https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Atmospheric Chemistry and Physics Discussions

Impacts of emission changes in China from 2010 to 2017 on domestic and intercontinental air quality and health effect

Yuqiang Zhang[1], Drew Shindell[1,2,3], Karl Seltzer[1], Lu Shen[4], Jean-Francois Lamarque[5], Qiang Zhang[6], Bo Zheng[7], Jia Xing[8], Zhe Jiang[9], Lei Zhang[10]

[1]Nicholas School of the Environment, Duke University, Durham, NC, USA
[2]Duke Global Health Initiative, Duke University, Durham, NC, USA
[3]Porter School of the Environment and Earth Sciences, Tel Aviv University, Tel Aviv, Israel
[4]John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA
[5]NCAR/UCAR, Boulder, CO 80301, USA
[6]Department of Earth System Science, Tsinghua University, Beijing 100084, China
[7]Institute of Environment and Ecology, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen 518055, China
[8]State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China
[9]School of Earth and Space Sciences, University of Science and Technology of China, Hefei, Anhui, 230026, China
[10]State Key Laboratory of Pollution Control & Resource Reuse, School of the Environment, Nanjing University, Nanjing 210023, China

Correspondence to: Yuqiang Zhang (Yuqiang.Zhang@duke.edu)

Abstract. China has seen dramatic emission changes from 2010, especially after the implementation of Clean Air Action in 2013, with significant air quality and human health benefits observed. Air pollutants, such as $PM_{2.5}$ and surface ozone, as well as their precursors, have long enough lifetime in the troposphere which can be easily transported downwind. So emission changes in China will not only change the regional air quality domestically, but also affect the air quality in downwind regions. In this study, we use a global chemistry transport model to simulate the influence on both domestic and foreign air quality from the emission change from 2010 to 2017 in China. By applying the health impact functions derived from epidemiology studies, we then quantify the changes in air pollution-related (including both $PM_{2.5}$ and O_3) mortality burdens at regional and global scales. The majority of air pollutants in China reach their peak values around 2012 and 2013. Compared with the year 2010, the population-weighted annual $PM_{2.5}$ in China increases till 2011 (94.1 μg m^{-3}), and then begins to decrease. In 2017, the population-weighted annual $PM_{2.5}$ decreases by 17.6%, compared with the values in 2010 (84.7 μg m^{-3}). The estimated national $PM_{2.5}$ concentration changes in China are comparable with previous studies using fine-resolution regional models, though our model tends to overestimate $PM_{2.5}$ from 2013 to 2017 when evaluated with surface observation in China during the same periods. The emission changes in China increased the global $PM_{2.5}$-related mortality burdens from 2010 to 2013, by 27,700 (95%CI: 23,900—31, 400) deaths yr^{-1} in 2011, and 13, 300 (11,400—15,100) deaths yr^{-1} in 2013, among which at least 93% occurred in China. The sharp emission decreases after 2013 bring significant benefits for reduced avoided premature mortality in 2017, reaching 108, 800 (92,800—124,800) deaths yr^{-1} globally, among which 92% happening in China. Different trend as $PM_{2.5}$, the annual maximum daily 8-hr ozone in China increased, and also the ozone-related premature deaths, ranging

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

from 3,600 (2,700—4,300) deaths yr^{-1} in 2011 (75% of global total increased premature deaths), and 8,500 (6,500—9,900) deaths yr^{-1} in 2017 (143% of the global total). Downwind regions, such as South Korea, Japan, and U.S. generally see a decreased O_3-related mortality burden after 2013 as a combination of increased export of ozone and decreased export of ozone precursors. In general, we conclude that the sharp emission reductions in China after 2013 bring benefits of improved air quality and reduced premature deaths associated with air pollution at global scale. The benefits are dominated by the $PM_{2.5}$ decreases since the ozone is shown to actually increase with the emission decrease.

1 Introduction

Fine particulate matter with an aerodynamic diameter of less than 2.5 μm ($PM_{2.5}$) has been of particular interest to the research community as it has been known to pose great threats to environment, such as visibility impairment and material damages (Hand et al., 2013, 2014; Wu and Zhang, 2018), and human health (e.g. Pope et al., 2002; Krewski et al., 2009). Increased relative risk were found between short-term $PM_{2.5}$ exposures with daily all-cause mortality, respiratory and cardiopulmonary mortality across 24 countries in 652 cities (Liu et al., 2019). Long-term exposure to $PM_{2.5}$ has been found to cause premature deaths from cardiopulmonary and respiratory disease (Krewski et al., 2009; Burnett et al., 2014). Short-term exposure to ozone were associated with hospital admissions, emergency room visits for respiratory causes and school absences (Katsouyanni et al., 2009), and long-term exposure to surface ozone were also related with premature deaths from respiratory disease (Jerrett et al., 2009; Turner et al., 2016).

$PM_{2.5}$, as well as its precursors, can travel long distances, affecting air quality and health in other receptor regions (Ewing et al. 2010; Pfister et al., 2011; Anenberg et al., 2014), despite its relatively short lifetime in the atmosphere (days to weeks). Tropospheric ozone has a much longer lifetime compared with $PM_{2.5}$, with approximately a global average of 23 days (Young et al., 2013), and the research community has expressed particular interest in studying its intercontinental transport (e.g. Zhang et al., 2008, 2014; Cooper et al., 2015; Lin M. et al., 2012, 2017; Parrish et al., 2014). Numerous studies have been carried out to investigate the source-reception relations on air quality and associated mortality burden from emission changes in one source region onto the others (West et al., 2009a,b; Fry et al., 2013; Crippa et al., 2019). The Task Force on Hemispheric Transport of Air Pollution (TF HTAP, Janssens-Maenhout et al., 2015), has made great effort to organize international scientists to study the effect of emissions changes on the intercontinental transport of air quality and human health (http://www.htap.org/, accessed April 11 2020). Liang et al. (2018) used the ensemble model outputs from the TF HTAP, and estimated the source-receptor relations for air quality and avoided premature deaths from 20% reductions of anthropogenic emissions in East Asia. They estimated that 96,600 premature deaths would be avoided globally associated with $PM_{2.5}$ reductions, with 6% (5, 500 deaths) in other regions, while for ozone, 11, 400 premature deaths would be avoided globally, with 15% (1,700 deaths) in foreign countries other than East Asia.

To tackle the severe air pollution problem in China, the Chinese government has implemented strict clean air policies in recent years (State Council of the People's Republic of China, 2013). Before 2013, the clean air control policies were mainly focusing

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

on the emission standards of industry and power sectors, and after 2013 eight more stringent control measures were developed after China committed to reducing $PM_{2.5}$ pollution (see Fig. 1 in Zheng B. et al., 2018b). Significant emission reductions have been observed in China, and the air quality has substantially improved especially after the "Air Pollution Prevention and Control Action Plan" (APPCAP) in 2013 (Zheng Y. et al., 2017; Zhang et al., 2019; UN Environment 2019). The relative change of China's anthropogenic emissions for specific air pollutants during 2010–2017 are estimated as follows: −35 % for primary $PM_{2.5}$, -62 % for SO_2, −27 % for BC, −35 % for OC, −17 % for NO_x, and −27 % for CO (Zheng B. et al., 2018b). Significant decreases were observed for the $PM_{2.5}$ concentrations from surface observations, satellite retrievals and model simulations (e.g. Song et al., 2017; Huang et al., 2018; Li Y. et al., 2015; Li C. et al., 2017; Lin C. et al., 2018; Zheng Y. et al., 2017; Zhang Q. et al., 2019). The rapid reductions of major air pollutants in China were also confirmed using a long-term, robust observational record at Fukue Island, Japan (Kanaya et al., 2020). A recent study using high-resolution regional air quality model showed that the estimated national population–weighted annual mean $PM_{2.5}$ concentrations decreased from 61.8 to 42.0 μg m^{-3} from 2013 to 2017 (Zhang Q. et al. 2019). Meanwhile, summertime daily maximum 8-h average ozone (MDA8) in China has been shown an increasing trend since 2013 (Lu et al., 2018, 2020). The increasing surface ozone trend may be partially explained by the slowing down of the aerosol sink of hydroperoxyl radicals which mainly reacts with NO to produce ozone that caused by the significant $PM_{2.5}$ reductions, though a field campaign result may not be consistent with model sensitivity results (Li et al., 2019; Tan et al., 2020). Recent studies have found that the major air pollutants, such as SO_2, NO_x, CO, which are precursors for $PM_{2.5}$ and surface ozone, already reach peak values in 2012 or 2013, depending on the pollutants (Zheng B. et al., 2018a,b).

Previous studies have evaluated the benefits from the APPCAP in China since 2013 on improved air quality including both $PM_{2.5}$ and O_3 and avoided premature deaths (e.g. Huang et al., 2018; Zhang et al., 2019; Lu et al., 2020). However, limited studies have been carried out to investigate the benefits of these actions on global air quality and air pollution related mortality burden. In this study, we use a global chemical transport model to simulate the global air quality changes from 2010 to 2017 as a result of emission changes in China at the same time. Then we will apply the concentration response function (CRF) to estimate the air pollution-related mortality burden changes in China and other countries. This particular time period was chosen because the emissions in China were seen slightly increases and then significantly decreases. By incorporating the contrasting emission trends in China, we want to compare effect of different emission trends in China on global air quality.

2 Data and Methods

2.1 Model simulation using the CAM-chem

The global air quality from 2010 to 2017 were simulated using the CAM-chem model (CAM version 4, Lamarque et al., 2012) at a horizontal resolution of 1.9◦ (latitude) × 2.5◦ (longitude), and 56 vertical levels between the surface and 4 hPa (≈40 km), driven by the NASA GEOS5 Global Atmosphere Forcing Data (Tilmes 2016, http://rda.ucar.edu/datasets/ds313.0/, last Accessed 20 April, 2020). The detailed configurations, including the lower boundary conditions for long-lived species such as

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

CO_2 and CH_4, the online biogenic emission inventory, and other natural emissions, are referred to Zhang et al. (2016). In this version of CAM-chem, the bulk aerosol model was applied based on the work of Tie et al. (2001, 2005), in which the sulfate aerosol is formed by the oxidation of SO_2 in both the gas and aqueous phase (Lamarque et al., 2012; Tilmes et al., 2016). The ammonium nitrate is also represented depending on the amount of sulfate present in the air mass following the parameterization of gas/aerosol partitioning by Metzger et al. (2002). A comprehensive evaluation for the model performance in simulating temporal and spatial distribution of global ozone and aerosols by comparing surface observation, balloon, aircraft and satellite were carried out in previous studies (Tilmes et al., 2015, 2016; Zhang et al., 2016). In this study, we used the same configurations as our previous one (Zhang et al., 2016), and in the Chemistry-Climate Model Initiative project (Tilmes et al., 2016). We focus on the model evaluation in simulating the surface ozone and $PM_{2.5}$ concentrations in China from 2013 to 2017. Base case simulations were run consistently from 2010 to 2017 with one year spin-up in 2009 using time-varying global anthropogenic emission inventory as a combination of the Community Emissions Data System (CEDS, v2017-05-18, Hoesly et al., 2018) developed by the Pacific Northwest National Laboratory, and the Multi-resolution Emission Inventory (MEIC, http://www.meicmodel.org/, last access 20 April 2020) developed by Tsinghua University, China. We performed first sensitivity simulation (CEDS_MEIC_ChinaFix) which keeps China emissions constant at the level of 2010, and the differences between the base and CEDS_MEIC_ChinaFix are the influences of anthropogenic emission changes in China since 2010 on global air quality and human health (Table 1). By making comparisons between these two scenarios, we can also rule out the influences of meteorological variabilities on the global distribution of ozone and $PM_{2.5}$. The global anthropogenic emissions other than China after 2014 were kept constant, since no global emissions inventory is available after 2014 when we first prepared the study. McDuffie et al. (2020) updated the global CEDS anthropogenic emissions through 2017 with continued update into 2019 (https://github.com/JGCRI/CEDS, last access 6 May 2021). However, this would have negligible effect on our conclusions since our focus is the emission changes from China on influence of domestic and international air transport. We also performed another sensitivity simulation with global anthropogenic emissions keeping constant at 2010 level (CEDS_MEIC_GlobalFix, Table 1). The air quality changes from 2011 to 2017 relative to 2010 from this sensitivity could allow us to take a look at the meteorological changes on $PM_{2.5}$ and O_3 changes in China at the same time.

It also came to our attention that the CEDS emissions tends to overestimate the magnitude of the Chinese emissions, and underestimate the emission decreasing trend in China (Zheng B. et al., 2018b; Paulot et al., 2018). From Fig. S1, we see that in 2014, the emissions from CEDS are at least 20% higher than the estimations from MEIC for most of the air pollutants, except for non-methane volatile organic compounds (1%). More specifically, the SO_2, OC and BC emissions estimated in CEDS are 84%, 81% and 58% higher individually than those estimated in MEIC. For the emission trend, CEDS estimated a continued increasing trend while MEIC estimated a peak for most of the air pollutants before the year 2012 (Liu et al., 2016; Zheng B. et al., 2018b). From Fig. S2 which shows the spatial patterns of the emission differences between CEDS and MEIC in 2014, we can see that the emissions in CEDS are higher in the western and south China, and lower in the eastern China. We performed another sensitivity simulation which applied CEDS global anthropogenic only from 2010 to 2014 (CEDS_Global),

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

to evaluate the model's performance in simulating $PM_{2.5}$ and O_3 in China and also discuss the relative air quality changes applying different emission inventory in China.

2.2 Surface observation for $PM_{2.5}$ and O_3 in China

Surface observation for hourly $PM_{2.5}$ and O_3 concentration were downloaded from China National Environmental Monitoring Center (CNEMC) Network (http://106.37.208.233:20035/) from 2013 to 2017, since data was not available before 2013. We evaluated the model's performance in simulating annual $PM_{2.5}$ and maximum daily 8 h average (MDA8 O_3) as these two metrics are related with the health impact analysis we performed in our study.

2.3 Health impact assessment for surface $PM_{2.5}$ and O_3

We applied the health impact function derived from long-term cohort studies, together with the baseline mortality rates and exposure population to quantify the air quality related mortality burden changes. The mortality burdens related to ambient air pollution including $PM_{2.5}$ and O_3 are calculated following Eq. (1):

$$\Delta Mort = Y_0 \times AF \times Pop\ , \qquad (1)$$

Where $\Delta Mort$ is mortality burden attributed to surface $PM_{2.5}$ and O_3, Y_0 is the baseline mortality rates for cause of specific disease, AF is the attribution fraction calculated as $1 - \frac{1}{RR}$ with RR as the relative risk, and Pop is the exposed population with ages greater than 25 years old. The RR for the $PM_{2.5}$ is calculated using the latest integrated exposure response model (IER, Burnett et al., 2014), following the previous methods in our group (Shindell et al., 2018). We used the 1000 simulations for the parameter distributions of α, β and γ retrieved from the latest GBD study (Stanaway et al., 2018) to derive the mean RR with 95% uncertainty intervals. The RR for the long-term exposure to surface ozone is from the updated cohort study (Turner et al., 2016), with RR of 1.12 (95 % confidence interval (CI): 1.08, 1.16) for respiratory disease. Country-age-specific baseline mortality rates (Y_0) in 2010 were retrieved from the latest GBD (Global Burden of Disease) project (Stanaway et al., 2018), and remapped to match the 10th International Statistical Classification of Diseases and Related Health Problems codes as used in the cohort study (Turner et al., 2016; Seltzer et al., 2020).

3 Results

3.1 Model evaluation with surface observation in China

From the model evaluation metrics, we see that the CAM-chem base simulation (CEDS_MEIC scenario, see Table 1) generally overestimates the annual $PM_{2.5}$ concentration in China, with mean bias (MB) of 19.3 μg m^{-3} and normalized mean bias (NMB) of 37.2% for all the 5 years. The MBs are around 20 μg m^{-3} from 2014 to 2017 with lowest values in year 2013 (MB of 7.6 μg m^{-3}) and highest in 2015 (21.7 μg m^{-3}). The lower MB and NMB in 2013 could be caused by the much less data available in

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

2013. The positive NMBs for all the years shows that the overestimations are systematic and may not affect our main conclusions since we focus on the changes among years. The higher modeling bias for surface $PM_{2.5}$ from the CAM-chem were also seen in other studies, for example, He and Zhang (2014) reported NMB of 37.6% and 41.85% for the contiguous United States and Europe in 2001. The bias in simulating the surface $PM_{2.5}$ in the CAM-chem (version 4) was mainly caused by the inaccurate prediction of SO_4^{2-}, NH_4^+, and organic aerosols, and missing major inorganic aerosol species such as nitrate and chloride (He and Zhang, 2014; Tilmes et al., 2016). By including advanced inorganic aerosol treatments, such as condensation of volatile species, explicit inorganic aerosol thermodynamics for sulfate, ammonium, nitrate, sodium, and chloride (He and Zhang, 2014), and more comprehensive secondary organic aerosols approach (Volatility Basis Set scheme, Times et al., 2019; Liu et al., 2020), the performance for simulating surface $PM_{2.5}$ could be significantly improved. The different metrics also exhibits small annual variabilities, with NMB around 40% and NME around 50% for each year, except for 2013 and 2014 which have smaller NMB values due to the limited size of the observations (Table 2). The CAM-chem can generally reproduce the spatial patterns of the annual $PM_{2.5}$ distributions, with correlation coefficient (R) greater than 0.7. With CEDS emissions applying globally (CEDS_Global scenario, see Table 1), we have better performance in both 2013 and 2014 for the annual $PM_{2.5}$ evaluation. Part of the reason is that we have less available data in these two years, while we suspect the main reason is that though total emissions in China are higher in CEDS than those in MEIC, CEDS tends to underestimate the emissions in eastern and central China (Fig. S2), where the majority of observations are available. With CEDSs emission applied, the annual $PM_{2.5}$ is lower in eastern China and higher in western and northwestern China (Fig. S3). The performance for CAM-chem in simulating annual MDA8 O_3 is slighter better than the surface $PM_{2.5}$, with NMB lower than 20% for all the years (Table 3). From Table 3, we can also see that the CAM-chem overestimates the annual MDA8 O_3 in China, which means our estimation for the O_3-related mortality burden will have positive bias. A high bias of about 10 ppb can be attributed to the coarse model resolution, which leads to an overestimate of ozone production, because of diluted emissions of ozone precursors (Tilmes et al, 2015). For annual MDA8 O_3, the CEDS simulation (CEDS_Global scenario) has a poorer performance compared with using MEIC emissions, which maybe caused by the overestimation of the surface ozone in western China (Fig. S4).

3.2 Air quality changes in China from 2010 to 2017

We first report the trends for the national annual population-weighted (Pop-weighted) $PM_{2.5}$ which decreased by by 17.6%, changing from 84.7 μg m^{-3} in 2010 to 69.8 μg m^{-3} in 2017 (Fig. 1). The Pop-weighted $PM_{2.5}$ was highest in 2011 (annual average of 94.1 μg m^{-3}), and decreases until 2017 (69.8 μg m^{-3}). From 2013 to 2017, the national annual Pop-weighted $PM_{2.5}$ decreased by 15.9 μg m^{-3}, comparable to the values (19.8 μg m^{-3}) estimated by Zhang Q. et al. (2019) which used a high-resolution regional air quality model. The area-weighted national average $PM_{2.5}$ concentrations shares a similar trend as the Pop-weighted average but with much lower values (Fig. 1a), demonstrating the fact that the high $PM_{2.5}$ pollutions happen in more density region. The annual average of area-weighted $PM_{2.5}$ concentration decreased by 7.6 μg m^{-3}, consistent with the estimations by Ding et al. (2019a) at 9.0 μg m^{-3}, which also demonstrates that overestimations of annual $PM_{2.5}$ from CAM-chem are systematic and will not affect our trend discussions. For the spatial patterns of the $PM_{2.5}$ changes, we see that

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

significant annual $PM_{2.5}$ changes (increases before 2013 and decreases after then) mainly occur in the eastern China (Fig. 2), which were the focused regions for China APPCAP (Ding et al., 2019a,b). When distinguishing the contributions from anthropogenic emissions vs. meteorology, we find that the annual $PM_{2.5}$ decreases in China are mainly dominated by the emission changes, consistent with previous studies (Dang and Liao, 2019; Ding et al., 2019a; Zhai et al., 2019; Zhang Q. et al., 2019). Compared with the year 2010, interannual meteorology led to annual $PM_{2.5}$ decreases by as high as 3.5 μg m^{-3} in 2012, and increases as high as 3.1 μg m^{-3} in 2015 (Fig. 4a).

Different from the $PM_{2.5}$ trend, the annual Pop-weighted average MDA8 O_3 has a continued increasing trend since 2010 (Fig. 1b), with peaks in 2014 (59.5 ppbv), and then decreases to 2017 (57.1 ppbv). The area-weighted MDA8 O_3 was comparable or even larger than the Pop-weighted (e.g. 2012), as a result of more uniform O_3 distribution in China or even higher ozone events in western China because of stratosphere-troposphere exchange with less population (Wang et al., 2011; Li et al., 2019). For the spatial patterns, the ozone increases mainly in Beijing–Tianjin–Hebei and Yangtze-River-Delta, and slightly decreases in the south (Fig. 3; Fig. S5). The anthropogenic emission reductions in China leads to ozone increases (Fig. 4b), which could be partially explained by aerosol sink of hydroperoxyl radicals slowing down due to $PM_{2.5}$ decreases (Li et al., 2018). The interannual meteorological condition changes have a much larger positive effect on the annual MDA8 O_3, compared with the anthropogenic emission changes, leading to ozone increases as high as 8.1 ppbv in 2014 and as low as 0.7 ppbv in year 2011. The meteorology-induced ozone increases can be attributed to increasing temperature which enhances the ozone production and biogenic NMVOCs emissions (Ding et al., 2019b; Liu and Wang, 2020), and the increases solar radiation (Wang et al., 2020; Ma et al., 2021).

3.3 Emission changes in China on global air quality and health

3.3.1 Global and regional air quality

The simulated global tropospheric ozone burden (total ozone below the chemical tropopause of 150 ppbv) from 2010 to 2017 calculated from the CEDS_MEIC simulation is 327.5 ± 5.2 Tg, agreeing well with the present tropospheric ozone burden estimated from previous ensemble models (ACCENT: 336 ± 27 Tg; ACCMIP:337 ± 23 Tg; TOAR: 340 ± 34 Tg, and CMIP6: 348 ± 15 Tg; Griffiths et al., 2021). From Fig. 5, the change for global tropospheric ozone burden from the emission changes in China ranges from 0.6 Tg (2011 and 2012) to -1.9 Tg (2017). The tropospheric ozone burden changes are not only seen in China, but also in downwind regions, especially in Northern Hemisphere, such as Pacific Ocean and U.S., as a result of the vertical transport of the air pollutants (Fig. S6).

Due to the prevailing western wind, the air pollutants in China could be easily transported to downwind regions, especially during springtime (Lin M. et al., 2012; Liang et al., 2019). From Fig. 6, we see that downwind regions, South Korea has the largest pop-weighted $PM_{2.5}$ changes, ranging from 0.7 μg m^{-3} in 2012 to -2.63 μg m^{-3} in 2017, following by Japan and U.S. (Fig. 6a). However, for ozone, we see that the emission changes in China have increased the surface ozone in South Korea from 2010 to 2017, mainly contributed by the increased export of ozone. Both Japan and U.S. are firstly seen an increase and

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

then decrease. However, the increases and the decreases are not usually following the same year as emissions and $PM_{2.5}$ concentration changes, demonstrating the non-linearity of the ozone productions.

3.3.1 Global and regional air pollution-related mortality burden changes

The global ambient $PM_{2.5}$-related mortality burden in 2010 is 4.08 million (95%CI: 2.15—6.0 million), similar with previous estimated in the same year applying the same IER method (3.6±1.0 million in 2010, Shindell et al., 2018). Compared with 2010, the emission changes in China (by comparing CEDS_MEIC and CEDS_MEIC_ChinaFix) lead to mortality increases by 27, 700 deaths yr^{-1} (95%CI: 23, 900—31, 400 deaths yr^{-1}) in 2011, with 93% occurring in China (25, 800, 95%CI: 22, 300—29, 200 deaths yr^{-1}; Table 4). From Table 4 (last column), we can see that China takes the largest majority of global $PM_{2.5}$-mortality burden changes from 2011 to 2017 (more than 90%), because of the relative linear relations between emission and concentration for $PM_{2.5}$, and its relative shorter lifetime in the troposphere. Relative to year 2010, the emission changes in China in 2017 leads to 108, 800 (95%CI: 92,800—124, 800) avoided premature deaths, with 92% (95%CI: 85, 900—114, 300) occurring in China. Among the three downwind regions, Japan displays the largest influences for the $PM_{2.5}$-related mortality burden changes, ranging from 197 deaths yr^{-1} in 2011 to -875 deaths yr^{-1} in 2017. The emission changes in China have comparable effect on the $PM_{2.5}$-related mortality burden change among South Korea and U.S. (Table 4).

The global ozone-related mortality in 2010 is 1.02 million (95% CI: 0.73—1.28 million), consistent with previous estimations using other global CTMs, such as GEOS-Chem (1.04—1.23 million) and GISS (0.8—1.3 million), applying the same relative risk (Mally et al., 2017; Shindell et al., 2018). In 2011, the emission changes in China increased the global ozone-related mortality by 4,900 (95%CI, 3,700—5,900) deaths yr^{-1} (Table 5), among which 73% is happening in China (3600 deaths yr^{-1}, 95%CI: 2,700—4,300). For the three downwind region, South Korea, Japan, and U.S., the added O_3-related mortality burdens in each country are 23, 200 and 131 deaths yr^{-1} individually. In 2017, the reduced anthropogenic emissions in China greatly increased the ozone-related mortality burden, by 8,500 deaths yr^{-1}, which is 43% higher than the global total added O_3-related mortality burden (5,920 deaths yr^{-1}). In general, the downwind regions have decreased O_3-related mortality burden (-65 and -289 deaths yr^{-1} for Japan and U.S. individually, Table 5), except for South Korea, which also has increased mortality burden by 17 deaths yr^{-1} (Table 5).

4 Discussion

Dramatic changes are observed for anthropogenic emissions in China since 2010, with majority of air pollutants, such as NO_x, SO_2, are CO peaks around 2012 and 2013, and decreases afterwards significantly. In this study, we use a global chemistry transport model (CAM-chem) to simulate the emission changes in China on domestic and global air quality and air pollution-related mortality burden changes from 2010 to 2017. Compared with surface $PM_{2.5}$ observations network in China, our model tends to overestimate the annual $PM_{2.5}$ concentrations, with normalized mean bias (NMB) of 37.2% and normalized mean error (NME) of 52.0% from 2013 to 2017. We also evaluated the model performances by applying the regional emission

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

inventory (MEIC) developed by Tsinghua University and the global emission inventory (CEDS) developed by PNNL and applied extensively in the CMIP6 experiments. For surface $PM_{2.5}$, we find that the CAM-chem with CEDS emissions tends to have a lower NMB and NME in 2013-2014 since CEDS emissions are lower in urban areas in China than that from MEIC though the national totals from CEDS are much higher than those from MEIC. For surface O_3, CAM-chem with MEIC emissions in China has a lower NMB (13.7%) and NME (21.9%) for the annual MDA8 O_3. The simulations with CEDS in China tend to have larger NMB (15.2%) and NME (36.6%) in 2013-2014.

From 2010 to 2017, we calculate that, the annual average population-weighted (Pop-weighted) $PM_{2.5}$ increases in China till 2011 (94.1 μg m^{-3}), and then decreases sharply afterwards. The annual Pop-weighted $PM_{2.5}$ in 2017 decreases by 17.6% (-14.9 μg m^{-3}), compared with the value in 2010 (84.7 μg m^{-3}), and 18.5% lower than 2013 (85.8 μg m^{-3}). Though CAM-chem overestimates the $PM_{2.5}$ concentrations in China, the simulated decreasing trend for annual $PM_{2.5}$ from 2013 to 2017 (-15.9 μg m^{-3} for Pop-weighted, and 7.6 μg m^{-3} for area-weighted) is comparable with previous studies using high resolution regional air quality models (-19.8 μg m^{-3} estimated from Zhang Q. et al. (2019), and -9.0 μg m^{-3} from Ding et al. (2019a)). The overestimation of the surface $PM_{2.5}$ concentration in China from CAM-chem were unlikely to affect our estimations of the trends for the $PM_{2.5}$-related health benefits, because of the high $PM_{2.5}$ concentration in China, as well as the non-linearity of the IER functions with the $PM_{2.5}$ concentration (Zhang Q. et al., 2019). The $PM_{2.5}$ overestimations in the CAM-chem model were likely caused by the uncertainties in the bottom-up emission inventories (Shen et al., 2019; Zhang Q. et al., 2019), and missing chemical mechanisms for the $PM_{2.5}$ components (Tilmes et al., 2016; Liu et al., 2020), and are in the same magnitudes as seen in other high resolution regional models (25%-30% in Shen et al., 2019; ~20% in Zhang Q. et al., 2019). The emission changes in China from 2010 to 2013 increased the global $PM_{2.5}$-related mortality burdens, varying from 27, 700 deaths yr^{-1} (95% confidence interval (CI): 23,900—31, 400) in 2011, and 13, 300 (95%CI: 11,420—15, 110) deaths yr^{-1} in 2013. Among the increased premature deaths, at least 93% occurring in China. The sharp emission decreases after 2013 bring significant benefits for reduced avoided premature mortality in 2017, reaching 108, 800 (92,800—124, 800) deaths yr^{-1}, among which 92% (100, 100 deaths yr^{-1} with 95%CI: 85, 900—114, 300) happening in China. Downwind regions, such as South Korea, Japan, and U.S. share the same $PM_{2.5}$ trend as China. The transport of $PM_{2.5}$ and its precursors could change the annual Pop-weighted $PM_{2.5}$ ranges from 0.7 μg m^{-3} in 2011 and -2.6 μg m^{-3} in 2017, leading to added 98 premature deaths in 2011, and 386 avoided premature deaths in 2017. Japan has a smaller change for the annual Pop-weighted $PM_{2.5}$, but much larger changes in $PM_{2.5}$-related mortality burden changes, ranging from 197 added premature deaths in 2011, and 875 avoided premature deaths in 2017. The influence for U.S. ranges from 44 added premature deaths in 2011, and 381 avoided premature deaths in 2017. Different trend as $PM_{2.5}$, the emission changes in China had an increasing trend for the annual maximum daily 8-hr ozone in China, and also the ozone-related premature deaths, ranging from 3, 600 deaths yr^{-1} in 2011 (75% of global total increased premature deaths), and 8, 500 deaths yr^{-1} in 2017 (143% of global total). Downwind regions, such as South Korea, Japan, and U.S. are generally seen decreased O_3-related mortality burden after 2013 as a combination of increased export of ozone and decreased export of ozone precursors. In general, we conclude that the sharp emission reductions in China after

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

2013 bring benefits of improved air quality and reduced premature deaths associated with air pollution at the global scale. The benefits are dominated by the $PM_{2.5}$ decreases.

Data availability. Global anthropogenic emissions data from CEDS are available from https://www.geosci-model-dev.net/11/369/2018/ (accessed May 4th, 2020). MEIC emission inventory is available from http://meicmodel.org/?page_id=560 (last access May 6th, 2021). Baseline health and population data are available from the World Health Organization and the United Nations, respectively. The CAM-chem model is available at http://www.cesm.ucar.edu/models/cesm1.2/ (accessed May 4th, 2020). Data from CESM modelling that support the findings of this study are available from the corresponding author upon request.

Author contributions. YZ and DS originally designed the study, and YZ conducted all simulations, created all figures and wrote the manuscript, with comments and edits from all authors. BZ and QZ contributed to develop the MEIC emission inventory.

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

Acknowledgements. Y.Z. and D.S. acknowledge the support by the NASA GISS grant 80NSSC19M0138. We gratefully acknowledge the CESM model which was developed and distributed by NCAR. We also appreciate the efforts from China Ministry of Ecology and Environment for maintaining the nationwide observation network and publishing hourly $PM_{2.5}$ and O_3 concentrations. We would like to thank the University of North Carolina at Chapel Hill and the Research Computing group for providing computational resources and support that have contributed to these research results.

Reference

Anenberg, S. C., West, J. J., Yu, H., Chin, M., Schulz, M., Bergmann, D., Bey, I., Bian, H., Diehl, T., Fiore, A., Hess, P., Marmer, E., Montanaro, V., Park, R., Shindell, D., Takemura, T. and Dentener, F.: Impacts of intercontinental transport of anthropogenic fine particulate matter on human mortality, Air Qual. Atmos. Heal., 7(3), 369–379, doi:10.1007/s11869-014-0248-9, 2014.

Burnett, R. T., Arden Pope, C., Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Singh, G., Hubbell, B., Brauer, M., Ross Anderson, H., Smith, K. R., Balmes, J. R., Bruce, N. G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M. C., Gapstur, S. M., Diver, W. R. and Cohen, A.: An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure, Environ. Health Perspect., 122(4), 397–403, doi:10.1289/ehp.1307049, 2014.

Cooper, O. R., Langford, A. O., Parrish, D. D., and Fahey, D. W.: Challenges of a lowered U.S. ozone standard, Science, 348, 1096-1097, 2015.

Crippa, M., Janssens-Maenhout, G., Guizzardi, D., Van Dingenen, R., and Dentener, F.: Contribution and uncertainty of sectorial and regional emissions to regional and global $PM_{2.5}$ health impacts, Atmos. Chem. Phys., 19, 5165–5186, https://doi.org/10.5194/acp-19-5165-2019, 2019.

Dang, R. and Liao, H.: Severe winter haze days in the Beijing–Tianjin–Hebei region from 1985 to 2017 and the roles of anthropogenic emissions and meteorology, Atmos. Chem. Phys., 19, 10801–10816, https://doi.org/10.5194/acp-19-10801-2019, 2019

Ding, D., Xing, J., Wang, S., Liu, K. and Hao, J.: Estimated Contributions of Emissions Controls, Meteorological Factors, Population Growth, and Changes in Baseline Mortality to Reductions in Ambient $PM_{2.5}$ and $PM_{2.5}$-Related Mortality in China, 2013-2017, Environ. Health Perspect., 127(6), 67009, doi:10.1289/EHP4157, 2019a.

Ding, D., Xing, J., Wang, S., Chang, X. and Hao, J.: Impacts of emissions and meteorological changes on China's ozone pollution in the warm seasons of 2013 and 2017, Front. Environ. Sci. Eng., 13(5), 1–9, doi:10.1007/s11783-019-1160-1, 2019b.

Ewing, S. A., Christensen, J. N., Brown, S. T., Vancuren, R. A., Cliff, S. S., and Depaolo, D. J.: Pb Isotopes as an Indicator of the Asian Contribution to Particulate Air Pollution in Urban California, Environ. Sci. Technol., 44, 8911–8916, https://doi.org/10.1021/es101450t, 2010.

Fry, M. M., Schwarzkopf, M. D., Adelman, Z. and West, J. J.: Air quality and radiative forcing impacts of anthropogenic volatile organic compound emissions from ten world regions, Atmos. Chem. Phys., 14, 523–535, doi:10.5194/acp-14-523-2014, 2014.

Griffiths, P. T., Murray, L. T., Zeng, G., Shin, Y. M., Abraham, N. L., Archibald, A. T., Deushi, M., Emmons, L. K., Galbally, I. E., Hassler, B., Horowitz, L. W., Keeble, J., Liu, J., Moeini, O., Naik, V., O'Connor, F. M., Oshima, N., Tarasick, D., Tilmes, S., Turnock, S. T., Wild, O., Young, P. J., and Zanis, P.: Tropospheric ozone in CMIP6 simulations, Atmos. Chem. Phys., 21, 4187–4218, https://doi.org/10.5194/acp-21-4187-2021, 2021.

Hand, J. L., Schichtel, B. A., Malm, W. C. and Frank, N. H.: Spatial and temporal trends in $PM_{2.5}$ organic and elemental carbon across the United States, Adv. Meteorol., 2013, doi:http://dx.doi.org/10.1155/2013/367674, 2013.

Hand, J. L., Schichtel, B. A., Malm, W. C., Copeland, S., Molenar, J. V., Frank, N. and Pitchford, M.: Widespread reductions in haze across the United States from the early 1990s through 2011, Atmos. Environ., 94, 671–679, doi:10.1016/j.atmosenv.2014.05.062, 2014.

He, J. and Zhang, Y.: Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments, Atmos. Chem. Phys., 14, 9171–9200, https://doi.org/10.5194/acp-14-9171-2014, 2014.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11, 369–408, https://doi.org/10.5194/gmd-11-369-2018, 2018.

Huang, J., Pan, X., Guo, X. and Li, G.: Health impact of China's Air Pollution Prevention and Control Action Plan: an analysis of national air quality monitoring and mortality data, Lancet Planet. Heal., 2(7), e313–e323, doi:10.1016/S2542-5196(18)30141-4, 2018.

Jerrett, M., Burnett, R. T., Pope, C. A., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E. and Thun, M.: Long-term ozone exposure and mortality, N. Engl. J. Med., 360, 1085–1095, doi:10.1056/NEJMoa0803894, 2009.

Kanaya, Y., Yamaji, K., Miyakawa, T., Taketani, F., Zhu, C., Choi, Y., Komazaki, Y., Ikeda, K., Kondo, Y., and Klimont, Z.: Rapid reduction in black carbon emissions from China: evidence from 2009–2019 observations on Fukue Island, Japan, Atmos. Chem. Phys., 20, 6339–6356, https://doi.org/10.5194/acp-20-6339-2020, 2020.

Katsouyanni, K., Samet, J., Anderson, H.R., Atkinson, R., Le Tertre, A., Medina, S., Samoli, E., Touloumi, G., Burnett, R.T., Krewski, D., Ramsay, T., Dominici, F., Peng, R.D., Schwartz, J., Zanobetti, A.: Air Pollution and Health: A European and North American Approach (APHENA). HEI Research Report 142. Health Effects Institute, Boston, MA, https://www.healtheffects.org/system/files/APHENA142.pdf, 2009.

Krewski, D., Jerrett, M., Burnett, R. T., Ma, R., Hughes, E., Shi, Y., Turner, M. C., Pope III, C. A., Thurston, G., Calle, E. E., and Thun, M. J.: Extended Follow-Up and Spatial Analysis of the American Cancer Society Study Linking Particulate Air Pollution and Mortality, HEI Research Report 140, Health Effects Institute, Boston, MA, 2009.

Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. a., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P. J. and Tyndall, G. K.: CAM-chem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, Geosci. Model Dev., 5, 369–411, doi:10.5194/gmd-5-369-2012, 2012.

Lawrence, M. G., Rolf, von K., Salzmann, M. and Rasch, P. J.: The balance of effects of deep convective mixing on tropospheric ozone, Geophys. Res. Lett., 30(18), 3–6, doi:10.1029/2003GL017644, 2003.

Li, J., Nagashima, T., Kong, L., Ge, B., Yamaji, K., Fu, J. S., Wang, X., Fan, Q., Itahashi, S., Lee, H.-J., Kim, C.-H., Lin, C.-Y., Zhang, M., Tao, Z., Kajino, M., Liao, H., Li, M., Woo, J.-H., Kurokawa, J., Wang, Z., Wu, Q., Akimoto, H., Carmichael, G. R., and Wang, Z.: Model evaluation and intercomparison of surface-level ozone and relevant species in East Asia in the context of MICS-Asia Phase III – Part 1: Overview, Atmos. Chem. Phys., 19, 12993–13015, https://doi.org/10.5194/acp-19-12993-2019, 2019.

Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q. and Bates, K. H.: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China, Proc. Natl. Acad. Sci. U. S. A., 116(2), 422–427, doi:10.1073/pnas.1812168116, 2019.

Li, Y., Lin, C., Lau, A. K. H., Liao, C., Zhang, Y., Zeng, W., Li, C., Fung, J. C. H. and Tse, T. K. T.: Assessing Long-Term Trend of Particulate Matter Pollution in the Pearl River Delta Region Using Satellite Remote Sensing, Environ. Sci. Technol., 49(19), 11670–11678, doi:10.1021/acs.est.5b02776, 2015.

Liang, C.-K., West, J. J., Silva, R. A., Bian, H., Chin, M., Davila, Y., Dentener, F. J., Emmons, L., Flemming, J., Folberth, G., Henze, D., Im, U., Jonson, J. E., Keating, T. J., Kucsera, T., Lenzen, A., Lin, M., Lund, M. T., Pan, X., Park, R. J., Pierce, R. B., Sekiya, T., Sudo, K., and Takemura, T.: HTAP2 multi-model estimates of premature human mortality due to intercontinental transport of air pollution and emission sectors, Atmos. Chem. Phys., 18, 10497–10520, https://doi.org/10.5194/acp-18-10497-2018, 2018.

Lin, C. Q., Liu, G., Lau, A. K. H., Li, Y., Li, C. C., Fung, J. C. H. and Lao, X. Q.: High-resolution satellite remote sensing of provincial $PM_{2.5}$ trends in China from 2001 to 2015, Atmos. Environ., 180 (March), 110–116, doi:10.1016/j.atmosenv.2018.02.045, 2018.

Lin, M., Fiore, A. M., Horowitz, L. W., Cooper, O. R., Naik, V., Holloway, J., Johnson, B. J., Middlebrook, A. M., Oltmans, S. J., Pollack, I. B., Ryerson, T. B., Warner, J. X., Wiedinmyer, C., Wilson, J. and Wyman, B.: Transport of Asian ozone pollution into surface air over the western United States in spring, J. Geophys. Res. Atmos., 117(4), 0–15, doi:10.1029/2011JD016961, 2012.

Lin, M., Horowitz, L. W., Payton, R., Fiore, A. M. and Tonnesen, G.: US surface ozone trends and extremes from 1980 to 2014: Quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, Atmos. Chem. Phys., 17(4), 2943–2970, doi:10.5194/acp-17-2943-2017, 2017.

Liu, C., Chen, R., Sera, F., Vicedo-Cabrera, A. M., Guo, Y., Tong, S., Coelho, M. S. Z. S., Saldiva, P. H. N., Lavigne, E., Matus, P., Ortega, N. V., Garcia, S. O., Pascal, M., Stafoggia, M., Scortichini, M., Hashizume, M., Honda, Y., Hurtado-Díaz, M., Cruz, J., Nunes, B., Teixeira, J. P., Kim, H., Tobias, A., Íñiguez, C., Forsberg, B., Åström, C., Ragettli, M. S., Guo, Y. L., Chen, B. Y., Bell, M. L., Wright, C. Y., Scovronick, N., Garland, R. M., Milojevic, A., Kyselý, J., Urban, A., Orru, H., Indermitte, E., Jaakkola, J. J. K., Ryti, N. R. I., Katsouyanni, K., Analitis, A., Zanobetti, A., Schwartz, J., Chen, J., Wu, T., Cohen, A., Gasparrini, A. and Kan, H.: Ambient particulate air pollution and daily mortality in 652 cities, N. Engl. J. Med., 381(8), 705–715, doi:10.1056/NEJMoa1817364, 2019.

Liu, F., Zhang, Q., Van Der A, R. J., Zheng, B., Tong, D., Yan, L., Zheng, Y. and He, K.: Recent reduction in NO_x emissions over China: Synthesis of satellite observations and emission inventories, Environ. Res. Lett., 11(114002), doi:10.1088/1748-9326/11/11/114002, 2016.

Liu, Y. and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 – Part 1: The complex and varying roles of meteorology, Atmos. Chem. Phys., 20, 6305–6321, https://doi.org/10.5194/acp-20-6305-2020, 2020.

Liu, Y., Dong, X., Wang, M., Emmons, L. K., Liu, Y., Liang, Y., Li, X., and Shrivastava, M.: Analysis of Secondary Organic Aerosol Simulation Bias in the Community Earth System Model (CESM2.1), Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2020-1182, in review, 2020.

Lu, X., Hong, J., Zhang, L., Cooper, O. R., Schultz, M. G., Xu, X., Wang, T., Gao, M., Zhao, Y. and Zhang, Y.: Severe Surface Ozone Pollution in China: A Global Perspective, Environ. Sci. Technol. Lett., 5(8), 487–494, doi:10.1021/acs.estlett.8b00366, 2018.

Lu, X., Zhang, L., Wang, X., Gao, M., Li, K., Zhang, Y., Yue, X. and Zhang, Y.: Rapid Increases in Warm-Season Surface Ozone and Resulting Health Impact in China Since 2013, Environ. Sci. Technol. Lett., doi:10.1021/acs.estlett.0c00171, 2020.

Ma, X., Huang, J., Zhao, T., Liu, C., Zhao, K., Xing, J., and Xiao, W.: Rapid increase in summer surface ozone over the North China Plain during 2013–2019: a side effect of particulate matter reduction control?, Atmos. Chem. Phys., 21, 1–16, https://doi.org/10.5194/acp-21-1-2021, 2021.

McDuffie, E. E., Smith, S. J., O'Rourke, P., Tibrewal, K., Venkataraman, C., Marais, E. A., Zheng, B., Crippa, M., Brauer, M., and Martin, R. V.: A global anthropogenic emission inventory of atmospheric pollutants from sector- and fuel-specific

sources (1970–2017): an application of the Community Emissions Data System (CEDS), Earth Syst. Sci. Data, 12, 3413–3442, https://doi.org/10.5194/essd-12-3413-2020, 2020.

Metzger, S., Dentener, F., Pandis, S., and Lelieveld, J.: Gas/aerosol partitioning: 1. A computationally efficient model, J. Geophys. Res., 107, 4312, doi:10.1029/2001JD001102, 2002.

Parrish, D. D., Lamarque, J.-F., Naik, V., Horowitz, L., Shindell, D. T., Staehelin, J., Derwent, R., Cooper, O. R., Tanimoto, H., Volz-Thomas, A., Gilge, S., Scheel, H.-E., Steinbacher, M., and Frühlich, M.: Long-term changes in lower tropospheric base- line ozone concentrations: Comparing chemistry-climate models and observations at northern midlatitudes, J. Geophys. Res., 119, 5719–5736, doi:10.1002/2013JD021435, 2014.

Paulot, F., Paynter, D., Ginoux, P., Naik, V., and Horowitz, L. W.: Changes in the aerosol direct radiative forcing from 2001 to 2015: observational constraints and regional mechanisms, Atmos. Chem. Phys., 18, 13265–13281, https://doi.org/10.5194/acp-18-13265-2018, 2018.

Pfister, G. G., Parrish, D. D., Worden, H., Emmons, L. K., Edwards, D. P., Wiedinmyer, C., Diskin, G. S., Huey, G., Oltmans, S. J., Thouret, V., Weinheimer, A., and Wisthaler, A.: Characterizing summertime chemical boundary conditions for air masses entering the US West Coast, Atmos. Chem. Phys., 11, 1769–1790, https://doi.org/10.5194/acp-11-1769-2011, 2011.

Pope III, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., and Thurston, G. D.: Lung cancer, cardiopulmonary motality, and long-term exposure to fine particulate air pollution, J. Am. Med. Assoc., 287, 1132–1141, 2002.

Tan, Z., Hofzumahaus, A., Lu, K., Brown, S. S., Holland, F., Huey, L. G., Kiendler-Scharr, A., Li, X., Liu, X., Ma, N., Min, K. E., Rohrer, F., Shao, M., Wahner, A., Wang, Y., Wiedensohler, A., Wu, Y., Wu, Z., Zeng, L., Zhang, Y. and Fuchs, H.: No Evidence for a Significant Impact of Heterogeneous Chemistry on Radical Concentrations in the North China Plain in Summer 2014, Environ. Sci. Technol., 54(10), 5973–5979, doi:10.1021/acs.est.0c00525, 2020.

Seltzer, K. M., Shindell, D. T., Kasibhatla, P., and Malley, C. S.: Magnitude, trends, and impacts of ambient long-term ozone exposure in the United States from 2000 to 2015, Atmos. Chem. Phys., 20, 1757–1775, https://doi.org/10.5194/acp-20-1757-2020, 2020.

Shen, G., Ru, M., Du, W. Zhu, X., Zhong, Q., Chen, Y., Shen, H., Yun, X., Meng, W., Liu, J., Cheng, h., Hu, J., Guan, D., and Tao, S.: Impacts of air pollutants from rural Chinese households under the rapid residential energy transition. Nat Commun 10, 3405, https://doi.org/10.1038/s41467-019-11453-w, 2019.

Shindell, D., Faluvegi, G., Seltzer, K., and Shindell, C.: Quantified, localized health benefits of accelerated carbon dioxide emissions reductions, Nat. Clim. Change, 8, 1–5, https://doi.org/10.1038/s41558-018-0108-y, 2018.

Song, C., He, J., Wu, L., Jin, T., Chen, X., Li, R., Ren, P., Zhang, L. and Mao, H.: Health burden attributable to ambient $PM_{2.5}$ in China, Environ. Pollut., 223(2017), 575–586, doi:10.1016/j.envpol.2017.01.060, 2017.

Stanaway, J. D., Afshin, A., Gakidou, E., Lim, S. S., Abate, D., Abate, K. H., Abbafati, C., Abbasi, N., Abbastabar, H., Abd-Allah, F., Abdela, J., Abdelalim, A., Abdollahpour, I., Abdulkader, R. S., Abebe, M., Abebe, Z., Abera, S. F., Abil, O. Z.,

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

Abraha, H. N., Abrham, A. R., Abu-Raddad, L. J., Abu-Rmeileh, N. M. E., Accrombessi, M. M. K., Acharya, D., Acharya, P., Adamu, A. A., Adane, A. A., Adebayo, O. M., Adedoyin, R. A., Adekanmbi, V., Ademi, Z., Adetokunboh, O. O., Adib, M. G., Admasie, A., Adsuar, J. C., Afanvi, K. A., Afarideh, M., Agarwal, G., Aggarwal, A., Aghayan, S. A., Agrawal, A., Agrawal, S., Ahmadi, A., Ahmadi, M., Ahmadieh, H., Ahmed, M. B., Aichour, A. N., Aichour, I., Aichour, M. T. E., Akbari, M. E., Akinyemiju, T., Akseer, N., Al-Aly, Z., Al-Eyadhy, A., Al-Mekhlafi, H. M., Alahdab, F., Alam, K., Alam, S., Alam, T., Alashi, A., Alavian, S. M., Alene, K. A., Ali, K., Ali, S. M., Alijanzadeh, M., Alizadeh-Navaei, R., Aljunid, S. M., Alkerwi, A., Alla, F., Alsharif, U., Altirkawi, K., Alvis-Guzman, N., Amare, A. T., Ammar, W., Anber, N. H., Anderson, J. A., Andrei, C. L., Androudi, S., Animut, M. D., Anjomshoa, M., Ansha, M. G., Antó, J. M., Antonio, C. A. T., Anwari, P., Appiah, L. T., Appiah, S. C. Y., Arabloo, J., Aremu, O., Ärnlöv, J., Artaman, A., Aryal, K. K., Asayesh, H., Ataro, Z., Ausloos, M., Avokpaho, E. F. G. A., Awasthi, A., Quintanilla, B. P. A., Ayer, R., Ayuk, T. B., et al.: Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks for 195 countries and territories, 1990–2017: A systematic analysis for the Global Burden of Disease Study 2017, Lancet, 1923–1994, https://doi.org/10.1016/S0140-6736(18)32225-6, 2018.

Tie, X., Brasseur, G., Emmons, L., Horowitz, L., and Kinnison, D.: Effects of aerosols on tropospheric oxidants: A global model study, J. Geophys. Res., 106, 22931–22964, 2001.

Tie, X., Madronich, S., Walters, S., Edwards, D. P., Ginoux, P., Ma- howald, N., Zhang, R., Lou, C., and Brasseur, G.: Assessment of the global impact of aerosols on tropospheric oxidants, J. Geo- phys. Res., 110, D03204, doi:10.1029/2004JD005359, 2005.

Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Ma, P.-L., Liu, X., Ghan, S., Bardeen, C., Arnold, S., Deeter, M., Vitt, F., Ryerson, T., Elkins, J. W., Moore, F., Spackman, J. R. and Val Martin, M.: Description and evaluation of tropospheric chemistry and aerosols in the Community Earth System Model (CESM1.2), Geosci. Model Dev., 8(5), 1395–1426, doi:10.5194/gmd-8-1395-2015, 2015.

Tilmes, S., 2016: GEOS5 Global Atmosphere Forcing Data. Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory, Boulder, CO. [Available online at http://rda.ucar.edu/datasets/ds313.0/.] last accessed 20 April, 2020.

Tilmes, S., Lamarque, J. F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., Smith, A. K., Neely, R. R., Conley, A., Vitt, F., Val Martin, M., Tanimoto, H., Simpson, I., Blake, D. R. and Blake, N.: Representation of the Community Earth System Model (CESM1) CAM4-chem within the Chemistry-Climate Model Initiative (CCMI), Geosci. Model Dev., 9(5), 1853–1890, doi:10.5194/gmd-9-1853-2016, 2016.

Tilmes, S., Hodzic, A., Emmons, L. K., Mills, M. J., Gettelman, A., Kinnison, D. E., Park, M., Lamarque, J. F., Vitt, F., Shrivastava, M., Campuzano-Jost, P., Jimenez, J. L., and Liu, X.: Climate Forcing and Trends of Organic Aerosols in the Community Earth System Model (CESM2), Journal of Advances in Modeling Earth Systems, 10.1029/2019ms001827, 2019.

490 Turner, M. C., Jerrett, M., Pope, C. A., Krewski, D., Gapstur, S. M., Diver, W. R., Beckerman, B. S., Marshall, J. D., Su, J., Crouse, D. L. and Burnett, R. T.: Long-Term Ozone Exposure and Mortality in a Large Prospective Study, Am. J. Respir. Crit. Care Med., 193(10), 1134–1142, doi:10.1164/rccm.201508-1633OC, 2016.

UN Environment 2019. A Review of 20 Years' Air Pollution Control in Beijing. United Nations Environment Programme, Nairobi, Kenya, 2019.

495 Wang, W., Parrish, D. D., Li, X., Shao, M., Liu, Y., Mo, Z., Lu, S., Hu, M., Fang, X., Wu, Y., Zeng, L., and Zhang, Y.: Exploring the drivers of the increased ozone production in Beijing in summertime during 2005–2016, Atmos. Chem. Phys., 20, 15617–15633, https://doi.org/10.5194/acp-20-15617-2020, 2020.

Wang, Y., Zhang, Y., Hao, J. and Luo, M.: Seasonal and spatial variability of surface ozone over China: Contributions from background and domestic pollution, Atmos. Chem. Phys., 11(7), 3511–3525, doi:10.5194/acp-11-3511-2011, 2011.

500 West, J. J., Naik, V., Horowitz, L. W. and Fiore, A. M.: Effect of regional precursor emission controls on long-range ozone transport – Part 1: Short-term changes in ozone air quality, Atmos. Chem. Phys., 9(16), 6077–6093, doi:10.5194/acp-9-6077-2009, 2009.

West, J. J., Naik, V., Horowitz, L. W. and Fiore, A. M.: Effect of regional precursor emission controls on long-range ozone transport – Part 2: Steady-state changes in ozone air quality and impacts on human mortality, Atmos. Chem. Phys., 9(16),

505 6095–6107, doi:10.5194/acp-9-6095-2009, 2009b.

Wu, W., Zhang, Y.: Effects of particulate matter ($PM_{2.5}$) and associated acidity on ecosystem functioning: response of leaf litter breakdown. Environ Sci Pollut Res 25, 30720–30727, 2018.

Young, P. J., Archibald, A. T., Bowman, K. W., Lamarque, J.-F., Naik, V., Stevenson, D. S., Tilmes, S., Voulgarakis, A., Wild, O., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V.,

510 Faluvegi, G., Horowitz, L. W., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R. B., Shindell, D. T., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 2063–2090, https://doi.org/10.5194/acp-13-2063-2013, 2013.

Zhai, S., Jacob, D. J., Wang, X., Shen, L., Li, K., Zhang, Y., Gui, K., Zhao, T., and Liao, H.: Fine particulate matter ($PM_{2.5}$)

515 trends in China, 2013–2018: separating contributions from anthropogenic emissions and meteorology, Atmos. Chem. Phys., 19, 11031–11041, https://doi.org/10.5194/acp-19-11031-2019, 2019.

Zhang, L., Jacob, D. J., Boersma, K. F., Jaffe, D. A., Olson, J. R., Bowman, K. W., Worden, J. R., Thompson, A. M., Avery, M. A., Cohen, R. C., Dibb, J. E., Flock, F. M., Fuelberg, H. E., Huey, L. G., McMillan, W. W., Singh, H. B. and Weinheimer, A. J.: Transpacific transport of ozone pollution and the effect of recent Asian emission increases on air quality

520 in North America: An integrated analysis using satellite, aircraft, ozonesonde, and surface observations, Atmos. Chem. Phys., 8(20), 6117–6136, doi:10.5194/acp-8-6117-2008, 2008.

Zhang, L., Jacob, D. J., Yue, X., Downey, N. V., Wood, D. A. and Blewitt, D.: Sources contributing to background surface ozone in the US Intermountain West, Atmos. Chem. Phys., 14(11), 5295–5309, doi:10.5194/acp-14-5295-2014, 2014.

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Zhang, Q., Zheng, Y., Tong, D., Shao, M., Wang, S., Zhang, Y., Xu, X., Wang, J., He, H., Liu, W., Ding, Y., Lei, Y., Li, J., Wang, Z., Zhang, X., Wang, Y., Cheng, J., Liu, Y., Shi, Q., Yan, L., Geng, G., Hong, C., Li, M., Liu, F., Zheng, B., Cao, J., Ding, A., Gao, J., Fu, Q., Huo, J., Liu, B., Liu, Z., Yang, F., He, K. and Hao, J.: Drivers of improved $PM_{2.5}$ air quality in China from 2013 to 2017, Proc. Natl. Acad. Sci. U. S. A., 116(49), 24463–24469, doi:10.1073/pnas.1907956116, 2019.

Zhang, Y., Cooper, O. R., Gaudel, A., Thompson, A. M., Nédélec, P., Ogino, S. Y. and West, J. J.: Tropospheric ozone change from 1980 to 2010 dominated by equatorward redistribution of emissions, Nat. Geosci., 9(12), 875–879, doi:10.1038/ngeo2827, 2016.

Zhang, X., Xu, X., Ding, Y., Liu, Y., Zhang, H., Wang, Y. and Zhong, J.: The impact of meteorological changes from 2013 to 2017 on $PM_{2.5}$ mass reduction in key regions in China. Sci. China Earth Sci. 62, 1885–1902, 2019+.

Zheng, Y., Xue, T., Zhang, Q., Geng, G., Tong, D., Li, X. and He, K.: Air quality improvements and health benefits from China's clean air action since 2013, Environ. Res. Lett., 12(114020), doi:https://doi.org/10.1088/1748-9326/aa8a32, 2017.

Zheng, B., Chevallier, F., Ciais, P., Yin, Y., Deeter, M. N., Worden, H. M., Wang, Y., Zhang, Q. and He, K.: Rapid decline in carbon monoxide emissions and export from East Asia between years 2005 and 2016, Environ. Res. Lett., 13(4), doi:10.1088/1748-9326/aab2b3, 2018a.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K. and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys., 18(19), 14095–14111, doi:10.5194/acp-18-14095-2018, 2018b.

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Atmospheric Chemistry and Physics Discussions

Table 1: Model simulation performed for this study.

Name	Anthropogenic Emissions	Meteorology
CEDS_MEIC	MEIC in China from 2010 to 2017 CEDS outside China from 2010 to 2017[1]	2010 to 2017
CEDS_MEIC_ChinaFix	MEIC in China constant as in 2010 CEDS outside China from 2010 to 2017[1]	2010 to 2017
CEDS_MEIC_GlobalFix	Emissions kept constant at 2010 level from both MEIC and CEDS	2010 to 2017
CEDS_Global	2010-2014 from CEDS globally	2010 to 2014

[1]The global emissions other than in China after 2014 are the same as in 2014 values since it is not available.

545

550

Table 2: Model performance for the annual $PM_{2.5}$ concentration compared with surface observation in China from 2013 to 2017, with mean bias (MB, μg m^{-3}), normalized mean bias (NMB, %), normalized mean error (NME, %), and root-mean-square error (RMSE, μg m^{-3}).

Year	# of stations	MB (μg m^{-3})	NMB (%)	NME (%)	RMSE (μg m^{-3})
2013	378	6.9	9.5	29.4	27.3
2014	537	21.8	34.3	45.6	39.7
2015	1438	21.6	41.3	54.0	39.5
2016	1431	19.2	40.0	54.5	36.5
2017	1461	19.4	42.2	55.7	34.8
2013-2017	5245	19.3	37.2	52.0	26.7
CAM_Chem using CEDS emissions only[1]					
2013[1]	378	0.8	1.0	23.5	20.1
2014[1]	537	19.5	30.1	42.1	34.3
2013-2014[1]	915	11.8	17.5	33.9	29.5

[1]The CAM-chem simulations applying global CEDS emissions only, which only has been ran from 2010 to 2014.

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Atmospheric Chemistry and Physics Discussions

555

Table 3: As Table 2 but for annual MDA8 O_3, with mean bias (MB, ppbv), normalized mean bias (NMB, %), normalized mean error (NME, %), and root-mean-square error (RMSE, ppbv).

Year	# of stations	MB (ppbv)	NMB (%)	NME (%)	RMSE (ppbv)
2013	1029	4.8	11.6	20.2	10.6
2014	1033	7.2	17.5	24.1	12.4
2015	1026	6.0	14.4	23.0	11.9
2016	1031	6.5	15.7	22.7	11.8
2017	1042	3.8	9.3	19.5	10.3
2013-2017	5161	5.7	13.7	21.9	11.4
CAM_Chem using CEDS emissions only[1]					
2013[1]	1029	14.1	33.8	34.5	16.4
2014[1]	1033	16.3	39.2	39.6	18.3
2013-2014[1]	2062	15.2	36.6	37.0	17.4

[1] The CAM-chem simulations applying global CEDS emissions only, which only has been ran from 2010 to 2014.

560

565

Table 4. The changes for the $PM_{2.5}$- mortality burden under the emission changes in China from 2010 to 2017 in China, as well as three other downwind regions—South Korea, Japan and U.S. The mortality burden changes at global level are also included. Positive values mean emission change in China increase the $PM_{2.5}$-related mortality burden in this region, and negative values
570 **mean decreases the $PM_{2.5}$-related mortality burden.**

Year	China	South Korea	Japan	U.S.	Global	% (China/Global)
2011	25,800	98	197	44	27,700	93%
2012	26,200	80	143	24	27,900	94%
2013	12,600	-21	-122	-39	13,300	95%
2014	-19,620	-147	-306	-138	-21,800	90%
2015	-47,670	-226	-541	-191	-51,600	92%
2016	-77,600	-264	-615	-284	-83,200	93%
2017	-100,100	-386	-875	-381	-108,800	92%

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021


Atmospheric Chemistry and Physics Discussions

Table 5. As Table 4 but for ozone-related mortality burden changes.

Year	China	South Korea	Japan	U.S.	Global	% (China/Global)
2011	3,600	23	172	131	4,900	73%
2012	3,400	15	113	140	4,900	70%
2013	5,500	17	115	93	6,600	83%
2014	6,400	25	68	-5	6,500	99%
2015	7,400	22	34	-100	6,500	113%
2016	7,500	11	-56	-188	5,600	133%
2017	8,500	17	-65	-289	5,900	143%

575

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Atmospheric Chemistry and Physics Discussions

Figure 1: National population-weighted (Pop-Weighted) and area-weighted annual $PM_{2.5}$ (a), and MDA8 O_3 (b) from 2010 to 2017 from our base model simulation (CEDS_MEIC). The units are μg m^{-3} for $PM_{2.5}$ and ppbv for ozone.

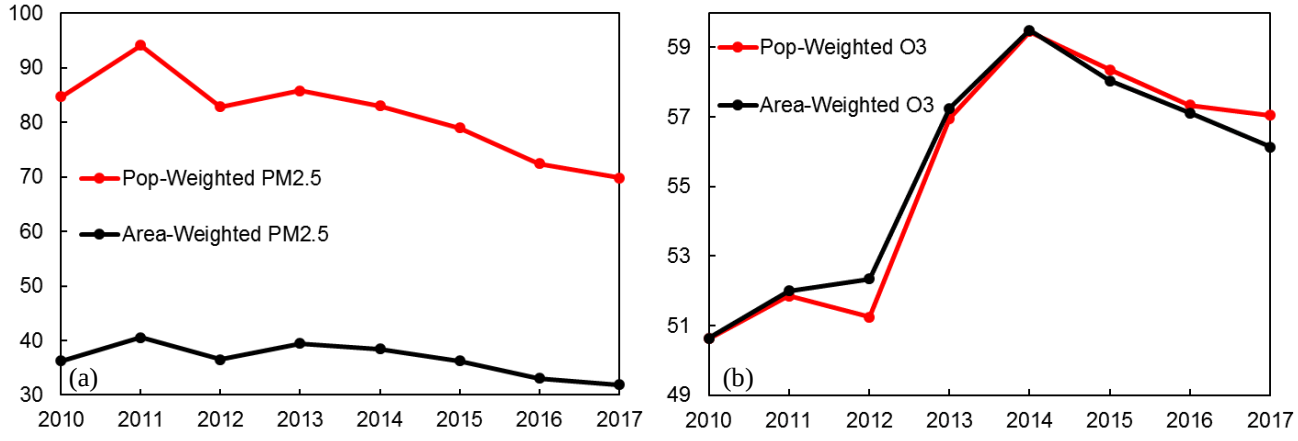
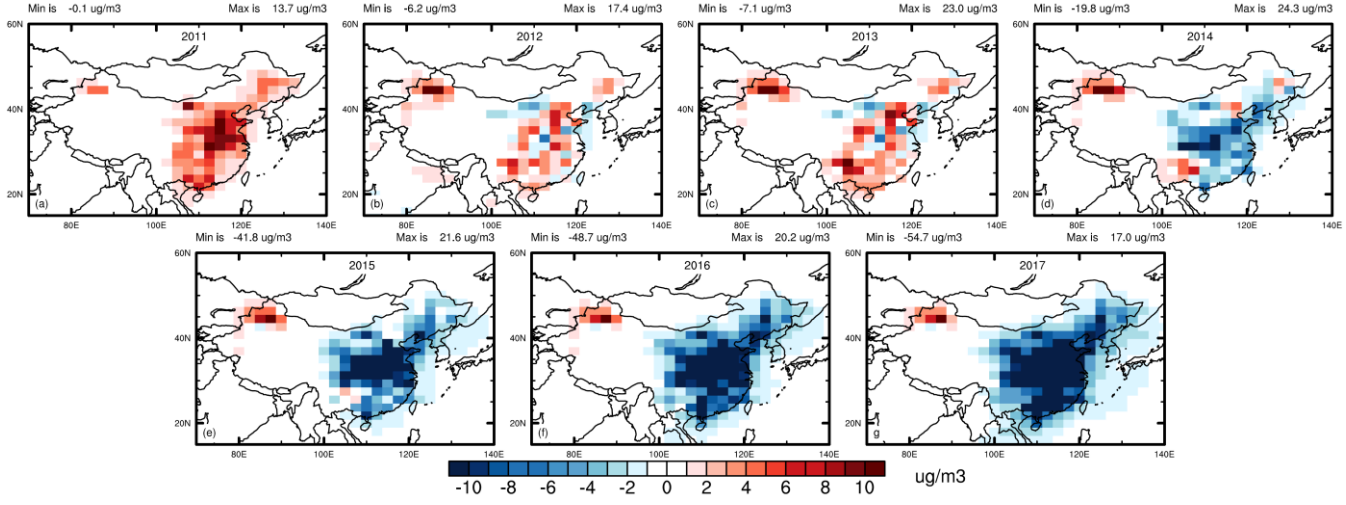


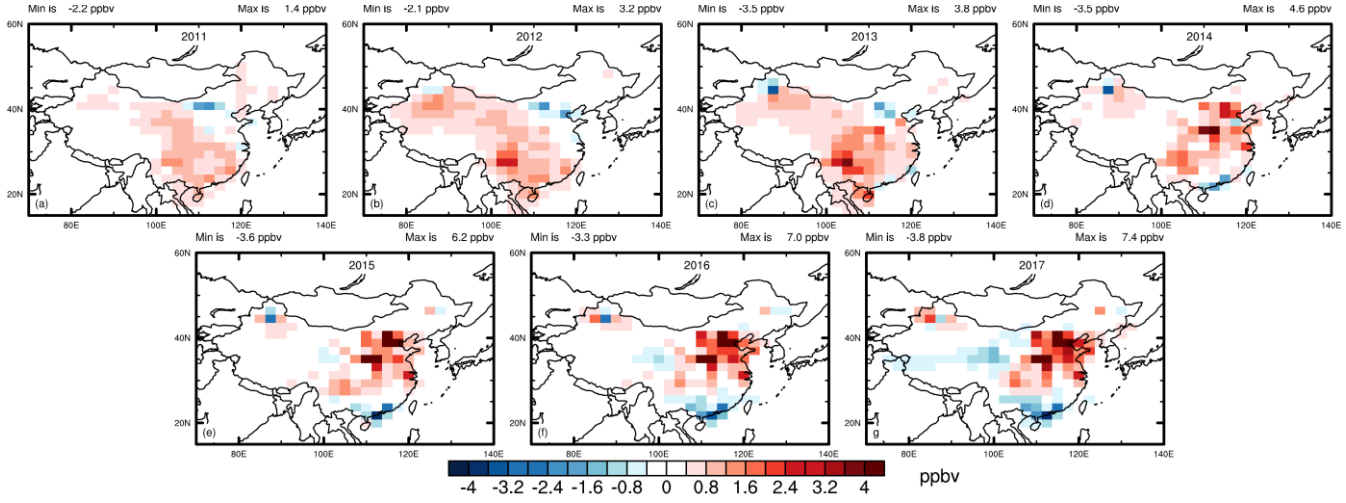
Figure 2: Annual $PM_{2.5}$ changes (unit of μg m^{-3}) from 2011 to 2017 due to anthropogenic emission changes in China only. The results are calculated as the differences between CEDS_MEIC and CEDS_MEIC_ChinaFix for each year.



https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Atmospheric Chemistry and Physics Discussions

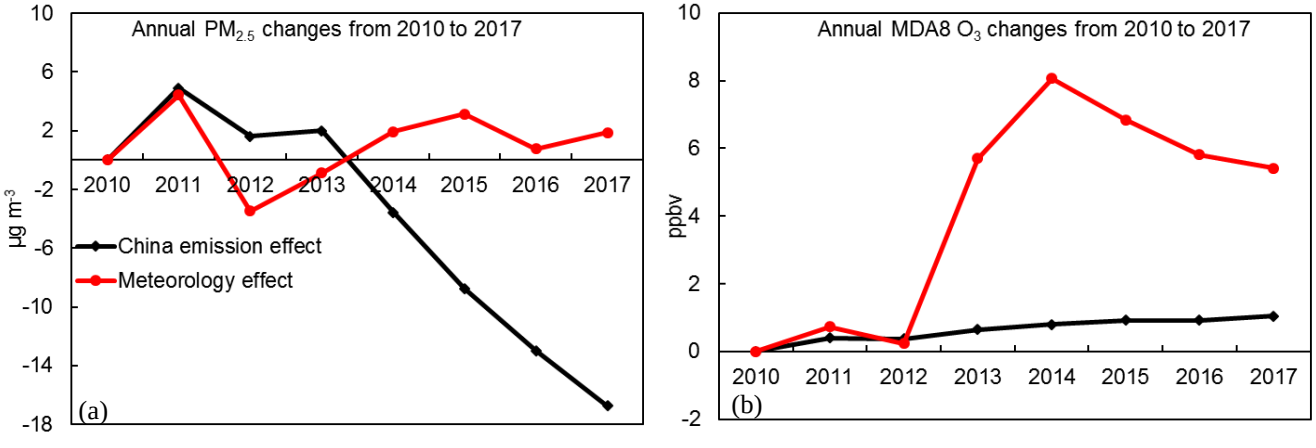
Figure 3: Same as Fig. 2 but for annual MDA8 O_3 changes.



595

600

Figure 4: The annual $PM_{2.5}$ and MDA8 O_3 changes in China due to emissions and meteorological changes from 2010 to 2017.

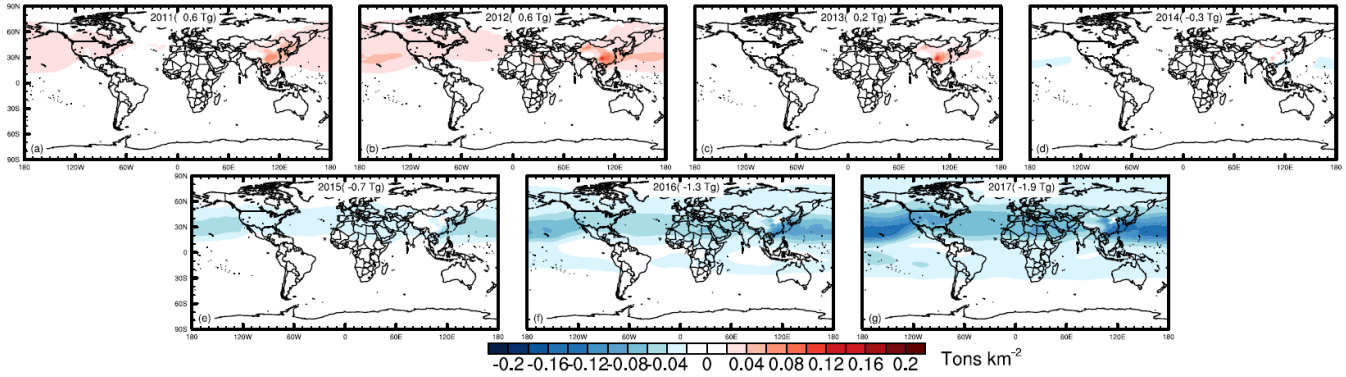


605

https://doi.org/10.5194/acp-2021-385
Preprint. Discussion started: 28 May 2021

Atmospheric Chemistry and Physics Discussions

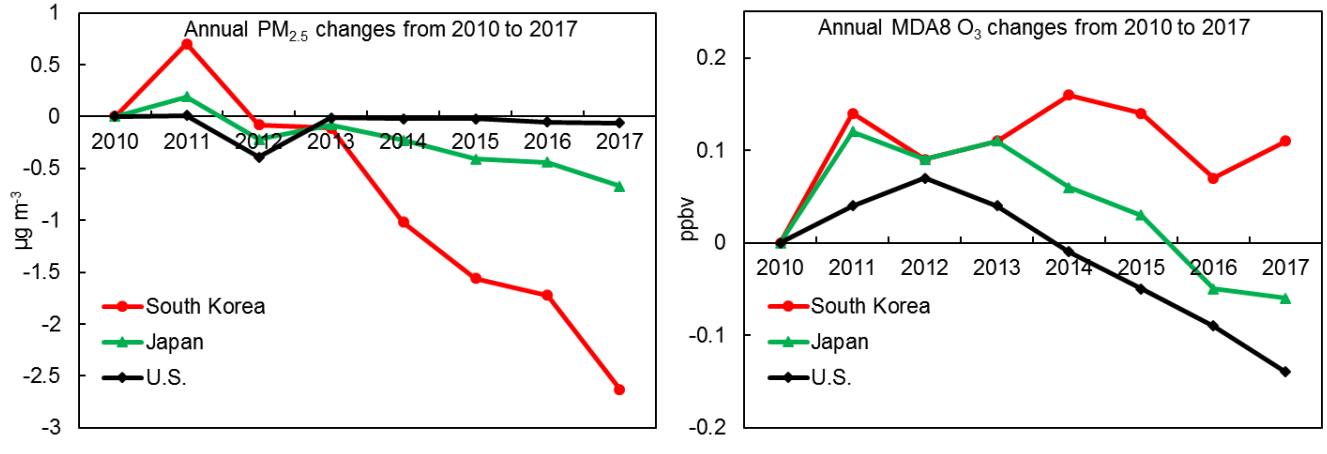
Figure 5. Spatial distribution for global tropospheric ozone burden changes from 2010 to 2017, as a result of emission changes in China.



610

615

Figure 6: The annual population-weighted $PM_{2.5}$ and MDA8 O_3 changes in South Korea, Japan, and U.S. from 2010 to 2017 on caused by Chinese emission changes.



620