Responses to Reviewers' comments

We would like thank the reviewers for the time and efforts in reviewing our manuscript. We have addressed all comments and revised the manuscript accordingly as detailed below (referee's comments in black, and responses in blue). Note that the line number refers to the revised manuscript.

Referee 1

Wang et al. analyze the surface ozone trends in China in 2014-2020, and use NOx and VOCs measurements in a box model to investigate the shift in ozone sensitivity regime in Beijing and Shanghai in this period. They find that ozone levels increased from 2014 to 2017, remained flat afterward, and decreased in 2020. They also find that summertime ozone sensitivity in Beijing and Shanghai has changed from a VOC-limited regime to a transition regime during 2014–2020. This study focuses on an important issue and is overall in high quality. The paper is well-written and easy to follow. I would recommend publication after the following comments being addressed.

My judgement is that the highlight of this paper is using 2014-2020 VOCs and NOx measurements in Beijing and Shanghai in the box model to identify the change of ozone sensitivity. The ozone trend in China itself has been documented in a number of studies as recognized by the authors and is well understood. I would suggest highlighting the novelty and finding of shift in ozone sensitivity in the title and introduction, instead of ozone trends.

Reply: We agree with the reviewer and we have emphasized the change of ozone sensitivity in the title and introduction accordingly. In addition, although the ozone trend in China has been reported in previous studies, the seasonal and spatial characteristics of ozone trend have not been fully understood. Therefore, we also emphasized them in the title and introduction.

Lines 1-3: Long-term trend of ozone pollution in China during 2014-2020: distinct seasonal and spatial characteristics and ozone sensitivity

Lines 122-142: Chinese government launched the Air Pollution Prevention and Control Action Plan in 2013–2017 and the Clean Air Action plan in 2018–2020 to reduce anthropogenic emissions (Cheng et al., 2019). In this case, ozone precursors decreased a lot while ozone pollution remained severe (Shao et al., 2021). Therefore, it is necessary to clearly understand the response of ozone to precursors' changes. The response of ozone to precursors' changes is primarily determined by the ozone precursor sensitivity. Wang et al. (2021) has analyzed the ozone precursor sensitivity using satellite observations of formaldehyde to NO_2. There are more other studies analyzing the ozone precursor sensitivity by using chemical transport models (Chen et al., 2021a;Kang et al., 2021;Li et al., 2019). Comprehensive measurements of O_3 precursors (VOCs and NO_X) and meteorological factors (photolysis frequencies, temperature and humidity) help to better identify the ozone precursor sensitivity (Kleinman et al., 1997;Kleinman, 2005). In this study, with comprehensive measurement data constrained in the observation-based box model, the ozone sensitivity regimes can be better diagnosed (Wang et al., 2020) and become complimentary to early studies. The goal of this study is to elucidate the spatial distribution, seasonal variation and temporal trends of ozone as well as the ozone precursor sensitivity in China by using comprehensive surface observations. Our study will provide a better understanding of the response of ozone to emission reductions, and inform the development of control measures to effectively mitigate ozone in the future.

Line 47: From Figure 5 it is clearly that ozone increases extend to 2019. I would suggest removing "reached a plateau after 2017".

Reply: Thank you. We have modified it. Ozone increase indeed extends to 2019. It is worth noting that the overall increasing rate of ozone after 2017 is slower than before 2017.

Lines 47-49: Although ozone precursors, including nitrogen oxides (NO_X) and carbon monoxide (CO), continuously decreased, ozone concentrations generally increased throughout the seven years with a slower increasing rate after 2017.

Line 95-97: In general, the authors can do better in catching up the more recent studies of ozone trends and ozone sensitivity in China. An example here, there are also studies pointing out the increases in tropospheric ozone in northern mid-latitudes extend to more recent years (e.g. 2017) than 2000 (Cooper et al., 2020; Gaudel et al., 2020).

Reply: Many thanks. To do better in catching up the more recent studies of ozone trends in China, we have moved the Table 1 and related discussion to the introduction in lines 84-102. We also included the more recent studies of ozone sensitivity in China in lines 125-131. We also cite the paper of Gaudel et al., 2020.

Lines 84-102: Measured ozone trends in previous studies are summarized in **Table 1**. For direct comparison of these results reported in different units, we have included estimated trends in units of % yr^{-1} for all studies. Xu and Lin (2011) and Verstraeten et al. (2015) have reported that tropospheric ozone concentrations increased in summer during 1979–2005 in the North China Plain and 2005–2010 in Eastern China at a rate of 1.1% and 3.0% yr^{-1}, respectively, based on satellite measurements. Urban ozone concentrations increased significantly in Beijing, Shanghai, Hongkong, Sichuan Basin and other cities during the past one to two decades at rates of 2.0 to 6.7% yr^{-1} (Gao et al., 2017;Cheng et al., 2016;Wang et al., 2020;Chen et al., 2021b;Li et al., 2020;Lu et al., 2018). A significant increase in ozone (+1.6% yr^{-1}) was detected at Shangdianzi, a rural site in the North China Plain (Ma et al., 2016). A moderate increase was detected at the global background site (Waliguan site) in western China (+0.44% yr^{-1}) (Xu et al., 2018;Xu et al., 2020). No significant trend was detected at either the eastern coastal Changdao site (Wang et al., 2020) or the Longfengshan site on the northeastern edge of China (Xu et al., 2020). In general, these studies show that ozone concentrations in China have risen in the past three to four decades.

Lines 117-118: Gaudel et al. (2020) reported that tropospheric ozone has increased above 11 regions of the Northern Hemisphere since the mid-1990s.

Lines 125-131: Therefore, it is necessary to clearly understand the response of ozone to precursors' changes. The response of ozone to precursors' changes is primarily determined by the ozone precursor sensitivity. Wang et al. (2021) has analyzed the ozone precursor sensitivity using satellite observations of formaldehyde to NO_2. There are more other studies analyzing the ozone precursor sensitivity by using chemical transport models (Chen et al., 2021a;Kang et al., 2021;Li et al., 2019).

Line 102-106: It might be a bit biased to state "characterization of ozone trends in China remain sparse" and "not yet well understood how changes in precursor emissions influence the trend of ozone in China". In fact studies of spatiotemporal ozone trends in China have been a lot as shown in Table 2. Wang et al. (2021) has addressed "how changes in precursor emissions influence the trend of ozone" using satellite observations, and there are even more studies testing the response by using chemical transport models (e.g. Chen et al., 2021) as mentioned in Section 3.3. The authors may want to soften the tune in the literature review and highlight the novelty of this study compared to the existing literatures.

Reply: Many thanks. We have softened the tune in the introduction and highlight the novelty of this study according to your suggestions. In this study, with comprehensive measurement data constrained in the observation-based box model, the ozone sensitivity regimes can be better diagnosed and become complimentary to early studies.

Lines 122-142: Chinese government launched the Air Pollution Prevention and Control Action Plan in 2013–2017 and the Clean Air Action plan in 2018–2020 to reduce anthropogenic emissions (Cheng et al., 2019). In this case, ozone precursors decreased a lot while ozone pollution remained severe (Shao et al., 2021). Therefore, it is necessary to clearly understand the response of ozone to precursors' changes. The response of ozone to precursors' changes is primarily determined by the ozone

precursor sensitivity. Wang et al. (2021) has analyzed the ozone precursor sensitivity using satellite observations of formaldehyde to NO_2. There are more other studies analyzing the ozone precursor sensitivity by using chemical transport models (Chen et al., 2021a;Kang et al., 2021;Li et al., 2019). Comprehensive measurements of O_3 precursors (VOCs and NO_X) and meteorological factors (photolysis frequencies, temperature and humidity) are key to identifying the ozone precursor sensitivity (Kleinman et al., 1997;Kleinman, 2005). In this study, with comprehensive measurement data constrained in the observation-based box model, the ozone sensitivity regimes can be better diagnosed (Wang et al., 2020) and become complimentary to early studies. The goal of this study is to elucidate the spatial distribution, seasonal variation and temporal trends of ozone as well as the ozone precursor sensitivity in China by using comprehensive surface observations. Our study will provide a better understanding of the response of ozone to emission reductions, and inform the development of control measures to effectively mitigate ozone in the future.

Line 130-137: Here urban and non-urban sites are distinguished by population density. It is a bit simple but fine. Nevertheless, I suggest the authors also refer to more comprehensive definition of urban/non-urban sites from the Tropospheric Assessment Report (Schultz et al., 2017) and a recent study by Gao et al. (2020), and see whether the urban/non-urban separation may influence the analyses.

Reply: Thank you. Population density is indeed a simple metric. Here the population density is used to roughly distinguish between urban and non-urban sites. Although the classification of urban and non-urban regions is not only determined by the population density, the population density can reflect the strength of human activity to a large extent.

In the Tropospheric Ozone Assessment Report (Fleming et al., 2018), the population density together with NOAA night-time lights were used for the urban and non-urban classifications. The addition of night-time light data may promote the urban/non-

urban separation compared with solely using population density. Given that the night-time light data is not easy to acquired, we didn't utilize this data in this study.

According to the requirements by the Ministry of Ecology and Environment (MEE, http://www.mee.gov.cn/), Gao et al (2020) divided the national ambient air quality monitoring sites into three types of sites, including evaluation, suburban and background sites. Evaluation sites are obliged to be built in urban areas and distributed equally to cover the whole city. Suburban sites are built more than 20 km away from main pollution sources and urban centers, and background sites are built even farther (> 50 km) away. Based on this method, we analyzed the ozone trend in urban and suburban areas as shown in the **Figure** below. The ozone levels in urban and suburban sites are similar. This result slightly differs from our results in Fig. 4 which indicate the ozone levels in urban areas are slightly higher than those in non-urban areas. This indicates different urban/non-urban separation methods can influence the analyses.

Please note that the Fig. 4 has been revised. In the original Fig. 4, we analyze data of all 1669 sites. However, this is not reasonable because the there is fewer sites in previous years than later years. Now I have updated the Fig. 4 by using the same number of sites for all years (about 960 sites). The updated Fig. 4 shows a smaller difference between urban and non-urban sites. We apologize for the misleading results in the previous Fig. 4. We have revised the Fig.4 and relevant description in the manuscript.

Lines 155-158: The Tropospheric Assessment Report (Fleming et al., 2018) used the population density together with NOAA night-time lights to classify the urban and non-urban sites. Given that it is not easy to acquire the night-time light data in China, urban and non-urban sites are distinguished by population density in this study.

Lines 311-314: The levels of these four metrics at urban sites were slightly higher than at non-urban sites with difference less than 8% (**Fig. 4**). These results in China

differ from those in Europe and North America, where the mean levels of these metrics at urban sites were slightly lower than those at non-urban sites (Fleming et al., 2018).

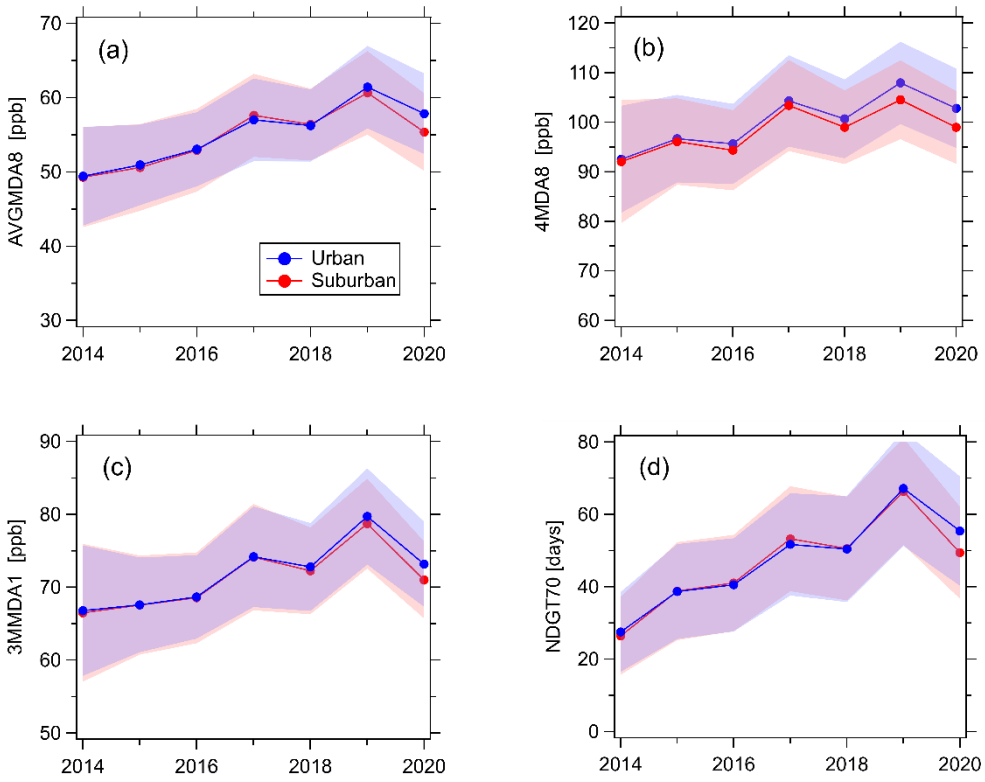

Figure. Variations in four ozone metrics (AVGMDA8, 4MDA8, 3MMDA1 and NDGT70) at urban and non-urban sites during 2014-2020. Here urban and non-urban sites are distinguished by the approach suggested by Gao et al (2020).

Line 148: Suggest citing Lefohn et al. (2018) for ozone metric information and implication.

Reply: Thanks. We have added the citation.

Lines 174-177: We derived all metrics from the hourly measurements that were filtered by data quality control procedures following the Tropospheric Ozone Assessment Report (TOAR) data completeness requirements and procedures (Lefohn et al., 2018).

Line 168-171: The use of in-situ long-term VOCs data is much appreciated and makes the study stand out from existing literatures. However, the VOCs measurements, in particular their trends, should be presented in figure to support the study.

Reply: As shown in the Fig. 8 below, total OH reactivity of VOCs decreased at a rate of -0.39 s^{-1} yr^{-1} (r^2=0.56) in Beijing and -0.46 s^{-1} yr^{-1} (r^2=0.59) in Shanghai. We have added Fig. 8 in the manuscript. The OH reactivity of VOCs is defined as the sum of all VOC concentrations multiplied by their respective reaction rate coefficients with OH. This parameter is more related to ozone production rate than the total VOC concentration. In addition, we have added the detailed measurement technique of VOCs in Section 2.1.

Lines 356-367: Figure 8 shows the trend of measured VOCs reactivity in Beijing and Shanghai in summertime during 2014-2020. The VOCs reactivity is defined as the sum of all VOCs concentrations multiplied by their respective reaction rate coefficients with OH, as shown in Eq. 3. The VOCs reactivity is more related to ozone production rate than VOCs concentrations (Zhang et al., 2014;Wang et al., 2020;Wang et al., 2021b). The summertime VOCs reactivity decreased at a rate of -0.39 s^{-1} (-7.5%) yr^{-1} (r^2=0.56) in Beijing and -0.46 s^{-1} (-8.4%) yr^{-1} (r^2=0.59) in Shanghai. It is notable that the trends of VOCs reactivity in Beijing and Shanghai are different from that of VOCs emissions across China.

$$VOCs\ reactivity = \sum_i^n k_{VOC_i}[VOC_i] \quad (3)$$

k_{VOC_i} represents the reaction rate coefficients between OH radicals and VOC species *i*. [VOC_i] is the concentration of VOC species *i*. n is the number of VOC species.

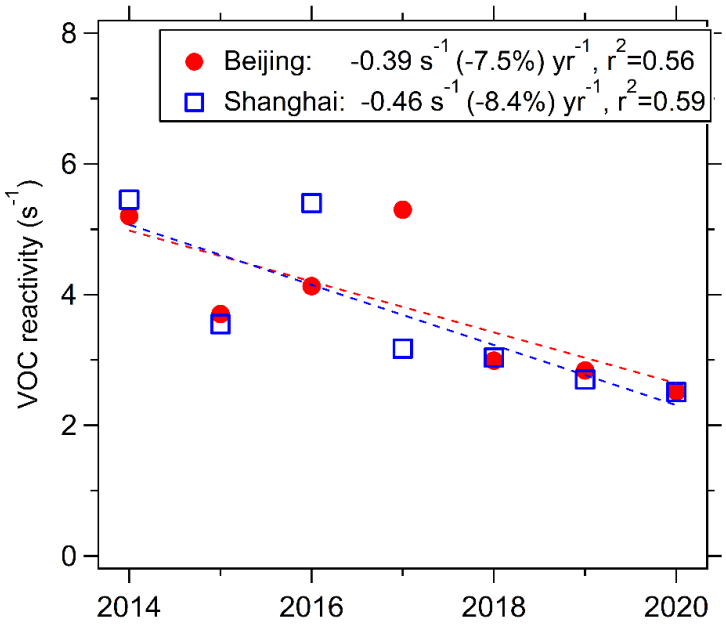

Figure 8. Variations in averages of daytime VOCs reactivity in Beijing and Shanghai, summertime during 2014-2020.

Line 331-333. Do VOCs measurement in Beijing and Shanghai show decrease from 2019 to 2020? This is critical for understanding ozone decrease in 2020. A recent study by Yin et al. (2021) suggests the ozone decrease in 2020 is also partly attributed to decrease in VOCs emissions. I wonder whether the authors can prove or disprove such statement from their observations and box model analyses.

Reply: As shown in Fig. 8, OH reactivity of VOCs decreased by 10% in Beijing and 8% in Shanghai from 2019 to 2020. Hence, we think that the decrease in VOCs can partly contribute to the ozone decrease in 2020. Our result can support the result of Yin et al. (2021).

Referee 2

This study investigated the long-term trend of ozone in China during 2014–2020 based on the surface observational data of O3, NO2 and CO, VOCs measurements at Beijing and Shanghai and a zero-dimension photochemical box model. The authors found that ozone levels increased from 2014 to 2017 and reached a plateau after 2017, then decreased in summer and increased in winter from 2019 to 2020. Using the photochemical box model, the changes in ozone sensitivity regime were reported to be the cause of the O3 changes. The methods are clearly outlined and the conclusions are solid. I have the following comments that can be addressed.

General comments

Many previous studies have examined the long-term ozone trend over China and revealed the similar patterns and causes as in this study. The authors did not fully emphasize the novelty of this study and should illustrate what are the new scientific findings or the new tools to explain the findings. In my view, the usage of VOCs measurements and the box model is probably the new data and tool to analyze the distinct seasonal and spatial O3 variations.

Reply: Many thanks. We agree with you that the usage of VOCs measurements and the box model is the new data and tool to analyze the distinct seasonal and spatial O_3 variations. By using comprehensive measurement data, we can identify the shift in ozone sensitivity accurately. According to your suggestions, we have emphasized our novelty in the introduction.

Lines 122-142:

Chinese government launched the Air Pollution Prevention and Control Action Plan in 2013–2017 and the Clean Air Action plan in 2018–2020 to reduce anthropogenic emissions (Cheng et al., 2019). In this case, ozone precursors decreased a lot while ozone pollution remained severe (Shao et al., 2021). Therefore, it is necessary to clearly understand the response of ozone to precursors' changes. The response of ozone to precursors' changes is primarily determined by the ozone precursor sensitivity. Wang et al. (2021) has analyzed the ozone precursor sensitivity using satellite observations of

formaldehyde to NO_2. There are more other studies analyzing the ozone precursor sensitivity by using chemical transport models (Chen et al., 2021a;Kang et al., 2021;Li et al., 2019). Comprehensive measurements of O_3 precursors (VOCs and NO_X) and meteorological factors (photolysis frequencies, temperature and humidity) help to better identify the ozone precursor sensitivity (Kleinman et al., 1997;Kleinman, 2005). In this study, with comprehensive measurement data constrained in the observation-based box model, the ozone sensitivity regimes can be better diagnosed (Wang et al., 2020) and become complimentary to early studies. The goal of this study is to elucidate the spatial distribution, seasonal variation and temporal trends of ozone as well as the ozone precursor sensitivity in China by using comprehensive surface observations. Our study will provide a better understanding of the response of ozone to emission reductions, and inform the development of control measures to effectively mitigate ozone in the future.

Also, the authors attributed the O3 trends to photochemistry along, which is incomplete. For the long-term trends, emissions exert a dominant role in the changes in pollutants, but for a short-term from 2019 to 2020, year-by-year changes in meteorological field can largely influence the O3 distribution through changes in both photochemistry and transboundary transport. From the analysis, the authors indicated that the shift of ozone sensitivity regime was the reason for the unique O_3 changes in 2019-2020, but they did not exclude the physical processes due to interannual variation in meteorological fields. It can be discussed in the manuscript.

Reply: Thank you for your comments. According to your suggestions, we have analyzed the levels of meteorological factors including temperature, relative humidity, wind speed and air pressure in Beijing and Shanghai during 2019-2020, and elucidated the influence of these meteorological factors on ozone.

Lines 442-453: Furthermore, the influence of meteorological factors on the ozone change from 2019 to 2020 was investigated. Table S1 shows the average values of primary meteorological factors including temperature, relative humidity, wind speed, wind direction, air pressure and photolysis frequency of NO_2 ($j(NO_2)$) in 2019 and 2020

in Beijing and Shanghai. Temperature increased in winter but decreased in summer from 2019 to 2020. Previous studies indicate that ozone concentrations show a positive correlation with temperature (He et al., 2017;Jacob and Winner, 2009). We surmise that the changes in temperature may partly contribute to the contrasting changes in ozone concentrations between summer and winter from 2019 to 2020. Besides temperature, the significant changes in relative humidity may also influence the ozone change. $j(NO_2)$ maintained stable in both winter and summer from 2019 to 2020, indicating a minor effect of photolysis frequencies on ozone changes.

Table S1. Variations in meteorological factors including temperature (T), relative humidity (RH), wind speed (WS), wind direction (WD), air pressure (P) and photolysis frequency of NO_2 ($J(NO_2)$) from 2019 to 2020 in Beijing and Shanghai.

City	Period	T (°C)	RH (%)	WS (m s^{-1})	WD (°)	P (hpa)	$J(NO_2)$ (10^{-3} s^{-1})
Beijing	Winter 2019	1.1	24	1.5	140	1019	0.0012
	Winter 2020	2.1	36	2.1	202	1018	0.0012
	Summer 2019	27.7	51	2.5	190	996	0.0025
	Summer 2020	26.7	57	2.2	198	997	0.0025
Shanghai	Winter 2019	7.6	73	3.7	184	1030	\
	Winter 2020	8.6	65	1.7	164	1054	\
	Summer 2019	30.6	74	4.0	144	1035	\
	Summer 2020	28.2	66	1.9	144	1044	\

Specific comments

Lines 168–175: Please descript the VOCs data in detail since that they are the key data for the conclusions.

Reply: Thanks. We have described the measurement of VOCs in detailed.

Lines 202-210: VOCs were measured using a commercial gas chromatography-mass spectrometer/flame ionization detector (GC-MS/FID) system coupled with a cryogen-

free preconcentration device (Wang et al., 2014). The system contains two-channel sampling and GC column separation which is able to measure C2–C5 hydrocarbons with the FID in one channel and measure C5–C12 hydrocarbons using MS detection in the other channel. The time resolution was 1 h, and ambient air was sampled during the first 5 min of each hour for both channels. The uncertainties for VOC measurements by GC–MS/FID are estimated to be 15 %–20 % (Wang et al., 2014).

Line 203: Four O_3 metrics were used in this study. What are the similarities and differences between the results using the four metrics?

Reply: Many thanks. We have included this discussion in the manuscript. The four metrics all generally increased from 2014 to 2020 with the increasing rate getting slower after 2017. During 2014-2017, AVGMDA8 and NDGT70 increased at rates of 7.4% yr^{-1} and 20% yr^{-1} respectively. 4MDA8 and 3MMDA1, which characterize extremely high ozone levels, increased at rates of 3.7% yr^{-1} and 3.5% yr^{-1} respectively. Obviously, the increasing rates of 4MDA8 and 3MMDA1 were significantly slower than those of AVGMDA8 and NDGT70.

Lines 316-317: The four metrics all generally increased from 2014 to 2020 with the increasing rate getting slower after 2017.

Lines 318-323: During 2014-2017, AVGMDA8 and NDGT70 increased at rates of 7.4% yr^{-1} and 20% yr^{-1} respectively. 4MDA8 and 3MMDA1, which characterize extremely high ozone levels, increased at rates of 3.7% yr^{-1} and 3.5% yr^{-1} respectively. Obviously, the increasing rates of 4MDA8 and 3MMDA1 were significantly slower than those of AVGMDA8 and NDGT70.

Line 214: The high O_3 over the North China Plain is also related to the high temperature extremes (e.g., Wang et al., 2022).

Reply: Thanks. We have added it in the manuscript.

Lines 251-253: In addition, the high O_3 concentration over the North China Plain is also related to the high temperature extremes (Wang et al., 2021a).

Lines 267-284: The description and table 2 are the O_3 trends reported in previous findings, which should be listed in introduction, unless they were compared with the data in this study in details.

Reply: Thank you. We have moved the description and table 2 to the introduction.

Lines 84-102: Measured ozone trends in previous studies are summarized in **Table 1**. For direct comparison of these results reported in different units, we have included estimated trends in units of % yr^{-1} for all studies. Xu and Lin (2011) and Verstraeten et al. (2015) have reported that tropospheric ozone concentrations increased in summer during 1979–2005 in the North China Plain and 2005–2010 in Eastern China at a rate of 1.1% and 3.0% yr^{-1}, respectively, based on satellite measurements. Urban ozone concentrations increased significantly in Beijing, Shanghai, Hongkong, Sichuan Basin and other cities during the past one to two decades at rates of 2.0 to 6.7% yr^{-1} (Gao et al., 2017;Cheng et al., 2016;Wang et al., 2020;Chen et al., 2021b;Li et al., 2020;Lu et al., 2018). A significant increase in ozone (+1.6% yr^{-1}) was detected at Shangdianzi, a rural site in the North China Plain (Ma et al., 2016). A moderate increase was detected at the global background site (Waliguan site) in western China (+0.44% yr^{-1}) (Xu et al., 2018;Xu et al., 2020). No significant trend was detected at either the eastern coastal Changdao site (Wang et al., 2020) or the Longfengshan site on the northeastern edge of China (Xu et al., 2020). In general, these studies show that ozone concentrations in China have risen in the past three to four decades.

Line 352: The author noted that transport effects were not considered, but they are important in the variations in O_3 concentrations in China (e.g., Yang et al. 2014, 2022).

Reply: Many thanks. The transport effect of ozone is indeed important in ozone pollution in China. However, our study haven't considered the transport effect which probably plays a crucial role in ozone trend and may also lead to uncertainties concerning the dignosis of ozone precursor sensitivities. We point out this issue in the Sec. 3.4. Please double check if the citation of Yang et al. is correct.

Lines 469-473: Lastly, the transport effect of ozone is important in ozone pollution in China (Han et al., 2018;Shen et al., 2022;Yang et al., 2022). However, our study has

not considered the transport effect which probably plays a crucial role in ozone trend and may also lead to uncertainties concerning the dignosis of ozone precursor sensitivities.

Line 408: I suggest the authors to add paragraphs to discussed the uncertainties and limitations in this study.

Reply: Many thanks. We have added a paragraph to discuss the limitations in this study.

Lines 454-473: There are several limitations of this study. One limitation is that the VOCs measurement data are only available in two megacities – Beijing and Shanghai. The trends of VOCs in Beijing and Shanghai cannot fully represent that in the whole country. As a result, the influence of the VOCs variation on the ozone trend across China is not completely elucidated. The diagnosis of ozone precursor sensitivity is also based on measurement data in the two megacities, which may not reflect the situation of the whole country. Another limitation of this study is that the photochemical box model is constrained by observations near the ground, hence may not accurately represent some aspects of the photochemistry throughout the boundary layer. The ozone precursor sensitivity in the upper layer of the boundary layer probably differs from that near ground under certain conditions due to varied VOCs and NO_X levels and meteorological factors with height (Zhang et al., 2018;Sun et al., 2018). Therefore, to acquire a more broaden and comprehensive diagnosis of ozone precursor sensitivity, the measurement of VOCs in more cities and over the whole boundary layer is required in the future. Lastly, the transport effect of ozone is important in ozone pollution in China (Han et al., 2018;Shen et al., 2022). However, our study has not considered the transport effect which probably plays a crucial role in ozone trend and may also lead to uncertainties concerning the dignosis of ozone precursor sensitivities.

Figure 6: Reframe the colorbar. Label and color confusion.

Reply: Many thanks. We have modified the color bar in Figure 6.

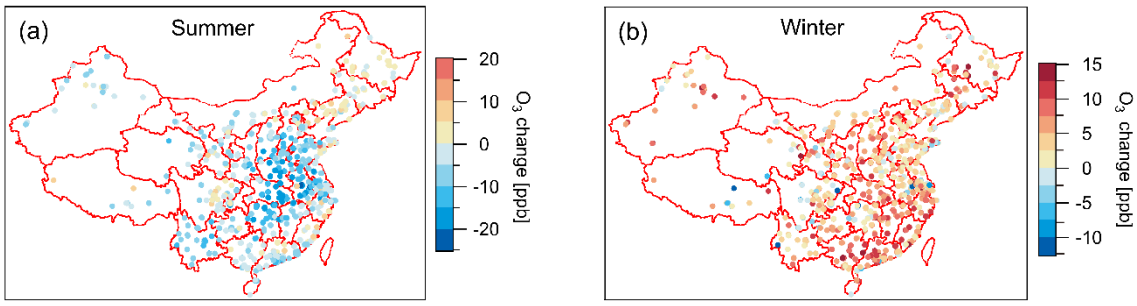

Figure 7: Why only the NO2 and CO were shown here without VOCs?

Reply: We have added the VOCs trend in Figure 8. As shown in the Fig. 8, total OH reactivity of VOCs decreased at a rate of -0.39 s^{-1} (-7.5%) yr^{-1} (r^2=0.56) in Beijing and -0.46 s^{-1} yr^{-1} (-8.4%) (r^2=0.59) in Shanghai.

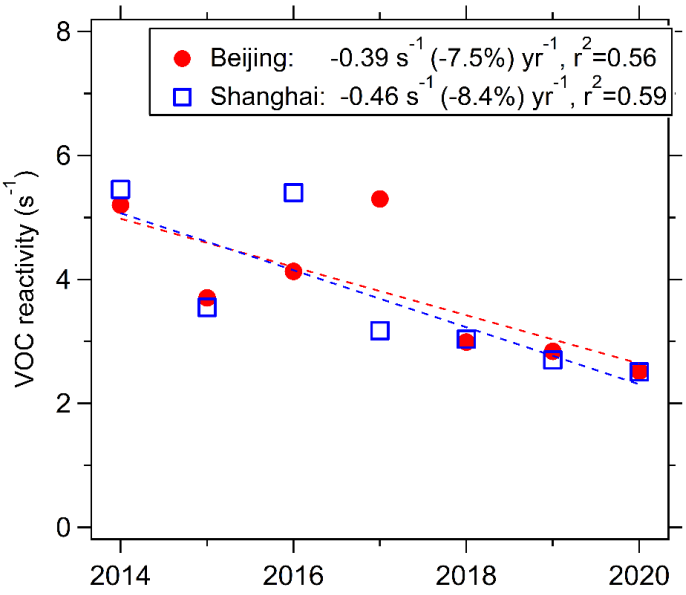

Figure 8. Variations in averages of daytime VOCs reactivity in Beijing and Shanghai, summertime during 2014-2020.

Lines 356-367: Figure 8 shows the trend of measured VOCs reactivity in Beijing and Shanghai in summertime during 2014-2020. The VOCs reactivity is defined as the sum of all VOCs concentrations multiplied by their respective reaction rate coefficients with

OH, as shown in Eq. 3. The VOCs reactivity is more related to ozone production rate than VOCs concentrations (Zhang et al., 2014;Wang et al., 2020;Wang et al., 2021b). The summertime VOCs reactivity decreased at a rate of -0.39 s^{-1} (-7.5%) yr^{-1} (r^2=0.56) in Beijing and -0.46 s^{-1} (-8.4%) yr^{-1} (r^2=0.59) in Shanghai. It is notable that the trends of VOCs reactivity in Beijing and Shanghai are different from that of VOCs emissions across China.

$$VOCs\ reactivity = \sum_i^n k_{VOC_i}[VOC_i] \quad (3)$$

k_{VOC_i} represents the reaction rate coefficients between OH radicals and VOC species i. [VOC_i] is the concentration of VOC species i. n is the number of VOC species.

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