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Long-term trends in the ambient $PM_{2.5}$- and O_3-related mortality burdens in the United States under emission reductions from 1990 to 2010

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Abstract. Concentrations of both fine particulate matter ($PM_{2.5}$) and ozone (O_3) in the United States (US) have decreased significantly since 1990, mainly because of air quality regulations. Exposure to these air pollutants is associated with premature death. Here we quantify the annual mortality burdens from $PM_{2.5}$ and O_3 in the US from 1990 to 2010, estimate trends and inter-annual variability, and evaluate the contributions to those trends from changes in pollutant concentrations, population, and baseline mortality rates. We use a fine-resolution (36 km) self-consistent 21-year simulation of air pollutant concentrations in the US from 1990 to 2010, a health impact function, and annual county-level population and baseline mortality rate estimates. From 1990 to 2010, the modeled population-weighted annual $PM_{2.5}$ decreased by 39 %, and summertime (April to September) 1 h average daily maximum O_3 decreased by 9 % from 1990 to 2010. The $PM_{2.5}$-related mortality burden from ischemic heart disease, chronic obstructive pulmonary disease, lung cancer, and stroke steadily decreased by 54 % from 123 700 deaths year^{-1} (95 % confidence interval, 70 800–178 100) in 1990 to 58 600 deaths year^{-1} (24 900–98 500) in 2010. The $PM_{2.5}$-related mortality burden would have decreased by only 24 % from 1990 to 2010 if the $PM_{2.5}$ concentrations had stayed at the 1990 level, due to decreases in baseline mortality rates for major diseases affected by $PM_{2.5}$. The mortality burden associated with O_3 from chronic respiratory disease increased by 13 % from 10 900 deaths year^{-1} (3700–17 500) in 1990 to 12 300 deaths year^{-1} (4100–19 800) in 2010, mainly caused by increases in the baseline mortality rates and population, despite decreases in O_3 concentration. The O_3-related mortality burden would have increased by 55 % from 1990 to 2010 if the O_3 concentrations had stayed at the 1990 level. The detrended annual O_3 mortality burden has larger inter-annual variability (coefficient of variation of 12 %) than the $PM_{2.5}$-related burden (4 %), mainly from the inter-annual variation of O_3 concentration. We conclude that air quality improvements have significantly decreased the mortality burden, avoiding roughly 35 800 (38 %) $PM_{2.5}$-related deaths and 4600 (27 %) O_3-related deaths in 2010, compared to the case if air quality had stayed at 1990 levels (at 2010 baseline mortality rates and population).

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1 Introduction

The 2015 Global Burden of Disease (GBD) study lists air pollution as the fourth-highest ranking global mortality risk factor (GBD 2016). The most recent GBD estimates that exposure to ambient particulate matter (particulate size less than 2.5 µm, $PM_{2.5}$) causes 4.2 million (95 % confidence interval (CI), 3.7–4.8 million) deaths globally, with an additional 254 000 (97 000–422 000) deaths globally caused by exposure to ozone (O_3) (Cohen et al., 2017). For the United States (US), the same study estimated a mortality burden attributable to ambient $PM_{2.5}$ of 88 400 (66 800–115 000) deaths, and 11 700 (4400–19 600) deaths attributable to O_3 in 2015 (Cohen et al., 2017). Adopting similar methods, the US burden of disease study lists ambient $PM_{2.5}$ and O_3 pollution as the 8th and 15th leading risk factors in the US in 2010 (Murray and Collaborators US Burden of Disease, 2013).

Over recent decades, emissions of air pollutants within the US have significantly decreased and air quality has improved. For example, between 1990 and 2010, total US anthropogenic emissions are estimated to have declined by 48 %, 49 %, 67 %, 60 % and 34 %, for NO_x ($= NO + NO_2$), non-methane volatile organic compounds (NMVOCs), SO_2, CO and $PM_{2.5}$, respectively (Xing et al., 2013). EPA observations showed that the US average annual $PM_{2.5}$ concentration declined by 42 % from 2000 to 2016, and maximum daily 8 h average (MDA8) O_3 declined by 22 % from 1990 to 2016 (US EPA, 2017). These air quality improvements were likely mainly driven by ambient air quality standards, and federal and state implementation of stationary and mobile source regulations, especially the 1990 Clean Air Act (CAA) Amendments, the 2002 NO_x State Implementation Plans (SIP) Call, and the Cross-State Air Pollution Rule (Chestnut and Mills, 2005; US EPA, 2011), together with other rules to reduce anthropogenic emissions from light duty, heavy duty, and nonroad vehicles (Fann et al., 2012b; US EPA, 2014). Other changes in energy and emission control technology that occurred concurrently with air quality regulations also helped to improve air quality. These decreased concentrations are expected to have brought substantial benefits for public health in the US, but assessing the health benefits requires quantification of changes in human exposure relating where air quality has improved to how population and baseline mortality are distributed.

Several recent studies have assessed the global (Anenberg et al., 2010; GBD, 2015, 2016; Lelieveld et al., 2015; Silva et al., 2013, 2016b) or national (Fann et al., 2012a; Punger and West, 2013) burdens of disease attributable to air pollution. However, less effort has been made to understand how these burdens evolve over time. Cohen et al. (2017) used estimates of air pollutant concentrations from a combination of air quality model simulations and satellite and surface observations to study the global and national temporal trends of the burdens of disease attributable to ambient $PM_{2.5}$ and O_3 (only model results were used for O_3), at 5-year intervals from 1990 to 2015. Two other studies (Butt et al., 2017; Wang et al., 2017) used coarse-resolution model simulations to study the global/hemispheric $PM_{2.5}$ mortality burdens for the past few decades. Fann et al. (2017) estimated the annual mean $PM_{2.5}$ concentration in the US from monitoring data and its all-cause mortality burden in 1980, 1990, 2000 and 2010. Epidemiological studies have also inferred how health effects have changed through time in the US (Correia et al., 2013; Pope et al., 2009). However, previous studies have not estimated mortality burdens for both $PM_{2.5}$ and O_3 in the US for several years in succession or investigated the different drivers for mortality trends or the inter-annual variability of the mortality burdens. The inter-annual variability analysis indicates whether the mortality burden at a given year, such as the results from Cohen et al. (2017) and Fann et al. (2017), is representative of years around it. In the US, such changes in the air pollution mortality burdens can support decision making on air pollution control policies. For the public, analyzing trends can effectively illustrate the benefits of past air pollution controls, as well as the challenges for future policy efforts.

Here we aim to quantify air pollution-related mortality in the continental US in each year from 1990 to 2010, mainly to understand the trends over this time period. We also analyze the contributions of changes in air pollutant concentrations, population, and baseline mortality rates to the overall trend, and analyze the inter-annual variability in mortality burden estimates. To achieve this, we use a 21-year (1990–2010) model simulation of $PM_{2.5}$ and O_3 concentrations over the continental US (CONUS) from 1990 to 2010. We also use annual county-level baseline mortality rates and population archived by the US Centers for Disease Control (CDC WONDER, https://wonder.cdc.gov/mortSQL.html, last access: 5 October 2018).

2 Methodology

2.1 Air quality simulations

We use simulations of air quality over the CONUS from 1990 to 2010 by Gan et al. (2015, 2016). These simulations used the WRF-CMAQ model which coupled the Weather Research and Forecasting (WRF, v3.4) model and the Community Multiscale Air Quality (CMAQ, v5.02) model (Wong et al., 2012), covering the CONUS at 36 km × 36 km. These simulations used an internally consistent emission inventory from 1990 to 2010 for the US developed by Xing et al. (2013), three-dimensional meteorological fields constrained by reanalysis of available surface and aloft measurements of the atmospheric state, and time-varying lateral boundary conditions provided by the hemispheric CMAQ (Mathur et al., 2017; Xing et al., 2015). The model system used the CB05 chemical mechanism with the AERO6 module for aerosols.

Gan et al. (2015, 2016) showed that the modeled trends of total $PM_{2.5}$ and its components generally matched both the CASTNET (Clean Air Status and Trend Network) and IMPROVE (Interagency Monitoring of Protection Visual) observational data from 1995 to 2010, with correlation coefficients usually larger than 0.87 for total $PM_{2.5}$ and its components. Moreover, the trends for $PM_{2.5}$ and its species were similar in direction and magnitude (Gan et al., 2015). There was a small or nearly no trend for $PM_{2.5}$ in the western US for both the model and observations, but a dramatic decreasing trend in the eastern US, with a larger decreasing trend from the model ($-0.44\,\mu g\,m^{-3}\,year^{-1}$) than from the IMPROVE observations ($-0.30\,\mu g\,m^{-3}\,year^{-1}$).

For O_3, Astitha et al. (2017) used dynamical evaluation methods, and showed that the simulated O_3 trends generally agreed very well with the observed downward trends, especially for the period from 2000 to 2010, albeit underestimating trends over some regions, for both the May to September average of daily maximum 8 h (MDA8) and annual 4th highest O_3. From 2000 to 2010, the regional trends for the 4th highest O_3 from the model (observations) were -0.80 ppbv $year^{-1}$ (-0.73 ppbv $year^{-1}$) for Southwest, -1.14 (-1.53) for Southcentral, -1.31 (-1.66) for Southeast, -1.46 (-1.61) for Midwest, -1.35 (-1.79) for Northeast, and -1.11 (-1.40) for CONUS.

2.2 Mortality burden attributable to ambient air pollution

The mortality burdens attributable to ambient $PM_{2.5}$ and O_3 (ΔMort) are estimated using the health impact function (HIF) following Eq. (1):

$$\Delta\text{Mort} = y_0 \times \text{AF} \times \text{Pop}, \quad (1)$$

where y_0 is the baseline mortality rate for specific diseases, AF is the attributable fraction calculated as $1 - 1/\text{RR}$, with RR as the relative risk of death from a specific disease, and Pop is exposed population age 25 years and greater.

For $PM_{2.5}$, RR is calculated using the integrated exposure–response (IER) model (Burnett et al., 2014), which has been extensively used by recent studies, including Liu et al. (2017), Silva et al. (2016a, b), Wang et al. (2017), and the World Health Organization (2016). The RR is calculated as a function of $PM_{2.5}$ concentration following Eq. (2):

$$\text{for } C < C_0 \text{RR}(C) = 1, \quad (2)$$

$$\text{for } C \geq C_0 \text{RR}(C) = 1 + \alpha \times \left(1 - \exp\left(-\gamma \times (C - C_0)^{\delta}\right)\right), \quad (3)$$

where C is the annual average ambient $PM_{2.5}$ concentration, C_0 is the $PM_{2.5}$ threshold concentration (5.8–8.0 $\mu g\,m^{-3}$), below which no additional risk is assumed, and the parameter values of α, γ, and δ are given by distributions (Burnett et al., 2014). For this study, the RRs are downloaded from the GBD website (Global Health Data Exchange (GHDx), 2013).

For the O_3-related mortality burden, $\text{RR} = \exp^{\beta \Delta X}$, where β is the concentration response factor, and ΔX is the difference in O_3 concentration (summertime 1 h daily maximum O_3) between the current year (1990 to 2010) and the low-concentration threshold. For RR, we use the value of 1.040 (with 95 % CI: 1.013, 1.067) from Jerrett et al. (2009) following recent studies (e.g., Cohen et al., 2017; GBD, 2015, 2016; Lim et al., 2012). Turner et al. (2016) found a larger RR for respiratory mortality (RR, 1.12; 95 % CI, 1.08–1.16) associated with the annual average of MDA8 O_3, and using these results would likely lead to a larger O_3 mortality burden (Malley et al., 2017). We account for all chronic respiratory disease (RESP), to be consistent with Jerrett et al. (2009). The counterfactual concentration of 37.6 ppbv (Lim et al., 2012; Lelieveld et al., 2015) is used in our study, to be comparable with Cohen et al. (2017). We use adults above 25 years, to be comparable with other calculations of $PM_{2.5}$ mortality burden following Silva et al. (2016a, b), even though the estimated RRs from Jerrett et al. (2009) were for adults above 30 years old only. Uncertainties in air pollution-related mortality burden calculations are based on the uncertainty in RRs only, ignoring those in modeled air pollutant concentrations, and population and baseline mortality rates, which may be larger than that from the RRs but are not in the scope of our study.

We use baseline mortality rates from each year to calculate deaths from air pollution in each year, as changes in baseline mortality rates from other socioeconomic determinants are likely more important than changes in deaths from air pollution. To estimate the annual baseline mortality rates (y_0) for each disease associated with $PM_{2.5}$ (chronic obstructive pulmonary disease, COPD; ischemic heart disease, IHD; lung cancer, LC; cerebrovascular disease and ischemic stroke, STROKE) and O_3 (RESP), we acquire US county-level specific causes of mortality data for each year from the National Center for Health Statistics (NCHS) (CDC, 2017). We then aggregate the county-level mortality data to each model grid cell at 36 km × 36 km. The specific causes of mortality data for some counties are sometimes suppressed when the total deaths are lower than 10 $year^{-1}$ to protect privacy (Jian et al., 2016), missing or considered "unreliable" when the total deaths are less than 20 $year^{-1}$, and are corrected following established procedures (BenMAP, 2017; Fann et al., 2017; also see the Supplement).

Definitions of each disease follow the GBD study (Lim et al., 2012; Table S1 in the Supplement). Note that the CDC changed the disease codes from the International Classification of Diseases 9th Revision (ICD 9) in 1998 to ICD10 in 1999, and there were discontinuities in the death counts of specific diseases (Anderson et al., 2001; Anderson and Rosenberg, 2003). To account for the discontinuities, we group the total deaths for each of the five diseases for ICD9 and ICD10 using the results of Anderson et al. (2001) and Anderson and Rosenberg (2003), who reported deaths for 135 specific causes in 1996 for both the ICD9 and ICD10

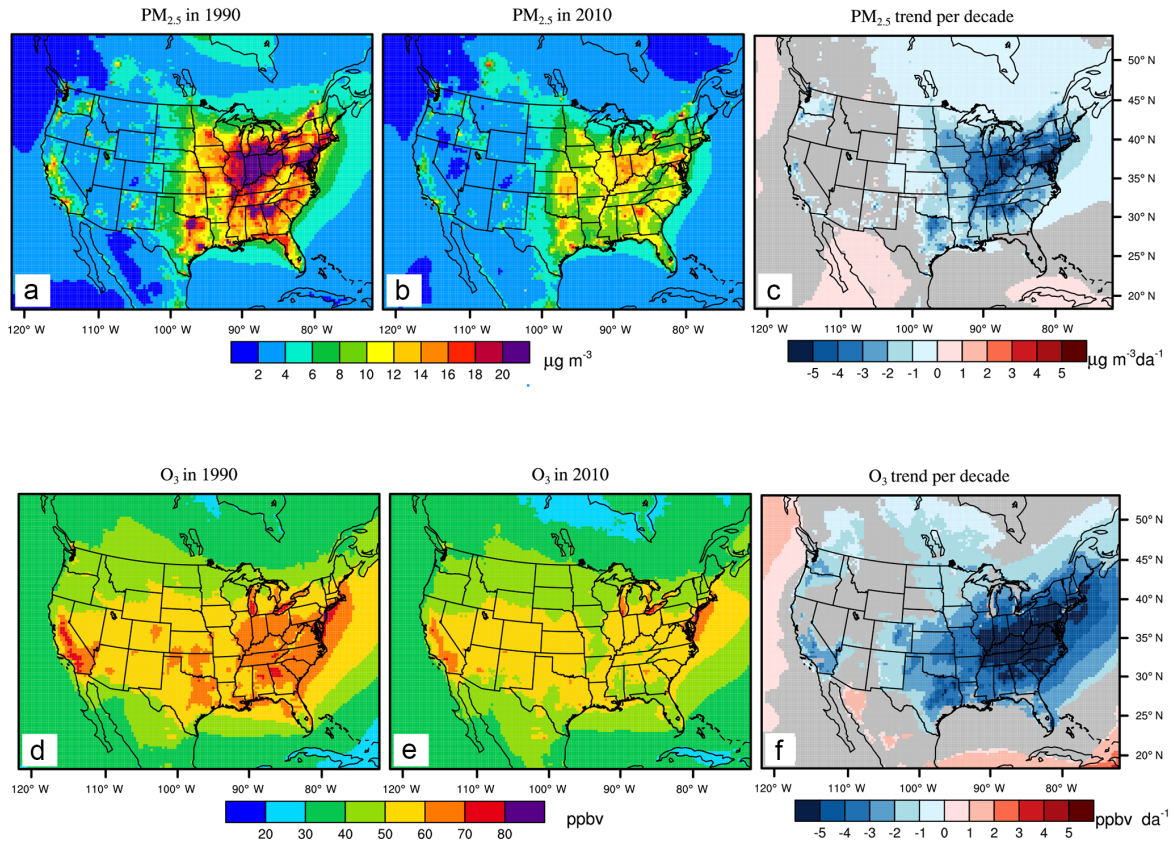

Figure 1. Annual mean $PM_{2.5}$ ($\mu g\,m^{-3}$) in 1990 **(a)** and 2010 **(b)**, and the 21-year trends (**c**, $\mu g\,m^{-3}$ decade^{-1} ($\mu g\,m^{-3}$ decade^{-1})) and summertime average of 1 h daily maximum O_3 in 1990 (ppbv) **(d)** and 2010 **(e)**, and the trend (**f**, ppbv decade^{-1} (ppbv decade^{-1})). The grey shaded areas in panels **c** and **f** indicate trends that are insignificant with p-values for the standard Student's t test larger than 0.05.

codes, and calculate comparability ratios (Table S1). We then recalculate comparability ratios for the five diseases (RESP, COPD, IHD, LC and STROKE) as the ratios of deaths for ICD9 and ICD10 (Table S2). Finally, we apply these ratios to the ICD9 baseline mortality rates from 1990 to 1998.

Annual population in the US at county level was taken from the US Bureau of Census, which reported populations associated with the 1990, 2000, and 2010 censuses and estimated population for each year in between (CDC 2017; https://www.cdc.gov/nchs/data/nvss/bridged_race/Documentation_bridge_postcenv2017.pdf, last access: 5 September 2018). The adult population above 25 years in the US has steadily grown between 1990 and 2010, with an average 1.23 % year^{-1} rate of increase (Fig. S1 in the Supplement).

2.3 The contribution of different factors to mortality trends

The overall trends in $PM_{2.5}$- and O_3-related mortality between 1990 and 2010 are a combination of contributions from trends in population, baseline mortality rates, and concentration. Here we separate the contributions of each factor by assuming that only a single factor was changing from 1990 to 2010, with the other two constant at 1990 levels. For example, the mortality burden change associated with air pollution changes in year y ($\Delta Mort_P^y$), relative to 1990, is calculated following Eq. (4):

$$\Delta Mort_P^y = y_0^{1990} \times AF^y \times Pop^{1990} - y_0^{1990} \times AF^{1990} \times Pop^{1990}. \quad (4)$$

Similarly, we also calculate the mortality burden change without accounting for ambient air pollution changes ($\Delta Mort_{noP}^y$) following Eq. (5):

$$\Delta Mort_{noP}^y = y_0^y \times AF^{1990} \times Pop^y - y_0^{1990} \times AF^{1990} \times Pop^{1990}. \quad (5)$$

3 Results

3.1 Air quality trends

From 1990 to 2010, annual average $PM_{2.5}$ in the model decreases significantly in the eastern US (Fig. 1c), but slightly decreases or even increases in the northwest, southwest and west (Fig. S2 and Table S3; also see Fig. S3 for the US 9 regions defined by the National Oceanic and Atmospheric Administration, Zhang et al., 2016). The dramatic

decreasing trends of $PM_{2.5}$ in the eastern US were also reported in previous studies (Gan et al., 2015; Xing et al., 2015) due to emission reductions. The increasing trend in the western central area is due in part to frequent wildfires (Dennison et al., 2014; Hand et al., 2013, 2014; Jaffe et al., 2008; Murphy et al., 2011; Spracklen et al., 2007). In general, the decadal decreasing trends in the east are larger than 2 $\mu g\,m^{-3}$ $decade^{-1}$ from 1990 to 2010, especially in the central area ($-3.48\,\mu g\,m^{-3}$ $decade^{-1}$) and northeast ($-3.14\,\mu g\,m^{-3}$ $decade^{-1}$). The summertime average of 1 h daily maximum O_3 decreases significantly in the central and eastern US, generally at a rate greater than 4 ppbv $decade^{-1}$. It also decreases in the western US, but at a much smaller rate than in the east, generally less than 1 ppbv $decade^{-1}$ (Fig. 1f; Table S3).

In Fig. 2, both the spatial average and population-weighted average (PWA) annual $PM_{2.5}$ exhibit smooth decreasing trends (Fig. 2, top): the spatial average of annual $PM_{2.5}$ has decreased by 29 %, from 9.07 $\mu g\,m^{-3}$ in 1990 to 6.45 $\mu g\,m^{-3}$ in 2010, with a decadal rate of decrease of 1.1 $\mu g\,m^{-3}$ $decade^{-1}$. The corresponding PWA $PM_{2.5}$ decreases by 39 %, from 17.61 $\mu g\,m^{-3}$ in 1990 to 10.73 $\mu g\,m^{-3}$ in 2010, with a decadal decreasing rate of 3.2 $\mu g\,m^{-3}$ $decade^{-1}$. Years with high $PM_{2.5}$, such as in 1994, 1996, and 2000, are mainly caused by increases in organic carbon due to large wildfires in the western US (Spracklen et al., 2007). Both the spatial average and PWA O_3 also exhibit decreasing trends over the past 2 decades, with greater inter-annual variability resulting from meteorological variability (Porter et al., 2017). The spatial average O_3 concentration decreases by 9 %, from 55.02 ppbv in 1990 to 49.99 ppbv in 2010, decreasing at a rate of 2.4 ppbv $decade^{-1}$. The PWA O_3 also decreased by 9 %, from 58.96 ppbv in 1990 to 53.57 ppbv in 2010, decreasing at a rate of 3.0 ppbv $decade^{-1}$. We also calculate the air quality and mortality burden trends separately for two 11-year periods, 1990 to 2000 and 2000 to 2010, following Astitha et al. (2017). Both $PM_{2.5}$ and O_3 decrease more strongly in the second decade than in the first decade for both spatial average and PWA (Table S5), consistent with previous findings (Astitha et al., 2017; Gan et al., 2015; Porter et al., 2017; Xing et al., 2015).

We then calculate trends in the number of days annually that exceed the daily $PM_{2.5}$ standard (35 $\mu g\,m^{-3}$), and the daily MDA8 O_3 standard (70 ppbv) (Fig. S4). The exceedance days decrease for both $PM_{2.5}$ and O_3, especially in the eastern US. In 2010, fewer than 5 days exceed the air quality standard for the majority of the US (Fig. S4b, e). We also calculate the population exposure exceedances by multiplying the population (adults >25 years old) by the number of air quality exceedance days in each grid cell. The $PM_{2.5}$ population exposure exceedances have decreased from 5340 million people-days in 1990 to 1042 million people-days in 2010, and the O_3 population exposure exceedances has decreased from 4691 million people-days in 1990 to 2236 million people-days in 2010 (Fig. S1). These decreases in population exposure exceedances occur despite population growth over this period.

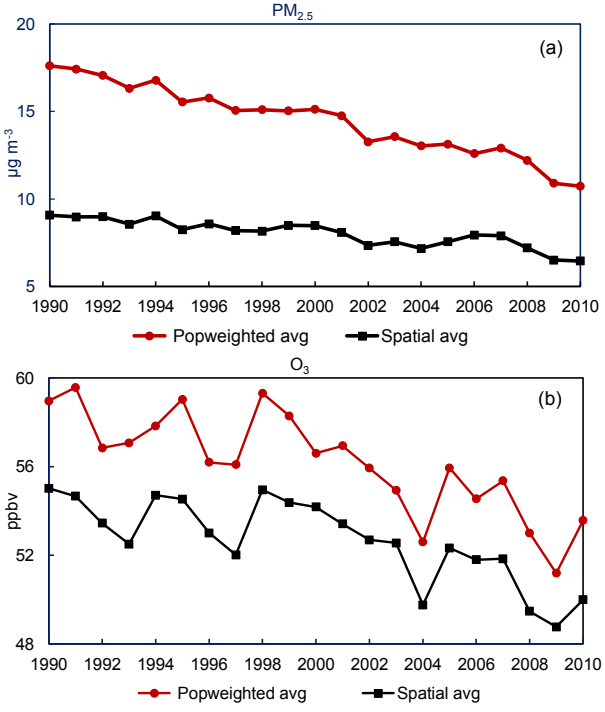

Figure 2. Population-weighted average (Popweighted avg) and spatial average over CONUS land areas of annual average $PM_{2.5}$ **(a)** and summertime average of 1 h daily maximum O_3 **(b)** concentration from 1990 to 2010. Population-weighted average concentrations are based on population in each year. Using the same population in each year yields estimates of population-weighted concentrations that are only slightly different (not shown).

3.2 Mortality burden trends and contributing factors

The mortality burdens associated with exposure to ambient $PM_{2.5}$ in the US steadily decreased by 54 %, from 123 700 (95 % confidence interval considering the uncertainty in relative risk only, 70 800–178 100) deaths $year^{-1}$ in 1990 to 58 600 (24 900–98 500) deaths $year^{-1}$ in 2010 (Fig. 3). The leading cause of $PM_{2.5}$-related mortality is IHD, which decreases by 55 %, from 96 500 (62 600–132 500) deaths $year^{-1}$ in 1990 to 43 600 (21 500–68 700) deaths $year^{-1}$, followed by LC, which has decreased by 44 %, from 12 500 (2500–21 000) deaths $year^{-1}$ in 1990 to 7000 (900–13 400) deaths $year^{-1}$ in 2010 (Table S4). The $PM_{2.5}$ mortality burden per 100 000 adults is much higher in the east than the west for both 1990 and 2010 (Fig. 4), due to the higher $PM_{2.5}$ concentrations (Fig. 1).

Table 1 shows the mortality burdens for $PM_{2.5}$ and O_3 in 2010, and also the burden changes since 1990 from different contributing factors. From the table, we see that the $PM_{2.5}$-

Table 1. The total mortality burdens in 2010 and the burdens in 2010 due to changes since 1990 in each of three factors (concentration, baseline mortality rates and population) and where the concentration change is excluded, for $PM_{2.5}$ and O_3, and the relative changes between 2010 and 1990. The relative changes are calculated as (2010–1990) / 1990. The mortality burdens in the US for $PM_{2.5}$ and O_3 in 1990 are 123 700 deaths year^{-1} (70 800–178 100) and 10 900 deaths year^{-1} (3700–17 500).

		2010 (deaths year^{-1})	Relative changes
$PM_{2.5}$	Mortality burden	58 600 (24 900–98 500)	−54 %
	Concentration change only	78 900 (35 700–129 200)	−36 %
	Mortality rate change only	68 300 (35 800–101 300)	−45 %
	Population change only	173 500 (99 900–250 000)	40 %
	Concentration change excluded	94 400 (50 300–140 000)	−24 %
O_3	Mortality burden	12 300 (4 100–19 800)	13 %
	Concentration change only	8100 (2700–13 100)	−25 %
	Mortality rate change only	13 100 (4400–21 000)	20 %
	Population change only	14 100 (4800–22 700)	30 %
	Concentration change excluded	16 900 (5700–27 000)	55 %

related mortality burden in 2010 would have decreased by only 24 % (94 400 deaths year^{-1} in 2010, 95 %CI, 50 300–139 800) compared with that in 1990, if the $PM_{2.5}$ concentrations had stayed constant over the period 1990–2010, due to decreases in the baseline mortality rates for the specific causes of death that $PM_{2.5}$ influences (Fig. 3), especially IHD (Fig. 5), despite the population increase. Therefore, the reduction in $PM_{2.5}$ concentrations from 1990 to 2010 significantly accelerates the decrease in the mortality burden. The decreased $PM_{2.5}$ concentration avoided roughly 35 800 (38 %) $PM_{2.5}$-related deaths in 2010, compared to the case if current air quality stays at level in 1990 (estimated as the 2010 mortality burden minus the "concentration change excluded" case in 2010). The benefit of the decreased $PM_{2.5}$ concentration could also be estimated as the "concentration change only" case in Fig. 3, yielding 78 900 (35 700–129 200) deaths year^{-1} in 2010, decreasing by 36 % (−44 800 deaths year^{-1}) compared with 1990. The population increases from 1991 to 2010 would lead to increases in the $PM_{2.5}$ mortality burden, but that increase is smaller than the combined reduction from decreasing $PM_{2.5}$ concentrations and baseline mortality rates (Figs. S5 and S6).

When separating the two 11-year periods, the $PM_{2.5}$-related mortality burden decreased by 45 % from 2000 to 2010 (decreasing trend of −4400 deaths year^{-1}), much higher than the 15 % decrease from 1990 to 2000 (decreasing trend of −2100 deaths year^{-1}) (Table S5). The detrended annual $PM_{2.5}$-related mortality burden has a coefficient of variation (CV, standard deviation divided by the average) of 4 %, mainly caused by inter-annual variation in $PM_{2.5}$ concentrations (Table S6 and Fig. S6).

We also calculate burdens and trends for each state individually (Table 2). The three states with the highest $PM_{2.5}$ mortality burden in 1990 are New York (NY, 13 700 deaths year^{-1}), California (CA, 9500 deaths year^{-1}) and Pennsylvania (PA, 9200 deaths year^{-1}); and in 2010, NY (5100 deaths year^{-1}), Texas (TX, 4200 deaths year^{-1}) and Ohio (OH, 3900 deaths year^{-1}). NY has seen the largest benefits of mortality burden decreases (−8500 deaths year^{-1}), followed by CA (−6100 deaths year^{-1}) and PA (−5500 deaths year^{-1}). For the relative mortality burden changes, generally large percent decreases in $PM_{2.5}$-related mortality are seen in western, northern, and northeastern states (including Nevada, Utah, Colorado, Montana, Maine and Vermont) (Fig. 6), because the $PM_{2.5}$ concentrations in

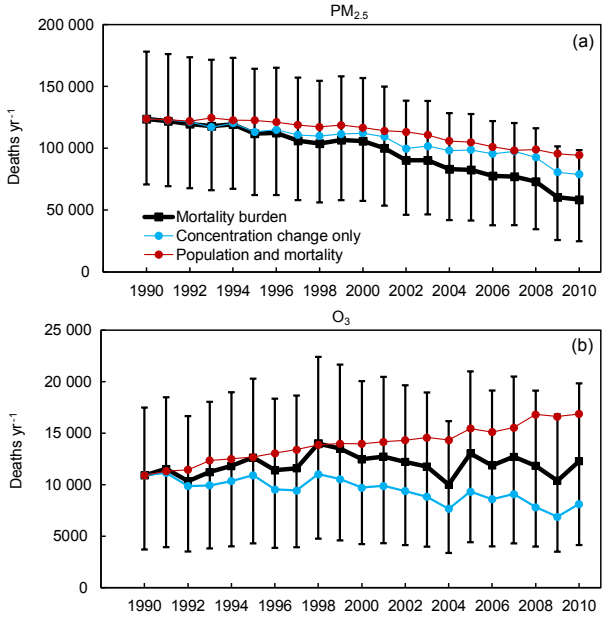

Figure 3. Trends in the total mortality burden (black) for $PM_{2.5}$ (**a**, as a total of ischemic heart disease (IHD) + stroke (STROKE) + chronic obstructive pulmonary disease (COPD) + lung cancer (LC)) and O_3 (**b**, chronic respiratory disease (RESP)), and mortality burdens considering the air quality change only (blue), and with air quality changes excluded (red). Units are deaths $year^{-1}$. The error bars are the 95 % CI for the total mortality burden (black).

2010 are very low or even fall below the low-concentration threshold in these states (Fig. 1), as confirmed by the mortality burden changes from concentration changes alone (Table S7). For other states in the eastern US with large relative mortality burden changes, the contributing factors are different. For example, for Connecticut, the relative mortality burden changes from the decrease in $PM_{2.5}$ concentration are larger than that from the decrease in the baseline mortality rates. However, for Massachusetts, NY and PA, the decreases in baseline mortality rates have a slightly larger effect than that from the decrease in $PM_{2.5}$ concentration. For CA, the effects from the decrease in baseline mortality rates and $PM_{2.5}$ concentration are comparable (Table S7).

The mortality burden associated with exposure to O_3 from RESP has increased by 13 %, from 10 900 (3700–17 500) deaths $year^{-1}$ in 1990 to 12 300 (4100–19 800) deaths $year^{-1}$ in 2010 (Fig. 3). The O_3 mortality burden per 100 000 adults is highest in the midwest and southwest (Fig. 4). The O_3-related mortality burden in 2010 would have increased by 55 % (10 600 deaths $year^{-1}$ in 2010, 95 % CI, 3600–17 100) compared with that in 1990 if the O_3 concentration had stayed constant over the period 1990–2010 (Fig. 3), due to increases in both population and baseline mortality rates (Fig. S5). The decreased O_3 concentration

Table 2. The mortality burden for 48 US states and the District of Columbia in 1990 and 2010, and the absolute changes from 1990 to 2010. Units are deaths $year^{-1}$.

	$PM_{2.5}$-related mortality			O_3-related mortality		
States	1990	2010	Diff	1990	2010	Diff
AL	2135	1166	−969	159	238	−12
AR	1127	752	−375	74	133	22
AZ	554	196	−358	125	329	138
CA	9515	3420	−6095	567	1272	359
CO	222	35	−187	115	230	64
CT	1795	458	−1337	93	129	−22
DC	250	157	−92	12	21	−6
DE	492	264	−227	26	54	14
FL	4688	2441	−2246	483	774	34
GA	3149	1954	−1195	221	413	51
IA	1500	756	−743	74	102	1
ID	174	120	−54	18	39	14
IL	7770	3547	−4223	280	500	38
IN	3821	2067	−1754	198	360	71
KS	1064	697	−367	84	147	26
KY	2420	1388	−1032	160	257	23
LA	1752	855	−898	109	195	4
MA	3417	1107	−2310	153	197	−57
MD	2893	1713	−1180	155	261	1
ME	347	5	−341	28	21	−19
MI	5894	2590	−3304	220	407	46
MN	1626	699	−927	61	107	16
MO	3135	1906	−1229	175	286	31
MS	1352	608	−743	75	124	6
MT	9	2	−7	12	19	1
NC	3321	1961	−1361	208	430	70
ND	75	23	−52	8	12	−1
NE	535	257	−278	55	81	7
NH	453	73	−380	24	25	−13
NJ	5332	2196	−3137	223	404	28
NM	245	180	−65	37	109	42
NV	10	0	−10	52	138	60
NY	13 712	5239	−8473	406	613	−88
OH	7876	3932	−3944	400	690	103
OK	1499	1058	−441	120	248	77
OR	633	219	−413	39	42	−15
PA	9238	3727	−5511	393	584	−70
RI	630	172	−457	32	44	−5
SC	1673	974	−699	109	218	30
SD	140	68	−72	14	23	3
TN	3097	1895	−1202	199	317	13
TX	6499	4178	−2321	417	896	228
UT	107	10	−96	25	72	29
VA	2806	1592	−1214	183	336	29
VT	196	14	−182	11	8	−9
WA	917	394	−522	71	75	−27
WI	2479	977	−1503	75	148	35
WV	1161	534	−627	84	122	−9
WY	1.2	0.4	−0.8	12	25	8

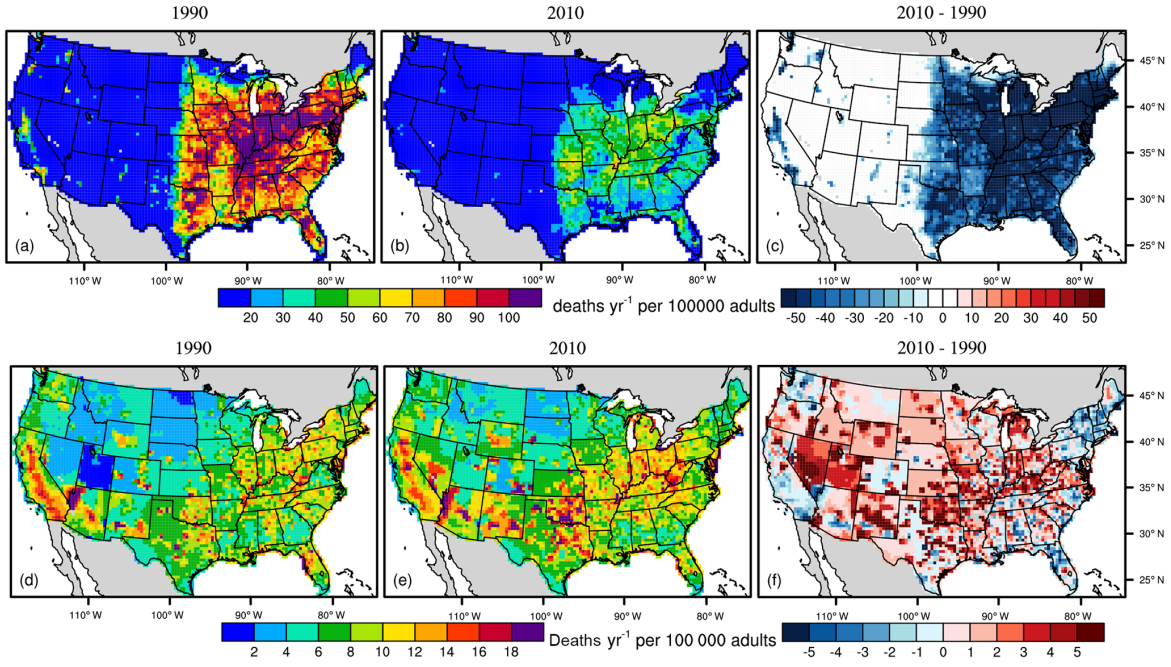

Figure 4. The mortality burdens associated with $PM_{2.5}$ **(a, b)**, O_3 **(d, e)** in 1990 **(a, c)** and 2010 **(b, d)**, and the differences (2010 minus 1990) **(c, f)** for each 36 km × 36 km grid cell. Units are deaths year^{-1} per 100 000 adults (above 25 years old).

would have avoided roughly 4600 (27 %) O_3-related deaths in 2010, compared to the case if ozone concentrations stay at level in 1990 (estimated as the 2010 mortality burden minus the "concentration change excluded" case in 2010). The benefit of the decreased O_3 concentration could also be estimated as the "concentration change only" case in Fig. 3, yielding 8100 (2700–13 100) deaths year^{-1} in 2010, decreasing by 25 % (−2800 deaths year^{-1}) compared with 1990. The change in O_3 generally reduces the mortality burden relative to 1990 with some inter-annual variation (Fig. S6) due to meteorology and wildfires (Porter et al., 2017), while the increases in population and baseline mortality rates generally increase the mortality burden, with a larger contribution from the population change (Fig. S6).

When separating the O_3 mortality trends into 2 decades, we find that the burdens decrease slightly (−70 deaths year^{-1}) from 2000 to 2010, compared with the increasing trend from 1990 to 2000 (240 deaths year^{-1}) (Table S5). The increasing trend in the first decade is caused by the combined effect of increases in baseline mortality rates and population, while the decreasing trend in the second decade is dominated by decreases in O_3 concentration (Fig. S6). The inter-annual variability for the detrended annual O_3 mortality burden from 1990 to 2010 (CV of 12 %) is larger than $PM_{2.5}$ (CV of 4 %), caused mainly by variations in O_3 concentrations from 1990 to 2010 (Table S6).

The three states with the highest O_3 mortality burden in 1990 are CA (910 deaths year^{-1}), Florida (FL, 740 deaths year^{-1}) and NY (700 deaths year^{-1}); and in 2010, CA (1270 deaths year^{-1}), TX (900 deaths year^{-1}) and FL (770 deaths year^{-1}) (Table 2). CA has seen the largest O_3 mortality burden increases (360 deaths year^{-1}), followed by TX (230 deaths year^{-1}) and Arizona (AZ, 140 deaths year^{-1}), with the greatest decrease in NY (−90 deaths year^{-1}). For the relative mortality burden changes, large percent decreases in O_3-related mortality are seen in the northwestern (Washington and Oregon) and northeastern US (Fig. 6), mainly caused by significant O_3 decreases (Table S7), while the greatest percent increases occur in the southwestern US driven mainly by large population increases, and also the baseline mortality rate increases.

Previous health impact assessments have used national baseline mortality rates (Cohen et al., 2017; Silva et al., 2016a, b, etc.), but baseline mortality rates can vary strongly within individual counties (Fig. 5; Dwyer-Lindgren et al., 2016). We performed sensitivity analyses by applying the national average baseline mortality rates for each disease to every county in the mortality burden calculations. We find that the $PM_{2.5}$ mortality burden calculated from the national average baseline mortality rates is lower than those calculated from the county-level baseline mortality rates, ranging among individual years from −2.2 % to −1.3 % (Table S8). For the O_3 mortality burden, the differences between using the national average baseline mortality rates and our best estimates range from −1.1 % to 2.0 % (Table S8). However, using the national average baseline mortality rates fails to capture regional mortality burden hotspots for both $PM_{2.5}$ and

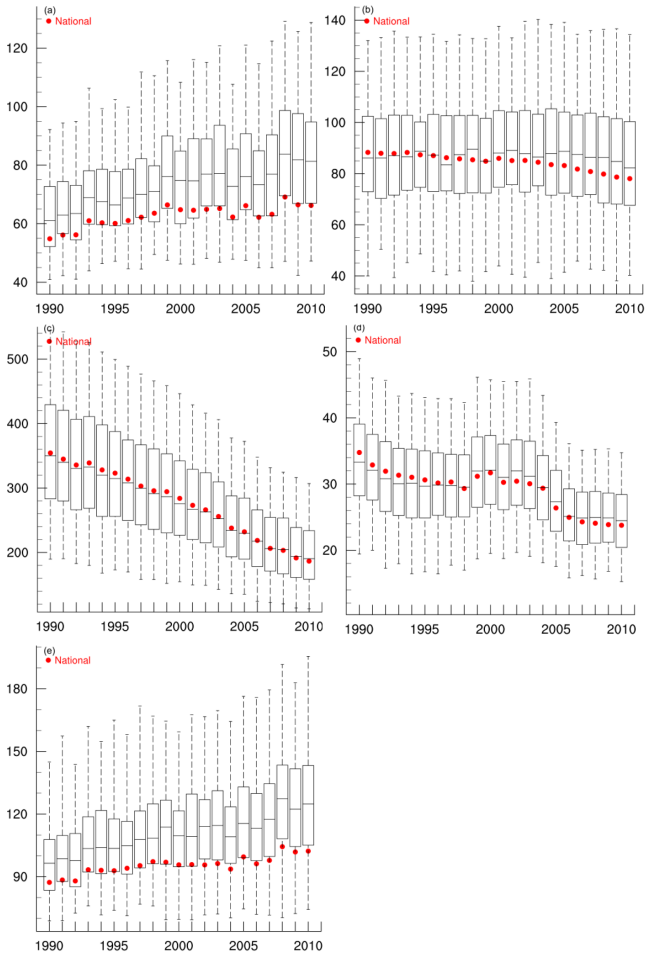

Figure 5. The baseline mortality rates for specific causes of death related to $PM_{2.5}$, including chronic obstructive pulmonary disease **(a)**, lung cancer **(b)**, ischemic heart disease **(c)** and stroke **(d)**, and respiratory diseases related to O_3 **(e)**. The bottom whiskers, bottom border, middle line, top border and top whiskers of the boxes indicate the 5th, 25th, 50th, 75th, and 95th percentiles, respectively, across all counties; the red circles are the national average rate. Baseline mortality rates are shown for 1990–1998 after they are corrected to ensure comparability between ICD9 and ICD10 codes. The units on the *y* axis are per 100 000 people.

O_3 (Figs. S7–S8), demonstrating the value of using county-level baseline mortality rates where possible.

3.3 Comparison with previous studies

The mortality burden associated with $PM_{2.5}$ calculated in our study generally aligns with several previous findings (Fig. 7; also Table S9). Our $PM_{2.5}$ mortality burden is higher than that reported by Cohen et al. (2017) in 1990 (17 % higher) and 1995 (4 % higher), and lower in 2000 (−0.5 %), 2005 (−17 %) and 2010 (−30 %) (Fig. 7). The overestimation of $PM_{2.5}$ mortality burdens in the early 2000s are likely due to the higher population-weighted $PM_{2.5}$ concentration simulated by WRF-CMAQ (Fig. 2), compared with Cohen et al. (2017), in which they estimated the $PM_{2.5}$ concentration based on data fusion of air quality model outputs, satellite retrievals and ground observations. The lower mortality burdens in the second decade (from 2000 to 2010) in our study likely reflect that Cohen et al. (2017) included hemorrhagic stroke and lower respiratory infections in the $PM_{2.5}$-related mortality burden, in addition to COPD, LC, IHD and STROKE, and used an updated integrated exposure–response function. While the absolute value is similar, our results show a stronger decreasing trend (-3000 deaths year^{-1}) than Cohen et al. (2017) (-1000 deaths year^{-1}), which may result from the overestimation of $PM_{2.5}$ decreasing trends in our model relative to ground observations (Gan et al., 2016). The $PM_{2.5}$ mortality burdens estimated in our study are much lower than those from Fann et al. (2017), but the temporal patterns are similar, mainly because Fann et al. (2017) estimated the total all-cause mortality with a different HIF.

To compare with Cohen et al. (2017), who reported the O_3 mortality burden from the COPD, which is a subset of RESP, we recalculate the O_3 mortality burden from the COPD (Table S4). The newly calculated O_3 mortality burden from the COPD is generally lower than the estimate of Cohen et al. (2017) by 8 %–30 % (Fig. 7). This could be caused by the fact that for the O_3 changes, we use the summertime (April to September) average of the 1 h daily maximum, while Cohen et al. (2017) used the 3-month average, which will be higher. The temporal trend for the O_3 mortality burdens from our study is similar to that from Cohen et al. (2017), except that the burden decreases after 2005 in our study but increases in Cohen et al. (2017). The O_3 mortality burden from the RESP disease in 2005 estimated from our study is much lower than two previous studies (Fann et al., 2012a; Punger and West, 2013; Table S10). As discussed in the methods, the lower US background O_3 concentration used in these two studies (22 ppb in the eastern US and 30 ppb in the western US) could lead to a higher O_3 mortality burden. We then did sensitivity analysis by using the pre-industrial O_3 concentration simulated by an ensemble of model outputs from the Atmospheric Chemistry and Climate Model Intercomparison Project (Lamarque et al., 2013; see Fig. S9 and Sect. 2 in the Supplement) as the counterfactual risk exposure factor, and recalculated the O_3 mortality burden with RESP. The new calculated O_3 mortality burdens are estimated to be 64 %–100 % higher than the current estimation from RESP using the low-concentration threshold (Table S10). In Fig. 7, we see that the new estimated O_3 mortality burden from RESP in 2005 (dashed line) is now comparable with the two previous studies.

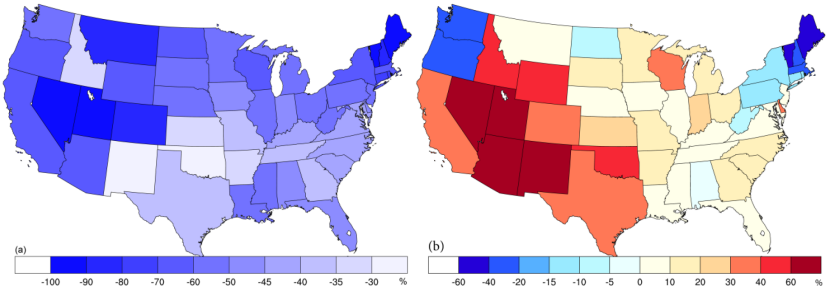

Figure 6. Relative mortality burden changes from 1990 to 2010 for the 48 states and the District of Columbia for $PM_{2.5}$ **(a)** and O_3 **(b)**. The relative changes are calculated as (2010–1990)/1990 × 100 %. Note the different color scales for the two plots. The values for the District of Columbia are −37 % for $PM_{2.5}$ and −23 % for O_3.

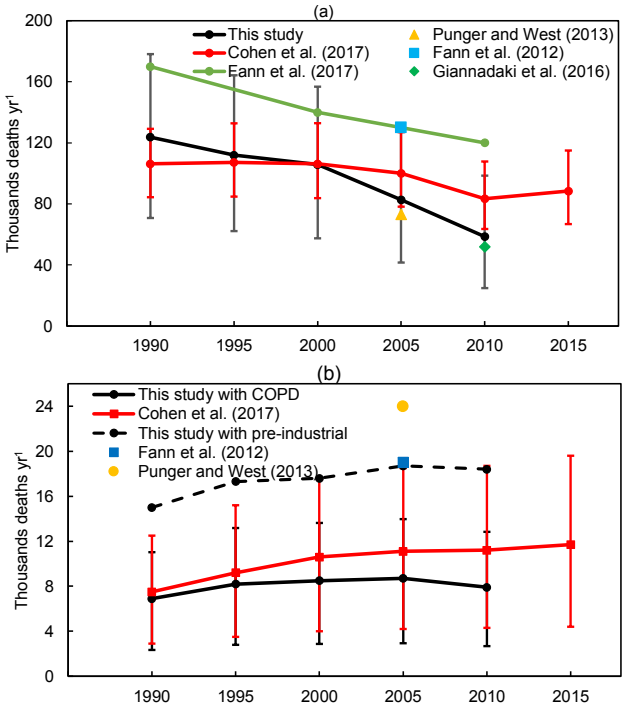

Figure 7. Comparisons of the US mortality burdens attributed to $PM_{2.5}$ **(a)** and O_3 **(b)** in this study, with Cohen et al. (2017), Fann et al. (2012a, 2017), Punger and West (2013), and Giannadaki et al. (2016). The black line for O_3 is the recalculated O_3 mortality burden from the COPD, and the black dashed line is the recalculated O_3 mortality burden from RESP using the pre-industrial O_3 concentration as the counterfactual risk exposure factor. The error bars show the 95 % CI from the RRs, shown for this study and Cohen et al. (2017).

4 Conclusions

Significant improvements in air quality occurred in the US from 1990 to 2010, which we estimate to have decreased the population-weighted annual average $PM_{2.5}$ by 39 %, and the summertime (April to September) 1 h daily maximum O_3 by 9 %. However, both $PM_{2.5}$ and O_3 are still a great threat to the public health in the US, with estimated mortality burdens of 58 600 (24 900–98 500) deaths year^{-1} and 12 300 (4100–19 800) deaths year^{-1} in 2010, respectively. The mortality burdens associated with exposure to ambient $PM_{2.5}$ have decreased by 54 % over the past 2 decades. However, if the annual $PM_{2.5}$ concentration levels had remained constant during 1990–2010, the associated mortality burden would have only decreased by 24 %, due to decreases in the baseline mortality rates of causes of death affected by $PM_{2.5}$ and despite population growth. The air quality improvements have significantly decreased the mortality burden, avoiding roughly 35 800 (38 %) $PM_{2.5}$-related deaths in 2010, compared to the case if air quality had stayed at 1990 levels.

The mortality burdens attributable to O_3 are estimated to have increased by 13 % during the same period. However, without the emission reductions associated with implementation of measures under the CAA and the NO_x SIP Call, the O_3 mortality burden would have increased by 55 % during 1990–2010. In calculating the O_3 mortality burdens, we use the average of 1 h daily maximum O_3 and the RR from Jerrett et al. (2009), but higher O_3 mortality burdens would likely have resulted had we used RRs from Turner et al. (2016). We estimate that the air quality improvements avoided 4600 (27 %) O_3-related deaths in 2010, compared to the case if air quality had stayed at 1990 levels.

We also estimate the inter-annual variability in mortality burdens considering air pollutant concentrations in individual years and annual county-level baseline mortality rates, and find that the O_3 mortality burdens are more variable (CV of 12 %) than for $PM_{2.5}$ (CV of 4 %), mainly because of inter-annual variability in concentrations. We found that the inter-annual variability is small for $PM_{2.5}$ but larger for ozone, which has not been shown previously that we are aware of.

The uncertainties in air pollution-related mortality estimates presented in this study are based on the uncertainty in relative risks for the specific causes of death only, and do not account for uncertainties in population and baseline mortality rates (which are likely small), nor for uncertainty in the modeled air pollutant concentration. Previous studies have shown that the uncertainties from the modeled air

pollutant concentrations may be greater than uncertainties in baseline mortality or relative risk, so the use of model ensembles is suggested to better quantify the uncertainty (Silva et al., 2016a; Liang et al., 2018). Uncertainties also exist due to the assumption of equal toxicity for different components of $PM_{2.5}$ (Li et al., 2016). For our analysis, we use modeled air pollutant concentration without any bias correction based on either in situ observation or satellite data (Brauer et al., 2015; Hogrefe et al., 2009; van Donkelaar et al., 2015; Xu et al., 2016). In our study, the $PM_{2.5}$ mortality burden trend may be overestimated, and O_3 mortality burdens underestimated, based on comparing the modeled air pollution trends with the observations. Despite these uncertainties, this study illustrates the importance of past air pollutant reductions for public health in the US, and of continued air pollution controls to reduce air pollution-related mortality.

Data availability. The 21-year model outputs for the coupled WRF-CMAQ model, as well as the annual county-level baseline mortality rates, can be obtained by contacting the corresponding author Yuqiang Zhang (yuqiangzhang.thu@gmail.com, yuqiang.zhang@duke.edu).

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Author contributions. YZ designed the study, JX, CG and DW performed the model simulations, and YZ performed the data analysis and prepared the figures and tables. YZ wrote the paper with comments from all the coauthors.

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

Disclaimer. Although this work has been reviewed and approved for publication by the US EPA, the views expressed in this paper are those of the authors and do not necessarily represent the views or policies of the US EPA.

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