Response to the Major Comments of Referee #1

We thank Reviewer #1 for the careful review of our work and would like to address your major concerns as follows. Our responses to your minor comments and the complete revision of our manuscript will be uploaded subsequently.

Major comments

This study uses a suite of chemical transport model experiments (GEOS-Chem) to examine the extent to which changes in anthropogenic emissions and meteorology influence the outflow of ozone from East Asia under present-day and future climate. The authors show that Asian NOx emissions almost doubled over the historical analysis period 1986-2006, along with increases in VOC emissions and global methane (Fig.1). However, their model with both emissions and meteorology varying over 1986-2006 shows little overall trend in the outflow of ozone from East Asia (Fig.6). This result contradicts with many prior studies suggesting that rising Asian emissions over the past 20-30 years contribute to raising baseline ozone downwind of Asia and over western North America. The referee believes that there are likely some fundamental flaws in the model experiments (or analysis approach). Further in-depth analyses are needed to evaluate the modeled ozone response to emission trends.

Response:

Thanks to the Reviewer for the careful review of our work. We would like to address your major concerns as follows:

1) In the manuscript, O_3 outflow from East Asia includes the effects of emissions in different regions of the world, other than Asian emissions alone, owing to the relatively long lifetime (~3 weeks) of O_3 (Fiore et al., 2002; Liao et al., 2006). Although the anthropogenic emissions of O_3 precursors in Asia increased a lot over 1986–2006, the global anthropogenic emissions of these precursors exhibited no significant trends over the past two decades (Figure 1(a) in the manuscript, similar to those reported by Lamarque et al., 2010), which explains in part the statistically insignificant decadal trend of O_3 outflow from East Asia.
2) The outflow flux of O_3 depends on both tropospheric O_3 concentrations and winds. The zonal winds exhibited large interannual variations over 1986–2006 (Figure 8(a) in the manuscript, also reported by Yuan and Ni, 2013; Du et al., 2016), which led to the large interannual variations in O_3 outflow flux. The emission-driven trend in O_3 flux is swamped by the large interannual variability in zonal winds. Therefore, with variations in both anthropogenic emissions and meteorological parameters, the simulated O_3 outflow fluxes showed statistically insignificant decadal trend.
3) Following the reviewer's suggestion (the following recommended analyses (1)), we have compared the trends in simulated O_3 concentrations with the observations. In fact, the simulated O_3 concentrations in our model exhibited statistically significant increasing trends over 1986–2006 (see our response to the following recommended analyses (1)), which verified the validity of the model experiments.

The referee recommends the following analyses:

(1) Does the model (MetEmis) simulate significant increases in surface and free

tropospheric ozone over East Asia during the period 1986–2006? How well do the modeled trends compare with observations? While long-term ozone observations over East Asia are very limited, there are some data available. Please see Section 3 and Figs 4-6 in the following manuscript and references therein:

Lin, M., Horowitz, L. W., Payton, R., Fiore, A. M., and Tonnesen, G.: US surface ozone trends and extremes from 1980–2014: Quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1093, in review, 2016, accessible at http://www.atmos-chemphys-discuss.net/acp-2016-1093/.

Response:

Following the reviewer's suggestion, we have calculated the trends in simulated (MetEmis) surface O_3 concentrations over East Asia, and compared with the observed trends collected from the above manuscript (Lin et al., 2016) and references therein. Figure R1 shows the comparison of simulated O_3 trends with observations. Although the model underestimates the observed trends, simulated O_3 concentrations at all stations exhibit statistically significant increasing trends. The modeled O_3 trends had low biases in previous studies (Tanimoto et al., 2009; Parrish et al., 2014); Parrish et al. (2014) compared O_3 trends simulated by three chemistry-climate models with observations at Asian sites, and reported that one model captured less than one third of the observed increasing trend, and the other two models suggested no significant increasing trends.

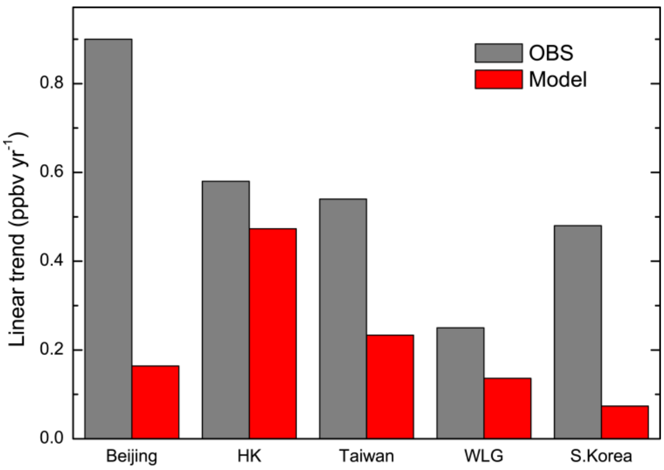

Figure R1. Comparison of modeled (MetEmis) trends in annual-mean O_3 concentrations with observations.

(2) This study defines the Asian ozone outflow as the ozone flux through the meridional plain along 135E from 20N-55N and from the surface to 100 hPa. If you restrict the calculation to the surface to 200-300 hPa or up to the tropopause, does the calculated O_3 flux change substantially? I wonder if the O_3 flux up to 100 hPa is overwhelmingly influenced by stratosphere-to-troposphere exchange (STE) and thus the emission-driven trend is swamped by interannual variability in STE.

Response:

1) As we describe in Section 2.2 of our manuscript, we do not consider the interannual variations in stratosphere–troposphere exchange (STE) of O_3 in this study. The model imposes a global annual mean cross-tropopause O_3 flux of 500

Tg yr^{-1}. We have added discussions on this issue in the text and in the Conclusion section of our revised manuscript.

2) Following the reviewer's suggestion, we have also calculated O_3 fluxes through the meridional plain along 135 °E from the surface to 200 hPa. Figure R2(a) and R2(b) show the evolutions of annual O_3 outflow fluxes across the meridional plane from the surface to 100 hPa and from the surface to 200 hPa, respectively. When we restrict the calculation to 200 hPa, the patterns of variations in O_3 fluxes are similar to those calculated from the surface to 100 hPa. With variations in both anthropogenic emissions and meteorological parameters (the MetEmis simulation), the simulated O_3 outflow shows large IAVs but a statistically insignificant (P > 0.05) trend.

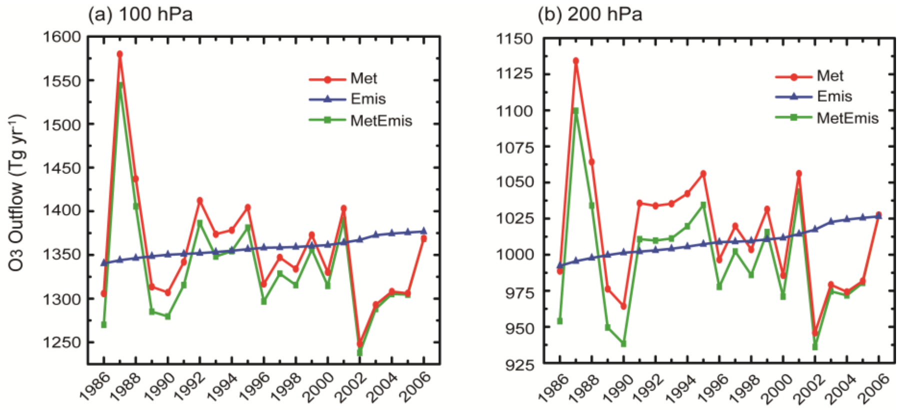

Figure R2. Evolution of annual O_3 outflow fluxes (Tg yr^{-1}) across the meridional plane along 135 ° E from 20 °N to 55 °N for (a) from the surface to 100 hPa and (b) from the surface to 200 hPa.

(3) This study uses tropospheric column ozone (TCO) retrieved from TOMS/SBUV to evaluate their model simulation of TCO seasonal cycle and long-term trends (Figs 3 and 4). But how good are the TOMS TCO retrievals? TOMS TCO is possibly representative of mid- and upper tropospheric ozone variability. It is not expected to resolve ozone variability in the lower troposphere. So why use TOMS to evaluate the model?

Response:

1) Tropospheric column ozone (TCO) can be retrieved from satellites TES, OMI/MLS, TOMS/SBUV, and so on. The TES and OMI are both on the EOS Aura satellite launched in July 2004. Therefore, TCO retrievals from TES or OMI/MLS are not available before year 2004 (the MetEmisB simulation in the manuscript is conducted over 1997–2006). However, TCO retrievals from TOMS/SBUV retrievals are available since 1979.

2) Fishman et al. (1996) compared tropospheric ozone fields derived from TOMS/SBUV with ozone measurements from aircraft, ozonesondes, and TOMS/SAGE. In general, TOMS/SBUV technique successfully captures the amount, large-scale gradients, and temporal variations of tropospheric column ozone. TOMS/SBUV technique has been extensively used to study the temporal-spatial distributions of TCO (Fishman and Balok, 1999; Fishman et al., 2003) and intercontinental transport of tropospheric ozone (Creilson et al., 2003).

(4) Fig.9 and associated discussions about the future changes. Changes in atmospheric circulation on regional scales under future climate scenarios are known to have large

uncertainty. The different models often yield different results and large ensemble members are typically required.

Theodore G. Shepherd, Atmospheric circulation as a source of uncertainty in climate change projections, Nature Geoscience 7, 703–708 (2014), doi:10.1038/ngeo2253.

How many ensembles are included in your experiments? Rather than just showing your results, a through literature review is needed in Section 5 to place your results into context. What is the robust conclusion across the models in the published literature regarding changes in zonal winds and other circulation aspects over Asia under future climate? Do your model agree with the published work?

Response:

1) The future simulation of O_3 outflow is driven by meteorological data from the Goddard Institute for Space Studies (GISS) general circulation model (GCM) 3. The GISS Model 3 is coupled with a "Q-flux" ocean as described in Wu et al. (2008). The GISS Model 3/GEOS-Chem combination has been used to project future aerosols and ozone over United States and China under future climate (Wu et al., 2008; Pye et al., 2009; Wang et al., 2013; Jiang et al., 2013).

2) Following the reviewer's suggestion, we have compared the future changes in zonal winds in our revised manuscript with those reported by previous studies: "The projected future changes in zonal winds are consistent with previous studies. By analyzing 18 CMIP5 models, Huang and Wang. (2016) assessed the future changes in atmospheric circulation during spring over East Asia. They found that although different models projected different changes (even in sign) in zonal winds, the ensemble mean of five better-skill models among the 18 CMIP5 models exhibited overall increases in zonal winds throughout the whole troposphere, which agrees with our simulation. Based on 31 (29)-model ensemble mean results, Jiang and Tian. (2013) showed that westerlies along 135 °E during winter (summer) were projected to weaken (strengthen) south of 40 °N. The projected patterns of future changes in westerlies during winter and summer are also captured by our model."

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