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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impacts of aerosol direct effects on tropospheric ozone through changes in atmospheric dynamics and photolysis rates" *by* Jia Xing et al.

Jia Xing et al.

xingjia@tsinghua.edu.cn

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We thank the referee for a thoughtful and detailed review of our manuscript. Incorporation of the reviewer's suggestions has led to a much improved manuscript. Below we provide a point-by-point response to the reviewer's comments and summarize the changes that have been incorporated in the revised manuscript.

[Comment]: In this paper, the authors have applied the WRF-CMAQ model to analyze the impact of aerosols on tropospheric ozone through their impacts on atmospheric dynamics and photolysis rates. Their results indicate that reducing aerosols may have negative impacts on ozone which need to be considered for air quality management

in China. The topic is of general interest given the focus on reducing PM2.5 pollution. The simulations have been designed appropriately to address the goals of the study.

[Response]: We thank the reviewer for the overall positive assessment of the manuscript and recognition of the implications of the results of the analysis presented.

[Comment]: However, the authors have not considered the prominent way aerosols impact tropospheric ozone formation - via heterogeneous reactions - which leads me to question the conclusions of this study. Several studies have highlighted the role of aerosols in modulating ozone via heterogeneous reactions (eg., Liao and Seinfeld, 2005; Ti et al., 2005; Pozzoli et al., 2008; Xu et al., 2012; Lou et al., 2014), which have largely been ignored in this study. The authors need to provide a strong justification for ignoring the impact of aerosols on ozone via heterogeneous reactions, before I can recommend this paper for publication.

[Response]: We agree with the reviewer that the heterogeneous reactions associated with aerosols have substantial impacts on ozone, including hydrolysis of N2O5, irreversible absorption of NO2 and NO3 as well as the uptake of HO2, and are well documented in the literature (Tang et al., 2004; Tie et al., 2005; Liao and Seinfeld, 2005; Pozzoli et al., 2008; Li et al., 2011; Xu et al., 2012; Lou et al., 2014). Our model contains comprehensive treatment of heterogeneous hydrolysis of N2O5 (Davis et al., 2008; Sarwar et al., 2012; Sarwar et al., 2014). While our model accounts for such a heterogeneous reaction, we have not quantified its impacts on ozone in this study.

However, in this study, we focused our analysis on another important aspect of aerosol influence (ADE, the aerosol direct effects), i.e., scattering and absorption of incoming solar radiation and how the subsequent effects of the associated cooling suppresses atmospheric ventilation. The assessment of impacts of aerosol direct effects (ADE) is an important aspect of designing emission reduction strategies that seek co-benefits associated with reductions in both particulate matter and ozone. In this study, we examine the ADE impacts which were not well quantified in previous studies. It may

be noted that all model calculations analyzed in this study included the heterogeneous N2O5 hydrolysis pathway. We agree with the reviewer that all influences of aerosols on ozone need to be addressed before definite policy-relevant conclusions regarding their overall impact can be reached. China plans to implement stringent control actions aimed at lowering the ambient concentrations of PM2.5 in the next two decades. It will be necessary to quantify all the possible influences resulting from this expected reduction of aerosols, including changes in heterogeneous reactions associated with aerosols, as well as the changes expected in ADE discussed in this manuscript. In addition, secondary aerosol and ozone comes from both NOx and VOCs emissions. Reducing aerosols by controlling gaseous precursors will also have substantial impacts on ozone (Xing et al., 2017), which needs further evaluation.

To address the reviewer's concern, we have clarified the scope of our analysis in the revised manuscript as below:

(Page 2 Line 21-25) "Many studies suggest that aerosols may have substantial impacts on ozone through heterogeneous reactions including hydrolysis of N2O5, irreversible absorption of NO2 and NO3, as well as the uptake of HO2 (Tang et al., 2004; Tie et al., 2005; Li et al., 2011; Lou et al., 2014). While our model contains comprehensive treatment of the heterogeneous hydrolysis of N2O5 (Davis et al., 2008; Sarwar et al., 2012; Sarwar et al., 2014), we have not quantified its impacts on ozone in this study. However, ADE impacts on ozone have not been well evaluated previously."

(Page 10 Line 15-19) "Reducing aerosols will have substantial impacts on ozone. Quantification of the aerosol influence on ozone is important to understand co-benefits associated with reductions in both particulate matter and ozone. This study focused on the evaluation of ADE impacts which were not well quantified previously. However, the heterogeneous reactions associated with aerosols, as well as the impacts of emission controls of gaseous precursors on both aerosols and ozone also need to be studied in order to fully understand the influence from reducing aerosols on ambient ozone."

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[Comment]: P2L10: Please provide a reference for "ADE modulate the temperature, atmospheric ventilation, cloud and rainfall".

[Response]: We have provided the references for this sentence as below:

(Page 2 Line 10-12) "ADE modulate the temperature (e.g., Hansen et al., 1997; Mitchell et al., 1995), atmospheric ventilation (e.g., Jacobson et al., 2007; Mathur et al., 2010;), cloud and rainfall (e.g., Albrecht, 1989; Liou and Ou, 1989; Twomey, 1977)"

Mitchell JFB, Davis RA, Ingram WJ, Senior CA. On Surface Temperature, Greenhouse Gases, and Aerosols: Models and Observations. Journal of Climate 1995; 8: 2364-2386.

Hansen J, Sato M, Ruedy R. Radiative forcing and climate response. J Geophys Res-Atmos 1997; 102: 6831-6864

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[Comment]: P3 Section 2.1: What meteorological fields are used to drive WRF-CMAQ?

[Response]: The meteorological inputs for WRF simulations were derived from the NCEP FNL (Final) Operational Global Analysis data which has 1 degree spatial and 6-hour temporal resolution. NCEP ADP Operational Global Surface Observations were used for surface reanalysis which is used for indirect soil moisture and temperature nudging in the Pleim-Xiu Land Surface Model (PX LSM). The strength of nudging coefficients for four dimensional data assimilation and indirect soil temperature nudging employed in WRF have been tested and chosen to improve model performance for meteorological variables without dampening the effects of radiative feedbacks. The nudging coefficient for both u/v-wind and potential temperature is set to 0.00005 sec-1, while 0.00001 sec-1 is used for nudging of water vapor mixing ratio.

We have clarified in the revised manuscript as below:

(Page 3 Line 8-13) "The meteorological inputs for WRF simulations were derived from the NCEP FNL (Final) Operational Global Analysis data which has 1 degree spatial and 6-hour temporal resolution. NCEP ADP Operational Global Surface Observations were used for surface reanalysis and four dimensional data assimilation. We have tested and chosen proper strength of nudging coefficients, i.e., 0.00005 sec-1 is used for nudging of both u/v-wind and potential temperature, 0.00001 sec-1 is used for nudging of water vapor mixing ratio, to improve model performance without dampening the effects of radiative feedbacks (Hogrefe et al., 2015; Xing et al., 2015)."

[Comment]: P3L5,6: Please define acronyms (e.g., AERO6, BHCOAT) before using them.

[Response]: We have clarified the acronyms in the revised manuscript as below

(Page 3 Line 13-18) “In the model version used here, concentrations of gaseous species and primary and secondary aerosols are simulated by using Carbon Bond 05 (CB05) gas-phase chemistry (Sarwar et al., 2008) and the sixth-generation CMAQ modal aerosol model (AERO6) (Appel et al., 2013). The aerosol optical properties were estimated by the coated-sphere module (i.e., BHCOAT, Bohren and Huffman, 1983) based on simulated aerosol composition and size distribution (Gan et al., 2015).”

[Comment]: P4L20: Please clarify if this is for observations. The observations are hardly visible in Figure 2.

[Response]: Yes, this is for observations. We have replotted Figure 2 in the revised manuscript to make the comparison more evident, as below

(Page 4 Line 28 - Page 5 Line 3) “In January, higher DM1O3 concentrations are seen in PRD where solar radiation is stronger than in the north. The model generally captured the spatial pattern with highest DM1O3 in PRD over the simulated domain. Simulated DM1O3 in YRD, SCH and HUZ are higher than observations. Such overestimation might be associated with the relative coarse spatial resolution in the model. NO titration effects in urban areas were not well represented in the model.”

[Comment]: P4L24: Please provide a reference for “In January, O3 production in north China is VOC-limited regime”

[Response]: Following reviewer’s suggestion, a reference has been added as below:

(Page 5 Line 6-8) “In January, O3 production in north China is occurring in a VOC-limited regime (e.g., Liu et al., 2010), thus increases in NOx at the surface stemming from the stabilized atmosphere by ADE inhibit O3 formation due to enhanced titration by NO.”

Liu, X.H., Zhang, Y., Xing, J., Zhang, Q., Wang, K., Streets, D.G., Jang, C., Wang, W.X. and Hao, J.M., 2010. Understanding of regional air pollution over China using CMAQ, part II. Process analysis and sensitivity of ozone and particulate matter to precursor emissions. Atmospheric Environment, 44(30), pp.3719-3727.

[Comment]: P4: It would also be helpful to see maps of PM2.5 to assess if the reductions in O3 due to aerosol feedbacks are co-located with PM concentrations.

[Response]: Following the reviewer’s suggestion, we made the plot of PM2.5 concentrations as Figure R1 It clearly shows that the ADE effects on O3 are co-located with PM concentrations. Besides, we calculated the O3 responses to ADE under low- and high-PM2.5 levels, summarized in Table 2. Mostly, the O3 responses to ADE are larger under high PM2.5 levels, indicating the positive correlations between O3 responses and PM levels.

We have added corresponding discussions to the revised manuscript, as below:

(Page 6 Line 3-5) “Comparing the O3 responses to ADE (see $\triangle$-ADE in Table 2) under low- and high- PM2.5 levels, reveals that O3 responses to ADE are larger under high PM2.5 levels, indicating the positive correlations between O3 responses and PM2.5 levels.”

[Comment]: P4-5: How significant are the changes in O3 in response to Dynamics and Photolysis

[Response]: Out results suggest that ADE reduced surface daily maxima 1h O3 (DM1O3) in China by up to 39 μg m-3 through the combination of $\triangle$Dynamics and $\triangle$Photolysis in January, but enhanced surface DM1O3 by up to 4 μg m-3 in July.

The enhancement in peak O3 in summer found in this study is different from the traditional expectation of a negative O3 response to ADE when solely considering the reduction in solar radiance.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2017-198/acp-2017-198-AC2-supplement.pdf

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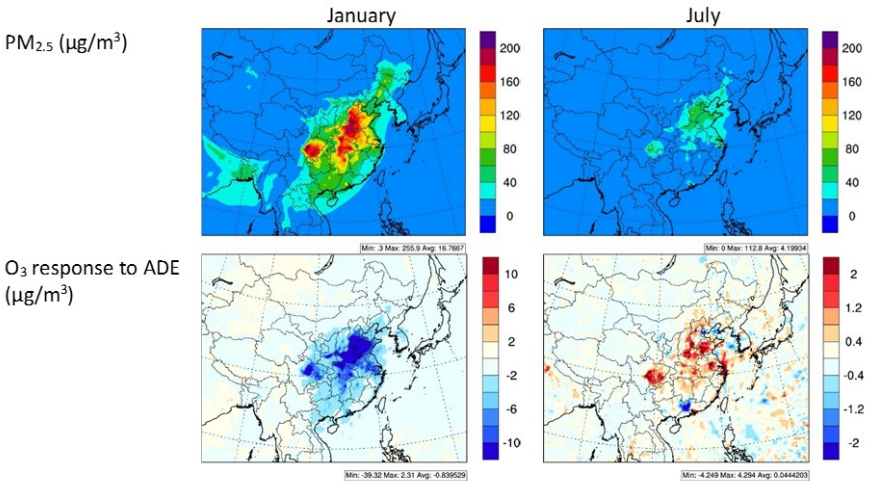

Figure R1 spatial distribution of $PM_{2.5}$ concentration and O_3 response to ADE

Fig. 1.