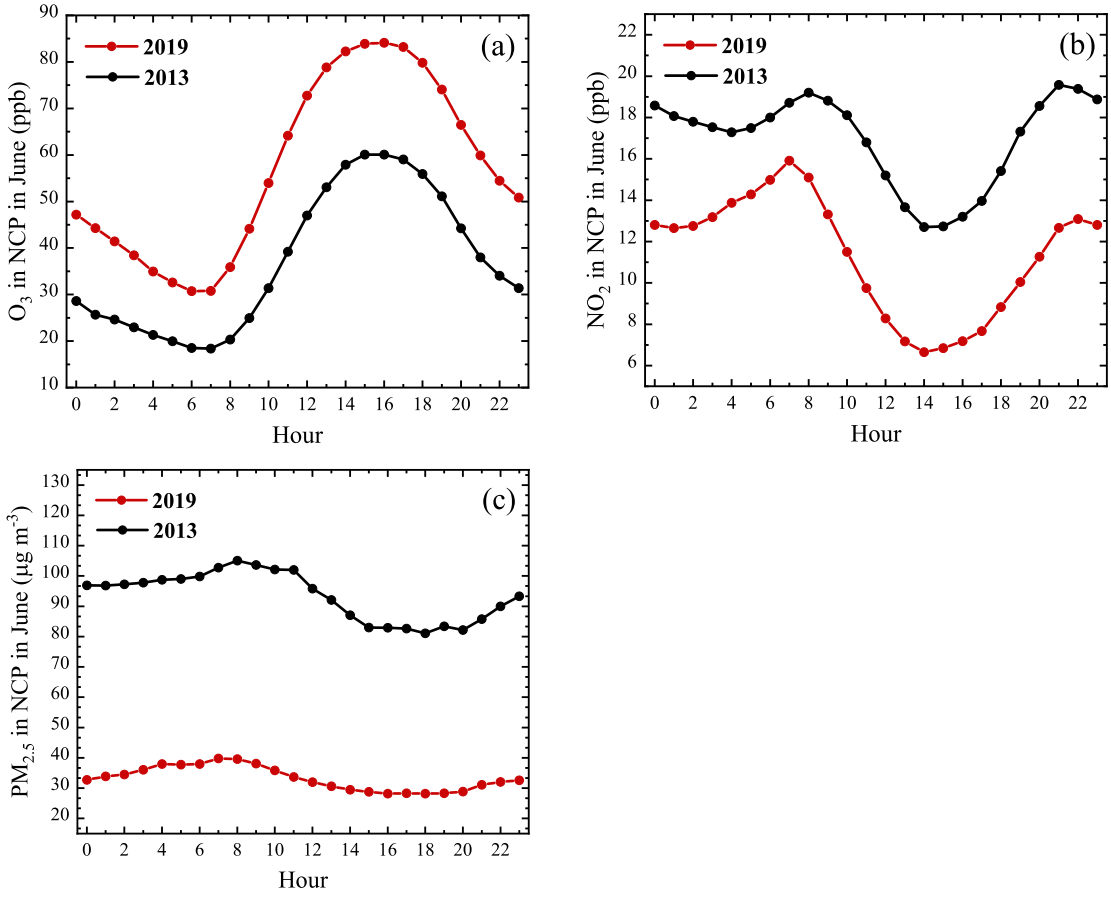

Figure T4. Average diurnal profiles of (a) O_3, (b) NO_2, (c) $PM_{2.5}$ in June of 2013 (black lines) and 2019 (red lines).

Technical comments

Page 26, line 665 (Fig. 3): PBLH (g, h).

Thanks. We will revise it.

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