

## **Responses to reviewers' comments on "Interannual variation, decadal trend, and future change in ozone outflow from East Asia" by J. Zhu et al. (MS No.: acp-2016-938)**

We would like to thank the reviewers for their comments that are helpful for improving the quality of our work. The manuscript has been revised accordingly, and our point-by-point responses are provided below. The reviewers' comments are shown with black font and our replies including the updates to the manuscript are highlighted in blue below. A marked-up manuscript version is also showing the changes made.

### **Response to Reviewer #1**

#### **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 NO<sub>x</sub> 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:**

We would like to address your major concerns as follows:

- 1) In the manuscript, O<sub>3</sub> 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<sub>3</sub> (Fiore et al., 2002; Liao et al., 2006). Although the anthropogenic emissions of O<sub>3</sub> precursors in Asia increased a lot over 1986–2006, the global anthropogenic emissions of these precursors (NO<sub>x</sub>, CO, and NMVOCs) exhibited no significant trends over the past two decades (see Figure 1(a) in the revised manuscript, the trends we present are similar to those reported by Lamarque et al. (2010)), which explains in part the statistically insignificant decadal trend of O<sub>3</sub> outflow from East Asia.
- 2) The outflow flux of O<sub>3</sub> depends on both tropospheric O<sub>3</sub> concentrations and winds. As pointed out by the Reviewer, O<sub>3</sub> concentrations in East Asia exhibited an increasing trend in the past two decades (Ding et al., 2008a; Wang et al., 2009b; Xu et al., 2016). However, the zonal winds exhibited large interannual variations over 1986–2006 (Figure 9(a) in the revised manuscript, also reported by Yuan and Ni (2013) and Du et al. (2016)), which led to the large interannual variations in O<sub>3</sub> outflow flux. The emission-driven trend in O<sub>3</sub> flux was swamped by the large interannual variability in zonal winds. Therefore, with variations in both anthropogenic emissions and meteorological parameters, the simulated O<sub>3</sub> outflow fluxes showed statistically insignificant decadal trend.
- 3) Following the Reviewer's suggestion (the recommended analysis (1) below), we have compared the trends in simulated O<sub>3</sub> concentrations with observations. In fact, the simulated O<sub>3</sub> concentrations in our model exhibited statistically significant increasing trends over 1986–2006 (see our responses to the recommended analyses below), which verified the validity of our model experiments.

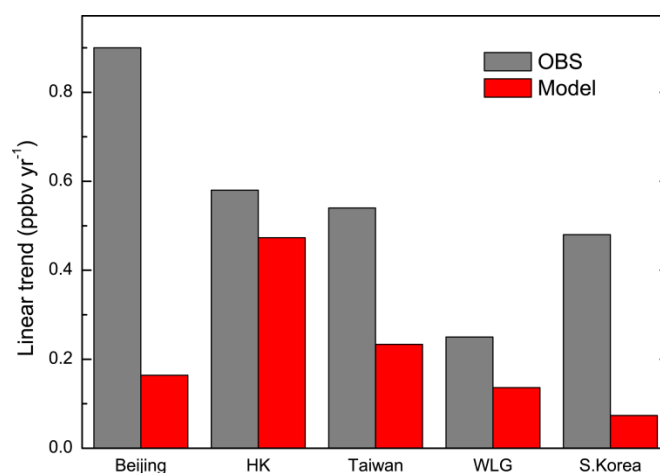
#### **The referee recommends the following analyses:**

(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

**Response:**

Following the Reviewer's suggestion, we have added new Figure 5 and the following descriptions on the comparison in the fourth paragraph of Section 3 of our revised manuscript: "The increasing trend in surface-layer O<sub>3</sub> in East Asia over the past two decades was reported by previous studies (Ding et al., 2008a; Wang et al., 2009b; Xu et al., 2016). Figure 5 compares the simulated trends in annual-mean surface-layer O<sub>3</sub> concentrations from the MetEmis experiment with the observed trends collected from Lin et al. (2016) and references therein. The observed trends of annual-mean O<sub>3</sub> at Beijing, Hongkong, Taiwan, Waliguan, and South Korea stations are +0.90 ppbv yr<sup>-1</sup>, +0.58 ppbv yr<sup>-1</sup>, +0.54 ppbv yr<sup>-1</sup>, +0.25 ppbv yr<sup>-1</sup>, +0.48 ppbv yr<sup>-1</sup>, respectively. Simulated O<sub>3</sub> concentrations at all stations exhibit statistically significant increasing trends, although the model underestimates the trends. The modeled O<sub>3</sub> trends were reported to have low biases in previous studies (Tanimoto et al., 2009; Parrish et al., 2014). Parrish et al. (2014) compared O<sub>3</sub> 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 whereas the other two models suggested no significant increasing trends."



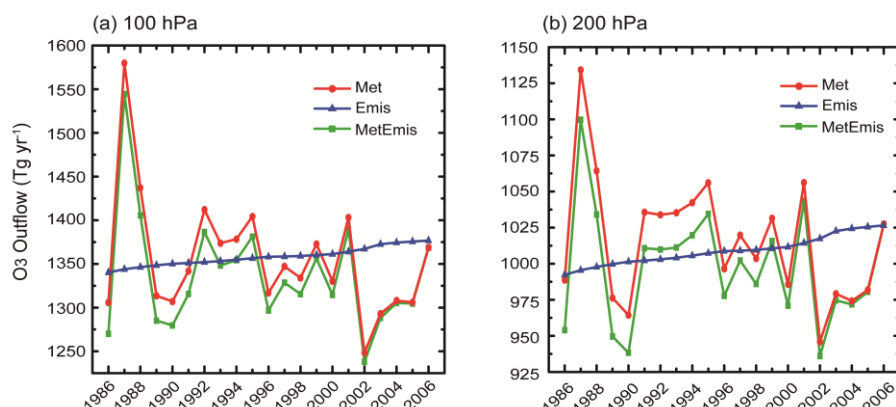
**Figure 5.** Comparison of simulated trends in annual-mean O<sub>3</sub> concentrations from the MetEmis experiment with observations for Beijing (location: 40.0 ° N, 116.5 ° E; years: 1995–2005; reference: Ding et al., 2008a), Hongkong (22.2 ° N, 114.3 ° E; 1994–2007; Wang et al., 2009b), Taiwan (23.5 ° N, 121.0 ° E; 1994–2007; Lin et al., 2010), Waliguan (36.3 ° N, 100.9 ° E; 1994–2013; Xu et al. 2016), and South Korea (37.3 ° N, 126.5 ° E; 1990–2010; Lee et al., 2014).

(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<sub>3</sub> flux change substantially? I wonder if the O<sub>3</sub> 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 described in the last paragraph of Section 2.2, the interannual variations in stratosphere–troposphere exchange (STE) of O<sub>3</sub> is not considered in this work. The model imposes a global annual mean cross-tropopause O<sub>3</sub> flux of 500 Tg yr<sup>-1</sup>. We have added the following discussions on this issue in the Uncertainty Discussion section (Section 6) of our revised manuscript: "There are some uncertainties in our simulations. First, the influence of interannual variation in stratosphere-troposphere exchange on tropospheric O<sub>3</sub> is not considered in this study. Terao et al. (2008) reported that the stratosphere-troposphere exchange had large impacts on the interannual variability of tropospheric O<sub>3</sub> over Canada and Europe but the impact was much smaller over East Asia."
- 2) Following the Reviewer's suggestion, we have calculated O<sub>3</sub> fluxes through the meridional plain along 135 ° E from the surface to 200 hPa. Figures R1(a) and R1(b) show the evolutions of annual O<sub>3</sub> 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<sub>3</sub> fluxes are similar to those calculated from the surface to 100 hPa. Both figures show that, with variations in both anthropogenic emissions and meteorological parameters (the MetEmis simulation), the simulated O<sub>3</sub> outflow has large IAVs but a statistically insignificant ( $P > 0.05$ ) trend.



**Figure R1.** Evolution of annual O<sub>3</sub> outflow fluxes (Tg yr<sup>-1</sup>) 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:**

As both reviewers pointed out, the TOMS/SBUV retrievals have large uncertainty in the lower troposphere O<sub>3</sub>. Therefore, we have changed the comparisons with satellite to the comparisons with surface and sondes observations in the revised manuscript (the second and the third paragraphs of Section 3). The data from WDCGG and EANET are used to evaluate the simulated surface-layer O<sub>3</sub> concentrations, and the ozonesonde data from WOUDC are used to evaluate the simulated O<sub>3</sub> concentrations for the boundary layer, middle and upper troposphere. See our response to Major Comment (2) of Reviewer #2 for further details of comparisons.

(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 thorough 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<sub>3</sub> outflow is driven by the archived 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 concentrations of tropospheric ozone and aerosols 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 now compare future changes in zonal winds projected by our model with those reported in previous studies in Section 5.2 of our revised manuscript: “Our 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 during spring, which agrees with our simulation. Based on 31 (29)-model ensemble mean results, Jiang and Tian (2013) showed that the westerlies along 135 °E during winter (summer) were projected to weaken (strengthen). Such projected patterns of future changes in westerlies during winter and summer are also captured by our model.” We have also added the following discussions on this issue in the Uncertainty Discussion section (Section 6) of our revised manuscript: “Finally, projecting future atmospheric circulation on regional scales has large uncertainty, which is undergoing continuing improvement.”

### **Other minor comments**

Page 2, Line 10: Also cite Jacob et al., 1999 - the first paper on Asian influence on US ozone.

Jacob, D. J., Logan, J. A. Murti, P. P. Effect of rising Asian emissions on surface ozone in the United States. *Geophys. Res. Lett.* 26, 2175-2178 (1999).

#### **Response:**

We have added the reference (Jacob et al., 1999) in the revised manuscript (the second paragraph of Section 1).

Page 3, Lines 9-10: Also cite Lin et al. (2015, GRL) - who found that measurement sampling biases substantially influence the ozone trends derived from sparse measurements over the western US originally reported by Cooper et al. (2010, Nature).

Lin, M., W. Horowitz, O.R. Cooper, D. Tarasick, S. Conley, L.T. Iraci, B. Johnson, T. Leblanc, I. Petropavlovskikh, E.L. Yates (2015): Revisiting the evidence of increasing springtime ozone mixing ratios in the free troposphere over western North America, *Geophysical Research Letter*, 42, doi:10.1002/2015GL065311.

#### **Response:**

We have added the reference (Lin et al., 2015) in the revised manuscript (the fourth paragraph of Section 1).

Page 3, Lines 13-15: Also cite Schnell, J. L., M. J. Prather, B. Josse, V. Naik, L. W. Horowitz, G. Zeng, D. T. Shindell, and G. Faluvegi (2016), Effect of climate change on surface ozone over North America, Europe, and East Asia, *Geophys. Res. Lett.*, 43, 3509-3518, doi: 10.1002/2016GL068060.

#### **Response:**

We have added the reference (Schnell et al., 2016) in the revised manuscript (the fourth paragraph of Section 1).

Page 3, Lines 20-30: Also explicitly discuss the results from Lin et al. (2014, Nature Geoscience). They found that interannual variability of springtime Asian ozone outflow is strongly influenced by ENSO-related shifts in the subtropical jet stream. Transport of Asian pollution towards the eastern North Pacific during spring has weakened in the 2000s due to more frequent La Nina-like conditions.

#### **Response:**

Following the Reviewer’s suggestion, we have added the following discussions in the fourth paragraph of Section 1 in the revised manuscript: “Lin et al. (2014) reported that interannual variability in springtime Asian O<sub>3</sub> transport, as inferred by the East Asian CO<sub>t</sub> (carbon-monoxide-like tracer), was strongly influenced by ENSO-related shifts in the subtropical jet stream, and that the decrease in ozone-rich Eurasian airflow reaching the eastern North Pacific during spring in the 2000s was attributed to more frequent La Nina events.”

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## Response to Reviewer #2

**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 O<sub>3</sub> outflow. See our response to your major comment (1);
  - More comprehensive validation on simulated surface-layer O<sub>3</sub> concentrations by using O<sub>3</sub> measurements from WDCGG and EANET, and the validation on simulated O<sub>3</sub> 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 O<sub>3</sub> (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 O<sub>3</sub> 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 O<sub>3</sub> outflow fluxes by changing O<sub>3</sub>

concentrations over East Asia (Yang et al., 2014; Lou et al., 2015), and by altering zonal winds (Kurokawa et al., 2009). The O<sub>3</sub> 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 O<sub>3</sub> fluxes and zonal winds is calculated to be +0.96 during summer when the APDM values of O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> concentrations, and the ozonesonde data from WOUDC are used to evaluate the simulated O<sub>3</sub> 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 O<sub>3</sub>. We use observations of tropospheric O<sub>3</sub> 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/](http://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](http://www.eanet.asia/product/index.html)), which are used to evaluate the simulated surface-layer O<sub>3</sub> concentrations. The four Japanese sites are “remote” sites in the downwind regions of China. Figure 3 compares the time series of monthly surface-layer O<sub>3</sub> mixing ratios simulated by MetEmisB with those measured by WDCGG and EANET. Simulated surface-layer O<sub>3</sub> levels agree well with observations at all the four stations. The model captures fairly well the seasonal cycles and interannual variations of surface O<sub>3</sub>, 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 O<sub>3</sub> outflow flux is the highest, but overestimates the low values during summer when Asian O<sub>3</sub> outflow is the minimum.

To evaluate the simulated O<sub>3</sub> 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](http://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 O<sub>3</sub> mixing ratios simulated by MetEmisB with



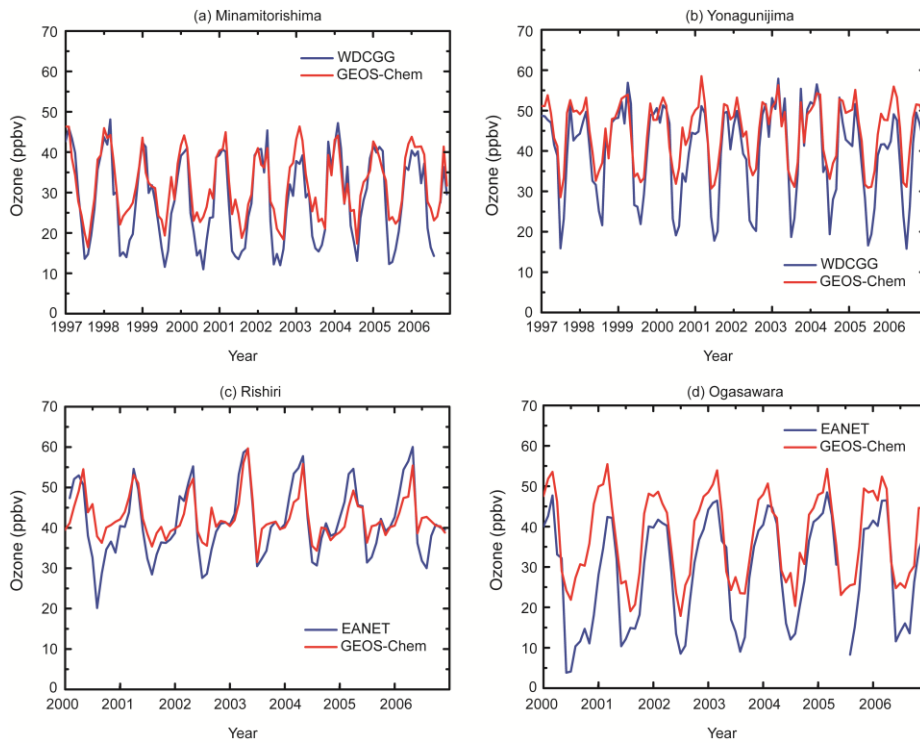
those measured by ozonesonde. Comparisons are shown for four altitudes in the troposphere. The GEOS-Chem model captures the seasonal cycles and interannual variations of tropospheric O<sub>3</sub> at all altitudes, with correlation coefficients ranging from 0.68 to 0.88 for Naha site, and from 0.55 to 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 O<sub>3</sub> measurements used in model evaluation.

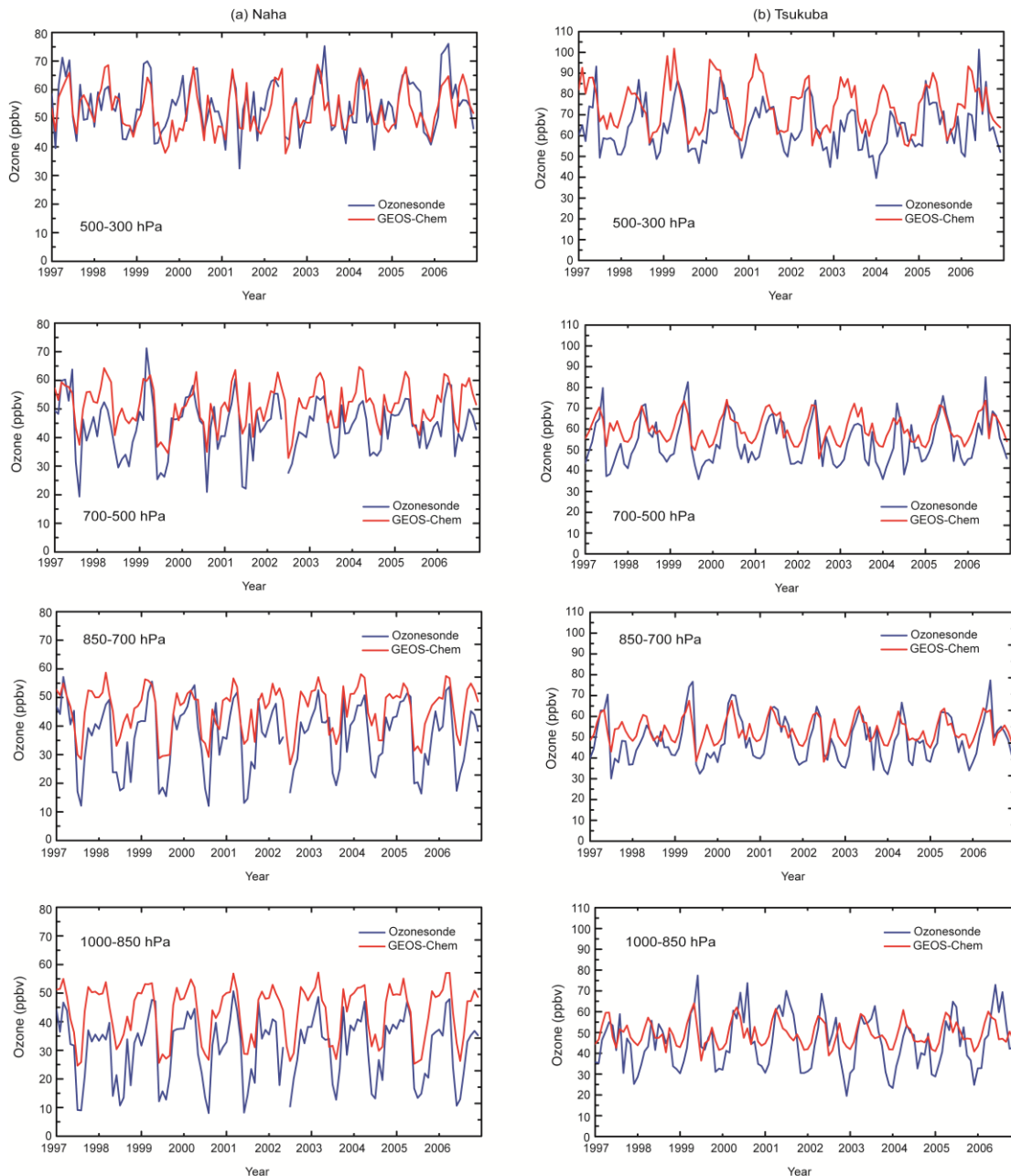
Site	Location	Database	Height	R <sup>a</sup>	NMB <sup>b</sup> (%)
Minamitorishima	24.3 N, 154.0 E	WDCGG	surface	0.92	+12.7
Yonagunijima	24.5 N, 123.0 E	WDCGG	surface	0.93	+12.6
Rishiri	45.1 N, 141.2 E	EANET	surface	0.82	+2.4
Ogasawara	27.1 N, 142.2 E	EANET	surface	0.90	+29.6
Naha	26.2 N, 127.7 E	WOUDC	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

<sup>a</sup> Correlation coefficient (R) between the observed and simulated monthly O<sub>3</sub> mixing ratios.

<sup>b</sup> Normalized mean bias (NMB, %) between the observed and simulated monthly O<sub>3</sub> mixing ratios.



**Figure 3.** Time series of monthly surface-layer O<sub>3</sub> 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.

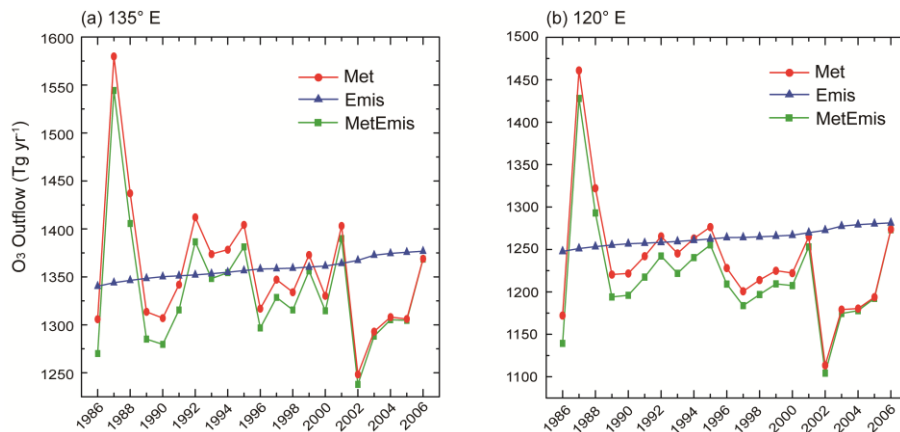
(3) Is 135 degE appropriate? - The authors mentioned in the title “ozone outflow from East Asia” and used a longitudinal transect at 135 degE to diagnose the eastward flux of ozone. I wonder why at 135 degE, not 120 degE, to be more close to ozone production region in central-eastern China. I think, 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 O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> outflow fluxes (Tg yr<sup>-1</sup>) 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 O<sub>3</sub> outflow during summer is not large, the percentage increase in O<sub>3</sub> 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 O<sub>3</sub> measurements from WDCGG and EANET, and with ozonesonde measurements from WOUDC for the boundary layer, middle and upper troposphere O<sub>3</sub> 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 O<sub>3</sub> outflow flux along 120° E, more close to the Asian continent. It is concluded from Figure H1 that the variations in O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> 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.



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**Response:**

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

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# Interannual variation, decadal trend, and future change in ozone outflow from East Asia

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**Abstract.** We examine the past and future changes in O<sub>3</sub> outflow from East Asia using a global three-dimensional chemical transport model GEOS-Chem. The simulations of Asian O<sub>3</sub> outflow for 1986–2006 are driven by the assimilated GEOS-4 meteorological fields, and those for 2000–2050 are driven by the meteorological fields archived from the Goddard Institute for Space Studies (GISS) General Circulation Model (GCM) 3 under the IPCC SRES A1B scenario. Sensitivity studies are conducted to examine the respective impacts of meteorological parameters and emissions on the variations in the outflow flux of O<sub>3</sub>. When both meteorological parameters and anthropogenic emissions varied during 1986–2006, the simulated Asian O<sub>3</sub> outflow fluxes exhibited a ~~small and~~ statistically insignificant decadal trend ~~of  $-2.2\%$  decade<sup>-1</sup>~~, but large interannual variations (IAVs) with seasonal absolute percent departure from the mean (APDM) values of 4–9 % and annual APDM value of 3.3 %. Sensitivity simulations indicated that the large IAVs of O<sub>3</sub> outflow fluxes were mainly caused by the variations in meteorological conditions. Variations in meteorological parameters drove the IAVs in O<sub>3</sub> outflow fluxes by altering O<sub>3</sub> concentrations over East Asia and by altering zonal winds, and the latter was identified to be the key factor since the O<sub>3</sub> outflow was highly correlated with zonal winds during 1986–2006. The simulations of the 2000–2050 changes show that the annual outflow flux of O<sub>3</sub> will increase by 2.0 %, 7.9 %, and 12.2 %, respectively, owing to climate change alone, emissions change alone, and changes in both climate and emissions. Therefore, climate change will aggravate the effects of the increases in anthropogenic emissions on future changes in the Asian O<sub>3</sub> outflow. Future climate change is predicted to greatly

increase Asian O<sub>3</sub> outflow in the spring and summer seasons as a result of the projected increases in zonal winds. Findings from the present study help to understand the variations in tropospheric O<sub>3</sub> in the downwind regions of East Asia on different timescales, and have important implications for long-term air quality planning [for the downwind regions of China, such as Japan and US](#).

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## 1 Introduction

Tropospheric ozone (O<sub>3</sub>) is an important air pollutant, which has a detrimental effect on human health (Fann et al., 2012; Jhun et al., 2014), crops (Wilkinson et al., 2011; Tai et al., 2014), and ecosystems (Fuentes et al., 2013; Yue and Unger, 2014). It is also an important greenhouse gas that directly contributes to global warming (IPCC, 2013). O<sub>3</sub> has a relatively long lifetime of weeks in the free troposphere (Young et al., 2013; Monks et al., 2015), which makes intercontinental transport of O<sub>3</sub> an important issue for understanding O<sub>3</sub> concentrations and planning emission control measures.

A number of previous studies have shown that Asian continental outflow impacts the global O<sub>3</sub> budget (Liu et al., 2002), and influences O<sub>3</sub> air quality in the downwind regions, such as [the western North Pacific through the western North America the United States and Canada](#) (Jacob et al., 1999; Tanimoto et al., 2005; Kim et al., 2006; Li et al., 2008; Zhang et al., 2008; Chiang et al., 2009; [Kurokawa et al., 2009](#); Huang et al., 2010; [Nagashima et al., 2010](#); Walker et al., 2010; Ambrose et al., 2011; Lin et al., 2012; Ou-Yang et al., 2013; [Han et al., 2015](#); [Pochanart et al., 2015](#)). Liu et al. (2002) reported that boundary-layer O<sub>3</sub> pollution was lifted into the upper troposphere by deep convection over the Asian maritime continent, from where it was transported northward along the upper branch of the local Hadley circulation and into the mid-latitude westerlies, influencing the global O<sub>3</sub> budget. Using a global 3-D chemical transport model GEOS-Chem, Zhang et al. (2008) estimated that Asian pollution enhanced surface-layer O<sub>3</sub> concentrations by 5–7 ppbv over western North America in spring 2006. Walker et al. (2010) used the same model (GEOS-Chem) to evaluate sensitivities of tropospheric O<sub>3</sub> over Canada to Asian anthropogenic emissions, and reported that the contribution from Asian emissions to O<sub>3</sub> profiles above Whistler, Canada, was 6–8 ppbv in spring 2006. Through an integrated analysis of in situ and satellite measurements in May–June 2010 with a global chemistry-climate model GFDL AM3, Lin et al. (2012) reported that Asian emissions could contribute 8–15 ppbv O<sub>3</sub> over the western United States on days when the observed daily maximum 8-h average O<sub>3</sub> (MDA8 O<sub>3</sub>) exceeded 60 ppbv, and that 20 % of MDA8 O<sub>3</sub> exceedances of 60 ppbv would not have occurred in the southwestern United States in the absence of Asian anthropogenic emissions.

Asian O<sub>3</sub> outflow exhibits seasonal variations (Liu et al., 2002; Han et al., 2015). Using a global 3-D chemical

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transport model GEOS-Chem, Liu et al. (2002) simulated the seasonal variations of the Asian outflow flux of O<sub>3</sub> over the Pacific, which was defined as the eastward flux integrated for the tropospheric column through a wall located at 150°E between 10°N and 60°N. They found that the Asian O<sub>3</sub> outflow flux reached the maximum in early spring (March) and the minimum in summer (July). Han et al. (2015) examined O<sub>3</sub> measurements at Ieodo Ocean Research Station, which was located in the East China Sea and regarded as an ideal place to observe Asian outflow without local effects. They reported that the seasonal variation of O<sub>3</sub> was distinct, with a minimum in August and two peaks in April and October, and was greatly affected by the seasonal wind pattern over East Asia.

Continental outflow of O<sub>3</sub> is expected to vary on interannual to decadal timescales, because tropospheric O<sub>3</sub> concentrations and meteorological parameters have variations on these timescales. Large interannual variations (IAVs) of tropospheric O<sub>3</sub> concentrations have been reported in previous observational studies (Kurokawa et al. 2009; Zhou et al., 2013). Analyzing 11 years of ozonesonde data over Hong Kong, Zhou et al. (2013) reported that observed tropospheric O<sub>3</sub> levels during 2000–2010 exhibited high IAV, with an annual averaged amplitude [defined as (maximum + 2nd maximum – minimum – 2nd minimum) × 0.5 / the average during 2000–2010] up to 30 % of the averaged concentrations at 3–8 km altitude. Kurokawa et al. (2009) analyzed observed springtime O<sub>3</sub> over Japan during 1985–2005, and found that the observed O<sub>3</sub> showed greater year-to-year variations than the annual rate of the long-term trend. Decadal trends of tropospheric O<sub>3</sub> concentrations have been reported for different locations on the basis of observations (Ding et al., 2008a; Xu et al., 2008; Tang et al., 2009; [Tanimoto, 2009](#); Wang et al., 2009b; Cooper et al., 2010; Wang et al., 2012; Lin et al., 2014, [2015](#); Zhang et al., 2014), such as –0.56 ppbv yr<sup>–1</sup> over Linan in eastern China (Xu et al., 2008), +0.58 ppbv yr<sup>–1</sup> over Hong Kong in southern China (Wang et al., 2009b), [+1.0 ppbv yr<sup>–1</sup> at Mt. Happo in Japan \(for springtime O<sub>3</sub>; Tanimoto, 2009\)](#), and +0.35 ppbv yr<sup>–1</sup> over Hawaii in North Pacific (for autumn O<sub>3</sub>; Lin et al., 2014). Future changes of tropospheric O<sub>3</sub> concentrations have also been predicted by modeling studies (Racherla and Adams, 2006, 2009; Lin et al., 2008; Wu et al., 2008a; Lam et al., 2011; Wild et al., 2012; Gao et al., 2013; Liu et al., 2013; Wang et al., 2013; Lee et al., 2015; Val Martin et al., 2015; [Schnell et al., 2016](#); Zhu and Liao, 2016). Wang et al. (2013), using the NASA GISS GCM/GEOS-Chem model combination, reported that the summer surface-layer O<sub>3</sub> levels averaged over China would increase by 11.9 ppbv due to the combined changes in climate and emissions over 2000–2050 under the SRES A1B scenario. Meteorological parameters, especially winds that are important for O<sub>3</sub> outflow, also exhibit variations on different time scales (Chang et al., 2000; Ding et al., 2008b; Sun et al., 2009; Zhang and Guo, 2010; Hirahara et al., 2012). Large IAVs of the East Asian summer monsoon (EASM) have been reported in previous studies (Zhu et al., 2012; Yang et al., 2014). The decadal-scale weakening of the EASM since the 1950s has also been reported by many previous studies, and anomalous northeasterlies during the weak



monsoon years were found over the western North Pacific near 40 °N, which did not favor the outflow of pollutants from northern China (Chang et al., 2000; Ding et al., 2008b; Zhu et al., 2012). On the basis of NCEP/NCAR reanalysis data, Sun et al. (2009) showed that the axis location of the East Asia subtropical westerly jet (EASWJ) had displaced southward since the end of the 1970s, intensifying the westerly wind over 25–35 °N and weakening it over 42–50 °N, and therefore influencing the outflow of pollutants. [Lin et al. \(2014\) reported that interannual variability in springtime Asian O<sub>3</sub> transport, as inferred by the East Asian COt \(carbon-monoxide-like tracer\), was strongly influenced by ENSO-related shifts in the subtropical jet stream, and that the decrease in ozone-rich Eurasian airflow reaching the eastern North Pacific during spring in the 2000s was attributed to more frequent La Nina events.](#) Most of the Coupled Model Intercomparison Project Phase 3 (CMIP3) models projected that the Asian jet would be intensified on its equatorward side by the end of the 21st century (Zhang and Guo, 2010; Hirahara et al., 2012).

Few previous studies have examined the IAVs, decadal trends, and future changes in O<sub>3</sub> outflow. In this work, we examine the historical (1986–2006) and future (2000–2050) changes of O<sub>3</sub> outflow from East Asia, and systematically quantify the roles of meteorological parameters and/or anthropogenic emissions on the changes. The descriptions of the model, emissions, and numerical simulations are presented in Sect. 2. Section 3 evaluates the model performance for tropospheric O<sub>3</sub>. Section 4 discusses the IAVs and decadal trends in the O<sub>3</sub> outflow from East Asia over 1986–2006. Future changes in O<sub>3</sub> outflow from East Asia for 2000–2050 are presented in Sect. 5.

## 2 Methods

### 2.1 Model description

We apply the global 3-D chemical transport model GEOS-Chem to simulate O<sub>3</sub> outflow fluxes. The GEOS-Chem model includes a detailed simulation of O<sub>3</sub>–NO<sub>x</sub>–hydrocarbon (~80 species, ~300 chemical reactions) (Bey et al., 2001) and aerosol chemistry. Aerosol species include sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), ammonium (NH<sub>4</sub><sup>+</sup>) (Park et al., 2004; Pye et al., 2009), black carbon (BC) and organic carbon (OC) (Park et al., 2003), mineral dust (Fairlie et al., 2007), and sea salt (Alexander et al., 2005). The simulations account for the impacts of aerosols on the distributions and concentrations of O<sub>3</sub> through heterogeneous reactions and changes in photolysis rates (Lou et al., 2014).

To simulate historical changes in the Asian outflow of O<sub>3</sub>, the GEOS-Chem model is driven by the assimilated GEOS-4 meteorological fields from the Goddard Earth Observing System (GEOS) of NASA Global Modeling and Assimilation Office (GMAO). We perform simulations for 1986–2006, which are the years with available GEOS-4 meteorological datasets. The version of the model used here has a horizontal resolution of 2 ° (latitude) × 2.5 °

(longitude), with 30 vertical layers.

To simulate future changes of O<sub>3</sub> outflow fluxes during 2000–2050, the GEOS-Chem simulation is driven by meteorological data from the National Aeronautics and Space Administration/Goddard Institute for Space Studies (NASA/GISS) general circulation model (GCM) 3 (Rind et al., 2007) for both the present day (1996–2005) and future (2046–2055), following Wu et al. (2008b), Pye et al. (2009), Wang et al. (2013), and Jiang et al. (2013). Both the GISS and GEOS-Chem models used here have a horizontal resolution of 4° latitude by 5° longitude, with 23 vertical layers.

## 2.2 Emissions

For simulations during 1986–2006, the global anthropogenic emissions of reactive nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO), and sulfur dioxide (SO<sub>2</sub>) in the model are from the Emission Database for Global Atmospheric Research (EDGAR) inventory (Olivier and Berdowski, 2001). The global emissions of non-methane volatile organic compounds (NMVOCs) are from the Global Emissions Inventory Activity (GEIA) inventory (Piccot et al., 1992). Global emissions of carbonaceous aerosols (BC and OC) follow Bond et al. (2007). Anthropogenic emissions of reactive NO<sub>x</sub>, CO, SO<sub>2</sub>, NH<sub>3</sub>, and NMVOCs over East Asia are overwritten by the emissions inventory of Streets et al. (2003) and Zhang et al. (2009). IAVs of anthropogenic emissions are represented by global-gridded annual scaling factors as described by van Donkelaar et al. (2008) for NO<sub>x</sub>, CO, and NMVOCs. Biomass burning emissions are taken from the Global Fire Emissions Database-3 (GFED-3) inventory (van der Werf et al., 2010) for 1997–2006. The biomass burning emissions before 1997 are unavailable because of the lack of datasets.

Figure 1 shows the evolution of anthropogenic and biomass burning emissions of O<sub>3</sub> precursors (NO<sub>x</sub>, CO, NMVOCs) summed over the globe and Asia (60–150° E, 10° S–55° N) over 1986–2006. Global anthropogenic emissions of these precursors exhibited no significant trends, while the Asian anthropogenic emissions showed large increases over the past two decades. Relative to year 1986, the Asian anthropogenic emissions of NO<sub>x</sub>, CO, and NMVOCs in 2006 increased by 70.0 %, 42.1 %, and 50.9 %, respectively. Compared with anthropogenic emissions, biomass burning emissions had greater IAVs during 1997–2006. Figure 1 also shows the pathway for the global CH<sub>4</sub> abundance used in our simulations of O<sub>3</sub>. The CH<sub>4</sub> mixing ratio in 1986 was 1672 ppb, which increased by 6.3 % in 2006.

For future simulations during 2000–2050, anthropogenic emissions of O<sub>3</sub> precursors, including NO<sub>x</sub>, CO, and NMVOCs, are taken from Wu et al. (2008b), and those of NH<sub>3</sub> and SO<sub>2</sub> follow those in Pye et al. (2009). The future anthropogenic emissions of O<sub>3</sub> precursors, aerosol precursors, and aerosols under the SRES A1B scenario are

generated by the Integrated Model to Assess the Greenhouse Effect (IMAGE) socioeconomic model using growth factors for different species and countries (Streets et al., 2004). Table 1 shows the present-day (year 2000) and future (year 2050) anthropogenic emissions of O<sub>3</sub> precursors under the SRES A1B scenario. The global emissions of NO<sub>x</sub>, CO, and NMVOCs are projected to increase by 78.4 %, 26.4 %, and 89.4 % over 2000–2050; and the Asian emissions are projected to increase by 159.6 %, 23.7 %, and 118.6 %, respectively. Present-day methane mixing ratios are specified as 1750 ppbv on the basis of observations (Wu et al., 2008b). The future methane concentrations are set to 2400 ppbv, following the SRES A1B scenario (Pye et al., 2009).

The natural emissions of O<sub>3</sub> precursors, including NO<sub>x</sub> from lightning and soil, and NMVOCs from vegetation, are calculated on the basis of the assimilated GEOS-4 meteorological fields and GISS Model 3 meteorological parameters. The lightning NO<sub>x</sub> emissions follow Price and Rind (1992), with the NO<sub>x</sub> vertical profile proposed by Pickering et al. (1998). The soil NO<sub>x</sub> emissions scheme in the GEOS-Chem model is based on the work of Yienger and Levy (1995) and Wang et al. (1998). Biogenic emissions of NMVOCs are calculated according to the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006). Figure 2 shows the evolution of natural emissions summed over the globe and Asia over 1986–2006. Both global and Asian NO<sub>x</sub> emissions from lightning exhibited large IAVs and significant increasing trends. It has been shown that warming climate leads to increased lightning NO<sub>x</sub> (IPCC, 2013). Compared with lightning NO<sub>x</sub> emissions, NO<sub>x</sub> emissions from soil showed smaller IAVs and no significant decadal trend. Both global and Asian biogenic emissions of NMVOCs have been shown to have large IAVs, as a result of the changes in both vegetation and meteorological parameters (Fu and Liao, 2012). For future simulations during 2000–2050, the natural emissions of O<sub>3</sub> precursors are listed in Table 2. The simulated emissions of lightning NO<sub>x</sub>, soil NO<sub>x</sub>, and biogenic VOCs are projected to increase by 18.8 %, 14.9 %, and 22.1 % for the globe, and by 16.7 %, 21.4 %, and 18.9 % for Asia, respectively.

The effects of changes in stratosphere–troposphere exchange (STE) of O<sub>3</sub> are not included in this study for both past and future simulations. The cross-tropopause O<sub>3</sub> flux is represented by the synthetic O<sub>3</sub> (Synoz) method (McLinden et al., 2000), which imposes a global annual mean cross-tropopause O<sub>3</sub> flux of 500 Tg yr<sup>-1</sup>.

### 2.3 Numerical experiments

To examine the respective and combined impacts of meteorological parameters, anthropogenic emissions, and biomass burning emissions on the IAVs and decadal trends of O<sub>3</sub> outflow from East Asia during 1986–2006, we perform simulations for four cases (Table 3):

(a) Met: The simulation of O<sub>3</sub> outflow for 1986–2006, to examine the effect of variations in meteorological parameters alone. The meteorological parameters vary from 1986 to 2006, and the anthropogenic emissions are fixed at year 2006 levels. Biomass burning emissions are turned off.

(b) Emis: The simulation of O<sub>3</sub> outflow for 1986–2006, to examine the effect of variations in anthropogenic emissions alone. The anthropogenic emissions vary from 1986 to 2006, and the meteorological parameters are fixed at year 2006 levels. Biomass burning emissions are turned off.

(c) MetEmis: The simulation of O<sub>3</sub> outflow for 1986–2006, with variations in both meteorological parameters and anthropogenic emissions during 1986–2006. Biomass burning emissions are turned off.

(d) MetEmisB: The simulation of O<sub>3</sub> outflow for 1997–2006, with variations in meteorological parameters, anthropogenic emissions, and biomass burning emissions during 1997–2006. Note that biomass burning emissions in the model are not available before 1997.

To identify the relative roles of future changes in meteorological parameters and emissions in 2000–2050 changes in Asian O<sub>3</sub> outflow flux, another four simulations are carried out: (a) Met2000Emis2000: present-day climate and emissions; (b) Met2050Emis2000: future climate and present-day anthropogenic emissions; (c) Met2000Emis2050: present-day climate and future anthropogenic emissions; and (d) Met2050Emis2050: future climate and emissions. Both the future climate and anthropogenic emissions follow the IPCC SRES A1B scenario.

The mass flux of O<sub>3</sub> through the vertical plane along 135 °E from 20 °N to 55 °N from the surface to 100 hPa (marked in Fig. 3a) is used to quantify Asian O<sub>3</sub> outflow. The metric of mass flux through a vertical plane was also used by Liu et al. (2002) to represent Asian O<sub>3</sub> outflow, and by Jiang et al. (2013) and Yang et al. (2015) to represent Asian aerosol outflow. It should be noted that the O<sub>3</sub> outflow flux from East Asia includes the effects of emissions in different regions of the world owing to the relatively long lifetime (~3 weeks) of O<sub>3</sub> (Fiore et al., 2002; Liao et al., 2006). However, Liu et al. (2002) found that anthropogenic sources in Asia made the largest contribution to the Asian outflow flux of O<sub>3</sub>.

### 3 Model evaluation

The GEOS-Chem simulations of O<sub>3</sub> have been evaluated extensively for the U.S. (Liu et al., 2006; Wu et al., 2008b; Zhang et al., 2008), Europe (Auvray and Bey, 2005; Liu et al., 2006; Kim et al., 2015), and China (Wang et al., 2013; Lou et al., 2014; Yang et al., 2014; Zhu and Liao, 2016). These studies showed that the GEOS-Chem model captured the magnitude and distribution of the surface-layer concentration and column burden of tropospheric O<sub>3</sub> fairly well.

The vertical distributions of O<sub>3</sub> have also been evaluated by aircraft campaigns and ozonesonde measurements (Zhang et al., 2008; Walker et al., 2010; Wang et al., 2011), showing that the GEOS-Chem model closely reproduced the observed O<sub>3</sub> profiles.

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 O<sub>3</sub>. We use observations of tropospheric O<sub>3</sub> 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/wdogg/](http://www.ds.data.jma.go.jp/gmd/wdogg/)), 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](http://www.eanet.asia/product/index.html)), which are used to evaluate the simulated surface-layer O<sub>3</sub> concentrations. The four Japanese sites are “remote” sites in the downwind regions of China. Figure 3 compares the time series of monthly surface-layer O<sub>3</sub> mixing ratios simulated by MetEmisB with those measured by WDCGG and EANET. Simulated surface-layer O<sub>3</sub> levels agree well with observations at all the four stations. The model captures fairly well the seasonal cycles and interannual variations of surface O<sub>3</sub>, 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 O<sub>3</sub> outflow flux is the highest, but overestimates the low values during summer when Asian O<sub>3</sub> outflow is the minimum.

To evaluate the simulated O<sub>3</sub> 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](http://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 O<sub>3</sub> 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 O<sub>3</sub> 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.

The increasing trend in surface-layer O<sub>3</sub> in East Asia over the past two decades was reported by previous studies (Ding et al., 2008a; Wang et al., 2009b; Xu et al., 2016). Figure 5 compares the simulated trends in annual-mean surface-layer O<sub>3</sub> concentrations from the MetEmis experiment with the observed trends collected from Lin et al. (2016) and references therein. The observed trends of annual-mean O<sub>3</sub> at Beijing, Hongkong, Taiwan, Waliguan, and South



Korea stations are  $+0.90 \text{ ppbv yr}^{-1}$ ,  $+0.58 \text{ ppbv yr}^{-1}$ ,  $+0.54 \text{ ppbv yr}^{-1}$ ,  $+0.25 \text{ ppbv yr}^{-1}$ ,  $+0.48 \text{ ppbv yr}^{-1}$ , respectively. Simulated  $\text{O}_3$  concentrations at all stations exhibit statistically significant increasing trends, although the model underestimates the trends. The modeled  $\text{O}_3$  trends were reported to have low biases in previous studies (Tanimoto et al., 2009; Parrish et al., 2014). Parrish et al. (2014) compared  $\text{O}_3$  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 whereas the other two models suggested no significant increasing trends.

In general, the GEOS-Chem model can capture fairly well the seasonal cycles and interannual variations of tropospheric  $\text{O}_3$ , although the model overestimates the low values during summer indicating an overestimation of Asian  $\text{O}_3$  outflow in summer. The increasing trends in surface-layer  $\text{O}_3$  in China over the past two decades can also be captured by GEOS-Chem model, although the modeled  $\text{O}_3$  trends have low biases.

We use tropospheric column  $\text{O}_3$  (TCO) products retrieved from TOMS/SBUV (Total Ozone Mapping Spectrometer/Solar Backscattered Ultraviolet instruments) to evaluate model performance for tropospheric  $\text{O}_3$  over China and regions downwind. The TOMS/SBUV retrievals are available for 1997–2005. In 2001, it was discovered that the TOMS had experienced instrument problems, and the quality of the total  $\text{O}_3$  measurements had degraded (Fishman et al., 2003). Therefore, we show in Fig. 3 the maps of TCO retrieved from TOMS/SBUV and those simulated by the model (in the MetEmisB simulation) averaged over 1997–2005 (excluding year 2001). The TOMS/SBUV retrievals exhibit high values exceeding 40 Dobson units (DU) over the North China Plain and Sichuan Basin, and low values of 25–35 DU over the Tibetan Plateau. The spatial distribution of TCO is mostly captured by the model. The differences between the GEOS-Chem simulation and TOMS/SBUV observation indicate that the model underestimates the TCO over the Tibetan Plateau by  $\sim 10$  DU and overestimates the TCO over central and eastern China by  $\sim 10$  DU. Zhang et al. (2010) attributed the overestimation of TCO in the northern subtropics in the GEOS-Chem model to an excessive stratospheric influx of  $\text{O}_3$ .

We then compare the simulated monthly and yearly variations in TCO averaged over a selected region ( $30\text{--}50^\circ\text{N}$ ,  $130\text{--}150^\circ\text{E}$ , downwind of China; Fig. 4) in the western Pacific Ocean in the MetEmisB simulation with those obtained from TOMS/SBUV. The GEOS-Chem model generally overestimates TCO values, especially during winter/spring months. Chandra et al. (2003) also reported that the TCO values simulated by the GEOS-Chem model tended to be higher by 5–10 DU compared with sonde values at Naha, Japan ( $26^\circ\text{N}$ ,  $128^\circ\text{E}$ ) during winter months. The satellite measurements suggest that TCO has a maximum in July and a minimum in January. The seasonality in TCO is reproduced fairly well by the model, with a correlation coefficient of +0.90 between simulated and TOMS/SBUV TCO. However, the maximum in the model occurs two months earlier than in TOMS/SBUV. Wang et al. (2011) also

~~reported that the maximum of TCO averaged over East China simulated by the GEOS Chem model occurred two months earlier than Tropospheric Emission Spectrometer (TES) retrievals. On the interannual scale, for 1997–2005, the model mostly captures the years with maximum or minimum TCO values, with a correlation coefficient of +0.56.~~

~~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 O<sub>3</sub> column burden over China and downwind regions, which lends us confidence to simulate the temporal evolutions of the Asian O<sub>3</sub> outflow.~~

## 4 Simulated Asian O<sub>3</sub> outflow during 1986–2006

### 4.1 Seasonal patterns of Asian O<sub>3</sub> outflow

Figure 56 shows the pressure–latitude cross-sections along 135 °E of the seasonal O<sub>3</sub> outflow fluxes averaged over 1997–2006 in the MetEmisB simulation. The maximum O<sub>3</sub> fluxes were found in the middle-upper troposphere, in consistent with Liu et al. (2002) and Wang et al. (2009a), and it could be attributed to the vertical distributions of both zonal winds and O<sub>3</sub> concentrations. The westerlies strengthen with altitudes with the strongest winds occurring around 200 hPa (known as the East Asia subtropical westerly jet) (Ren et al., 2011). Concentrations of O<sub>3</sub> are high in the upper troposphere over the mid-latitudes (Wang, 1999).

The seasonal mass fluxes through the meridional plane (along 135 °E from 20 °N to 55 °N, and from the surface to 100 hPa) were calculated to be 509.6, 437.6, 126.6, and 268.7 Tg season<sup>-1</sup> for December–January–February (DJF), March–April–May (MAM), June–July–August (JJA), and September–October–November (SON), respectively.

Although the seasonal flux was highest in DJF, the monthly O<sub>3</sub> flux through the panel peaked in March and reached the nadir in July (not shown in Fig. 56). Such monthly variations of the Asian O<sub>3</sub> outflow flux agreed with those in Liu et al. (2002). The maximum O<sub>3</sub> outflow in March was caused by the combined effects of meteorological conditions, biomass burning emissions, and stratospheric O<sub>3</sub> intrusion. The “warm conveyor belt” (WCB) airstreams that export pollution from the Asian boundary layer to the free troposphere, and the mid-latitude prevailing westerly winds in the free troposphere that transport pollution from Asia to the Northwest Pacific, were strongest during the early spring period (Eckhardt et al., 2004; Pochanart et al., 2004). The contribution from Asian biomass burning emissions on O<sub>3</sub> outflow was maximum in the spring and insignificant during other seasons (Liu et al., 2002). The stratospheric O<sub>3</sub> intrusion was also found to be most effective in late winter and early spring (Danielsen and Mohnen, 1977; Mahlman and Moxim, 1978).

## 4.2 IAVs and decadal trends of Asian O<sub>3</sub> outflow

Figure 67a shows the simulated annual O<sub>3</sub> outflow fluxes through the meridional plane along 135 °E from 20 °N to 55 °N, from the surface to 100 hPa, during 1986–2006 in the Met, Emis, and MetEmis simulations, and Fig. 67b shows the associated deviations from the mean (DEV). The simulations of the O<sub>3</sub> outflow in Met, Emis, and MetEmis examined, respectively, the effects of variations in meteorological parameters alone, anthropogenic emissions alone, and both meteorological parameters and anthropogenic emissions. The outflow fluxes of O<sub>3</sub> with changes in anthropogenic emissions alone (the Emis simulation) exhibited a statistically significant ( $P < 0.001$ ) increasing trend. However, the magnitude of the increasing trend was very small; the decadal trend of the Asian O<sub>3</sub> outflow flux in the Emis simulation was calculated to be +16.7 Tg decade<sup>-1</sup> (i.e., +1.2 % decade<sup>-1</sup>) using the linear fit with least-squares method. The DEV, defined as

$$\text{DEV} = 100\% \times \left( C_i - \frac{1}{n} \sum_{i=1}^n C_i \right) / \left( \frac{1}{n} \sum_{i=1}^n C_i \right),$$

where  $n$  is the number of years examined ( $n = 21$  for 1986–2006) and  $C_i$  is the simulated O<sub>3</sub> outflow flux in year  $i$ , changed from -1.3 % (in 1986) to +1.4 % (in 2006), also indicating a small increasing trend in the O<sub>3</sub> outflow flux.

With variations in meteorological parameters alone (the Met simulation), simulated O<sub>3</sub> outflow fluxes exhibited large IAVs, but a statistically insignificant ( $P > 0.05$ ) decadal trend of -3.4 % decade<sup>-1</sup>. The DEV values in the Met simulation ranged from -8 % to +16.5 %. With variations in both anthropogenic emissions and meteorological parameters (the MetEmis simulation), the simulated O<sub>3</sub> outflow showed large IAVs, but a statistically insignificant ( $P > 0.05$ ) decadal trend of -2.2 % decade<sup>-1</sup>.

To analyze the IAVs of O<sub>3</sub> outflow fluxes, the decadal trend obtained from the linear fit was removed from the time series of simulated O<sub>3</sub> outflow fluxes, following the approach used in previous studies that examined IAVs of aerosol outflow fluxes (Yang et al., 2015) and O<sub>3</sub> concentrations (Camp et al., 2003). The deviations from the mean of the detrended O<sub>3</sub> outflow fluxes from the Met, Emis, and MetEmis simulations over 1986–2006 are shown in Fig. 67c. While the detrended outflow fluxes of O<sub>3</sub> in Met and MetEmis simulations showed large IAVs with DEV values in the range of -7.5 % to +13.5 %, the DEV values in the Emis simulation were very small (in the range of ±0.3 %). The two deviation curves from the Met and MetEmis simulations almost coincided with each other, indicating the dominant role of variations in meteorological parameters in the IAVs of the Asian O<sub>3</sub> outflow flux.

The IAVs in the O<sub>3</sub> outflow fluxes were further quantified with statistical variables of mean absolute deviation (MAD) and absolute percent departure from the mean (APDM), which have been used in previous IAV studies, such

as Mu and Liao (2014), Lou et al. (2015), and Yang et al. (2015). The absolute IAVs of the O<sub>3</sub> outflow fluxes can be quantified by MAD, defined as

$$\text{MAD} = \frac{1}{n} \sum_{i=1}^n |C_i - \frac{1}{n} \sum_{i=1}^n C_i|,$$

while the IAVs relative to the multi-year average outflow flux can be quantified by APDM, defined as

$$\text{APDM} = 100\% \times \text{MAD} / \left( \frac{1}{n} \sum_{i=1}^n C_i \right),$$

where  $n$  is the number of years examined ( $n = 21$  for years 1986–2006) and  $C_i$  is the detrended O<sub>3</sub> outflow flux in year  $i$ . The MAD and APDM values of the detrended seasonal and annual O<sub>3</sub> outflow fluxes across the meridional plane along 135 °E from 20 °N to 55 °N, from the surface to 100 hPa, are shown in Fig. 78. The seasonal MAD and APDM values in the Emis simulation were close to zero, while those in the Met and MetEmis simulations were relatively large.

The APDM values in the Met and MetEmis simulations were maximum in JJA and minimum in SON. The MAD and APDM values in the Met simulation were almost identical to those in the MetEmis simulation, which indicated again that the IAVs of the O<sub>3</sub> outflow fluxes were mainly dependent on the variations in meteorological conditions, rather than the variations in anthropogenic emissions. With variations in both meteorological parameters and anthropogenic emissions, the seasonal APDM values were in the range of 4–9 % and the annual APDM value was 3.3 %.

Figure 89 shows the pressure–latitude cross-sections of MAD values along 135 °E for detrended annual O<sub>3</sub> outflow fluxes from the Met, Emis, and MetEmis simulations. The O<sub>3</sub> outflow in the Met simulation exhibited large IAVs throughout the whole troposphere, with MAD values greater than 0.2 kg yr<sup>-1</sup> m<sup>-2</sup>. The MAD values increased with altitude, which could be attributed to the vertical distributions of the IAVs in westerly winds (see MAD values of winds in Fig. 89a). The variations in anthropogenic emissions led to very small IAVs, with MAD values less than 0.2 kg yr<sup>-1</sup> m<sup>-2</sup> (Fig. 89b) throughout the troposphere. With both variations in meteorological parameters and anthropogenic emissions, the MAD values (Fig. 89c) showed almost identical magnitudes and spatial distributions to those in the Met simulation (Fig. 89a), indicating the dominant role of variations in meteorological conditions in the IAVs of the O<sub>3</sub> outflow.

Variations in meteorological conditions can influence the IAVs of the O<sub>3</sub> outflow fluxes by changing O<sub>3</sub> concentrations over East Asia (Yang et al., 2014; Lou et al., 2015), and by altering zonal winds (Kurokawa et al., 2009). The O<sub>3</sub> 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 O<sub>3</sub> fluxes and zonal winds is calculated to be +0.96 during summer when the APDM values of O<sub>3</sub> 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 O<sub>3</sub> outflow fluxes.](#)

### 4.3 Effect of variations in biomass burning emissions

The biomass burning emissions of the O<sub>3</sub> precursors exhibited large IAVs during 1997–2006 (Fig. 1). To analyze the impacts of biomass burning emissions on IAVs of O<sub>3</sub> outflow fluxes, we compare the MAD and APDM values of detrended O<sub>3</sub> outflow fluxes during 1997–2006 in the MetEmis and MetEmisB simulations. The MAD (APDM) was calculated to be 31.17 Tg yr<sup>-1</sup> (2.35 %) in the MetEmis simulation and 31.82 Tg yr<sup>-1</sup> (2.36 %) in the MetEmisB simulation. The minor influence of biomass burning emissions on the IAVs of the O<sub>3</sub> outflow fluxes from East Asia was also supported by Voulgarakis et al. (2015). Furthermore, Lin et al. (2014) reported that meteorological variability, compared with the variability in biomass burning, was much more important for driving the IAVs in springtime O<sub>3</sub> at the Mauna Loa Observatory, a remote North Pacific site sensitive to Asian pollution outflow.

## 5 Future changes in Asian O<sub>3</sub> outflow for 2000–2050

In this part of the study, we quantify future decadal changes in Asian O<sub>3</sub> outflow during 2000–2050 under the SRES A1B scenario, and examine the relative impacts of variations in climate and anthropogenic emissions on the changes. We conduct each simulation for 10 years, driven by 1996–2005 meteorology to represent the present-day (year 2000) climate, and by 2046–2055 meteorological fields to represent the future (year 2050) climate, following 1 year of model spin-up. All the results presented below are 10-year averages. Simulated present-day and future changes in seasonal and annual fluxes of O<sub>3</sub> across the vertical plane along 135 °E from 20 °N to 55 °N are summarized in Table 45.

### 5.1 Present-day O<sub>3</sub> outflow

The pressure–latitude cross-sections along 135 °E of the simulated present-day (Met2000Emis2000) seasonal O<sub>3</sub> outflow fluxes, driven by the meteorological inputs provided by GISS GCM 3, are shown in Fig. 910a. The magnitudes, spatial distributions, and seasonal variations agree closely with those driven by the assimilated GEOS-4 meteorological fields (Fig. 56). The O<sub>3</sub> outflow flux through the vertical plane is simulated to be 1877.1 Tg yr<sup>-1</sup> with GISS GCM 3 meteorology, and 1342.5 Tg yr<sup>-1</sup> with the GEOS-4 assimilated meteorological fields, which indicates the reliability of the simulated present-day O<sub>3</sub> outflow fluxes.

### 5.2 Effect of future changes in climate alone

Relative to the present-day value, year 2050 annual outflow of O<sub>3</sub> is estimated to increase by 2.0 % (Table 45) as a result of climate change alone (Met2050Emis2000 minus Met2000Emis2000). The outflow of O<sub>3</sub> shows a slight decrease of 1.8 % in DJF and of 3.8 % in SON, but a large increase of 14.5 % in JJA and of 7.3 % in MAM. The spatial distributions of projected changes in O<sub>3</sub> fluxes are well consistent with those of changes in zonal winds (Fig. 910b). The wind speed of the westerlies in DJF and SON decreases across the troposphere over 30–45 °N, leading to the reductions in the O<sub>3</sub> outflow fluxes. In contrast, the increases in zonal winds in JJA and MAM lead to the increases of O<sub>3</sub> outflow fluxes throughout the troposphere over 30–45 °N. Our 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 during spring, which agrees with our simulation. Based on 31 (29)-model ensemble mean results, Jiang and Tian (2013) showed that the westerlies along 135 °E during winter (summer) were projected to weaken (strengthen). Such projected patterns of future changes in westerlies during winter and summer are also captured by our model. Changes in O<sub>3</sub> concentrations also contribute to the changes in O<sub>3</sub> outflow; although the zonal winds are projected to increase north of 40 °N in the upper troposphere during SON, the O<sub>3</sub> outflow fluxes are simulated to decrease because of the significant decreases of O<sub>3</sub> levels north of 40 °N in the upper troposphere (Fig. S1).

### 5.3 Effect of future changes in anthropogenic emissions alone

The annual outflow of O<sub>3</sub> through the vertical plane is simulated to increase by 7.9 % relative to the present-day value (Table 45) as a result of the changes in anthropogenic emissions alone (Met2000Emis2050 minus Met2000Emis2000). Considering that the O<sub>3</sub> outflow with changes in anthropogenic emissions alone exhibits an increasing trend of 1.2 % decade<sup>-1</sup> over 1986–2006 (Sect. 4.2), the increasing trend of 1.2 % decade<sup>-1</sup> (i.e., 6.0 % half-century<sup>-1</sup>) is close to the value of 7.9 % over the future half-century.

The projected future O<sub>3</sub> fluxes show increases during all seasons, which can be attributed to the increases in O<sub>3</sub> concentrations at all altitudes over Asia and upwind regions (i.e., Europe and Central Asia; Fig. S1) as a result of the increases in anthropogenic emissions of the O<sub>3</sub> precursors (NO<sub>x</sub> and NMVOCs) and CH<sub>4</sub> concentrations. NO<sub>x</sub> emissions in 2050 are projected to increase by 159.6% over Asia and by 78.4% globally, while NMVOCs emissions are projected to increase by 118.6% over Asia and by 89.4% globally under the SRES A1B scenario (Table 1). The

CH<sub>4</sub> mixing ratios are projected to increase by 37.1% relative to the present-day value. The largest increases of O<sub>3</sub> outflow fluxes are located in the middle-upper troposphere (Fig. 910c) owing to the strong westerlies located here. It is noted that, in spite of the significant increases of emissions, the simulated surface-layer O<sub>3</sub> concentrations show slight decreases over the North China Plain in DJF, which subsequently leads to the small decreases of O<sub>3</sub> outflow fluxes at the surface layer over 30–40° N. In DJF, biogenic VOC emissions are especially low over the North China Plain, whereas anthropogenic NO<sub>x</sub> emissions are fairly high due to the residential heating, leading to a low VOCs/NO<sub>x</sub> ratio in this region (Lou et al., 2010; Fu et al., 2012). Therefore, increases in NO<sub>x</sub> emissions lead to decreases in surface-layer O<sub>3</sub> concentrations over the North China Plain.

#### 5.4 Effect of future changes in both climate and anthropogenic emissions

The annual outflow of O<sub>3</sub> through the vertical plane is projected to increase by 12.2 % (Table 45) during 2000–2050 as a consequence of future changes in both climate and anthropogenic emissions (Met2050Emis2050 minus Met2000Emis2000). Climate change in DJF and SON slightly offsets the effects of changes in anthropogenic emissions, while climate change in MAM and JJA enhances the effects of variations in anthropogenic emissions. When considering future changes in both emissions and climate, the projected O<sub>3</sub> outflow fluxes show increases throughout almost the entire troposphere along 135° E during all seasons (Fig. 910d).

### 6 Uncertainty discussion

There are some uncertainties in our simulations. First, the influence of interannual variation in stratosphere-troposphere exchange on tropospheric O<sub>3</sub> is not considered in this study. Terao et al. (2008) reported that the stratosphere-troposphere exchange had large impacts on the interannual variability of tropospheric O<sub>3</sub> over Canada and Europe but the impact was much smaller over East Asia. The second is the uncertainty associated with the selection of longitudinal transect. We calculate O<sub>3</sub> flux through the vertical plane along 135° E, because 135° E is the easternmost boundary of China (i.e., Wusuli River in Northeastern China). We also calculate the O<sub>3</sub> outflow flux along 120° E, more close to ozone production region in central-eastern China, and find that the variations in O<sub>3</sub> 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 O<sub>3</sub> outflow shows large IAVs but a statistically insignificant ( $P > 0.05$ ) trend. The conclusion is consistent with that drawn from the variations in O<sub>3</sub> outflow calculated at 135° E. Finally, projecting future atmospheric circulation on regional scales has large uncertainty, which



is undergoing continuing improvement.

## **6.7 Conclusions**

We quantify the past and future changes in the O<sub>3</sub> outflow from East Asia using the global 3-D chemical transport model GEOS-Chem. The historical (1986–2006) simulations are driven by the assimilated GEOS-4 meteorological fields, and the future (2000–2050) simulations under the IPCC SRES A1B scenario are driven by the meteorological fields archived from GISS GCM 3. Sensitivity studies are conducted to examine the respective impacts of meteorological parameters and emissions on the variations in the outflow flux.

~~The TCO products retrieved from TOMS/SBUV are used to evaluate model performance in simulating tropospheric O<sub>3</sub>. Simulated spatiotemporal distributions of TCO over China and the downwind regions agree fairly well with TOMS/SBUV measurements, which gives us confidence to simulate the temporal evolutions of Asian O<sub>3</sub> outflow.~~

The measurements from WDCGG and EANET are used to evaluate the simulated surface-layer O<sub>3</sub> concentrations; the ozonesonde data from WOUDC are used to evaluate the simulated O<sub>3</sub> concentrations for the boundary layer, middle and upper troposphere. Generally, the seasonal cycles and interannual variations of tropospheric O<sub>3</sub> are captured fairly well by the GEOS-Chem model, although the model overestimates the low values during summer. The increasing trends in surface-layer O<sub>3</sub> in China over the past two decades can also be captured by GEOS-Chem model, although the modeled O<sub>3</sub> trends have low biases. Simulated Asian O<sub>3</sub> outflow flux peaks in early spring, and reaches the nadir in summer. The maximum O<sub>3</sub> fluxes are located in the middle-upper troposphere.

The IAVs and decadal trends of Asian O<sub>3</sub> outflow are examined over 1986–2006. Simulated O<sub>3</sub> outflow fluxes showed large IAVs, but an insignificant decadal trend; with variations in both meteorological parameters and anthropogenic emissions, the seasonal APDM values were in the range of 4–9 % ~~and the decadal trend was  $-2.2\%$  decade<sup>-1</sup> (statistically insignificant).~~ Sensitivity simulations showed that the large IAVs of the O<sub>3</sub> outflow fluxes were mainly caused by the variations in meteorological conditions, rather than the variations in anthropogenic and biomass burning emissions. Although variations in meteorological parameters could influence the IAVs of the O<sub>3</sub> outflow fluxes by changing O<sub>3</sub> concentrations over East Asia and by altering zonal winds, the latter was identified to be the key factor because of the high correlation coefficient of +0.71 between the annual fluxes and zonal winds.

The decadal changes in Asian O<sub>3</sub> outflow are also examined during 2000–2050. The present-day annual O<sub>3</sub> flux through the vertical plane is calculated as 1877.1 Tg, which is projected to change over 2000–2050 by +2.0 %, +7.9 %, and +12.2 %, respectively, due to climate change alone, emissions change alone, and changes in both climate and emissions. During MAM and JJA, climate change plays a larger role in the future changes in O<sub>3</sub> outflow compared

with emissions changes, owing to the significant increases in zonal winds during these two seasons. It is noted that climate change will aggravate the impacts of increases in anthropogenic emissions on the O<sub>3</sub> outflow from East Asia over 2000–2050 under the SRES A1B scenario.

These findings are helpful for understanding the temporal evolutions of tropospheric O<sub>3</sub> on different timescales in the downwind regions of East Asia. Observed IAVs of tropospheric O<sub>3</sub> on a relatively short timescale can be attributed to variations in meteorological parameters. Furthermore, conclusions from this study will have important implications for long-term air quality planning for the downwind regions of China, such as Japan and US. Since future climate change will increase O<sub>3</sub> outflow from East Asia, extra efforts are needed to reduce anthropogenic emissions of O<sub>3</sub> precursors to offset the adverse effects caused by climate change.

## **7.8 Data availability**

GEOS-Chem is an open-access model developed collaboratively at Harvard University and other institutes in North America, Europe, and Asia. The source codes can be downloaded from <http://acmg.seas.harvard.edu/geos/>. ~~The TCO (tropospheric column O<sub>3</sub>) data are retrieved from TOMS/SBUV; TOMS total O<sub>3</sub> measurements are available from <http://toms.gsfc.nasa.gov/>; and SBUV stratospheric O<sub>3</sub> measurements are available from <http://orbit.net.nesdis.noaa.gov/orad/sit/ozone>. The O<sub>3</sub> measurements at Minamitorishima and Yonagunijima are available from the World Data Centre for Greenhouse Gases (WDCGG, [www.ds.data.jma.go.jp/gmd/wdcgg/](http://www.ds.data.jma.go.jp/gmd/wdcgg/)), and those at Rishiri and Ogasawara are available from the Acid Deposition Monitoring Network in East Asia (EANET, [www.eanet.asia/product/index.html](http://www.eanet.asia/product/index.html)). The ozonesonde data at Naha and Tsukuba are available from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC, [www.woudc.org](http://www.woudc.org)).~~ All data presented in this study are available upon request from the corresponding author.

*Author contributions.* H. Liao and J. Zhu conceived the study and designed the experiments. J. Zhu performed the simulations, carried out the data analysis, and prepared the manuscript. Y. Mao provided useful comments on the paper. Y. Yang and H. Jiang helped with performing the experiments.

*Competing interests.* The authors declare that they have no conflicts of interest.

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**Table 1.** Anthropogenic emissions<sup>a</sup> of O<sub>3</sub> precursors for the present day (year 2000) and future (year 2050, under the SRES A1B scenario).

Species	Global			Asian <sup>b</sup>		
	2000	2050	Change (%)	2000	2050	Change (%)
NO <sub>x</sub> (Tg N yr <sup>-1</sup> )	33.4	59.6	+78.4	10.9	28.3	+159.6
CO (Tg CO yr <sup>-1</sup> )	1054.2	1332.0	+26.4	393.7	487.2	+23.7
NMVOCS (Tg C yr <sup>-1</sup> )	70.8	134.1	+89.4	28.5	62.3	+118.6
CH <sub>4</sub> (ppbv)	1750	2400	+37.1	1750	2400	+37.1

<sup>a</sup> Biomass burning emissions are included.

<sup>b</sup> Asia covers the domain of 60–150 °E, 10 °S–55 °N.

**Table 2.** Natural emissions of O<sub>3</sub> precursors for the present day (year 2000) and future (year 2050, under the SRES A1B scenario).

Species	Global			Asian <sup>a</sup>		
	2000	2050	Change (%)	2000	2050	Change (%)
Lightning NO <sub>x</sub> (Tg N yr <sup>-1</sup> )	4.8	5.7	+18.8	1.2	1.4	+16.7
Soil NO <sub>x</sub> (Tg N yr <sup>-1</sup> )	6.7	7.7	+14.9	1.4	1.7	+21.4
Biogenic VOCs (Tg C yr <sup>-1</sup> )	614.5	750.2	+22.1	106.1	126.2	+18.9

<sup>a</sup> Asia covers the domain of 60–150 °E, 10 °S–55 °N.

**Table 3.** Experimental design of the simulations for 1986–2006.

Simulation	Simulated years	Meteorological parameters	Anthropogenic emissions	CH <sub>4</sub> abundance	Biomass burning emissions
Met	1986–2006	Vary over 1986–2006	Fixed at 2006	Fixed at 2006	Turn off
Emis	1986–2006	Fixed at 2006	Vary over 1986–2006	Vary over 1986–2006	Turn off
MetEmis	1986–2006	Vary over 1986–2006	Vary over 1986–2006	Vary over 1986–2006	Turn off
MetEmisB <sup>a</sup>	1997–2006	Vary over 1997–2006	Vary over 1997–2006	Vary over 1997–2006	Vary over 1997–2006

<sup>a</sup> The MetEmisB simulation is conducted for 1997–2006 owing to the unavailability of biomass burning emissions before 1997.

**Table 4.** Information for the sites with O<sub>3</sub> measurements used in model evaluation.

Site	Location	Database	Height	R <sup>a</sup>	NMB <sup>b</sup> (%)
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
			500–300 hPa	0.55	+15.8
Tsukuba	36.1 N, 140.1 E	WOUDC	700–500 hPa	0.76	+12.3
			850–700 hPa	0.76	+8.61
			1000–850 hPa	0.60	+8.5

<sup>a</sup> Correlation coefficient (R) between the observed and simulated monthly O<sub>3</sub> mixing ratios.

<sup>b</sup> Normalized mean bias (NMB, %) between the observed and simulated monthly O<sub>3</sub> mixing ratios.

**Table 45.** Simulated present-day O<sub>3</sub> fluxes and projected changes from the present day (1996–2005) to the future (2046–2055) through the vertical plane along 135 °E from 20 °N to 55 °N, and from the surface to 100 hPa, due to future climate change alone, change in anthropogenic emissions alone, and changes in both climate and emissions.

	O <sub>3</sub> Fluxes <sup>a</sup>			
	Met2000Emis2000 (present day)	Met2050Emis2000 (climate)	Met2000Emis2050 (emissions)	Met2050Emis2050 (climate + emissions)
DJF	792.9	778.3 (−1.8%)	850.5 (+7.3%)	853.7 (+7.7%)
MAM	597.0	640.4 (+7.3%)	639.0 (+7.0%)	698.0 (+16.9%)
JJA	146.5	167.7 (+14.5%)	161.3 (+10.1%)	187.2 (+27.8%)
SON	340.7	327.8 (−3.8%)	374.1 (+9.8%)	368.1 (+8.0%)
Annual	1877.1	1914.1 (+2.0%)	2024.9 (+7.9%)	2106.9 (+12.2%)

<sup>a</sup> The units are Tg season<sup>−1</sup> for seasonal fluxes and Tg yr<sup>−1</sup> for annual fluxes. Values in parentheses are percentage changes relative to the present-day fluxes.

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## Figure captions

**Figure 1.** Evolution of annual anthropogenic and biomass burning emissions summed over the globe and Asia (60–150 ° E, 10 ° S–55 ° N) for NO<sub>x</sub> (Tg N yr<sup>-1</sup>), CO (Tg CO yr<sup>-1</sup>), and NMVOCs (Tg C yr<sup>-1</sup>) over 1986–2006. Blue squares represent anthropogenic emissions, and red circles represent the sum of anthropogenic emissions and biomass burning emissions. The last panel shows the evolution of global CH<sub>4</sub> abundance (ppbv) during 1986–2006.

**Figure 2.** Evolution of annual natural emissions summed over the globe and Asia (60–150 ° E, 10 ° S–55 ° N) for lightning NO<sub>x</sub> (Tg N yr<sup>-1</sup>), soil NO<sub>x</sub> (Tg N yr<sup>-1</sup>), and biogenic VOCs (Tg C yr<sup>-1</sup>) over 1986–2006.

**Figure 3.** Time series of monthly surface-layer O<sub>3</sub> 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<sub>3</sub> 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.

**Figure 5.** Comparison of simulated trends in annual-mean O<sub>3</sub> concentrations from the MetEmis experiment with observations for Beijing (location: 40.0 ° N, 116.5 ° E; years: 1995–2005; reference: Ding et al., 2008a), Hongkong (22.2 ° N, 114.3 ° E; 1994–2007; Wang et al., 2009b), Taiwan (23.5 ° N, 121.0 ° E; 1994–2007; Lin et al., 2010), Waliguan (36.3 ° N, 100.9 ° E; 1994–2013; Xu et al. 2016), and South Korea (37.3 ° N, 126.5 ° E; 1990–2010; Lee et al., 2014).

~~**Figure 3.** Comparison of tropospheric column ozone (TCO, units: DU) from the MetEmisB simulation with those retrieved from TOMS/SBUV. Both the model results and the TOMS/SBUV retrievals are averaged over 1997–2005 (excluding year 2001). Also shown in (a) is the location of the meridional plane through which the Asian O<sub>3</sub> outflow flux is calculated.~~

~~**Figure 4.** Monthly and yearly variations of tropospheric column ozone (TCO, units: DU) retrieved from TOMS/SBUV (blue line) and simulated by MetEmisB (red line). Both the retrieved and simulated TCO values are averaged over the selected region (30–50 ° N, 130–150 ° E) in the western Pacific Ocean.~~

**Figure 56.** The pressure–latitude cross-sections along 135 ° E of the simulated seasonal O<sub>3</sub> outflow fluxes and zonal winds during four seasons averaged over 1997–2006 in the MetEmisB simulation. The O<sub>3</sub> mass fluxes are shown by shades (units: kg season<sup>-1</sup> m<sup>-2</sup>), and winds are represented by contours (units: m s<sup>-1</sup>). Positive fluxes represent eastward fluxes, and negative values represent westward fluxes.

**Figure 67.** Evolution of (a) annual O<sub>3</sub> outflow fluxes (Tg yr<sup>-1</sup>) across the meridional plane along 135 ° E from 20 ° N to 55 ° N, and from the surface to 100 hPa, over 1986–2006 in the Met, Emis, and MetEmis simulations; (b) the associated deviations from the mean (%); and (c) deviations from the mean (%) of the detrended O<sub>3</sub> outflow fluxes. The deviation from the mean (DEV) is defined in Sect. 4.2.

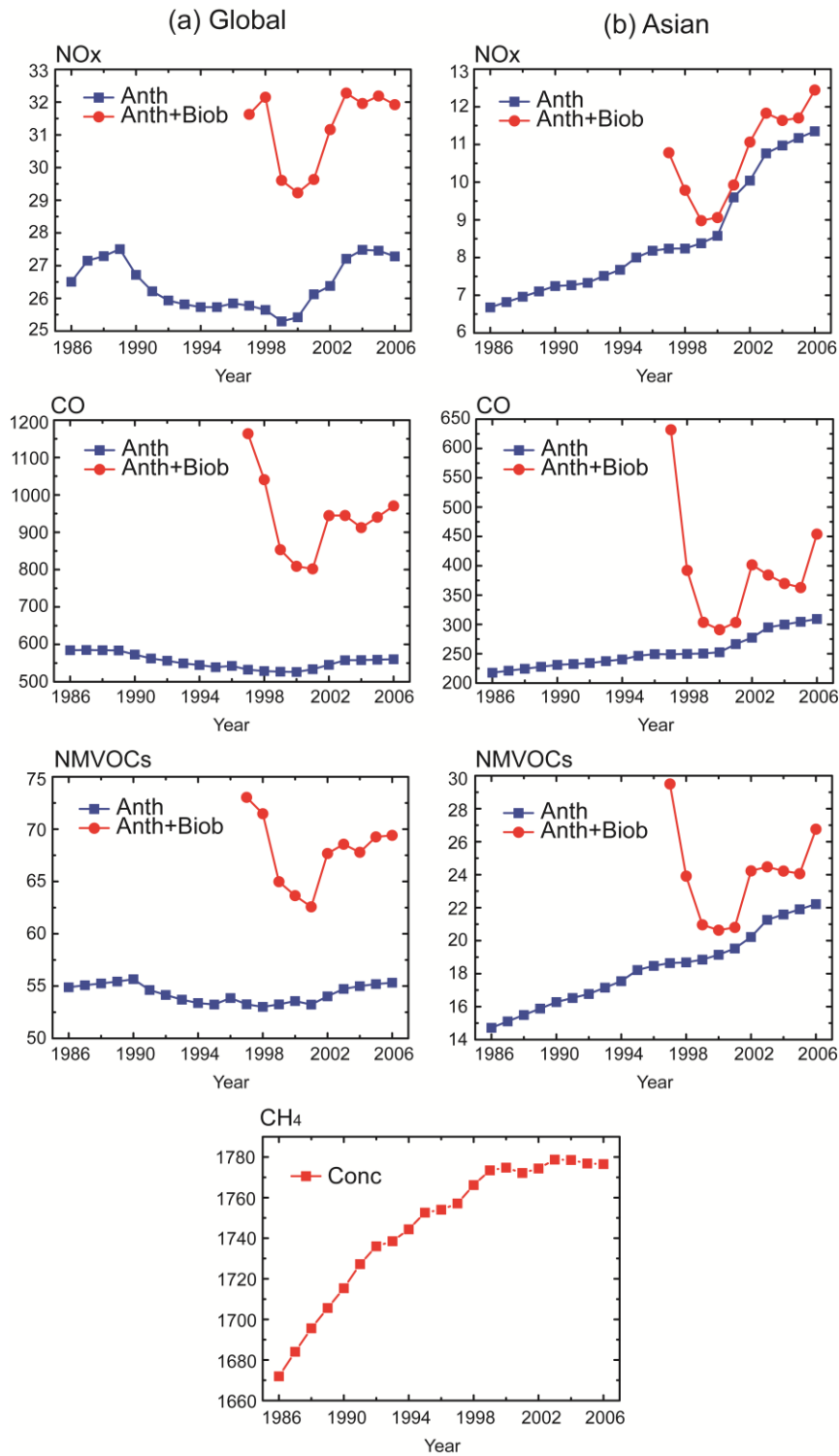
**Figure 78.** The MAD and APDM values of the detrended seasonal and annual O<sub>3</sub> outflow fluxes across the meridional plane along 135 ° E from 20 ° N to 55 ° N, and from the surface to 100 hPa, over 1986–2006 in Met, Emis, and MetEmis simulations. Both the MAD and APDM are defined in Sect. 4.2. The units of MAD are Tg season<sup>-1</sup> for seasonal fluxes

and  $\text{Tg yr}^{-1}$  for annual fluxes.

**Figure 89.** The pressure–latitude cross-sections along  $135^\circ\text{E}$  of MAD values for detrended annual  $\text{O}_3$  outflow fluxes and zonal winds over 1986–2006 in the Met, Emis, and MetEmis simulations. The MAD values for  $\text{O}_3$  mass fluxes are shown by shades (units:  $\text{kg yr}^{-1} \text{m}^{-2}$ ), and the MAD values for winds are represented by contours (units:  $\text{m s}^{-1}$ ).

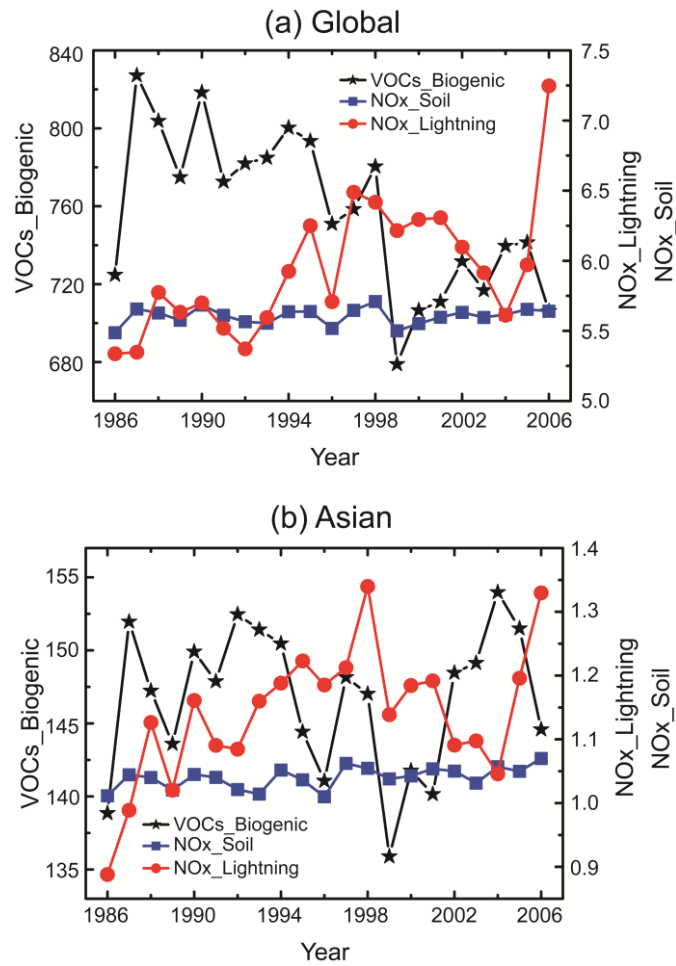
5 **Figure 910.** (a) The pressure–latitude cross-sections along  $135^\circ\text{E}$  of simulated present-day  $\text{O}_3$  mass fluxes and zonal winds (Met2000Emis2000). Projected changes in  $\text{O}_3$  mass fluxes and zonal winds from the present day (1996–2005) to the future (2046–2055) caused by (b) climate change alone (Met2050Emis2000 minus Met2000Emis2000); (c) changes in anthropogenic emissions alone (Met2000Emis2050 minus Met2000Emis2000); and (d) changes in both climate and anthropogenic emissions (Met2050Emis2050 minus Met2000Emis2000). The  $\text{O}_3$  mass fluxes are shown  
10 by shades (units:  $\text{kg season}^{-1} \text{m}^{-2}$ ), and winds are represented by contours (units:  $\text{m s}^{-1}$ ). The dotted areas are statistically significant at the 95 % level, as determined by a two-sample Student's  $t$ -test.



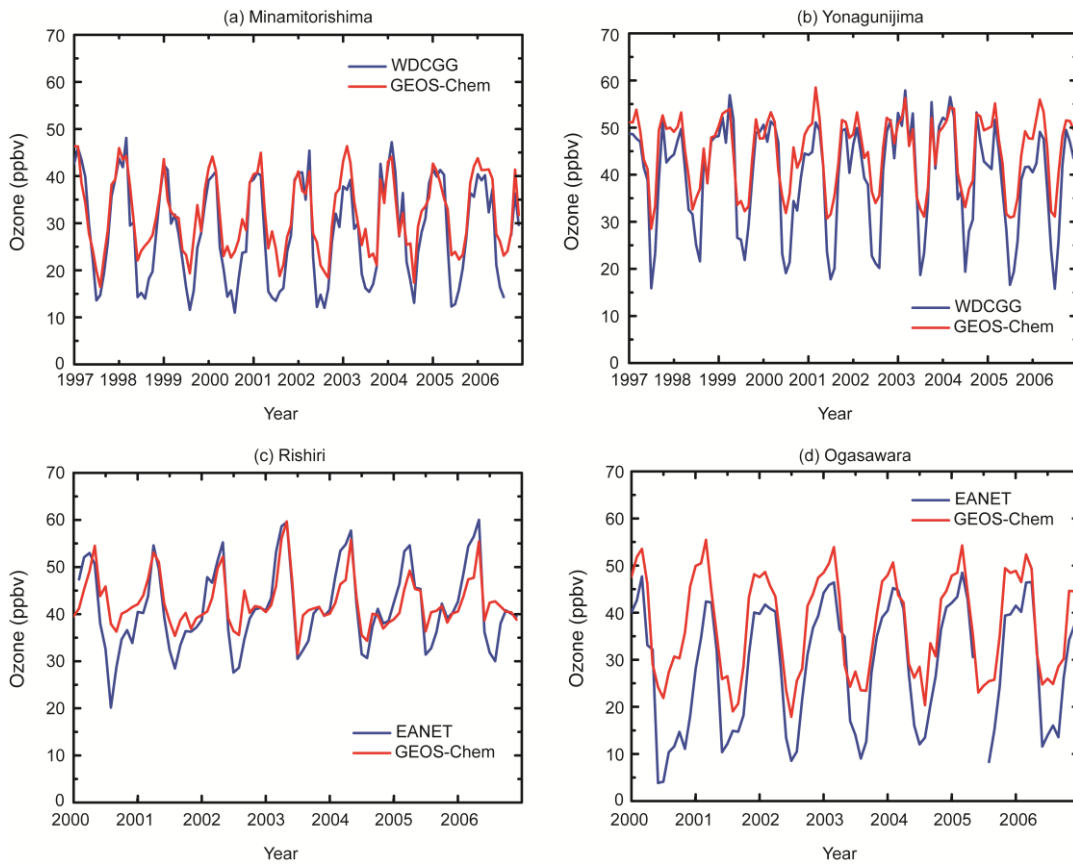


**Figure 1.** Evolution of annual anthropogenic and biomass burning emissions summed over the globe and Asia (60–150 ° E, 10 ° S–55 ° N) for NO<sub>x</sub> (Tg N yr<sup>-1</sup>), CO (Tg CO yr<sup>-1</sup>), and NMVOCs (Tg C yr<sup>-1</sup>) over 1986–2006. Blue squares represent anthropogenic emissions, and red circles represent the sum of anthropogenic emissions and biomass burning emissions. The last panel shows the evolution of global CH<sub>4</sub> abundance (ppbv) during 1986–2006.

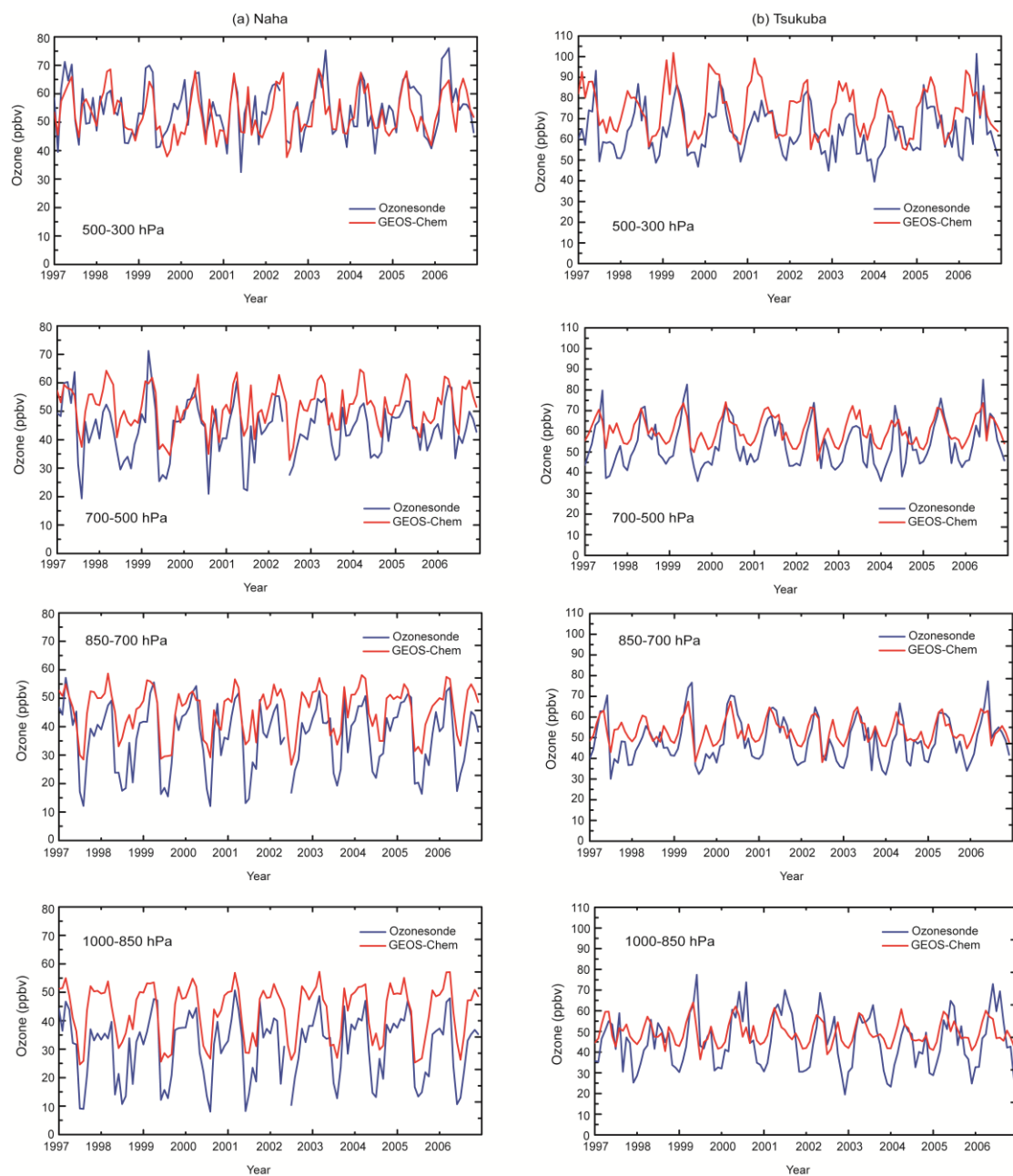
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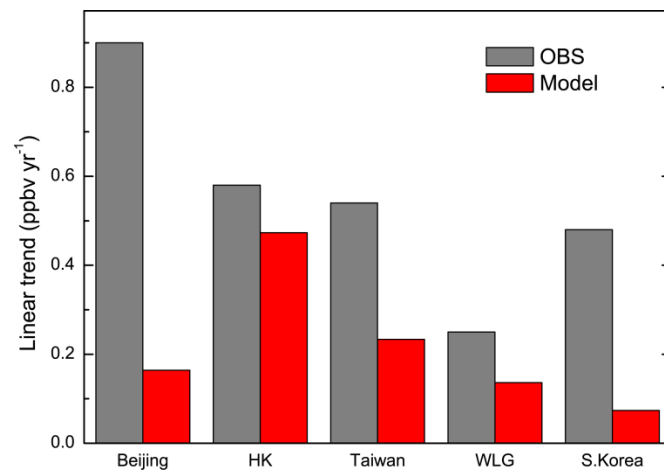
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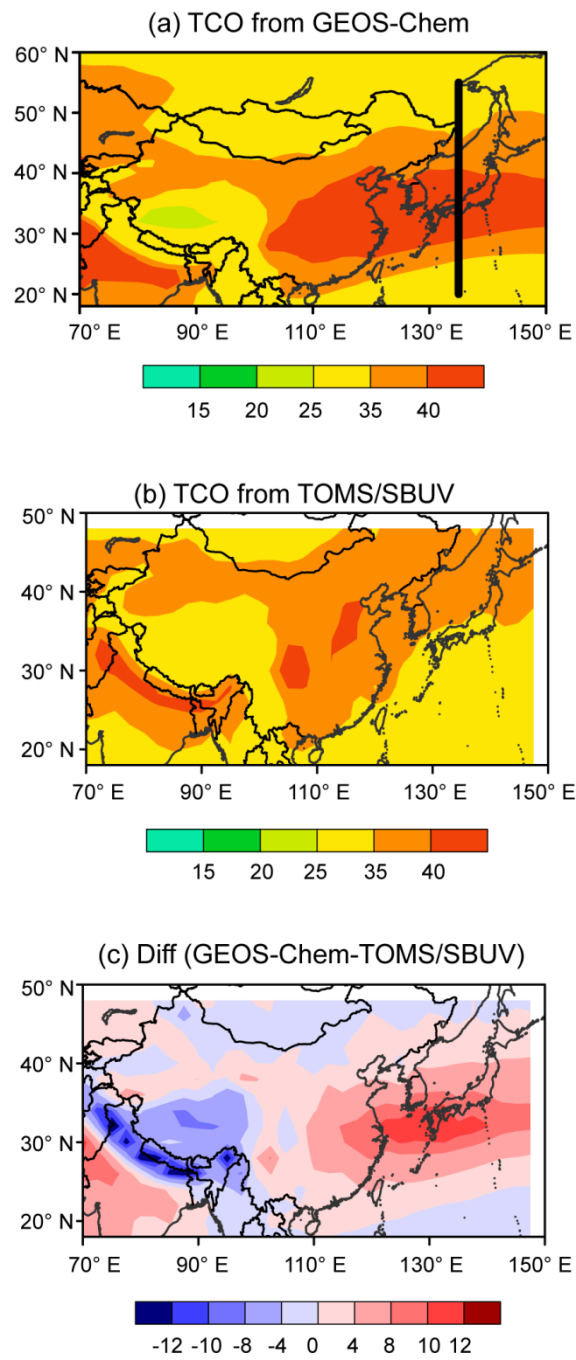


**Figure 4.** Time series of monthly  $O_3$  mixing ratios measured by ozonesonde (blue line), and simulated by MetEmissB (red line). (a) Naha and (b) Tsukuba are ozonesonde sites from WOUDC. Comparisons are shown for four altitude levels in the troposphere.



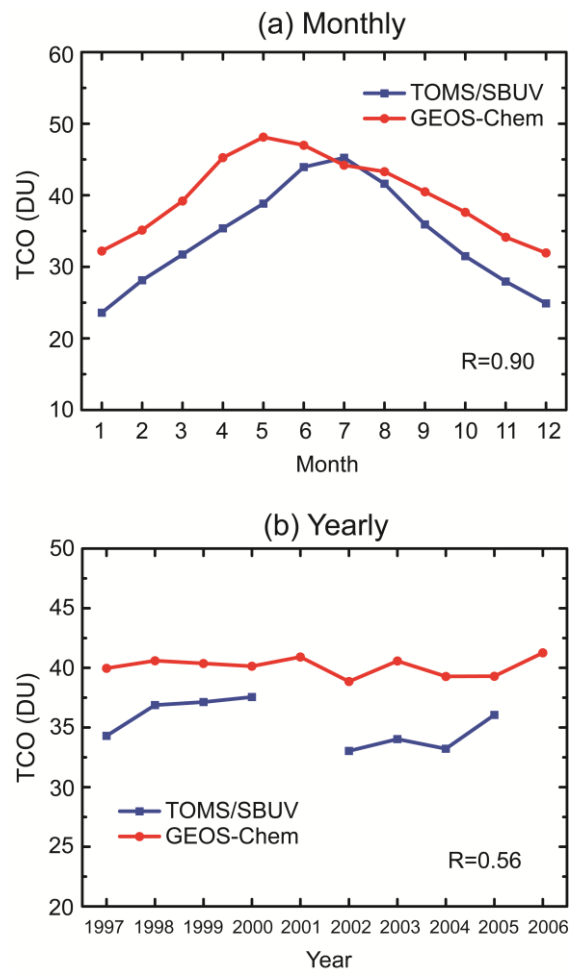
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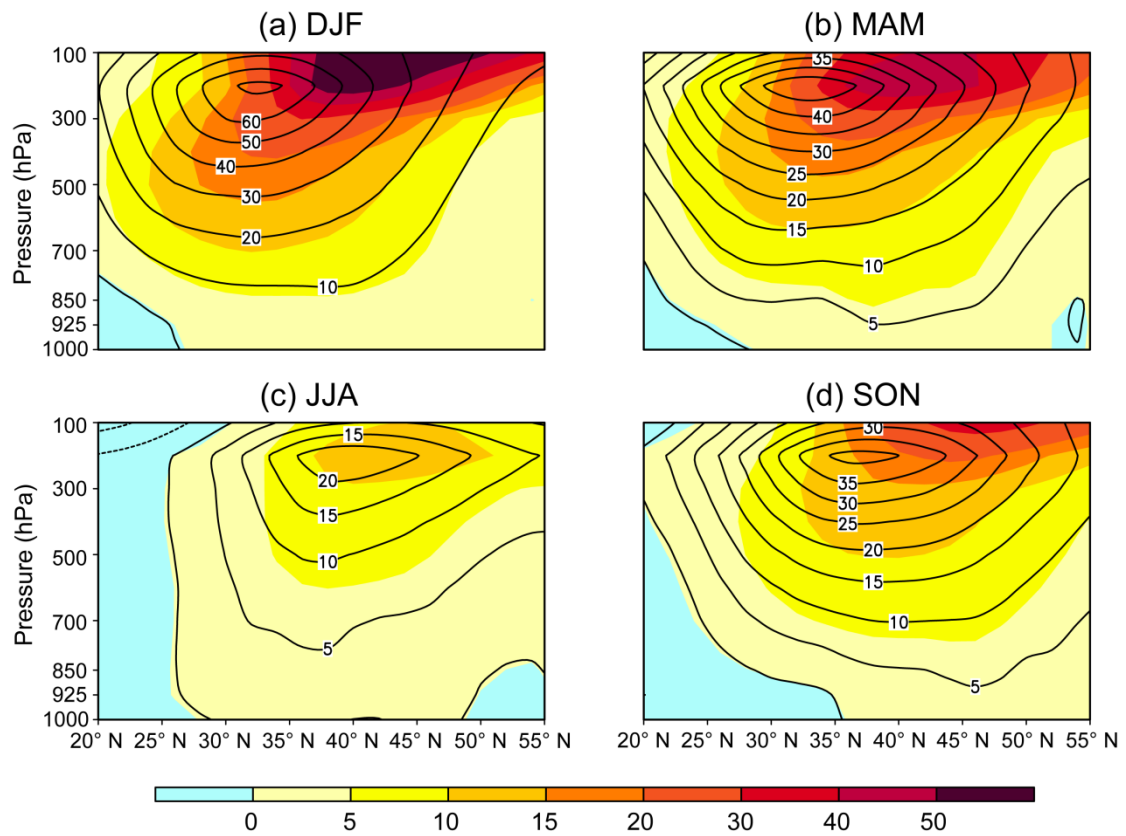


**Figure 3.** Comparison of tropospheric column ozone (TCO, units: DU) from the MetEmisB simulation with those retrieved from TOMS/SBUV. Both the model results and the TOMS/SBUV retrievals are averaged over 1997–2005 (excluding year 2001). Also shown in (a) is the location of the meridional plane through which the Asian O<sub>3</sub> outflow flux is calculated.

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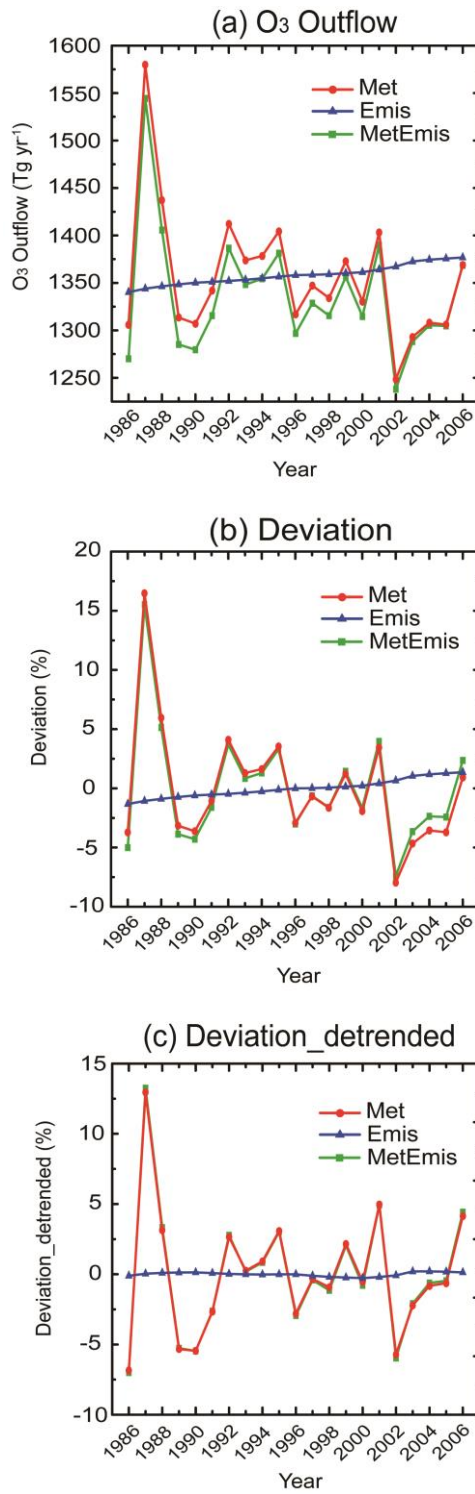
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**Figure 56.** The pressure–latitude cross-sections along 135° E of the simulated seasonal O<sub>3</sub> outflow fluxes and zonal winds during four seasons averaged over 1997–2006 in the MetEmisB simulation. The O<sub>3</sub> mass fluxes are shown by shades (units: kg season<sup>-1</sup> m<sup>-2</sup>), and winds are represented by contours (units: m s<sup>-1</sup>). Positive fluxes represent eastward fluxes, and negative values represent westward fluxes.

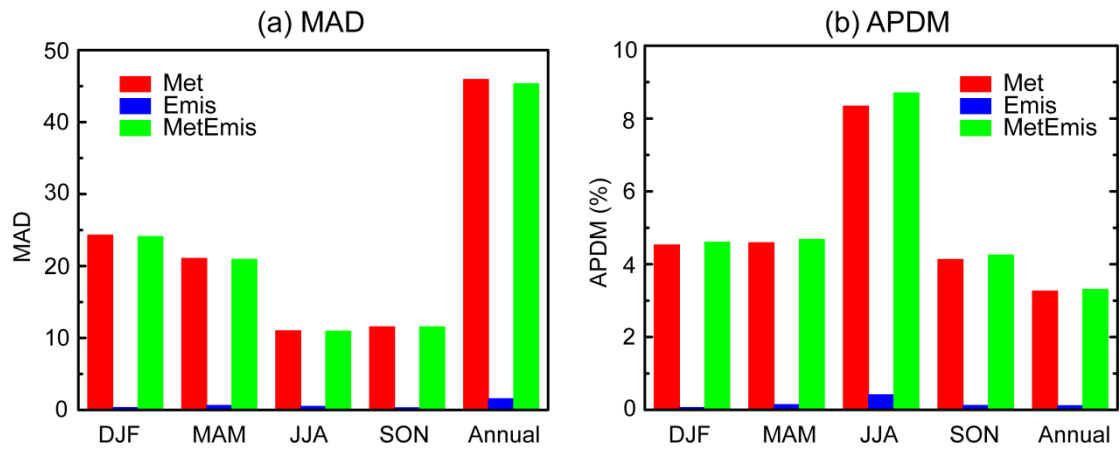
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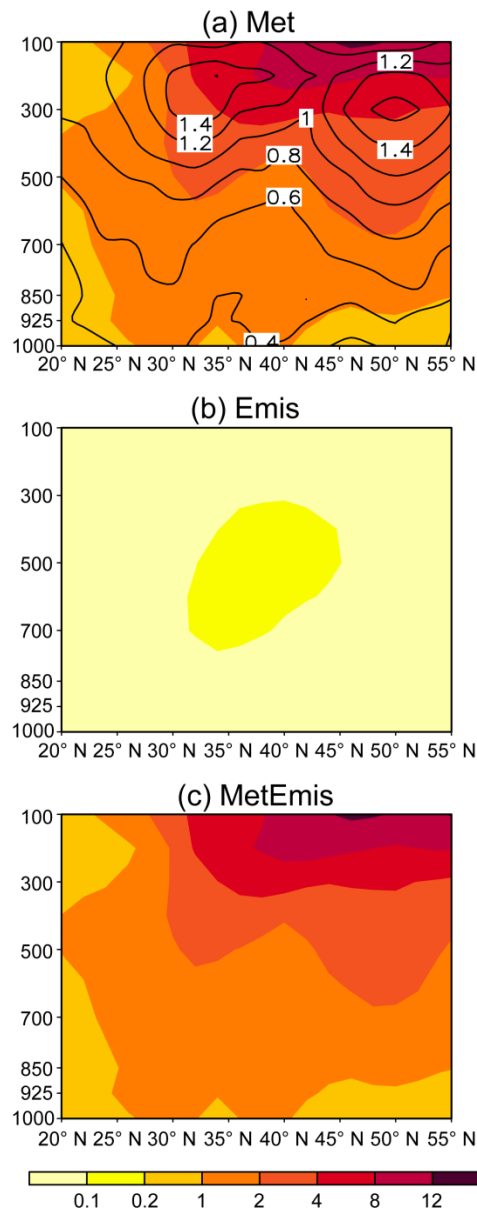
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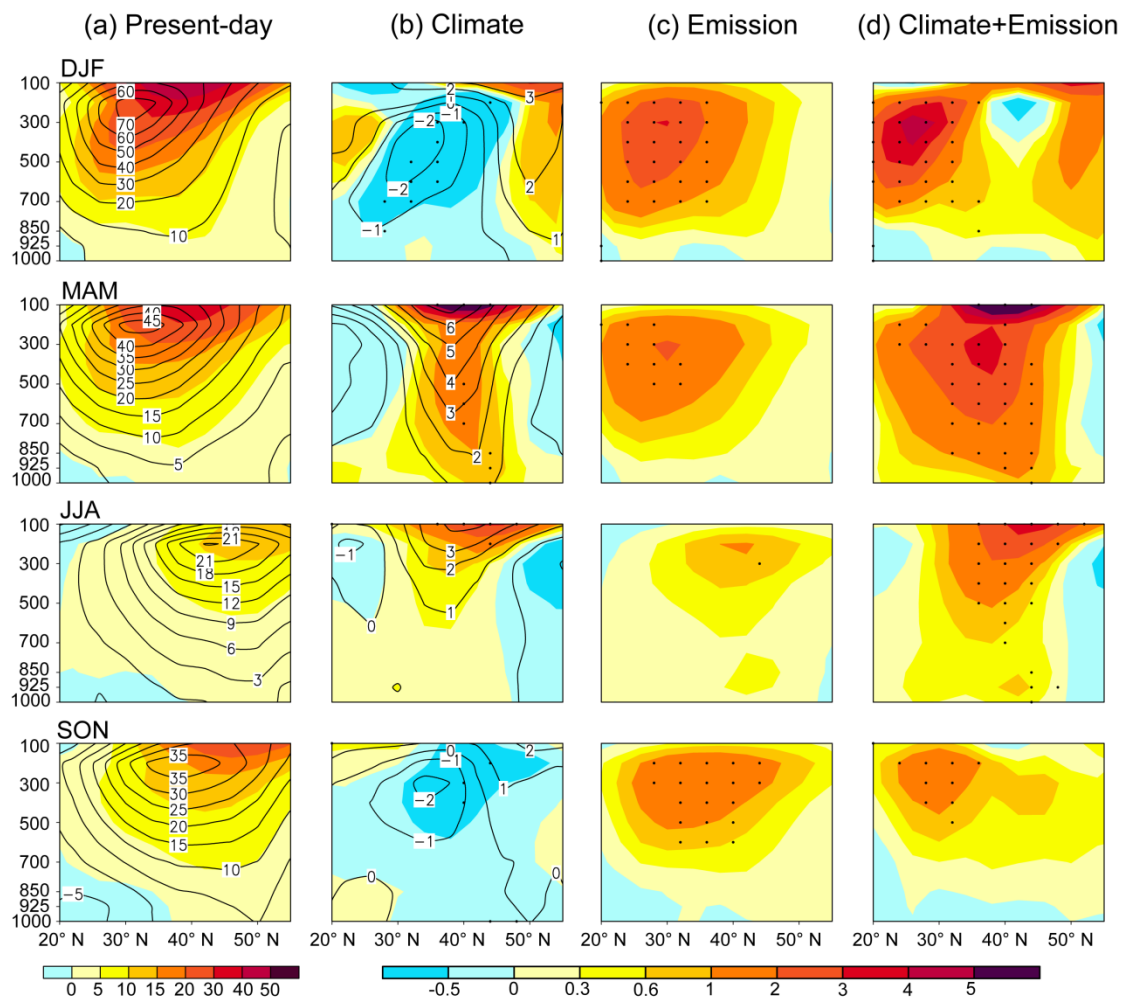


**Figure 78.** The MAD and APDM values of the detrended seasonal and annual O<sub>3</sub> outflow fluxes across the meridional plane along 135 °E from 20 °N to 55 °N, and from the surface to 100 hPa, over 1986–2006 in Met, Emis, and MetEmis simulations. Both the MAD and APDM are defined in Sect. 4.2. The units of MAD are Tg season<sup>-1</sup> for seasonal fluxes and Tg yr<sup>-1</sup> for annual fluxes.

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**Figure 89.** The pressure–latitude cross-sections along 135° E of MAD values for detrended annual O<sub>3</sub> outflow fluxes and zonal winds over 1986–2006 in the Met, Emis, and MetEmis simulations. The MAD values for O<sub>3</sub> mass fluxes are shown by shades (units: kg yr<sup>-1</sup> m<sup>-2</sup>), and the MAD values for winds are represented by contours (units: m s<sup>-1</sup>).



**Figure 910.** (a) The pressure–latitude cross-sections along 135 °E of simulated present-day O<sub>3</sub> mass fluxes and zonal winds (Met2000Emis2000). Projected changes in O<sub>3</sub> mass fluxes and zonal winds from the present day (1996–2005) to the future (2046–2055) caused by (b) climate change alone (Met2050Emis2000 minus Met2000Emis2000); (c) changes in anthropogenic emissions alone (Met2000Emis2050 minus Met2000Emis2000); and (d) changes in both climate and anthropogenic emissions (Met2050Emis2050 minus Met2000Emis2000). The O<sub>3</sub> mass fluxes are shown by shades (units: kg season<sup>-1</sup> m<sup>-2</sup>), and winds are represented by contours (units: m s<sup>-1</sup>). The dotted areas are statistically significant at the 95 % level, as determined by a two-sample Student’s *t*-test.