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Key Points:

- Tropospheric ozone burden increased from 1980 to 2010, driven mainly by increases in emissions from Southeast Asia, East Asia, and South Asia, as well as global methane concentration increases
- Among regions, the greatest ozone burden influence came from Southeast Asia despite smaller emission increases, highlighting the much greater sensitivity for this region

Supporting Information:

- supporting Information S1

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Contributions of World Regions to the Global Tropospheric Ozone Burden Change From 1980 to 2010

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Abstract We investigate the contributions of emission changes from 10 world regions, as well as the global methane concentration change, on the global tropospheric ozone burden change from 1980 to 2010. The modeled global tropospheric ozone burden has increased by 28.1 Tg, with 26.7% (7.5 Tg) of this change attributed to the global methane increase. Southeast Asia (5.6 Tg) and South Asia (4.0) contribute comparably to the global ozone burden change as East Asia (5.6), even though NO_x emission increases in each region are less than one-third of those in East Asia, highlighting the greater sensitivity of global ozone to these regions. Emission decreases from North America, Europe, and Former Soviet Union have led to ozone burden decreases of 2.8, 1.0, and 0.3 Tg. The greater sensitivity of the global ozone burden to emission changes in tropical and subtropical regions emphasizes the importance of controlling emissions in these regions for global ozone.

Plain Language Summary The global tropospheric ozone burden is highly sensitive to emission changes in tropical and subtropical regions, due to high temperature, strong sunlight, and convection which are favorable for ozone production and accumulation. Through model sensitivity simulations, we show that emission increases in Southeast Asia, South Asia, and East Asia contribute over half of the global tropospheric ozone burden increase from 1980 to 2010. Southeast Asia and South Asia contribute about as much to the ozone increase as East Asia, even though emission increases were much smaller from these regions, showing the high ozone sensitivity in these regions.

1. Introduction

Ozone (O_3) at the surface is detrimental to human health, crop yields, and ecosystems (Cooper et al., 2014; Fowler et al., 2009; Mills et al., 2018; Monks et al., 2015; Silva et al., 2013; Zhang et al., 2018). Tropospheric ozone is recognized as the third most important contributor to the positive radiative forcing, based on its increases since 1750, following carbon dioxide (CO_2) and methane (CH_4; Myhre et al., 2013). Ozone is a secondary air pollutant, which is not emitted directly, but is produced through chemical reactions of precursor gases in the atmosphere, such as nitrogen oxides (NO_x), carbon monoxide (CO), methane, and non-methane volatile organic compounds (NMVOCs). Ozone precursors are mainly emitted by human activities, such as fossil fuel combustion, residential burning, oil and gas production, agriculture, and biomass burning. Observations from aircraft, ozonesondes, and different satellites show that the tropospheric ozone burden has been increasing in the second half of the 20th century (Gaudel et al., 2018; Hemispheric Transport of Air Pollution [HTAP], 2010). Both satellite ozone measurements and global chemical transport models have found that the largest ozone burden increases—about +6 to +7 Dobson units (i.e., ~15%–20% of average background ozone) from 1980 to 2016—are over India, Southeast Asia, and East Asia (Ziemke et al., 2019).

Previous studies have demonstrated that methane emissions affect global ozone with little dependence on the location of emissions (Fiore et al., 2008). For short-lived ozone precursors, the global tropospheric ozone burden (B_{O3}) responds differently to emission changes from different world regions, with generally

much greater sensitivity to emissions in tropical and subtropical regions (Fry et al., 2012, 2013, 2014; Naik et al., 2005; West et al., 2009a). Since about 1980, global anthropogenic emissions of ozone precursors have been shifting toward the equator, particularly decreasing in North America and Europe, and increasing in East and South Asia (Duncan et al., 2016; Granier et al., 2011; Lamarque et al., 2010; Richter et al., 2005; Xing et al., 2013). In our previous study (Zhang et al., 2016), we investigated for the first time the influences of changes in the spatial distribution of global anthropogenic emissions of short-lived ozone precursors, the magnitude of these emissions, and the global atmospheric methane concentration on the global B_{O3} change from 1980 to 2010. We found that the spatial distribution change of emissions is most important for the increase in B_{O3}, slightly exceeding the combined influences of the increased emission magnitude and global methane (Zhang et al., 2016). We also found that B_{O3} has increased most strongly over Southeast, East, and South Asia, a conclusion that was supported by satellite and ozonesonde observations. Based on previous studies that found a much greater sensitivity of B_{O3} to emissions in tropical and subtropical regions and especially Southeast Asia, we hypothesized that emission increases from these regions were particularly important for the global B_{O3} increase, because of the strong sunlight, high temperature, and strong convection (Gupta et al., 1998; Lawrence et al., 2003; West et al., 2009a). However, the effects of emission changes over recent decades from individual world regions on the global B_{O3} has not been previously quantified.

Here we build on our previous study (Zhang et al., 2016) by investigating how emission changes from different world regions, as well as the global methane concentration changes, have contributed to global B_{O3} changes (ΔB_{O3}) from 1980 to 2010. We are particularly interested in quantifying the contributions of emissions from tropical and subtropical regions including Southeast Asia, South Asia, and Central and South America, as models and observations now suggest that tropospheric ozone is increasing fastest over these regions (Gaudel et al., 2018, 2020). We also calculate B_{O3} changes from multimodel experiments from the second phase of the Task Force on Hemispheric Transport of Air Pollutants (HTAP2; Galmarini et al., 2017), which have not been reported previously, to investigate the sensitivity of B_{O3} to emissions from different world regions.

2. Methods

The global chemistry-climate model CAM-chem is used in this study, which is based on the global Community Atmosphere Model (CAM) version 4, the atmospheric component of the Community Earth System Model (v1.2.2; Lamarque et al., 2012; Tilmes et al., 2015, 2016). Model simulations are constructed to be consistent with those in our previous study (Zhang et al., 2016). The model uses a horizontal grid with a resolution of 2.5° × 1.9° (longitude × latitude), and 56 vertical levels between the surface and 4 hPa (≈40 km) with a time step of 1800 s. The NASA Global Modeling and Assimilation Office GEOS-5 meteorology from 2008 to 2012 is used to drive the model as a chemical transport model, such that meteorological inputs for all simulations are identical. For all simulations, the first year is spin-up and results are presented as 4-year averages. By using consistent meteorology among all the simulations, we focus on the effects of changes in anthropogenic emissions on B_{O3}, and ignore other influences, such as possible influences of climate change. Our previous study showed that the interannual variability in ozone burden due to meteorological changes over the 4 years modeled is small compared with the overall ozone burden change (Table S2; Zhang et al., 2016). Monthly mean distributions of chemically active stratospheric species (such as O_3, NO, NO_2, and N_2O_5) are prescribed using the climatology from the Whole Atmospheric Community Climate Model simulations (Garcia et al., 2007; Lamarque et al., 2012). Global anthropogenic emissions of all short-lived species including ozone precursors, aerosols, and aerosol precursors, from all anthropogenic sectors including biomass burning, ships and aircraft, are from ACCMIP for 1980 (Lamarque et al., 2010) and RCP8.5 for 2010 (Riahi et al., 2011), which are compatible with one another. Monthly temporal variations for the anthropogenic air pollutant emissions are added by using monthly emission factors from RETRO (Schultz et al., 2008) and the NMVOCs are respeciated into CAM-chem chemical species following previous methods (Fry et al., 2014; Silva et al., 2016). All natural emissions, such as biogenic, lightning NO_x, volcano, soil NO_x, and ocean emissions used the same configuration as in our previous study (Lamarque et al., 2012; Zhang et al., 2016), and are constant across all simulations. Natural biogenic and lightning NO_x emissions are calculated online as functions of meteorology and other factors, and therefore represent emissions from 2009 to 2012 (MEGAN2.1 for biogenic, Guenther et al., 2012, and Price parameterization for lightning NO_x,

Price & Rind, 1992; Price et al., 1997). Other natural emissions (volcano, ocean, and soil) were created as an average over recent decades, not considering yearly variability, following Lamarque et al. (2010).

We use three base simulations from our previous study (Zhang et al., 2016), the first two of which have global anthropogenic emissions and methane concentrations for 1980 (S_1980) and 2010 (S_2010), and a third in which CH_4 concentration is set to the 1980 level and all other parameters stay the same as S_2010 (named S_CH_4). The global CH_4 concentration is set globally uniform to 1,798 ppbv in 2010 (S_2010), and 1,567 ppbv in 1980 (S_1980 and S_CH_4; see Table 1 in Zhang et al., 2016). In this study, we conduct another 10 sensitivity simulations; for each of these, we replace the anthropogenic emissions of all air pollutants in 2010 with their emissions in 1980, for 10 world regions individually, holding all other regions and the global CH_4 concentration at the 2010 levels (Table S1). The differences between S_2010 and the 10 sensitivity runs (S_2010—sensitivity) are the ΔB_{O3} from that region's emission changes from 1980 to 2010. B_{O3} is defined as the total ozone mass below the chemical tropopause of annual average of 150 ppbv ozone for each grid cell in the S_2010 simulation, with the same tropopause applied to all other simulations. Large uncertainties exist for the global B_{O3} calculations due to different definitions of the tropopause (Gaudel et al., 2018; Griffiths et al., 2020; Young et al., 2018), and the calculated B_{O3} in 2010 in our study is well within the range derived from multimodel mean and multi-satellite products (see the model evaluation section).

The 10 world regions follow the definitions introduced by HTAP2, except that we reduce the 13 land regions from HTAP2 to 10 regions here, grouping Northern Africa and Sub-Saharan together as a new region Africa (AFR), grouping Mexico and Central America and South America to give Central South America (CSA), and grouping Russia, and Belarus, Ukraine and Central Asia to give the Former Soviet Union (FSU). The other seven regions include North America (NAM), Europe (EUR), South Asia (SAS), East Asia (EAS), South East Asia (SEA), Pacific, Australia and New Zealand (PAN), and the Middle East (MDE; Figure S1). Since in HTAP2 definitions the region classification number for each grid cell is defined by the largest area fraction contributed by individual regions (Janssens-Maenhout et al., 2015), we found that when these region definitions are applied to ACCMIP and RCP8.5 emissions, some coastal cells with emissions were treated as ocean. To ensure we account for the relevant emissions from each region, we extend the 10 land mass regions outward into the oceans by two grid cells at 0.5° × 0.5° horizontal resolution. By doing this, the inland region emissions increase by 2%–30% depending on the region and pollutant, compared with the case when we do not include the two extra cells (Tables S2–S4).

To evaluate model performance in simulating the surface, vertical and long-term ozone trends from 1980 to 2010, we thoroughly compared the model results in S_1980 and S_2010 with long-term surface observations, ozonesonde, aircraft, and satellites in our previous work (Zhang et al., 2016). Compared with surface ozone observations, S_2010 overestimates ozone by 5.8 ppbv averaged over all stations in the United States (average from 2009 to 2012 from the US CASTNET network), and 0.7 ppbv over Europe (average from 2009 to 2011 from the EMEP network), but captures the seasonal cycles very well. Our model also captures very well the vertical distribution of ozone from ozonesondes, although it is biased high between 30°S and 30°N, particularly in the upper troposphere. The B_{O3} in 2010 (342.7 ± 4.5 Tg/yr) simulated by CAM-Chem is in the range of multimodel simulations (ACCENT: 336 ± 27 Tg; ACCMIP: 337 ± 23 Tg; TOAR: 340 ± 34 Tg, and CMIP6: 348 ± 15 Tg; Griffiths et al., 2020; Young et al., 2013, 2018), and is also comparable with satellite observations (Ziemke et al., 2011, 2019). The estimated net increase of B_{O3} of about 28 Tg from 1980 to 2010 is also consistent with OMI/MLS satellite retrievals between October 2004 and December 2016, which indicate a 21.8 Tg increase in tropospheric ozone over 60°S-60°N (Blunden & Arndt, 2017). Zhang et al. (2016) evaluated the modeled 1980–2010 ozone trend at six rural or remote sites globally, showing that the model captures the trend well at five these sites (Figure S19 in Zhang et al., 2016). We also evaluated the modeled ozone trends by comparing the In-Service Aircraft for a Global Observing System (IAGOS) commercial aircraft and SHADOZ ozonesondes in South, East, and Southeast Asia, and found that the ozone changes from 1994–2004 to 2005–2014 from these observations are similar to the modeled 1980 to 2010 changes (Figures S13–S15 and Table 4 in Zhang et al., 2016). We update this evaluation by comparing with updated IAGOS trends from more than 34,600 measured ozone profiles between 1994 and 2016, which showed statistically significant increases in observed tropospheric ozone above 11 regions of the Northern Hemisphere (NH) since the mid-1990s (Gaudel et al., 2020). The model captures well the ozone increases over these 11 regions and the latitudinal gradient of these increases (maximum increases near the equator), though the

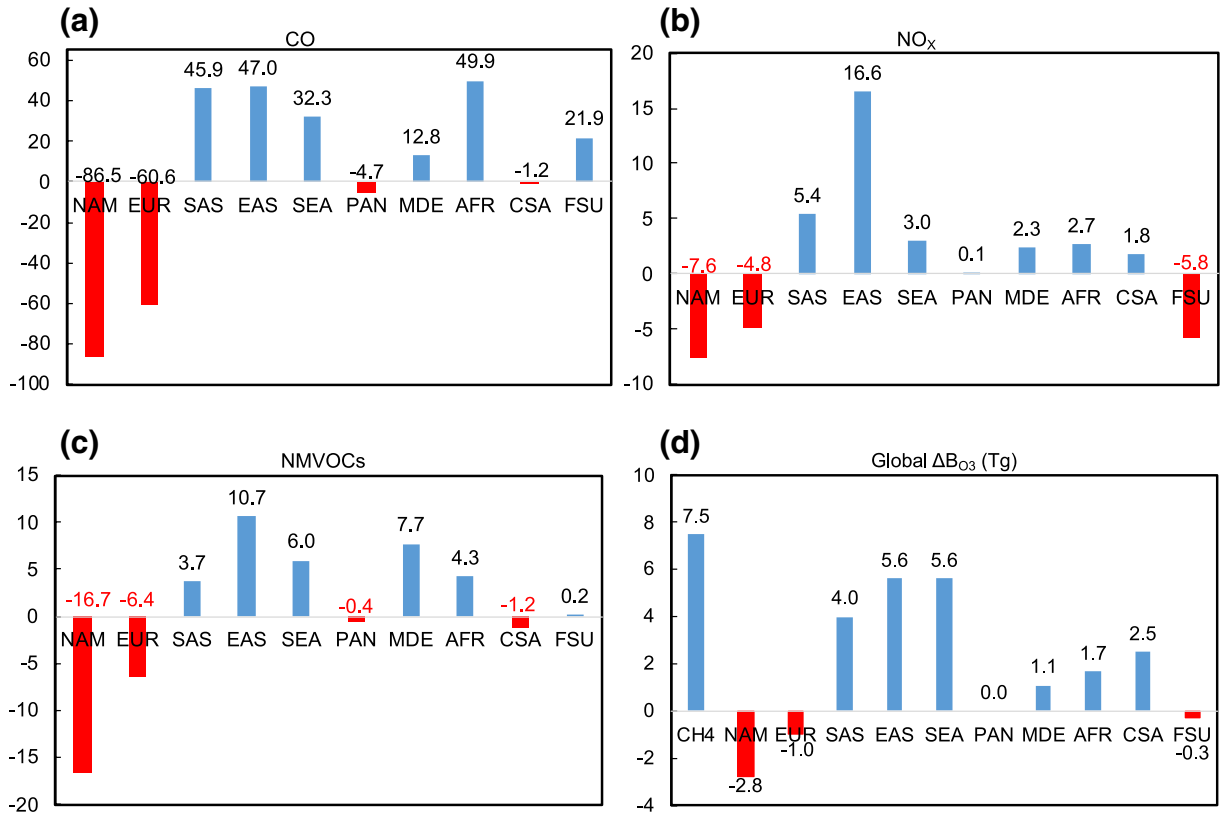

Figure 1. Emission changes from 1980 to 2010 for CO (a, Tg CO), NO_x (b, Tg NO_2), and NMVOCs (c, Tg NMVOCs), and global tropospheric ozone burden changes (d, Tg O_3) from global methane increases as well as emission changes from the 10 world regions. NMVOC, non-methane volatile organic compound.

model underestimates the increasing trends in the lower troposphere, particularly over India, Malaysia, North China and Korea, and Southeast Asia. We also evaluated the ozone burden trend simulated in our model with ozone burden changes from 1979 to 2016 observed from the composite record of using TOMS/OMI/MLS/OMPS satellite measurements, and our model captures the spatial distribution and magnitude changes very well between 25°S and 25°N (Figure 7a in Ziemke et al., 2019, and Figure S6).

3. Results

3.1. Regional Emission Changes

From 1980 to 2010, EAS had by far the largest NO_x emissions increase (16.6 Tg), triple the emissions in 1980, mainly from industry and transportation, as well as the largest increase in VOCs emissions (Figure 1 for absolute changes in Tg; Figures S2 and S3 for relative changes in %). The largest increases in CO emissions occurred in AFR (49.9 Tg, 22% higher than 1980; Figure S2), EAS (47.0 Tg, 40%), and SAS (45.9 Tg, 70%), due to residential biomass burning and industrial emissions (Hoesly et al., 2018). SAS and SEA, with more than 50% of the increases from biomass burning, see Figure S4) also have large emission increases for these pollutants. NAM and EUR had the largest emission decreases of 62% and 69% for CO, 36%, 32% for NO_x, and 67%, 47% for NMVOCs (Figure S2). The overall decline of CO emissions resulted from motor vehicle emission controls (Granier et al., 2011; Hoesly et al., 2018), while the NO_x decreases were from the implementation of emission control devices on thermal power plants, the shuttering of inefficient plants, and stricter vehicle emission standards in these regions (Duncan et al., 2016; Lamsal et al., 2015). NO_x emissions in FSU have also decreased by 43%, but CO increased by 52% (Figure S2), largely from residential emissions (Hoesly et al., 2018; Popovicheva et al., 2014; also see Figure S4).

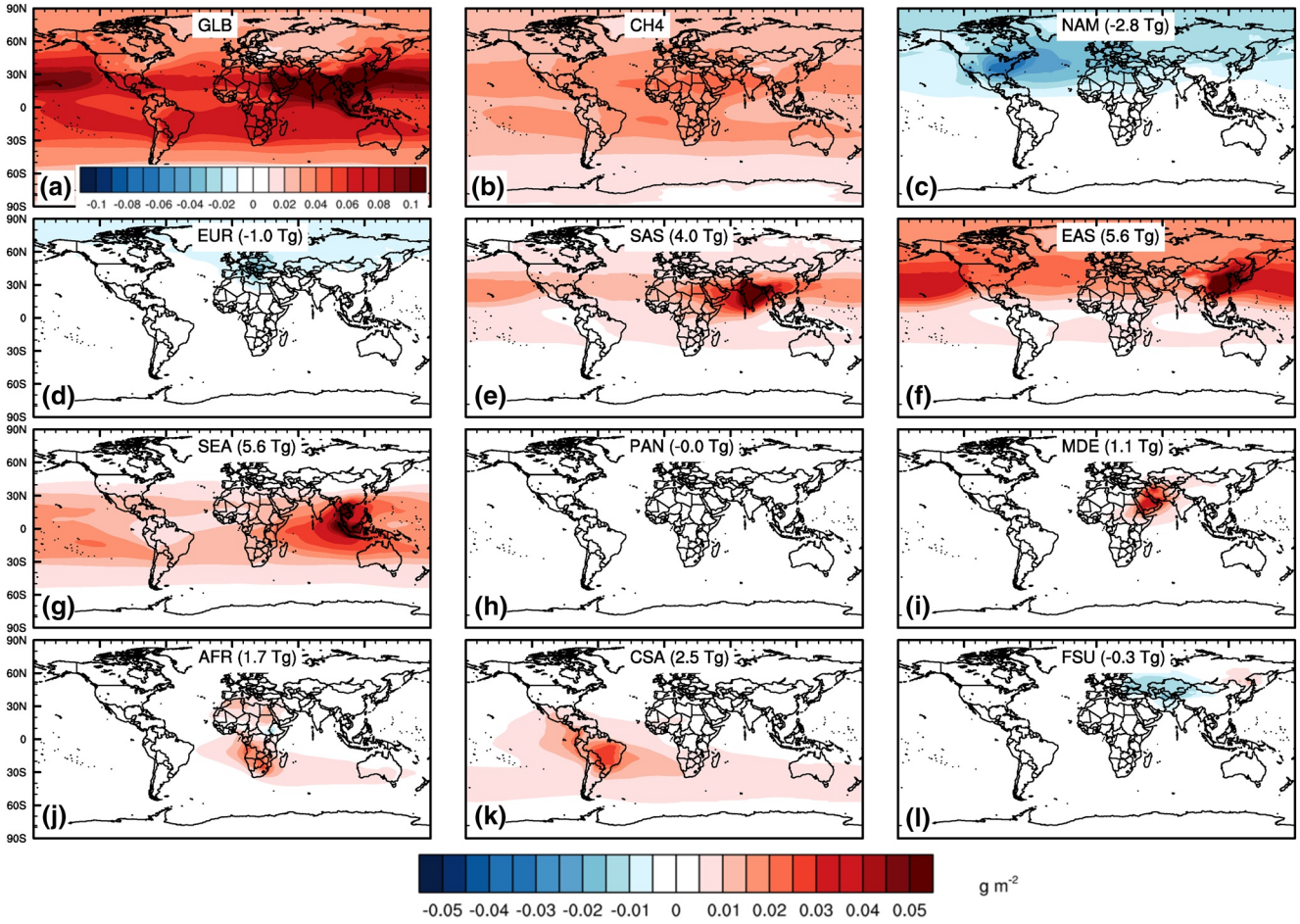

Figure 2. Spatial distributions for annual ΔB_{O3} (g/m^2) from 1980 to 2010, for (a) total emission changes from 1980 to 2010, (b) global CH_4 concentration change, and (c)–(l) emission changes in 10 world regions. Note the different colorbar used in panel (a).

3.2. Global Tropospheric Ozone Changes

The global B_{O3} is modeled to have increased 28.1 Tg from 1980 to 2010, with the largest increase from the global CH_4 increase (7.5 Tg; Figure 1d). Among the 10 regions, the global ΔB_{O3} is estimated to increase most from emission changes in SEA (5.6 Tg), EAS (5.6 Tg), and SAS (4.0 Tg). These three regions together accounted for 54% of the global ΔB_{O3}. Emission decreases from NAM and EUR contributed ΔB_{O3} decreases of −2.8 and −1.0 Tg (Figure 1d). Emission changes in FSU also contributed global ΔB_{O3} decreases (−0.3 Tg), mainly caused by the NO_x decreases (Figure 1d). Other regions contributed to the global ΔB_{O3} from negligibly (~0 Tg from PAN) to considerably (2.5 Tg from CSA). The total global ΔB_{O3} summed from the global CH_4 concentration change and the emission changes in the 10 world regions (23.9 Tg) are slightly lower than difference between S_2010 and S_1980 (28.1 Tg), mainly because of the nonlinear response of ozone to the precursors, but also because we do not account for emission changes over the oceans (Tables S2–S4). Although EAS has much larger NO_x and NMVOCs increases from 1980 to 2010 than that in SAS and SEA (Figure 1), the ΔB_{O3} are comparable between these three regions, as a result of the large sensitivity of ΔB_{O3} to NO_x emissions in SAS and SEA (Fry et al., 2012; Naik et al., 2005; West et al., 2009a).

The spatial pattern of the modeled ΔB_{O3} also suggests a strong influence of emission increases from SEA, EAS, and SAS, and decreases from NAM and EUR, and this pattern is consistent with satellite observations (Ziemke et al., 2019). The global CH_4 concentration increase has contributed more uniformly to the global ΔB_{O3} (Figure 2b), but does not explain the pattern of ΔB_{O3}.

The global zonal ΔB_{O3} increases are more notable in the NH than that in the Southern Hemisphere extending from the surface to 100 hPa (Figure 3). The global zonal ΔB_{O3} increases show a strong influence of global CH_4, which is more spatially uniform than in the regional scenarios. Emission increases from SEA and SAS cause large ozone increases over the tropics, extending to high elevation, which shows the strong convection over these regions. This convection lifts ozone precursors to high elevations where they have a longer lifetime to form and accumulate ozone, reflecting the higher temperature and strong sunlight in these regions (Lawrence et al., 2003; Zhang et al., 2016). Although the tropics have greater water vapor, the source of HO_x radicals that destroy ozone, less HO_x is present at higher elevation, making the ozone lifetime longer. In contrast, ozone reductions over NAM and EUR stay at high latitude, with little transport toward the equator, and do not reach high altitude. Much of the emissions from EAS are far enough north that they are mainly not transported toward the equator, or to high altitude, helping to explain the lower sensitivity for emissions from EAS relative to SEA and SAS.

We also analyzed spatial and zonal ΔB_{O3} in each season (Figures S7–S15). In JJA and SON, there is greater sensitivity to emissions from EAS and SAS, as the intertropical convergence zone is further north, and emissions from EAS and SAS are transported more effectively toward the tropics and high elevation. In contrast, emissions from SEA do not cause large differences in B_{O3} in different seasons. NAM and EUR have slightly larger ΔB_{O3} decreases in JJA (Figure S11).

3.3. Comparisons with HTAP2 Sensitivity Experiments

To further investigate the greater sensitivity of ΔB_{O3} to emissions from tropical and subtropical regions, we calculated the global ΔB_{O3} for regional reductions from the HTAP2 multimodel experiment, which simulated 2010. Previous HTAP2 studies have analyzed regional emission perturbations on surface air quality and radiative forcing changes, but here we present ΔB_{O3} for experiments which simulated 20% reductions in all anthropogenic air pollutant emissions globally and from six source regions analyzed here: NAM, EUR, SAS, EAS, RBU (here RBU in the HTAP2 experiment equivalent to the FSU region in our study), and MDE (Galmarini et al., 2017; Janssens-Maenhout et al., 2015; Stjern et al., 2016). We chose to analyze the six CTMs (Table S5) that simulated the base experiment, the global 20% reduction, and the 20% reductions from at least four of six regions.

Whereas the HTAP2 experiments reduced emissions of multiple precursors by the same percentage, our experiments changed emissions by different percentages for different precursors based on the changes from 1980 to 2010. To compare the modeled sensitivities, we normalize the global ΔB_{O3} by the NO_x emission changes (Tg O_3/[Tg N yr^{-1}]), since previous studies found that percent changes in NO_x produce greater B_{O3} changes compared with CO and NMVOCs (Fry et al., 2012). For HTAP2 experiments, the global ΔB_{O3} (Figure S16) is most sensitive to changes in emissions from SAS and MDE (Figure 4). In our experiments, SAS and MDE also had the greatest sensitivities of the six regions that HTAP2 studies by perturbing emissions. However, we also find that the highest sensitivities occur in three regions that HTAP2 did not simulate, and which are mainly in tropical and subtropical regions—SEA (6.1 Tg O_3/[Tg N yr^{-1}]), CSA (4.7), and PAN (3.7)—and we also show high sensitivity to emissions from AFR. The HTAP2 results for SAS and MDE provide supportive evidence for our conclusion of greater sensitivity from tropical and subtropical regions, suggesting that future experiments analyzing ozone like HTAP2 should include more regions and give a greater priority to studying the impact of emissions from the tropics. From Figure 4, we also see that the global ΔB_{O3} sensitivity to 20% global emission perturbations (GLO) is lower (ensemble mean of 1.5 Tg O_3/[Tg N yr^{-1}]) than that in our study (3.1 Tg O_3/[Tg N yr^{-1}]), mostly caused by the different percent changes of other air pollutants (CO and NMVOCs), and nonlinear response of O_3 at different NO_x levels.

4. Conclusions and Discussion

The global ozone burden is modeled to have increased from 1980 to 2010 by 28.1 Tg, with global CH_4 concentration increases contributing 26.7% of this total (7.5 Tg). Among world regions, emission increases in Southeast Asia (5.6 Tg), East Asia (5.6 Tg), and South Asia (4.0 Tg) are most important for the global ozone burden, together accounting for 54% of the total change. East Asia has much larger NO_x and NMVOCs increases from 1980 to 2010 than those in Southeast Asia and South Asia, but the global ozone burden

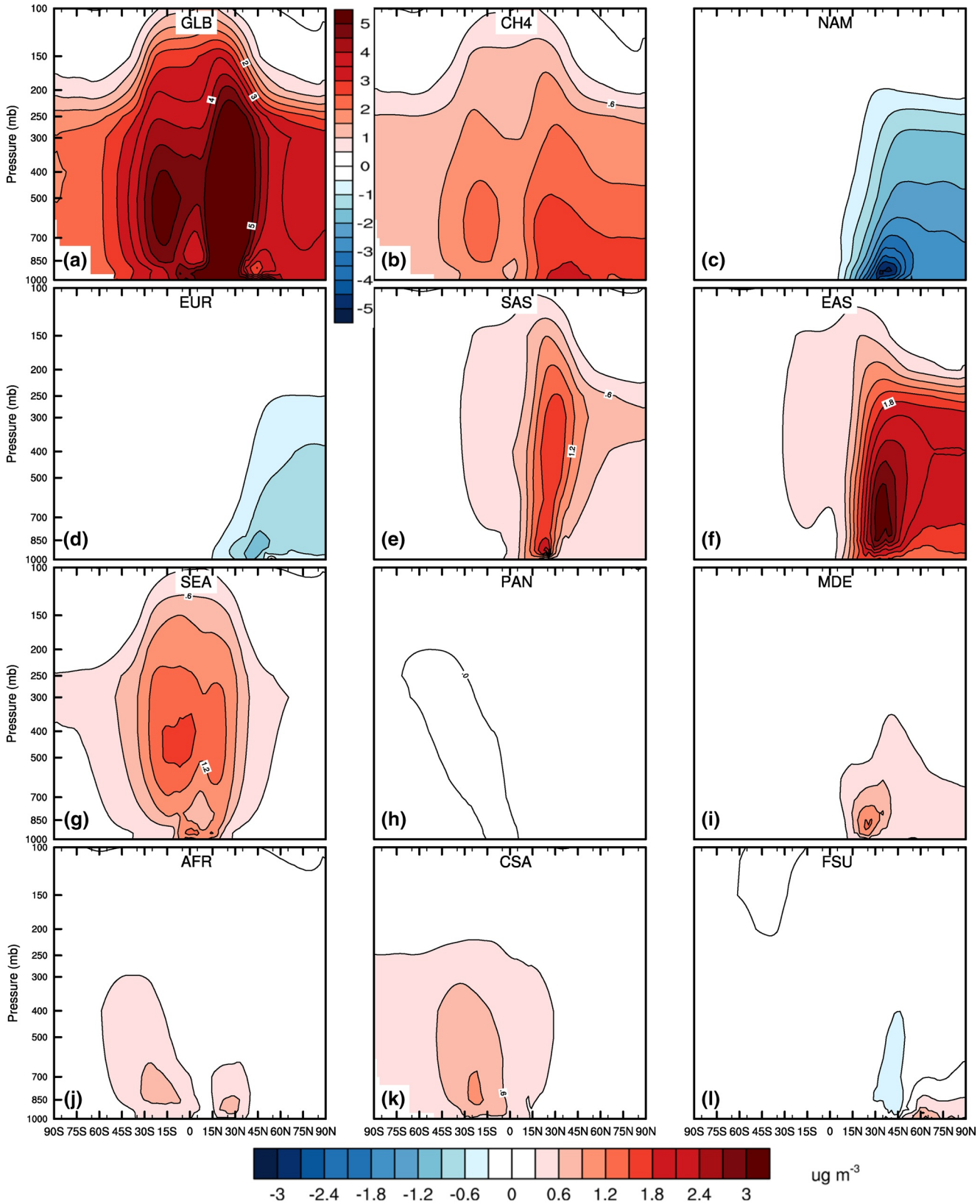

Figure 3. Zonal annual average O_3 change ($\mu g/m^3$) from 1980 to 2010, for (a) total emission changes, (b) global CH_4 concentration change, and (c)–(l) emission changes in 10 world regions. Note the different colorbar used in panel (a).

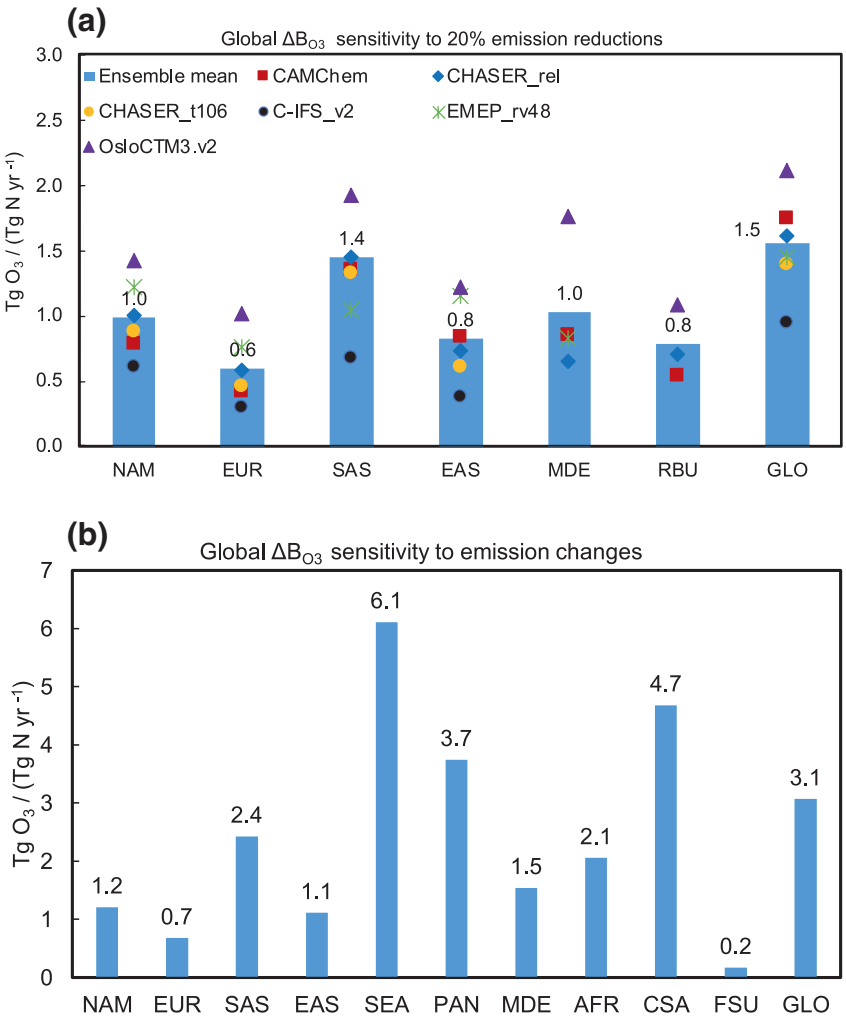

Figure 4. The sensitivity of global tropospheric ozone burden changes, normalized per unit NO_x emissions, to (a) regional and global 20% emission reductions in 2010 for all anthropogenic air pollutants from the HTAP2 experiments (blue columns are the ensemble mean from the six models), (b) regional and global emission changes from 1980 to 2010 in all anthropogenic air pollutants simulated in our study (unit of Tg O_3/[Tg N yr^{-1}]). Note for the HTAP2 results in panel a, the CHASER_t106 and C-IFS_v2 models did not perform the MDE and RBU perturbation experiments, and the EMEP_rv48 model did not perform the RBU experiment. In (b), for the GLO (3.1 Tg O_3/[Tg N yr^{-1}]) we do not consider the B_{O3} changes caused by CH_4 concentration changes from 1980 to 2010 (S_CH_4 − S_1980), to compare with the HTAP2 results.

change is comparable between these three regions, as a result of large strong sensitivity of ozone burden and convection over these tropical and subtropical regions. The emission reductions in North America and Europe contribute to global ozone burden decreases, by 2.8 and 1.0 Tg. We further calculate the sensitivity of ΔB_{O3} to regional emission from the HTAP2 multimodel experiment, which also simulated 2010. From HTAP2 experiments, we find that the global ΔBO_3 also has large sensitivity to changes in emissions from SAS and MDE regions (the HTAP2 experiments did not simulate perturbations from SEA), consistent with our findings.

Changes in emissions of NO_x, VOCs, and CO affect concentrations of the hydroxyl radical (OH), which is the major sink for CH_4 (Fiore et al., 2002; Wang & Jacob, 1998; Wild & Prather, 2000). The changes in CH_4 lifetime are important for climate forcing and in turn affect global tropospheric ozone concentration in the long-term (West et al., 2007, 2009b; Stevenson et al., 2006, 2013). We did not include this long-term ozone influence, since simulations used observed CH_4 concentrations in 1980 and 2010. But changes in ozone precursor emissions from different world regions affected this growth of methane. Future work should consider

incorporating feedback from changes in ozone precursors from different world regions on global methane concentration changes, and resulting changes in long-term ozone via changes in OH (West et al., 2009b). Here, we investigated the contributions of regional emission changes on the global tropospheric ozone burden. Future work could extend analysis of these simulations to consider effects on ground-level ozone air pollution, health impacts, and tropospheric ozone radiative forcing (RF). Unlike the surface ozone response which was mostly determined by regional and local emission changes (Liang et al., 2018; also Figure S12a in Zhang et al., 2016), changes in ozone radiative forcing may have spatial patterns different from the global tropospheric ozone burden, since the ozone RF is also affected by temperature and relative humidity (Fry et al., 2012; Kuai et al., 2017). It should be noted that other effects may influence the long-term trends in the global tropospheric ozone burden, such as stratosphere-troposphere exchange, interannual-to-decadal climatological variations, and natural sources (biogenic), which were not simulated in our study as we focus on the effects of anthropogenic emission changes.

We conclude that, in addressing the growing global ozone tropospheric burden, special attention should be paid in both research and environmental policy to low latitude regions, such as Southeast Asia and South Asia because of the greater sensitivity to emissions. NO_x emissions from these two regions increased only 18% and 33% of the NO_x increases in East Asia from 1980 to 2010, but their effects on the global ozone burden are comparable. Since 2010, global emissions have continued to evolve, as China is now reducing emissions (M. Li et al. 2018, M. Li, Liu, et al., 2017; Zheng, Chevallier et al., 2018; Zheng, Tong et al., 2018). However, ozone concentrations have worsened recently in China and it remains an important issue (Lu et al., 2018, 2020). Meanwhile, emissions in India and other South Asia regions have continued to grow (Koplitz et al., 2017; Kumar et al., 2018; C. Li, McLinden et al., 2017), and emissions from Africa are expected to accelerate (Liousse et al., 2014). For example, emissions of CO, NO_x, and NMVOCs in South Asia are projected to increase by 116%, 6%, and 18% in 2050 under the RCP8.5 scenario, and 72%, 4%, and 12% under the RCP6.0 scenario, relative to 2,000 (Kumar et al., 2018). Overall, the global shift of emissions toward the equator, where global ozone sensitivity is greater, is expected to continue. More efforts to reduce ozone precursor emissions domestically and internationally, including through methane reductions (West et al., 2006), are therefore needed to combat ozone as a global issue.

Data Availability Statement

Datasets for the HTAP2 multimodel results used in this research are available through the AeroCom servers (http://aerocom.met.no/data.html, accessed July 23, 2020, Labonne et al., 2017). The up-to-date IAGOS data are publicly available at https://doi.org/10.25326/20 (last accessed September 29, 2020). The simulated monthly mean ozone concentration from all the simulations can be accessed via DOI (https://doi.org/10.7924/r40p13p11).

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