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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.

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We thank the reviewer for the detailed and thoughtful review of our manuscript. Incorporation of the reviewer's suggestion has led to a much improved manuscript. Detailed below is our response to the issues raised by the reviewer. We also detail the specific changes incorporated in the revised manuscript in response to the reviewer's comments.

[Comment]: The simulations appear to have been done carefully and the results are of some moderate interest, but the presentation is very difficult to wade through because of laborious analyses of model results that are of little interest and because of postage-

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stamp figures that seem like core dumps of obscure information.

[Response]: We thank the reviewer for the suggestion. To improve the flow of the manuscript, we have reduced the amount of information presented and also moved some of the information into the supplementary material. Furthermore, we have increased the size of several figures to better present the information in the revised manuscript.

[Comment]: it appears that the authors did not examine (or mention, unless I missed it) the aerosol effect on ozone through heterogeneous chemistry but I would expect this effect to be at least as large as the effects from dynamics and photolysis. Not accounting for the effect of aerosols on heterogeneous chemistry (for example through N2O5 hydrolysis) compromises in my opinion the policy-relevant conclusions about the sensitivity of ozone to aerosol reductions.

[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

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order to fully understand the influence from reducing aerosols on ambient ozone."

Reference

Li J., Wang Z., Wang X., Yamaji K., et al. Impacts of aerosols on summertime tropospheric photolysis frequencies and photochemistry over Central Eastern China. Atmospheric Environment, 2011, 45: 1817-1829.

Liao, H., Seinfeld, J.H., Global impacts of gas-phase chemistry–aerosol interactions on direct radiative forcing by anthropogenic aerosols and ozone, J. Geophys. Res., 2005, 110, D18208

Tang Y. H., Carmichael G. R., Kurata G., Uno I., et al. Impacts of dust on regional tropospheric chemistry during the ACE-Asia experiment: A model study with observations. Journal of Geophysical Research-Atmospheres, 2004, 109

Tie X. X., Madronich S., Walters S., et al. Assessment of the global impact of aerosols on tropospheric oxidants. Journal of Geophysical Research-Atmospheres 2005, 110

Lou S. J., Liao H., Zhu B. Impacts of aerosols on surface-layer ozone concentrations in China through heterogeneous reactions and changes in photolysis rates. Atmospheric Environment, 2014, 85: 123-138.

Pozzoli, L., Bey, I., Rast, S., Schultz, M.G., Stier, P., Feichter, J., Trace gas and aerosol interactions in the fully coupled model of aerosol-chemistry-climate ECHAM5-HAMMOZ: 1. Model description and insights from the spring 2001 TRACE-P experiment, J. Geophys. Res., 2008, 113, D07308

Xu, J., Zhang, Y.H., Zheng, S.Q., He, Y.J., Aerosol effects on ozone concentrations in Beijing: a model sensitivity study, J. Environ. Sci., 2012, 24 (4), 645–656

Davis, J. M., Bhave, P. V., and Foley, K. M.: Parameterization of N2O5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate, Atmos. Chem. Phys., 8, 5295–5311, doi:10.5194/acp-8-5295-2008, 2008.

Sarwar, G., Simon, H., Bhave, P., and Yarwood, G.: Examining the impact of heterogeneous nitryl chloride production on air quality across the United States, Atmospheric Chemistry & Physics, 12, 1-19, 2012.

Sarwar, G., Simon, H., Xing, J., Mathur, R.: Importance of tropospheric ClNO2 chemistry across the Northern Hemisphere, Geophysical Research Letters, 41, 4050-4058, 2014.

Xing, J., Wang, S. X., Jang, C., et al. Overview of ABaCAS: an air pollution control cost-benefit and attainment assessment system and its applications in China, EM of Air & Waste Management Association, 2017 April.

[Comment]: Page 2: what meteorological data assimilation is done in the WRF simulation and how would it affect the sensitivity of dynamics to aerosols?

[Response]: We followed our previous coupled model design (Xing et al., 2015). 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 s-1, while 0.00001 s-1 is used for nudging of the water vapor mixing ratio.

We have clarified it in the revised manuscript as below:

(Page 3 Line 7-12) “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).”

Reference

Hogrefe, C., Pouliot, G., Wong, D., Torian, A., Roselle, S., Pleim, J. and Mathur, R.: Annual application and evaluation of the online coupled WRF–CMAQ system over North America under AQMEII phase 2. Atmospheric Environment, 115, 683-694, 2015.

Xing, J., Mathur, R., Pleim, J., Hogrefe, C., Gan, C.M., Wong, D.C. and Wei, C.: Can a coupled meteorology–chemistry model reproduce the historical trend in aerosol direct radiative effects over the Northern Hemisphere?. Atmospheric Chemistry and Physics, 15(17), 9997-10018, 2015.

[Comment]: Page 4: the authors present as established fact that ADE increases boundary layer stability. I'm not necessarily disputing that but a few references would be helpful.

[Response]: Both aerosol scattering and absorption of incoming solar radiation result in reduced solar radiation impinging the ground causing reduced ground temperatures, while light-absorbing carbon aloft increases the temperature in the upper boundary layer, but cools the surface. This cooling increases stability of the boundary layer and reduces ventilation of pollutants in the boundary layer.

Following the reviewer's suggestion, in the revised manuscript we have included a few references analyzing this process chain. The manuscript discussion has been updated as follows:

(Page 5 Line 6-8) "In January, O3 production in north China is occurring in a VOC-limited regime, thus increases in NOx at the surface stemming from the stabilized atmosphere by ADE (Jacobson et al., 2007; Mathur et al., 2010; Ding et al., 2013; Xing et al., 2015) inhibit O3 formation due to enhanced titration by NO."

Reference

Jacobson, M. Z.; Kaufman, Y. J.; Rudich, Y. Examining feedbacks of aerosols to urban climate with a model that treats 3-D clouds with aerosol inclusions. J. Geophys. Res.

2007, 112, D24205

Mathur, R.; Pleim, J. E.; Wong, D. C.; Otte, T. L.; Gilliam, R. C.; Roselle, S. J.; Young, J. O.; Binkowski, F. S.; Xiu, A. The WRF-CMAQ Integrated On-Line Modeling System: Development, Testing, and Initial Applications. In Air Pollution Modeling and its Applications XX; Steyn, D. G.; Rao, S. T., Eds.; Springer: Netherlands, Netherlands, 2010; pp 155−159.

Ding AJ, Fu CB, Yang XQ, Sun JN, Petaja T, Kerminen VM, et al. Intense atmospheric pollution modifies weather: a case of mixed biomass burning with fossil fuel combustion pollution in eastern China. Atmospheric Chemistry and Physics 2013; 13: 10545-10554.

Xing, J.; Mathur, R.; Pleim, J.; Hogrefe, C.; Gan, C. M.; Wong, D. C.; Wei, C.; Wang, J. Air pollution and climate response to aerosol direct radiative effects: a modeling study of decadal trends across the northern hemisphere. J. Geophys. Res. 2015, 120, 12,221−12,236.

[Comment]: Page 5, line 8: the authors find that aerosols decrease photolysis rates in winter but increase them in summer, and it's not clear why there is this seasonal difference. I suppose indeed that scattering aerosol could increase photolysis rates in summer, but then why not in winter?

[Response]: We agree with the reviewer that the scattering aerosol could increase photolysis rates in both seasons, while absorbing aerosol act oppositely. The overall impacts on photolysis rates are depend on the combined effects of all types of aerosols. Similar results were found in Tie et al (2005) who reported that surface-layer photolysis rates in eastern China were reduced less significantly in summer than in winter.

In this study, we found that the response of photolysis rates to ADE presents a strong diurnal pattern, which shows reductions in the early morning and late afternoon, but shows a slight increase at noon. The reason might be associated with the enhanced

ambient precursor concentrations (due to less reaction in early morning) at noon when O3 reaches the daily maximum.

To address the reviewer's concern, we have clarified this discussion in the revised manuscript as below:

(Page 5 Line 20-21) "Similar results were found in Tie et al (2005) who reported that surface-layer photolysis rates in eastern China were reduced less significantly in summer than in winter."

(Page 5 Line 24-26) "This increase in precursor concentrations then leads to enhanced O3 formation later in the day which compensates for or even overwhelms the disbenefit from the reduced solar radiances."

Reference

Tie X. X., Madronich S., Walters S., et al. Assessment of the global impact of aerosols on tropospheric oxidants. Journal of Geophysical Research-Atmospheres 2005, 110

[Comment]: Line 11 further seems to contradict the statement on line 8 by saying that photolysis rates decrease in summer

[Response]: The photolysis reaction rate depends on solar radiances and precursor levels. We agree with the reviewer that our original statement is ambiguous. We clarified it as below:

(Page 5 Line 24-26) "This increase in precursor concentrations then leads to enhanced O3 formation later in the day which compensates for or even overwhelms the disbenefit from the reduced solar radiances."

[Comment]: Figures 3-6 seem like core dumps. I didn't see a colorbar legend for figures 5-6.

[Response]: We have reduced the content of these figures to focus on the most important aspects and added a colorbar legend for figures 5-6 in the revised manuscript.

[Comment]: Page 6: I don't understand why dynamics decreases ozone deposition velocity in summer. During the daytime the ozone deposition velocity should be more limited by the surface resistance.

[Response]: The dry deposition velocity is computed as the reciprocal of the sum for aerodynamic resistance (Ra), quasi-laminar resistance (Rb), and surface resistance (Rc). The Ra is a function of wind speed and turbulence. Since the changes in dynamics decrease the wind speed, they thus increase Ra and consequently reduce dry deposition."

We have clarified this in the revised manuscript as below:

(Page 6 Line 23-25) "The stabilization of the atmosphere due to Dynamics leads to lower dry deposition rates (due to lower dry deposition velocity from the enhanced aerodynamic resistance) and thus increases surface O3."

[Comment]: Page 6: Page 9, line 1: summary states that aerosol effects improve the ozone simulation but I didn't see this demonstrated in the text.

[Response]: Following the reviewer's suggestion, we have reduced the content of Figure 3 and summarized the comparison in Table 2 in the revised manuscript, as below:

(Page 5 Line 28- Page 6 Line 10) "The impact of the ADE on O3 is further explored by examining the relationship between the observed and simulated O3 concentrations (DM1O3, daily values of the cities located in China) as a function of the observed PM2.5 concentrations (observed daily averaged values in those cities), as displayed in Figure 3. The predicted ozone concentrations under both low- and high- PM2.5 levels are compared in Table 2. In regards to model performance for DM1O3 simulations, the model generally exhibits a slight high bias in January but a low bias in July across the 5 regions. The inclusion of ADE moderately reduced O3 concentrations in January and slightly increased O3 in July, resulting in reduction in bias and improved performance for DM1O3 simulation in both January and July for most of regions.

Interestingly, from low to moderate PM2.5 levels (i.e., PM2.5 < 120 ug m-3), higher O3 concentration occur with higher PM2.5 concentrations, which is evident in both observations and simulations, suggestive of common precursors (e.g., NOx), source sectors, and/or transport pathways contributing to both O3 and PM2.5 in these regions. However, a negative correlation between O3 and PM2.5 is evident in winter when the PM2.5 can reach high levels larger than 120 ug m-3, indicating the strong ADE impacts on O3 through both feedbacks to dynamics and photolysis which significantly reduced O3."

[Comment]: Page 9: the summary presents as a punch line that one should decrease NOx emissions to improve both ozone and PM but this is hardly an original result.

[Response]: Traditionally, the co-benefits from NOx controls for ozone and PM reductions were thought of resulting from the fact that NOx acts as a common precursor for both O3 and PM2.5, thus a decrease of NOx emissions would be expected to reduce both ozone and PM. The analyses presented in this study reveals that the consideration of NOx controls is not only beneficial for directly reducing PM2.5 and O3, but also because of indirect benefits in reducing peak O3 through the weakening of the ADE from the reduced PM2.5, increasing the co-benefits from NOx controls for achieving both O3 and PM2.5 reductions.

To address the reviewer's concern, we have clarified this in the revised manuscript as below:

(Page 10 Line 11-14) "Traditionally, the co-benefits from NOx control for ozone and PM reduction are mostly thought of as resulting from the fact that NOx is a common precursor for both O3 and PM2.5. This study suggests that effective controls on NOx will not only gain direct benefits for O3 reduction, but also can indirectly reduce peak O3 through weakening the ADE from the reduced PM2.5, increasing the estimated co-benefits from NOx controls for achieving both O3 and PM2.5 reductions."

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2017-198/acp-2017-198-AC1-supplement.pdf

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