

Response to Referee #2

We would like to thank Referee #2 for the careful review of our work and address your concerns as follows. The reviewer's comments are shown with black font and our replies including the updates to the manuscript are highlighted in blue below.

General comments: The paper describes a model analysis of the past (1986-2006) and future (2000 vs. 2050) changes in the continental outflow of tropospheric ozone from East Asia. For the past and future changes, the authors ran the GEOS-Chem model driven by meteorological fields from GEOS4 and GISS GCM3 (under SRES A1B scenario), respectively. Basically the topics of the paper are of substantial interest. However, I found that the paper is rather descriptive and the discussion is not thorough. In many parts of the paper the authors show statistical results and interpretation rather than in-depth analyses that they could do with such a suite of model simulations. My another concern is that the authors' approach using the SREA A1B scenario now sounds old model sets, and I wonder why the authors did not try the simulations with RCP scenarios. Well, reserving this criticism, the paper still needs to be more focused on a new science with respect to continental outflow of ozone from East Asia that the authors can deliver from the current model runs.

Response:

- 1) We have added more detailed discussions and analyses in the revised manuscript, including:
 - Analyses on the key factor that drove the large interannual variations in continental O₃ outflow. See our response to your major comment (1);
 - More comprehensive validation on simulated surface-layer O₃ concentrations by using O₃ measurements from WDCGG and EANET, and the validation on simulated O₃ concentrations for the boundary layer, middle and upper troposphere by using the ozonesonde data from WOUDC. See our response to your major comment (2);
 - Comparisons with published literature regarding future changes in zonal winds. See our response to the recommended analyses (4) of Reviewer #1;
 - "Uncertainty Discussion" section (Section 6) to discuss the uncertainties associated with our model results.
- 2) The RCP scenarios have been compared with the SRES scenarios in previous studies (Lamarque et al., 2011; Riahi et al., 2011; van Vuuren et al., 2011; Fiore et al., 2012). For future air pollutant emissions, the RCPs assume uniformly an aggressive reduction, whereas the SRES scenarios allow unconstrained growth (Fiore et al., 2012). These two sets of projections likely bracket possible futures (Fiore et al., 2012). Therefore, the SRES A1B scenario is still used in recent studies to project future climate and O₃ (Lee et al., 2015; Redmond et al., 2015; Glotfelty et al., 2016; Sanderson and Ford, 2016).

Major comments:

(1) Why SRES A1B scenario? - I think this scenario is now out of date and would not be realistic for the future, suggesting the model studies less useful than before, say several years ago. If the authors stick to the SRES scenario, they would need to justify why they used this scenario not the RCP one. Also, I found that the discussions read a bit superficial, with a lot of interpretations by referring to previously published papers based on similar model settings with the SRES A1B scenario (i.e., Wu et al., Pye et al., Jiang et al.). The authors should focus on a new science with respect to continental outflow of ozone from East Asia, provide in-depth analysis in terms of meteorological and climatic mechanisms or key factors. In Abstract, the authors mentioned "Sensitivity simulations indicated that the large IAVs of O₃ outflow fluxes were mainly caused by the variations in meteorological conditions.", but this statement reads rather general. What meteorological factors or mechanisms are key for IAV? The authors showed statistical analysis but the mechanisms behind the large IAV is much more informative to the community.

Response:

- 1) We have justified the use of SRES A1B scenario in our response to your general comments.

2) Following the Reviewer's suggestion, we have added the following discussions on the key factor for the large IAVs in the revised manuscript (the last paragraph of Section 4.2): "Variations in meteorological conditions can influence the IAVs of the O₃ outflow fluxes by changing O₃ concentrations over East Asia (Yang et al., 2014; Lou et al., 2015), and by altering zonal winds (Kurokawa et al., 2009). The O₃ outflow flux is simulated to correlate positively with zonal wind averaged over 20°–55° N along 135° E, with a high correlation coefficient of +0.71 for annual fluxes and zonal winds. The correlation coefficient between O₃ fluxes and zonal winds is calculated to be +0.96 during summer when the APDM values of O₃ outflow fluxes are maximum. The high correlation coefficients indicate that the variation in zonal winds is the key factor that leads to the large IAVs of O₃ outflow fluxes."

(2) More robust model validation (Section 3) - Because of the large uncertainty in the retrieval of tropospheric ozone, comparison to satellite is not a robust way to quantitatively evaluate the model performance for the lower tropospheric ozone, in particular. The authors can make satellite comparisons with the reasons they mentioned in the Reply to the other reviewer, but why don't the authors evaluate the model by comparing to surface and sondes observations available in East Asia? I strongly believe that the model validation should be intensively made on seasonal basis since the authors are discussing the past and future ozone flux based on the model runs. The data from EANET are often used in evaluating the regional and global models by many groups in Asia (e.g., MICS-Asia) and in the international projects (e.g., HTAP) (e.g., Nagashima et al., ACP, 2010; Li et al., 2008). In Figures 3 and 4, the model overestimated the satellite-derived TCO over central-eastern China through the western North Pacific, and the phase of the seasonal cycle in TCO is not as great as the current state-of science models could be. I do not see the model doing a good job in reproducing the distributions and seasonal cycles, so cannot be positive to support the further analysis. The model overestimates TCO in spring, so this would give the overestimates in the calculated eastward flux. On the other hand, maybe the satellite-derived TCO is not too low (Figure 3), or the maximum shifts later than should be (Figure 4). I would encourage the authors to examine the model-observation comparison for the boundary layer, and middle and upper troposphere. Recent paper by Tanimoto, Zbinden, et al. (2015) showed robust observations for the seasonal cycles and interannual variations over Japan, and would be useful for this comparison.

Response:

Following the Reviewer's suggestion, we have changed the comparisons with satellite data to the comparisons with surface and sondes observations (added new Table 4, Figure 3, and Figure 4) in the revised manuscript. The data from WDCGG and EANET are used to evaluate the simulated surface-layer O₃ concentrations, and the ozonesonde data from WOUDC are used to evaluate the simulated O₃ concentrations for the boundary layer, middle and upper troposphere. We have added the following descriptions on the comparisons in the second and the third paragraphs of Section 3: "Here, we conduct comparisons with measurements to evaluate whether the version of the GEOS-Chem model used in this study can capture the temporal variations of tropospheric O₃. We use observations of tropospheric O₃ available in East Asia as summarized in Table 4. Observations at two sites (Minamitorishima and Yonagunijima) are from the World Data Centre for Greenhouse Gases (WDCGG, www.ds.data.jma.go.jp/gmd/wdcgg/), and those at another two sites (Rishiri and Ogasawara) are from the Acid Deposition Monitoring Network in East Asia (EANET, www.eanet.asia/product/index.html), which are used to evaluate the simulated surface-layer O₃ concentrations. The four Japanese sites are "remote" sites in the downwind regions of China. Figure 3 compares the time series of monthly surface-layer O₃ mixing ratios simulated by MetEmisB with those measured by WDCGG and EANET. Simulated surface-layer O₃ levels agree well with observations at all the four stations. The model captures fairly well the seasonal cycles and interannual variations of surface O₃, with high correlation coefficients of 0.82–0.93 (Table 4). Generally, the GEOS-Chem model can capture the high values during early spring or winter when Asian O₃ outflow flux is the highest, but overestimates the low values during summer when Asian O₃ outflow is the minimum.

To evaluate the simulated O₃ concentrations for the boundary layer, middle and upper troposphere,

we use the ozonesonde data at two Japanese sites from World Ozone and Ultraviolet Radiation Data Centre (WOUDC, www.woudc.org). The information for the two sites (Naha and Tsukuba) is listed in Table 4. Figure 4 compares the time series of monthly O₃ mixing ratios simulated by MetEmisB with those measured by ozonesonde. Comparisons are shown for four altitudes in the troposphere. The GEOS-Chem model captures the seasonal cycles and interannual variations of tropospheric O₃ at all altitudes, with correlation coefficients ranging from 0.68 to 0.88 for Naha site, and from 0.55 to 0.76 for Tsukuba site. However, the agreement with ozonesonde in the lowermost layer (1000–850 hPa) seems to be poorer than that with WDCGG or EANET. It is noted that, the ground-based measurements (WDCGG or EANET) and simulation results are calculated from continuous data, while the ozonesondes are regularly launched at a fixed local time with a typical frequency of 1–2 weeks (Tanimoto et al., 2015). The inconsistency in sampling time may be responsible for the poorer agreement with ozonesonde.”

Table 4. Information for the sites with O₃ measurements used in model evaluation.

Site	Location	Database	Height	R ^a	NMB ^b (%)
Minamitorishima	24.3 °N, 154.0 °E	WDCGG	surface	0.92	+12.7
Yonagunijima	24.5 °N, 123.0 °E	WDCGG	surface	0.93	+12.6
Rishiri	45.1 °N, 141.2 °E	EANET	surface	0.82	+2.4
Ogasawara	27.1 °N, 142.2 °E	EANET	surface	0.90	+29.6
Naha	26.2 °N, 127.7 °E	WOUDC	500–300 hPa	0.68	–2.61
			700–500 hPa	0.77	+16.4
			850–700 hPa	0.85	+24.3
			1000–850 hPa	0.88	+39.5
Tsukuba	36.1 °N, 140.1 °E	WOUDC	500–300 hPa	0.55	+15.8
			700–500 hPa	0.76	+12.3
			850–700 hPa	0.76	+8.61
			1000–850 hPa	0.60	+8.5

^a Correlation coefficient (R) between the observed and simulated monthly O₃ mixing ratios.

^b Normalized mean bias (NMB, %) between the observed and simulated monthly O₃ mixing ratios.

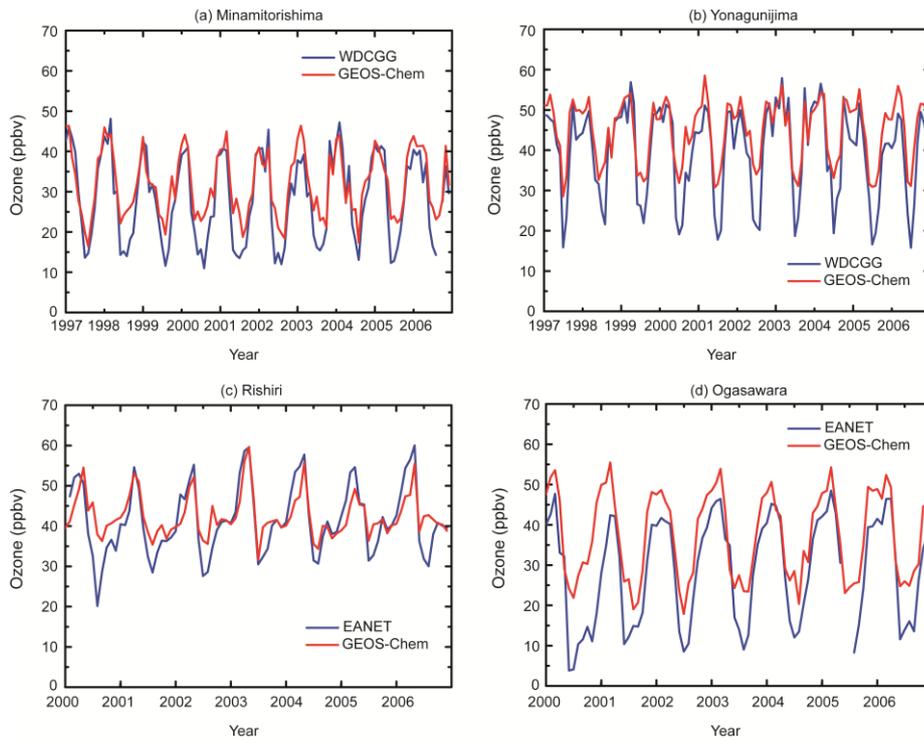


Figure 3. Time series of monthly surface-layer O₃ mixing ratios measured by WDCGG and EANET (blue line), and simulated by MetEmisB (red line). (a) Minamitorishima and (b) Yonagunijima are WDCGG sites, and (c) Rishiri and (d) Ogasawara sites are EANET sites.

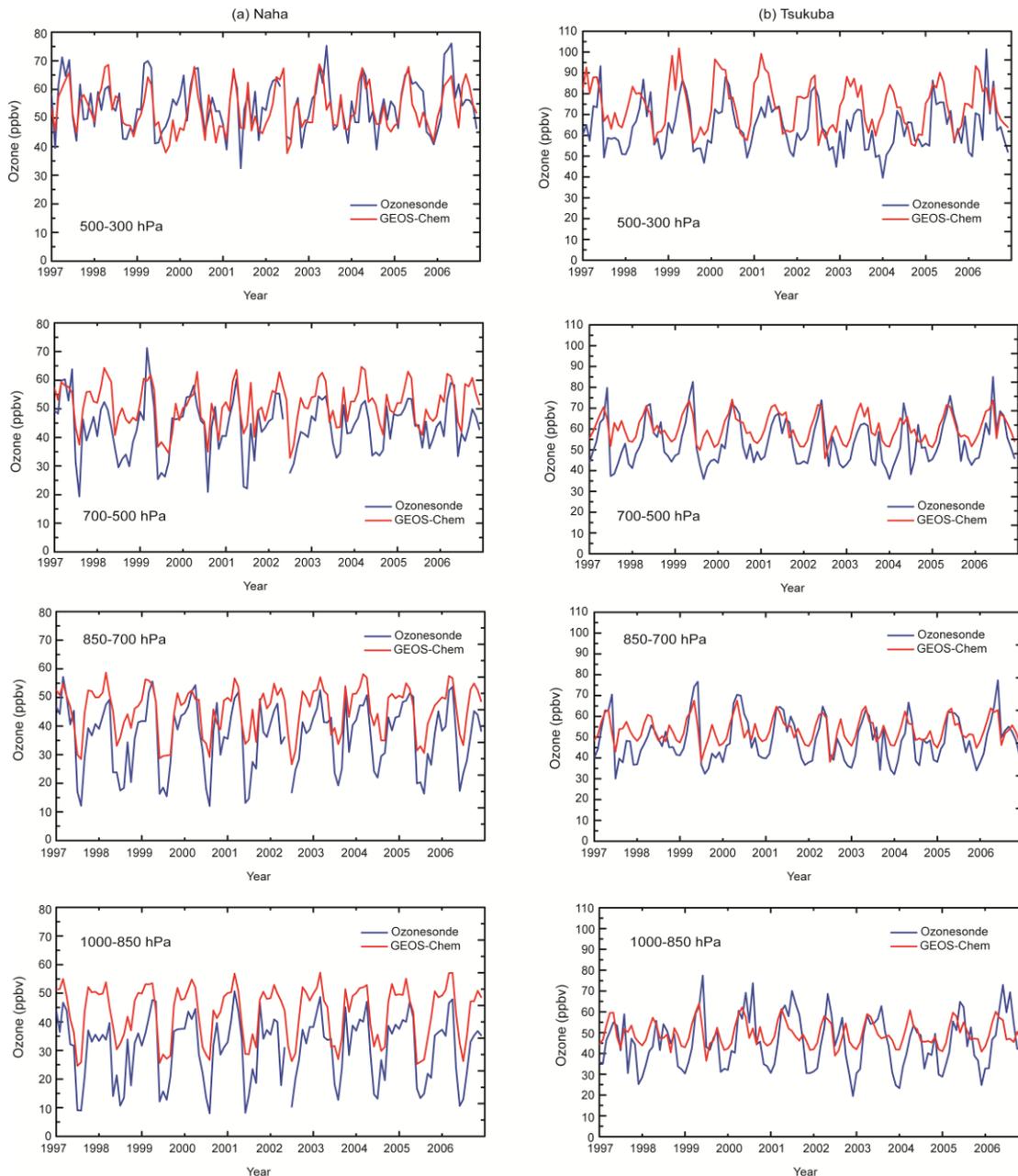


Figure 4. Time series of monthly O_3 mixing ratios measured by ozonesonde (blue line), and simulated by MetEmisB (red line). (a) Naha and (b) Tsukuba are ozonesonde sites from WOUDC. Comparisons are shown for four altitude levels in the troposphere.

(3) Is 135 degE appropriate? - The authors mentioned in the title “ozone outflow from East Asia” and used a longitudinal transect at 135 degE to diagnose the eastward flux of ozone. I wonder why at 135 degE, not 120 degE, to be more close to ozone production region in central-eastern China. I think, if the authors look at the flux at 120 degE, they would obtain higher signals in the ozone flux, and this would be much more direct in interpreting the model simulations. Also, the authors mainly discuss central-eastern China or North China Plain, rather than whole East Asia. This should be explicitly phrased, for example, “outflow from central-eastern China”, since this paper is not looking at the impacts on the western North America but focusing on export region.

Response:

We calculate O_3 flux through the vertical plane along 135 °E, because 135 °E is the easternmost boundary of China (i.e., Wusuli River in Northeastern China). Following the Reviewer’s suggestion, we have also calculated the O_3 outflow flux along 120 °E. Figure H1 shows the evolutions of annual O_3

outflow fluxes across the meridional plane along (a) 135 °E and (b) 120 °E over 1986–2006 in the Met, Emis, and MetEmis simulations. The variations in O₃ fluxes calculated at 120 °E are similar to those calculated at 135 °E. Both figures show that, with variations in both anthropogenic emissions and meteorological parameters (the MetEmis simulation), the simulated O₃ outflow shows large IAVs but a statistically insignificant ($P > 0.05$) trend. We have added the above discussion in the Uncertainty Discussion section (Section 6) of our revised manuscript.

Because the variations in O₃ fluxes calculated at 135 °E are similar to those calculated at 120 °E, we retain the calculations along 135 °E and the description of “ozone outflow from East Asia” in the revised manuscript.

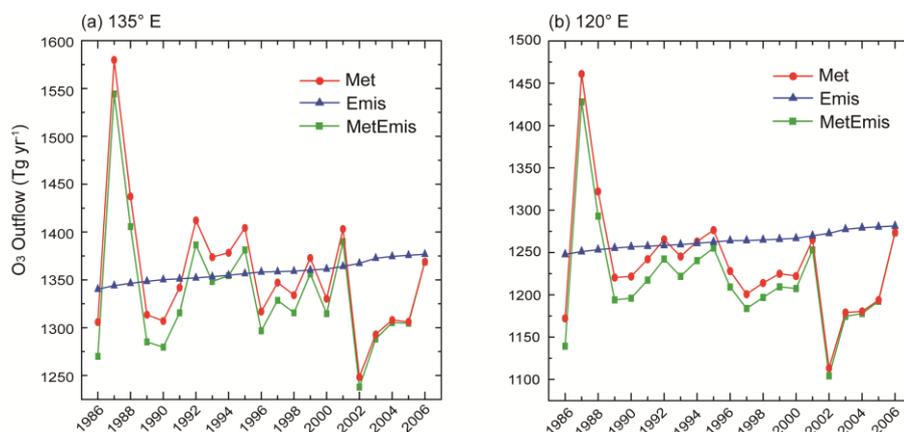


Figure H1. Evolution of annual O₃ outflow fluxes (Tg yr⁻¹) across the meridional plane along (a) 135 °E, and (b) 120 °E, from 20 °N to 55 °N and from the surface to 100 hPa over 1986–2006 in the Met, Emis, and MetEmis simulations.

Specific comments:

Abstract, L22: insignificant decadal trend of -2.2%/decade. Add +/- uncertainty, or just delete the number here.

Response:

We have deleted the number in our revised manuscript.

L28-29: spring and summer. The maritime flow from the Pacific Ocean is predominant in summer. Is summer really effective in the enhancement of continental outflow? I do not see strong enhancement in summer in Figure 9.

Response:

Although the absolute value of the increase in O₃ outflow during summer is not large, the percentage increase in O₃ outflow during summer is 14.5% (Table 5 in the revised manuscript). The large percentage increase can be mainly attributed to the enhancement in zonal winds. Based on 29-model ensemble mean results, Jiang and Tian (2013) also showed that the westerlies along 135 °E during summer would strengthen in future climate.

L31: important implications for long-term air quality planning. For whom? For US? For northern midlatitudes? For China? For East Asia?

Response:

We have revised the sentence as “have important implications for long-term air quality planning for the downwind regions of China, such as Japan and US” in the Abstract of our revised manuscript.

P2, L8-11: . . . influences ozone air quality in the downwind regions, such as the US and Canada. Downwind regions are not only the western US, but should include the neighboring regions and the Pacific Ocean. Ou-Yang et al. paper is already cited here, so the sentence should be rephrased to be something like “. . . such as the western North Pacific through the western North America”, and add some other references, for example, papers reporting long-range transport to Korea, Japan, and the

Northern Pacific (Han et al., ACP, 2015; Tanimoto et al., GRL, 2005; Pochanart et al., 2015, and many others!).

Response:

We have replaced it by “such as the western North Pacific through the western North America”, and added the following references: “Tanimoto et al., 2005; Kim et al., 2006; Li et al., 2008; Kurokawa et al., 2009; Nagashima et al., 2010; Han et al., 2015; Pochanart et al., 2015” (the second paragraph of Section 1).

P3, L8-12: Tanimoto, AE, 2009 should be cited here (decadal trends of . . .)

Response:

We have cited the reference (Tanimoto, 2009) in the revised manuscript (the fourth paragraph of Section 1).

Figure 4: The authors showed comparison of TCO for GEOS-Chem and TOMS/SBUV, suggesting the biases in the model. Again, why don't the authors make comparison to the surface and sonde observations?

Response:

As suggested by the Reviewer, we have conducted comparisons with surface-layer O₃ measurements from WDCGG and EANET, and with ozonesonde measurements from WOUDC for the boundary layer, middle and upper troposphere O₃ in the revised manuscript. See our response to your major comment (2).

Also, in P8, L10-13, the authors state that “although GEOS-Chem overestimates TCO values over eastern China and the western Pacific Ocean, the model exhibits reasonable performance in simulating the spatiotemporal distributions of the tropospheric ozone column burden over China and downwind regions, which lends us confidence to simulate the temporal evolutions of the Asian ozone outflow.” I would not agree with this statement, since model and satellite are quite different in the tail of outflow from China (the region of >40 DU, shown in orange), and this difference would lead to large biases in calculating outflow flux in particular, as the authors set the diagnosis line at 135 degE, off China, and over Japan. Also, technically, the authors said “the western Pacific Ocean” here and also in the Figure 4 caption, but the region where the authors pointed is mostly Japan, so the description must be accurately modified.

Response:

As the Reviewer pointed out in major comment (2), because of the large uncertainty in the retrieval of tropospheric ozone, comparison to satellite is not a robust way to quantitatively evaluate the model performance. Therefore, we have changed the comparisons with satellite to the comparisons with surface and sondes observations in the revised manuscript. See our response to your major comment (2). The old Figure 4 and associated description have been deleted and replaced by the comparisons with surface and sondes observations.

P9, Section 4.2 IAV and decadal trends: The authors basically said that the influence of Met. is larger than Emiss., which makes sense if they diagnosed at 135 degE, off the Asian continent, where Asian monsoon impacts are substantial.

Response:

Following your suggestions in major comment (3), we have calculated the O₃ outflow flux along 120° E, more close to the Asian continent. It is concluded from Figure H1 that the variations in O₃ fluxes calculated at 120° E are similar to those calculated at 135° E. With variations in both anthropogenic emissions and meteorological parameters (the MetEmis simulation), the simulated O₃ outflow along 120° E also shows large IAVs but a statistically insignificant ($P > 0.05$) trend. The two curves from the Met and MetEmis simulations almost coincide with each other, indicating the dominant role of variations in meteorological parameters in the IAVs of the Asian O₃ outflow flux.

A number of important references are missing:

Pochanart, P. et al, 2015, Boundary Layer Ozone Transport from Eastern China to Southern Japan: Pollution Episodes Observed during Monsoon Onset in 2004, Asian J. Atmos. Environ. 9, 48-56.

- Tanimoto, H., et al., 2005, Significant latitudinal gradient in the surface ozone spring maximum over East Asia, *Geophys. Res. Lett.* 32, L21805.
- Tanimoto, H., 2009, Increase in springtime tropospheric ozone at a mountainous site in Japan for the period 1998–2006, *Atmos. Environ.*, 43, 1358–1363.
- Nagashima, T., et al., 2010, The relative importance of various source regions on East Asian surface ozone, *Atmos. Chem. Phys.*, 10, 11305–11322, doi:10.5194/acp-10-11305-20109077-9120.
- Li, J., et al., 2008, Regional-scale modeling of near-ground ozone in the Central East China, source attributions and an assessment of outflow to East Asia - The role of regional-scale transport during MTX2006, *Atmos. Chem. Phys.*, 8, doi: 10.5194/acpd-8-13159-2008.
- Tanimoto, H., et al., 2015, Consistency of tropospheric ozone observations made by different platforms and techniques in the global databases, *Tellus B*, 67, 27073, doi:10.3402/tellusb.v67.27073.

Response:

We have added the above references in proper places of the revised manuscript.

References:

- Fiore, A. M., et al.: Global air quality and climate, *Chem. Soc. Rev.*, 41, 6663–6683, doi:10.1039/c2cs35095e, 2012.
- Glotfelty, T., Zhang, Y., Karamchandani, P., and Streets, D. G.: Changes in future air quality, deposition, and aerosol-cloud interactions under future climate and emission scenarios, *Atmos. Environ.*, 139, 176–191, doi:10.1016/j.atmosenv.2016.05.008, 2016.
- Han, J., Shin, B., Lee, M., Hwang, G., Kim, J., Shim, J., Lee, G., and Shim, C.: Variations of surface ozone at Jeodo Ocean Research Station in the East China Sea and the influence of Asian outflows, *Atmos. Chem. Phys.*, 15, 12611–12621, doi:10.5194/acp-15-12611-2015, 2015.
- Jiang, D. B., and Tian, Z. P.: East Asian monsoon change for the 21st century: Results of CMIP3 and CMIP5 models, *Chinese Sci. Bull.*, 58, 1427–1435, doi:10.1007/s11434-012-5533-0, 2013.
- Kim, J. H., Lee, H. J., and Lee, S. H.: The characteristics of tropospheric ozone seasonality observed from ozone soundings at Pohang, Korea, *Environ. Monit. Assess.*, 118, 1–12, doi:10.1007/s10661-006-0772-7, 2006.
- Kurokawa, J., Ohara, T., Uno, I., Hayasaka, M., and Tanimoto, H.: Influence of meteorological variability on interannual variations of springtime boundary layer ozone over Japan during 1981–2005, *Atmos. Chem. Phys.*, 9, 6287–6304, doi:10.5194/acp-9-6287-2009, 2009.
- Lamarque, J. F., Kyle, G. P., Meinshausen, M., Riahi, K., Smith, S. J., van Vuuren, D. P., Conley, A. J., and Vitt, F.: Global and regional evolution of short-lived radiatively-active gases and aerosols in the Representative Concentration Pathways, *Clim. Change*, 109, 191–212, doi:10.1007/s10584-011-0155-0, 2011.
- Lee, J. B., Cha, J. S., Hong, S. C., Choi, J. Y., Myoung, J. S., Park, R. J., Woo, J. H., Ho, C., Han, J. S., and Song, C. K.: Projections of summertime ozone concentration over East Asia under multiple IPCC SRES emission scenarios, *Atmos. Environ.*, 106, 335–346, doi:10.1016/j.atmosenv.2015.02.019, 2015.
- Li, J., Wang, Z., Akimoto, H., Yamaji, K., Takigawa, M., Pochanart, P., Liu, Y., Tanimoto, H., and Kanaya, Y.: Near-ground ozone source attributions and outflow in central eastern China during MTX2006, *Atmos. Chem. Phys.*, 8, 7335–7351, doi:10.5194/acp-8-7335-2008, 2008.
- Lou, S., Liao, H., Yang, Y., and Mu, Q.: Simulation of the interannual variations of tropospheric ozone over China: Roles of variations in meteorological parameters and anthropogenic emissions, *Atmos. Environ.*, 122, 839–851, doi:10.1016/j.atmosenv.2015.08.081, 2015.
- Nagashima, T., Ohara, T., Sudo, K., and Akimoto, H.: The relative importance of various source regions

- on East Asian surface ozone, *Atmos. Chem. Phys.*, 10, 11305–11322, doi:10.5194/acp-10-11305-2010, 2010.
- Pochanart, P., Wang, Z., and Akimoto, H.: Boundary Layer Ozone Transport from Eastern China to Southern Japan: Pollution Episodes Observed during Monsoon Onset in 2004, *Asian J. Atmos. Environ.*, 9, 48–56, doi:10.5572/ajae.2015.9.1.048, 2015.
- Redmond, G., Hodges, K. I., Mcsweeney, C., Jones, R., and Hein, D.: Projected changes in tropical cyclones over Vietnam and the South China Sea using a 25 km regional climate model perturbed physics ensemble, *Clim. Dyn.*, 45, 1983–2000, doi:10.1007/s00382-014-2450-8, 2015.
- Riahi, K., Rao, S., Krey, V., Cho, C., Chirkov, V., Fischer, G., Kindermann, G., Nakicenovic, N., and Rafaj, P.: RCP 8.5—A scenario of comparatively high greenhouse gas emissions, *Clim. Change*, 109, 33–57, doi:10.1007/s10584-011-0149-y, 2011.
- Sanderson, M. G., and Ford, G. P.: Projections of severe heat waves in the United Kingdom, *Clim. Res.*, 71, 63–73, doi:10.3354/cr01428, 2016.
- Tanimoto, H.: Increase in springtime tropospheric ozone at a mountainous site in Japan for the period 1998–2006, *Atmos. Environ.*, 43, 1358–1363, doi:10.1016/j.atmosenv.2008.12.006, 2009.
- Tanimoto, H., Sawa, Y., Matsueda, H., Uno, I., Ohara, T., Yamaji, K., Kurokawa, J., and Yonemura, S.: Significant latitudinal gradient in the surface ozone spring maximum over East Asia. *Geophys. Res. Lett.*, 32, L21805, doi:10.1029/2005GL023514, 2005.
- Tanimoto, H., Zbinden, R. M., Thouret, V., and Nédélec, P.: Consistency of tropospheric ozone observations made by different platforms and techniques in the global databases, *Tellus B*, 67, 27073, doi:10.3402/tellusb.v67.27073, 2015.
- van Vuuren, D. P., et al.: The representative concentration pathways: an overview, *Clim. Change*, 109, 5–31, doi:10.1007/s10584-011-0148-z, 2011.
- Yang, Y., Liao, H., and Li, J.: Impacts of the East Asian summer monsoon on interannual variations of summertime surface-layer ozone concentrations over China, *Atmos. Chem. Phys.*, 14, 6867–6880, doi:10.5194/acp-14-6867-2014, 2014.