Response to Reviewers' Comments

Dear Editor and Reviewers,

Thank you very much for your efforts in handling and evaluating our submission.
The review comments are very helpful for improving the original manuscript. We have carefully considered and tried to address all of these comments in the revised manuscript. Below are the detailed point-by-point responses to the review comments. For clarity, the reviewer's comments are listed below in *black italics*, while our responses and changes in the manuscript are highlighted in blue and red, respectively.
We look forward to receiving a further evaluation of our work.

Best regards,
Guy Brasseur and co-authors

Summary

The authors perform four sets of model simulations over China: a base case, a 50% NOx emission reduction, a 50% AVOC emission reduction, and a combined 50% NOx and 50% AVOC emission reduction. The science presented is largely sound. However, the paper currently reads as a lengthy report rather than a scientific manuscript. There is extensive repetition of conclusions in different sections of the paper, and a lot of the more impactful conclusions get lost in the details. I recommend that the authors restructure the paper around their main conclusions and science questions, rather than organizing by metric as is done currently. I also suggest that many of the minor details presented in the main text be moved to the supplement so that only text in support of the main conclusions of the paper is presented in the main text.

Major comments

As is currently written, a lot of the main conclusions and the primary storyline get lost in the details presented. In addition, a lot of conclusions get repeated in different sections. For example, a lot of the same conclusions are drawn when assessing ozone production regime (Section 3.1), odd oxygen production and destruction (Section 3.2.2), and ozone concentrations (Section 3.3.2). Uniting these sections will reduce the overall manuscript length, help highlight whether these different lines of analysis lead to consistent conclusions and help to bring the overall conclusions of the study to the forefront.

I wonder if the authors could comment on the relevance of broad NOx and AVOC reductions in China, in contrast to reductions that vary by sector or by region. For example, are transportation sector reductions more/less likely than stationary emissions, and what might this imply for the chemistry discussed? In addition, a lot of the analysis presented highlighted changes in radical cycling related to changes in OVOC emissions. Do we expect AVOC emission reductions to be consistent across classes of VOCs, or could the effectiveness of emission reductions of OVOCs vs hydrocarbons differ? And, similarly, do we expect consistent emission reductions for VOCs with higher and lower HCHO yields? How might this impact the conclusions presented here?

Author's reply: Thank you very much for your comments that are very helpful. Based on your comments, we significantly condensed our paper, with a shortened description of the text on our model results and are focusing on insights about how to mitigate ozone increase in urban China. For the metrics related to photochemical activity, we limited our analysis to the Atmospheric Oxidative Capacity (AOC) in the main text and moved other calculated metrics to the Supplement. The structure of this paper is as follows.

"This paper is structured as follows. Section 2 introduces the setups of the model system and describes the simulations performed for specified reductions in the emissions of

primary pollutants. In Section 3, we first analyzed the response in the near-surface concentration of ozone precursors and intermediate to primary emission reductions. Then, we also discuss the changes in the ozone formation regime. Further, we derive the associated changes in ozone, and aerosols to emission reductions. Finally, we describe the sensitivity of the atmospheric oxidative capacity (*AOC*) to the reduction in emissions. A summary and implication for policy making of our study is provided in Sec. 4."

Regarding policy to mitigate ozone, we highlight that the reduction in emissions should be implemented by regions and by the type of environment, with different strategies for the south and in the north of China and for the urban versus non-urban areas. We suggest that the reduction in NO_x emissions be coordinated with the reduction in AVOCs emissions, especially with reduction of alkenes, aromatics and unsaturated OVOC emissions, including methanol or ethanol. This conclusion is based on the contribution of these different species to the daytime oxidative capacity of atmosphere and to the secondary formation of OVOCs. The summary of our policy implication [also shown in Table 1 and Figure 1 (Table 2 and Figure 10 in Text)] is shown below.

"*Paths to mitigation.* We conclude this paper by highlighting a few chemical paths that should be considered when designing a mitigation policy for a reduction of ozone in the urban areas of China. Figure 10 presents a schematic description of the chemical mechanisms involved in the chemical production of atmospheric ozone and highlights how different reaction paths tend to change the ozone abundance in response to a reduction in NO_x and anthropogenic VOC (AVOCs) emissions. This graph shows that a reduction in NO_x emissions tends to increase the ozone concentration by (1) reducing the rate of the $NO + O_3$ reaction (ozone titration); (2) by increasing the rate of the $HO_2 + NO$ reaction due to an increase in the HO_2 level associated with the reduced uptake of this radical by a lowered aerosol load; (3) by an increase in the atmospheric oxidizing capacity (*AOC*) through OH- and ozone-related reactions. The graph also shows that a decrease in AVOCs emissions tends (1) to reduce the level of the HO_x radical and hence the ozone production by the $HO_2 + NO$ reaction; (2) to enhance the level of HO_x due to the reduced aerosol uptake and (3) to reduce the *AOC* with a negative effect on the ozone concentration. The relative importance of these different chemical mechanisms varies with location and environmental conditions.

We conclude that, in winter when the background ozone concentration is low, the reduction of NO_x emissions tends to increase the level of near-surface ozone, while the reduction in AVOC emissions has the opposite effect. This conclusion applies both in rural and in urban areas. A combined reduction in the emissions of these two primary pollutants tends to decrease the level of ozone in rural areas, but to increase ozone in urban areas. Thus, in urban areas during winter, an effective approach to reduce the surface ozone concentration is through a strong limitation in the emissions of volatile organic compounds.

In summer when the ozone level is generally high, the reduction of NO_x emissions is an effective action to reduce the ozone concentration in rural areas, but this measure is counterproductive in the NOx-saturated urban areas where ozone is controlled by VOCs. In fact, in urban areas during this season, the mechanisms involved in ozone mitigation are complex. For example, when NO_x emissions are reduced, the atmospheric OH concentration is enhanced because of its reduced destruction by NO_2. Following this increase in the OH concentration, an increase in the level of OVOCs, whose photolysis is an important source of HO_x radicals, also leads to accelerated ozone production and further amplifies the oxidation of VOCs. In addition, the increase in *AOC*, linked to the reaction of OH and ozone with alkenes and the reactions of OH with OVOCs also contribute to an increase in the ozone production. Further, the reduction in the aerosol load resulting from a reduction in the emissions of aerosol precursors promotes the ozone formation by decreasing the aerosol extinction and by reducing the uptake of HO_2. If combined with a 50% reduction in AVOCs, the increase in OVOCs and *AOC*, due to reduced NO_x emissions, can be offset. However, the aerosol-related promotion of the level of OH and HO_2 radicals can be enhanced, highlighting the complexity of summertime ozone mitigation in urban areas.

Table 2 provides quantitative information on the response of ozone at different urban locations for January and July. In urban areas, the reduction in the level of surface ozone requires a reduction in the emissions of anthropogenic VOCs. However, for practical reasons, a 50% reduction in AVOCs emissions, as assumed in our study, is difficult to implement over a short period of time. With the known contribution of the VOCs-related reactions to the *AOC*, the reduction in the emissions of alkenes, aromatics, and unsaturated OVOCs, especially the aldehydes and alcohols, should be a priority. The development of efficient mitigation strategies based on the reduction of AVOCs emissions requires, however, more detailed investigations on the reactivity of individual VOCs and on their potential impact on the ozone formation."

Table 1. Ozone changes due to reduction in emissions in urban sites (in percentage)

Location	Sites name	Ozone changes in winter condition (Mean ± SD)			
		NOx[a]	AVOCs[b]	N+A[c]	TOTAL[d]
North	Beijing	25.0 ± 25.2[e]	-2.5 ± 1.3	22.0 ± 32.8	20.0 ± 19.5
East	Shanghai	33.2 ± 35.3	-18.2 ± 13.5	21.8 ± 20.5	22.7 ± 18.8
South	Guangzhou	21.4 ± 22.6	-17.1 ± 11.2	7.1 ± 3.2	10.0 ± 3.5
West	Chengdu	21.3 ± 23.8	-9.4 ± 8.5	14.1 ± 8.3	20.3 ± 13.5
Location	Sites name	Ozone changes in summer condition (Mean ± SD)			
		NOx	AVOCs	N+A	TOTAL
North	Beijing	6.4 ± 3.8	-21.8 ± 19.2	-5.5 ± 4.2	-7.3 ± 5.0
East	Shanghai	17.1 ± 12.8	-22.9 ± 20.8	-2.9 ± 2.1	-2.6 ± 1.5
South	Guangzhou	15.0 ± 13.1	-14.5 ± 13.5	1.3 ± 1.0	1.3 ± 0.9
West	Chengdu	5.5 ± 4.5	-14.5 ± 10.2	-5.5 ± 2.0	-4.5 ± 1.9

a-d. Sensitivity cases with a 50% reduction in NO_x emissions (NOx), AVOCs emissions (AVOCs), NOx and AVOCs (N+A), and other species (NOx, AVOCs, CO, NH_3, SO_2) under consideration (TOTAL).

e. Values are displayed in the average ozone changes during daytime (06:00-19:00) in percentage with the standard deviation as the error bar. (ozone changes = (case value -base-line case) *100).

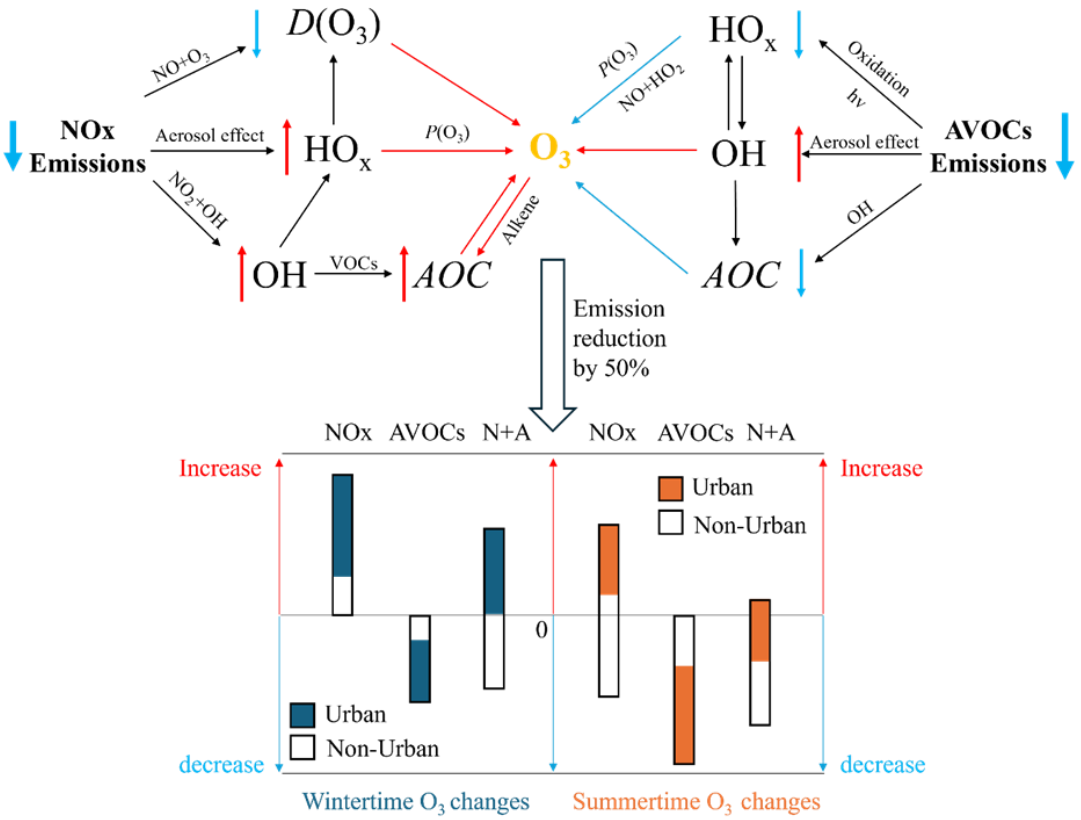

Figure 1. Schematics show the responses of oxidative processes, associated with ozone formation, to the reduction in primary emissions of NO_x and AVOCs in urban areas (VOC-limited) in winter and summer. Arrows besides the chemicals represent the changes associated with the reduction in emission. (decrease trend shown in blue; increase trend shown in red) Blue and red arrows closing to O_3 represent the positive and negative contributions to the ozone formations. *AOC*, $P(O_3)$, and $D(O_3)$ are the abbreviations of the Atmospheric Oxidative Capacity, production of ozone, and destruction of ozone. Bar figure shows the ranges of ozone changes in whole of China (black bar), in non-urban areas (white part in the bar), and in urban areas (colored part in the bar) in three emissions cases (NOx, AVOCs, and N+A represent the case with emission reduction in NO_x, Anthropogenic VOCs (AVOCs), and the combined NO_x and AVOCs emissions, respectively) relative to BASE cases in winter and summer conditions.

Minor comments

Line 110: typo (nitration vs titration)

Author's reply: Revised.

Line 236: Zhang et al, 2009 get these numbers from Tonnesen and Dennis, 2000, so Tonnesen and Dennis, 2000 should be cited here as the original citation.

Author's reply: Changed.