Response to review #1 on acp-2018-498

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

Yuqiang Zhang, J. Jason West, Rohit Mathur, Jia Xing, Christian Hogrefe, Shawn J. Roselle, Jesse O. Bash, Jonathan E. Pleim, Chuen-Meei Gan, David C. Wong

General comments: This is an interesting and useful contribution which evaluates the contribution of underlying factors to long-term air pollution-related mortality trends in the continental US, with the aim of highlighting the importance of concentration reductions. This appears to be the first application of a multi-decadal air quality modeling exercise to analyse such a question.
The manuscript is clearly written, with balanced arguments and inclusion of recent, relevant literature. The methods employed are appropriate, clearly described, and well supported.

We thank referee #1 for the very positive comments on our manuscript. We also appreciate the reviewer for the constructive suggestions, which have helped us improve the manuscript. All referee comments (in blue below) have been carefully addressed, and changes incorporated in the revised manuscript are shown using the track-changes option.

It would be helpful to have additional information on the accuracy of the linear interpolation method for population across Census years. Can they discuss the accuracy of this, perhaps with reference to sources that estimate inter-Census population? There are a variety of sources for this, with more sophisticated methods than linear interpolation that rely, e.g., on the American Community Survey. Geolytics Inc. has annual products for population, or perhaps LandScan Global population.
Response: The interannaul population between two censuses (1990 and 2000; 2000 and 2010) was not directly linear interpolated by us, instead they were derived from the Population Estimates project by US Census Bureau. The intercensal population estimates are estimates made for the years between two completed censuses which take into account the census at both the beginning and end of the decade.
(https://www.cdc.gov/nchs/data/nvss/bridged_race/Documentation_bridge_postcenv2017.pdf, accessed 5 September 2018).

From their documentation, the population products from the American Community Survey of Geolytics Inc. and the LandScan Global population both adopted the annual mid-year national population estimates from the US Bureau of Census (https://landscan.ornl.gov/documentation, accessed 5 September 2018)
(http://www.geolytics.com/USCensus,AmericanCommunitySurvey(ACS),Data,Features,Products.asp, accessed 5 September 2018).

To avoid confusion, we rewrote the sentence in line 17-19 in page 5 (page and line numbers are in the revised manuscript):
"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, accessed 5 September 2018)"

Specific comments: Page 10, Lines 1-3: The authors are careful to talk about "reduced mortality burden", but here they mention 'avoided deaths'. Perhaps add "premature" in front of deaths.

Response: We thank the reviewer for the suggestion. In the revised manuscript, we add the word "premature" as the referee suggested.

"The air quality improvements have significantly decreased the mortality burden, avoiding roughly 35,800 (38%) $PM_{2.5}$-related premature deaths in 2010, compared to the case if air quality had stayed at 1990 levels."

Page 7, line 1: recommend introducing Table 1 here, since this seems to be the first time its results are mentioned.

Response: We add the introduction to Table 1 in the beginning of this paragraph, and also rewrite the first sentence:

"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 $PM_{2.5}$-related mortality burden in 2010 would have decreased by only 24% (94,400 deaths yr^{-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,"

Figure 5(a) color bars took a moment to interpret, since for (b) using symmetric saturation with cool = reduction and warm increase, but for (a) these are all reductions. Despite the note, it still took a moment. Perhaps consider only using cool colors for 5(a)?

Response: This figure is now Figure 6, after we added a new figure 5 to the paper. We have changed panel a of this figure to use only cool colors in different shades, as suggested by the reviewer. Thank you for this very good suggestion.

Figure S1 – can you add a legend, perhaps, and/or indicate color of population increase line in caption?

Response: We now add a legend in Figure S1, and also in the caption, we wrote:

"The red line is the US total adult population > 25 yrs old from 1990 to 2010 with the y-axis on the right."

Technical corrections: Page 6 Line 18 - refers to the split decadal trend, which is in Table S5 not Table 1.

Response: We thank the reviewer for noticing this. We now changed "Table 1" to "supporting Table S5".

Page 6 Line 28: Table 1 says 54% not 53%

Response: The reviewer is correct. We have updated the number in the revised manuscript.

Page 5 Line 29: "Zhang" instead of Zhaneg.

Response: We thank the reviewer for pointing this out. We now made the changes in the revised manuscript.

Response to review #2 on acp-2018-498

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

Yuqiang Zhang, J. Jason West, Rohit Mathur, Jia Xing, Christian Hogrefe, Shawn J. Roselle, Jesse O. Bash, Jonathan E. Pleim, Chuen-Meei Gan, David C. Wong

The manuscript by Zhang et al. examines trends in premature mortalities associated with exposure to ambient $PM_{2.5}$ and O_3 in the US over 3 decades. The work is valuable towards understanding the different factors driving these trends and in contributing to a body of evidence documenting the public health benefits of air pollution controls. I have some questions and suggestions that relate to how the authors present uncertainty in their analysis, how they distinguish their work from previous studies, and how they present the trends in different components of the premature mortality calculations (i.e. concentrations vs baseline mortality rates). Addressing these will amount to minor revisions, after which I believe this manuscript will be suitable for publication in ACP.

Response: We thank referee #2 for the positive and constructive suggestions, which have helped us improve the manuscript. We have responded to each comment below and have noted the page and line number for each revision to the manuscript. (blue colors are for referee's comments).

Major comments:

1.26 and throughout: These confidence intervals only account for a subset of the uncertainties inherent in these estimates (i.e., they ignore any inaccuracies in the air pollution model). Thus, it should be clearly stated up front what these ranges do and do not represent. The same comment applies to other places where these numbers are prominently presented, such as e.g. Table 1.

Response: The reviewer is right that the confidence intervals we reported only consider the uncertainty from the RRs. We rephrase the sentence in page 4 line 28-29 (the pages and the numbers are referring the new draft) to clarify this:

"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 not in the scope of our study."

We also clarify the uncertainties in page 7 line 3 when we first report the mortality burdens:

"The mortality burdens associated with exposure to ambient $PM_{2.5}$ in the US steadily decrease by 54%, from 123,700 (95% confidence interval considering the uncertainty in relative risk only, 70,800-178,100) deaths yr-1 in 1990 to 58,600 (24,900-98,500) deaths yr-1 in 2010 (Fig. 3)."

As reported in several recent studies (Silva et al., 2016a; Liang et al., 2018), the uncertainties from the modeled air pollutants concentration may be greater than those from RRs and baseline mortality rates. So we suggest that ensembles of air quality models be used to quantify uncertainties where plausible. We then add the following sentence into the discussion at page 10 line 15:

"Previous studies have shown that the uncertainties from the modeled air pollutants 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)."

References:

Silva, R. A., West, J. J., Lamarque, J. F., Shindell, D. T., Collins, W. J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L. W., Nagashima, T., Naik, V., Rumbold, S. T., Sudo, K., Takemura, T., Bergmann, D., Cameron-Smith, P., Cionni, I., Doherty, R. M., Eyring, V., Josse, B., MacKenzie, I. A., Plummer, D., Righi, M., Stevenson, D. S., Strode, S., Szopa, S. and Zengast, G.: The effect of future ambient air pollution on human premature mortality to 2100 using output from the ACCMIP model ensemble, Atmos. Chem. Phys., 16(15), 9847–9862, doi:10.5194/acp-16-9847-2016, 2016a.

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., Tronstad Lund, M., 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(14), 10497–10520, doi:10.5194/acp-18-10497-2018, 2018.

2.25 - 3.6: I'm not sure I appreciate the significance of the differences between the work in this manuscript and the previous works of Cohen et al. (2017) and Fann et al. (2017), who estimated trends in premature mortalities associated with $PM_{2.5}$ (and O_3– Cohen) in the US since 1980. Yes, their analysis was only once every 5 years, not every year, but does that really make a big difference in the overall conclusions? I'm not sure what the importance of studying successive years is, or interannual variability. The authors results shown in Fig 3 would seem to indicate the answer is "not much", at least for $PM_{2.5}$. I further think works such as Fann 2017 also do discuss different drivers of the trends (mortality rates, population: : :). So, I suggest the authors could go into more detail here about what these previous studies found, including their quantitative results, and also how the present work goes beyond these previous studies methodologically. Update: upon reading section 3.3 (Comparison to previous studies), I'm more informed about how these results differ. Yet still, it would probably benefit the authors to put some more of this content up front in the introduction for motivation. At the very least, Section 3.3 could be alluded to in outlining the contents of what is to come (3.7-12).

Response: We thank the reviewer for pointing this out. The interannual variability analysis in our study 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. Indeed we found that the interannual varability is small for $PM_{2.5}$, but larger for ozone, which we are not aware has been shown previously. In addition, after carefully reading Fann et al., 2017, we do not think they provided the drivers analysis, which provides another innovation of our study.

We add one sentence in page 3 line 5:

"The interannual 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."

We then also add one sentence into the discussion on page 10 line 21-22:
"We found that the interannual varability is small for $PM_{2.5}$, but larger for ozone, which we are not aware has been shown previously."

General: I have some confusion about how to separately interpret the impacts of changing concentrations from changing mortality rates. At present the manuscript seems to imply that changes in baseline mortality rates are not benefits of improving air pollution. However, if reductions in concentrations improve air quality, wouldn't this lead to reductions in mortality rates? To what extent are the evolution of these two terms in the health impact function separate? Could the authors comment on an explain this a bit more?

Response: We agree with the reviewer that improving air quality could help to reduce the baseline mortality rates (Correia, et al., 2013; Pope et al., 2009), but the changes in baseline mortality rates were more caused by other factors such as changes in social-economic development, heathcare expenditures, and so on, than the air pollution level. The baseline mortality rates for lung cancer, ischemic heart disease and stroke decreased from 1990 to 2010, as a result of the exposed population becoming more resilient and healthier, and less susceptible to the risk of air pollution, while baseline mortality for chronic obstructive pulmonary and respiratory disease increased at the same time (see Figure S5 in our supporting materials).

To clarify this, we add the following sentence in page 4 line 31-32:
"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."

Reference:

Correia, A. W., Arden Pope, C., Dockery, D. W., Wang, Y., Ezzati, M. and Dominici, F.: Effect of air pollution control on life expectancy in the United States: An analysis of 545 U.S. Counties for the period from 2000 to 2007, Epidemiology, 24(1), 23–31, doi:10.1097/EDE.0b013e3182770237, 2013.

Pope, C. A., Ezzati, M. and Dockery, D. W.: Fine-Particulate Air Pollution and Life Expectancy in the United States, N. Engl. J. Med., 360(4), 376–386, doi:10.1056/NEJMsa0805646, 2009.

4.23: What was the basis for using an average value for the threshold? What impacts does this value have on the overall findings, quantitatively and qualitatively?

Response: The counterfactual_concentration_of 37.6 ppb for O_3 premature mortality calculations was used in the recent Global Burden Disease estimates (Cohen et al., 2017). We used this value for consistency with the GBD. Adopting modeled pre-industrial O_3 instead of the counterfactual concentration could lead to higher estimates of O_3 mortality burden at present day, as discussed in Lelieveld et al. (2013, 2015), and also Table S10 in our supporting materials.

To avoid confusion, we rewrite the sentence in page 4 line 25:
"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).

Reference:

Lelieveld, J., Barlas, C., Giannadaki, D. and Pozzer, A.: Model calculated global, regional and megacity premature mortality due to air pollution, Atmos. Chem. Phys., 13(14), 7023–7037, doi:10.5194/acp-13-7023-2013, 2013.
Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D. and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525(7569), 367–71, doi:10.1038/nature15371, 2015.

4.27: Again, what is the basis for this, and how does it affect the findings?
Response: We used one single model results for the mortality burden estimations, and so we could not include uncertainties from the modeled air pollution without further assumptions about those uncertainties. However, from previous findings, the uncertainties from the modeled air pollutants concentration are likely larger than larger than those from RRs and baseline mortality rates. So we suggest that ensemble air quality model results could be used to quantify the overall uncertainties. We now add discussion of this at the end of the manuscript in page 10 line 25-27:
"Previous studies have shown that the uncertainties from the modeled air pollutants 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)."

Results: Please provide as separate figures (there is plenty of room for this within the main body of an ACP paper): (1) trends in baseline mortality rates, (2) trends in concentrations, (3) trends in AF, (4) trends in mortalities. Providing these pieces of information separately would make parsing the results of this paper so much more straightforward. At less than 10 pages of text and only 6 figures, presently, there's no reason to place any of this in the SI.
Response: We thank the reviewer for the suggestion. We now move Figure S5 (the trends for baseline mortality rates) from the supporting materials into the main text as a new Figure 5. The numbers of the other figures in the main text as well as in the supporting are all updated as well. Trends in concentration are shown in Figure 2, trends in AF will be similar as trends in concentration, and trends in deaths are our main results in Figures 3 and 4.

Fig 5: I strongly object to the choice of color scale for panel (a). The potential for misunderstanding the results is quite high. Please use red-blue colors scales as commonly understood, which blue being negative and red being positive. Or pick an entirely different color scheme for these results that are strictly negative.
Response: We thank the reviewer for pointing this out. Now we updated the plot by using blue meaning negative only. See Figure 6 in the new manuscript.

Minor comments and corrections:
18: Perhaps, more precisely, "exposure to these pollutants are associated with…."
Response: We now rewrite this sentence:
"Exposure to these air pollutants is associated with premature death."

abstract: From the perspective of air quality control and the audience of ACP, it seems more interesting to report how many premature mortalities would have been avoided by PM2.5 and O3 reductions in the absence of changes in baseline mortality rates (rather than the other way around). That being said, perhaps these are the numbers that are reported in the last few lines of the abstract? It's not clear if these are / are not

accounting for changes in mortality rates (or population).

Response: The numbers reported in page 2 line 1-3 in the abstract were calculated using the mortality burden in 2010 considering all three factors (concentration, mortality rates and population) minus the mortality burden in 2010 considering two factors only (mortality rates and population; also see Figure 3 the black and red line). So here we are reporting the premature deaths which would have been avoided by $PM_{2.5}$ and O_3 reductions only, in the absence of changes in baseline mortality rates and population. To clarify this, we modify the sentence in page 2 line 1-3: "We conclude that air quality improvements have significantly decreased the mortality burden, avoiding roughly 35,800 (38%) $PM_{2.5}$-related deaths and 4,600 (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)."

2.15: observations sites –> observations

Response: We now deleted the "sites" in this sentence.

2.20: it's note entirely clear how the authors are separating the benefits of the NAAQS from those of the technologies put in place to meet the NAAQS – seems like these are perhaps two sides of the same coin..

Response: We rephrase this sentence following the reviewer's suggestions:
"Other changes in energy and emission control technology occurred concurrently with air quality regulations also helped to improve air quality."

2.24: I understand the wording, but if one accept the health impact analysis framework used here, then does it matters not where people live as much as where they die?

Response: Here we make the point that one needs to understand how air quality has improved in spatial relation to how population is distributed. The reviewer is correct that baseline mortality is used directly in our calculations ("where people die"), and so we have updated the language from "where people live" to "how population and baseline mortality are distributed".

2.15 - 2.24: Note the difference in tone between the assertiveness of this work ("improvements were mainly driven by ambient air quality standards…", and that of Fann 2017 who state "it is difficult to attribute this reduction to specific policy interventions….many factors are likely to have contributed….federal air quality policies are likely to have played an important role". It then seems that different federal regulations are cited, such as Acid Rain program. Thoughts about why the present work is a bit more sure of the role of regulations in this regard? Were EPA authors just being more cautious with their wording for professional reasons?

Response: Following the reviewer's suggestion, we now rewrote the sentence from 2.16 to 2.19:
"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 NOx State Implementation Plans (SIP) Call, and the Cross-State Air Pollution Rule (Chestnut and Mills, 2005; U.S. EPA, 2011), together with other rules to reduce anthropogenic emissions from light duty, heavy duty, and nonroad vehicles (Fann et al.2012b; U.S. EPA2014). Other changes in energy and emission control technology occurred concurrently with air quality regulations also helped to improve air quality."

References:

Chestnut, L. G. and Mills, D. M.: A fresh look at the benefits and costs of the US acid rain program, J. Environ. Manage., 77(3), 252–266, doi:10.1016/j.jenvman.2005.05.014, 2005.

Fann, N., Baker, K. R. and Fulcher, C. M.: Characterizing the $PM_{2.5}$-related health benefits of emission reductions for 17 industrial, area and mobile emission sectors across the U.S., Environ. Int., 49(2012), 141–151, doi:10.1016/j.envint.2012.08.017, 2012b.

US EPA. 2014. "Control of Air Pollution from Motor Vehicles: Tier3 Motor Vehicle Emission and Fuel Standards Final Rule." https://www.epa.gov/regulations-emissions-vehicles-and-engines/final-rule-control-air-pollution-motor-vehicles-tier-3 (accessed 5 September 2018).

4.16: I doubt this is what the authors meant to say. If RRs were downloaded from the GBD web site, then there would be no role for the simulations of O_3 and $PM_{2.5}$ concentrations described in section 2.1.

Response: The relative risks (RR) for $PM_{2.5}$ from the IER function (Burnet et al., 2014) that we downloaded from the GBD website are functions of $PM_{2.5}$ concentration, and we need the gridded concentration fields to determine the RR. Also, the RRs for O_3 are calculated (page 4 line 17) instead of downloaded.

4.25: The justification here seems a bit odd, as if the authors decided that sticking with the mismatched population age-ranges from their previous work would take precedent over correctly matching age ranges with the epidemiological study of Jerrett 2009.

Response: By aligning exposed population >=25 yrs for both $PM_{2.5}$ and O_3, we want to determine the magnitude differences for the mortality burdens of $PM_{2.5}$ and O_3. This method was also adopted by Lim et a., 2012; Cohen et al., 2017. In the revised manuscript, we have rewritten the sentence as:

"We use adults above 25 years, to be comparable with other calculations of $PM_{2.5}$ mortality burdens (Lim et al., 2012; Silva et al., 2016a,b; Cohen et al., 2017), even though the estimated RR from Jerrett et al. (2009) were for adults above 30 old only."

Reference:

Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., Amann, M., Anderson, H. R., Andrews, K. G., Aryee, M., Atkinson, C., Bacchus, L. J., Bahalim, A. N., Balakrishnan, K., Balmes, J., Barker-Collo, S., Baxter, A., Bell, M. L., Blore, J. D., Blyth, F., Bonner, C., Borges, G., Bourne, R., Boussinesq, M., Brauer, M., Brooks, P., Bruce, N. G., Brunekreef, B., Bryan-Hancock, C., Bucello, C., Buchbinder, R., Bull, F., Burnett, R. T., Byers, T. E., Calabria, B., Carapetis, J., Carnahan, E., Chafe, Z., Charlson, F., Chen, H., Chen, J. S., Cheng, A. T.-A., Child, J. C., Cohen, A., Colson, K. E., Cowie, B. C., Darby, S., Darling, S., Davis, A., Degenhardt, L., Dentener, F., Des Jarlais, D. C., Devries, K., Dherani, M., Ding, E. L., Dorsey, E. R., Driscoll, T., Edmond, K., Ali, S. E., Engell, R. E., Erwin, P. J., Fahimi, S., Falder, G., Farzadfar, F., Ferrari, A., Finucane, M. M., Flaxman, S., Fowkes, F. G. R., Freedman, G., Freeman, M. K., Gakidou, E., Ghosh, S., Giovannucci, E., Gmel, G., Graham, K., Grainger, R., Grant, B., Gunnell, D., Gutierrez, H. R., Hall, W., Hoek, H. W., Hogan, A., Hosgood, H. D., Hoy, D., Hu, H., Hubbell, B. J., Hutchings, S. J., Ibeanusi, S. E., Jacklyn, G. L., Jasrasaria, R., Jonas, J. B., Kan, H., Kanis, J. a, Kassebaum, N., Kawakami, N., Khang, Y.-H., Khatibzadeh, S., Khoo, J.-P., Kok, C., et al.: A comparative risk assessment of burden of disease and injury attributable to 67 risk

factors and risk factor clusters in 21 regions, 1990-2010: a systematic analysis for the Global Burden of Disease Study 2010., Lancet, 380(9859), 2224–60, doi:10.1016/S0140-6736(12)61766-8, 2012.

4.28 - 5.15: Would it be possible to avoid these types of inconsistencies in reporting to use heath impact functions associated with all-cause rather than cause-specific mortality rates, even if the epidemiological evidence of the responses isn't as robust?

Response: It is possible to avoid inconsistencies between the two standards of ICD9 and IC10 by quantifying the total all-cause mortality as used in Fann et al. (2017). However, by doing that we would assume a log-linear association between $PM_{2.5}$ and mortality burden (Krewski et al., 2009), which will have larger uncertainties when the $PM_{2.5}$ concentration is higher than 22 μg m^{-3} (Burnett et al., 2014; Pope et al., 2009). From the model simulation, the $PM_{2.5}$ concentration were higher than this maximum value in 1990 in eastern US (see Figure 1a).

7.24: in the eastern

Response: We now add "the" in the revised manuscript.
"For other states in the eastern US"

Title: should indicate this is about ambient AQ? Or that is obvious?

Response: We add "ambient" before "$PM_{2.5}$" and "O_3" in the title as the reviewer suggested.

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

Yuqiang Zhang[1,*], J. Jason West[2], Rohit Mathur[3], Jia Xing[4], Christian Hogrefe[3], Shawn J. Roselle[3], Jesse O. Bash[3], Jonathan E. Pleim[3], Chuen-Meei Gan[5], David C. Wong[3]

[1]Oak Ridge Institute for Science and Education (ORISE) Fellowship Participant at US Environmental Protection Agency, Research Triangle Park, NC 27711, USA

[2]Department of Environmental Sciences and Engineering, University of North Carolina at Chapel Hill, Chapel Hill, NC 27599, USA

[3]Computational Exposure Division, National Exposure Research Laboratory, Office of Research and Development, US Environmental Protection Agency, Research Triangle Park, NC 27711, USA

[4]State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

[5]CSC Government Solutions LLC, A CSRA Company, Research Triangle Park, NC, 27709, USA

[*]now at: Nicholas School of the Environment, Duke University, Durham, NC 27710, USA

Correspondence to: Yuqiang Zhang (yuqiangzhang.thu@gmail.com; yuqiang.zhang@duke.edu)

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) 1hr 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 53% from 123,700 deaths yr^{-1} (95% confidence interval, 70,800-178,100) in 1990 to 58,600 deaths yr^{-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 yr^{-1} (3,700-17,500) in 1990 to 12,300 deaths yr^{-1} (4,100-19,800) 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

Deleted: T

Deleted: are

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 4,600 (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).

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 (4,400–19,600) 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 (Murry et al., 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 (NMVOC), 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; U.S. EPA 2014). Other changes in energy and emission control technology 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 with how population and baseline mortality are distributed.

Several recent studies have assessed the global (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, 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-yr intervals from 1990 to 2015. Two other studies (Butt et al., 2017; Wang et al., 2017) used coarse resolution model

Deleted: sites

Deleted: mainly

Deleted: Other factors that improved air quality include technological development that made emission control technolog more effective and less expensive, and changes in the energy syst such as the recent growth of natural gas and wind that has displac coal for electricity generation

Deleted: where people live

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, investigated the different drivers for mortality trends, or the inter-annual variability of the mortality burdens. The interannual variability analysis indicate 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 burdens 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).

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 36km×36km. 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 western US for both the model and observations, while a dramatic decreasing trend in the eastern US, with a larger decreasing trend from the model (-0.44 $\mu g\ m^{-3}\ yr^{-1}$) than from the IMPROVE observations (-0.30 $\mu g\ m^{-3}\ yr^{-1}$).

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-hr (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 yr^{-1} (-0.73 ppbv yr^{-1}) for the 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 ($\Delta Mort$) are estimated using the health impact function (HIF) following Eq. (1):

$$\Delta Mort = y_0 \times AF \times Pop \quad (1)$$

where y_0 is the baseline mortality rate for specific diseases, AF is the attributable fraction calculated as $1 - 1/RR$, 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 World Health Organization (2016). The RR is calculated as a function of $PM_{2.5}$ concentration following Eq. (2):

$$for\ C < C_0, RR(C) = 1$$
$$for\ C \geq C_0, RR(C) = 1 + \alpha \times (1 - \exp(-\gamma \times (C - C_0)^{\delta})) \quad (2)$$

where C is the annual average ambient $PM_{2.5}$ concentration, C_0 is the $PM_{2.5}$ threshold concentration (5.8-8.0 μg/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 GBD website (Global Health Data Exchange (GHDx) 2013).

For the O_3-related mortality burden, RR= $exp^{\beta \Delta X}$, where β is the concentration response factor, and ΔX is the difference in O_3 concentration (summertime 1hr 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 RR from Jerrett et al. (2009) were for adults above 30 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 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.

Deleted: low-concentration threshold is the average of 33.3-41.9 ppbv O_3 indicated by Lim et al., (2012), that is,

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 (chronic respiratory disease, 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 36km×36km. The specific causes of mortality data for some counties are sometimes suppressed when the total deaths are lower than 10 per year to protect privacy (Jian et al., 2016), missing or considered as "unreliable" when the total deaths are less than 20 per year, and are corrected following established procedures (BenMAP, 2017; Fann et al., 2017; also see Supporting Information).

Definitions of each disease follow the Global Burden of Disease (GBD) study (Lim et al., 2012; supporting Table S1). 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 & 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 & Rosenberg (2003) who reported deaths for 135 specific causes in 1996 for both the ICD9 and ICD10 codes, and calculate comparability ratios (supporting Table S1). We then recalculate comparability ratios for the 5 diseases (RESP, COPD, IHD, LC and STROKE) as the ratios of deaths for ICD9 and ICD10 (supporting 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, accessed 5 September 2018). The adult population above 25 yrs in the US has steadily grown between 1990 and 2010, with an average 1.23% yr^{-1} rate of increase (supporting Fig. S1).

Deleted: regrid

Deleted: We use archived US population in individual counties from the CDC, which are from the US Census Bureau in 1990, 20 and 2010. The population from 1991 to 1999 was interpolated in each county between the 1990 and the 2000 censuses, and the population from 2001 to 2009 was interpolated between 2000 and 2010

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. (3):

$$\Delta Mort_P^y = y_0^{1990} \times AF^y \times Pop^{1990} - y_0^{1990} \times AF^{1990} \times Pop^{1990} \quad (3)$$

Similarly, we also calculate the mortality burden change without accounting for ambient air pollution changes ($\Delta Mort_{noP}^y$) following Eq. (4):

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

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. 1-c), but slightly decreases or even increases in the northwest, southwest and west (supporting Fig. S2 and Table S3; also see supporting Fig. S3 for the US 9 regions defined by 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 west central 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 $\mu g\ m^{-3}\ da^{-1}$ ($decade^{-1}$) from 1990 to 2010, especially in the central (-3.48 $\mu g\ m^{-3}\ da^{-1}$) and northeast (-3.14 $\mu g\ m^{-3}\ da^{-1}$). The summertime average of 1hr daily maximum O_3 decrease significantly in the central and eastern US, generally at a rate greater than 4 ppbv da^{-1}. It also decreases in the western US, but at a much smaller rate than in the east, generally less than 1 ppbv da^{-1} (Fig. 1-f; supporting 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 $\mu g\ m^{-3}\ da^{-1}$. 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 $\mu g\ m^{-3}\ da^{-1}$. 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 da^{-1}. The PWA O_3 also decrease by 9%, from 58.96 ppbv in 1990 to 53.57 ppbv in 2010, decreasing at a rate of 3.0 ppbv da^{-1}. We also calculate the air quality and mortality burden trends separately for two 11-yr 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 (supporting 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) (supporting 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 (supporting Fig. S4, b,e). We also calculate the population exposure exceedances by multiplying the population (adults > 25 yrs 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 (supporting Fig. S1). These decreases in population exposure exceedances occur despite population growth over this period.

Deleted: Zhaneg

Deleted: Table 1

3.2 Mortality burdens trends and contributing factors

The mortality burdens associated with exposure to ambient $PM_{2.5}$ in the US steadily decrease by 54%, from 123,700 (95% confidence interval considering the uncertainty in relative risk only, 70,800-178,100) deaths yr^{-1} in 1990 to 58,600 (24,900-98,500) deaths yr^{-1} in 2010 (Fig. 3). The leading cause for $PM_{2.5}$-related mortality is IHD, which decreases by 55%, from 96,500 (62,600-132,500) deaths yr^{-1} in 1990 to 43,600 (21,500-68,700) deaths yr^{-1}, followed by LC which has decreased by 44%, from 12,500 (2,500-21,000) deaths yr^{-1} in 1990 to 7,000 (900-13,400) deaths yr^{-1} in 2010 (supporting 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 $PM_{2.5}$-related mortality burden in 2010 would have decreased by only 24% (94,400 deaths yr^{-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 Figure 3, yielding 78,900 (35,700-129,200) deaths yr^{-1} in 2010, decreasing by 36% (-44,800 deaths yr^{-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 (supporting Figs. S5 and S6).

When separating the two 11-yr periods, the $PM_{2.5}$-related mortality burden has decreased 45% from 2000 to 2010 (decreasing trend of -4400 deaths yr^{-1}), much higher than the 15% decrease from 1990 to 2000 (decreasing trend of -2100 deaths yr^{-1}) (supporting Table S5). The detrended annual $PM_{2.5}$-related mortality burden has a coefficient of variation (CV, standard deviation divided by average) of 4%, mainly caused by inter-annual variation in $PM_{2.5}$ concentrations (supporting 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 yr^{-1}), California (CA, 9,500 deaths yr^{-1}) and Pennsylvania (PA, 9,200 deaths yr^{-1}); and in 2010, NY (5,100 deaths yr^{-1}), Texas (TX, 4,200 deaths yr^{-1}) and Ohio (OH, 3,900 deaths yr^{-1}). NY has seen the largest benefits of mortality burden decreases (-8,500 deaths yr^{-1}), followed by CA (-6,100 deaths yr^{-1}) and PA (-5,500 deaths yr^{-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 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 (supporting Table S7). For other states in the

Deleted: 53
Deleted: T
Deleted: (Table 1)
Deleted: supporting
Deleted: S5
Deleted: S6
Deleted: S7
Deleted: S7
Deleted: 5

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 of $PM_{2.5}$ concentration are larger than that from the decrease of the baseline mortality rates. However, for Massachusetts, NY and PA, the decreases of baseline mortality rates have a slightly larger effect than that from the decrease of $PM_{2.5}$ concentration. For CA, the effects from the decrease of baseline mortality rates and $PM_{2.5}$ concentration are comparable (supporting Table S7).

The mortality burden associated with exposure to O_3 from chronic respiratory disease (RESP) has increased by 13%, from 10,900 (3,700-17,500) deaths yr^{-1} in 1990 to 12,300 (4,100-19,800) deaths yr^{-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 yr^{-1} in 2010, 95%CI, 3,600-17,100) 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 (supporting Fig. S5). The decreased O_3 concentration would have avoided roughly 4,600 (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 Figure 3, yielding 8,100 (2,700-13,100) deaths yr^{-1} in 2010, decreasing by 25% (-2800 deaths yr^{-1}) compared with 1990. The change in O_3 generally reduces the mortality burden relative to 1990 with some inter-annual variation (supporting Fig. S6) due to meteorology and wildfires (Porter et al., 2017), while the increases of population and baseline mortality rates generally increase the mortality burden, with a larger contribution from the population change (supporting Fig. S6).

When separating the O_3 mortality trends into two decades, we find that the burdens decrease slightly (-70 deaths yr^{-1}) from 2000 to 2010, compared with the increasing trend from 1990 to 2000 (240 deaths yr^{-1}) (supporting 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 (supporting 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 (supporting Table S6).

The three states with the highest O_3 mortality burden in 1990 are CA (910 deaths yr^{-1}), Florida (FL, 740 deaths yr^{-1}) and NY (700 deaths yr^{-1}); and in 2010, CA (1270 deaths yr^{-1}), TX (900 deaths yr^{-1}) and FL (770 deaths yr^{-1}) (Table 2). CA has seen the largest O_3 mortality burden increases (360 deaths yr^{-1}), followed by TX (230 deaths yr^{-1}) and Arizona (AZ, 140 deaths yr^{-1}), with the greatest decrease in NY (-90 deaths yr^{-1}). For the relative mortality burden changes, large percent decreases in O_3-related mortality are seen in the northwest (Washington and Oregon) and northeast US (Fig. 6), mainly caused by significant O_3 decreases (supporting Table S7), while the greatest percent increases occur in the southwest 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, 2016b, 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

Deleted: S6

Deleted: S7

Deleted: S7

Deleted: S7

Deleted: supporting Figure S5

mortality burden calculations. We find that the $PM_{2.5}$ mortality burden calculated from the national average baseline mortality rates are lower than those calculated from the county-level baseline mortality rates, ranging among individual years from -2.2% to -1.3% (supporting Table S8). For the O_3 mortality burden, the difference between using the national average baseline mortality rates and our best estimates range from -1.1% to 2.0% (supporting Table S8). However, using the national average baseline mortality rates fails to capture regional mortality burden hotspots for both $PM_{2.5}$ and O_3 (supporting Figs. S7-S8), demonstrating the value of using county-level baseline mortality rates where possible.

Deleted: S8

Deleted: S9

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 supporting 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 yr^{-1}) than Cohen et al. (2017) (-1000 deaths yr^{-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.

Deleted: 6

Deleted: 6

To compare with Cohen et al. (2017), who reported the O_3 mortality burden from COPD, which is a subset of RESP, we recalculate O_3 mortality burden from COPD (supporting 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 1-hr daily maximum, while Cohen et al. (2017) used the three-month average, which will be higher. The temporal trend for the O_3 mortality burdens from our study is similar with 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; supporting Table S10). As discussed in the methods, the lower US background O_3 concentration used in these two studies (22ppb in the eastern US and 30ppb in the western US) could lead to 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 (supporting Fig. S9) as the counterfactual risk exposure factor, and recalculated the O_3 mortality burden with the RESP. The new calculated O_3 mortality burdens are estimated to be 64%-100% higher than current estimation from RESP using the

Deleted: 6

Deleted: S10

low-concentration threshold (supporting 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.

Deleted: 6

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 summertime (April to September) 1-hr daily maximum O_3 by 9%. However, both $PM_{2.5}$ and O_3 are still a great threat to the public health in US, with estimated mortality burdens of 58,600 (24,900-98,500) deaths yr^{-1} and 12,300 (4,100-19,800) deaths yr^{-1} in 2010, respectively. The mortality burdens associated with exposure to ambient $PM_{2.5}$ have decreased by 54% over the past two 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 1hr-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 have avoided 4,600 (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 interannual varability is small for $PM_{2.5}$, but larger for ozone, which we are not aware has been shown previously.

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 pollutants 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., 2015). 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 burdens 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-yrs 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 (Y. Zhang, yuqiangzhang.thu@gmail.com, Yuqiang.zhang@duke.edu).

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.

Acknowledgement: This research was supported in part by an appointment to the Research Participation Program at the U.S. EPA, Office of Research and Development (ORD), administered by the Oak Ridge Institute for Science and Education (ORISE) through an interagency agreement between the U.S. Department of Energy and the U.S. EPA, and also in part by National Aeronautics and Space Administration Health and Air Quality Applied Science Team #NNX16AQ80G. This work was also supported in part by China Ministry of Science and Technology National Key R & D program (2016YFC0207601). We thank Raquel Silva and Neal Fann from U.S. EPA for useful discussions. We greatly acknowledge Ana Rappold and Geoffrey Peterson from the U.S. EPA for their comments and suggestions on the initial version of this manuscript. We also appreciate the free usage of the county level baseline mortality and population data from US Centers for Disease Control and Prevention (CDC).

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

References

Anderson, R. N., Miniño, A. M., Hoyert, D. L. and Rosenberg, H. M.: Comparability of cause of death between ICD-9 and ICD-10: preliminary estimates., Natl. Vital Stat. Rep., 49(2), 1–32, 2001.

Anderson, R. N. and Rosenberg, H. M.: Disease classification: Measuring the effect of the Tenth Revision of the International Classification of Diseases on cause-of-death data in the United States, Stat. Med., 22(9), 1551–1570, doi:10.1002/sim.1511, 2003.

Anenberg, S. C., Horowitz, L. W., Tong, D. Q. and West, J. J.: An Estimate of the Global Burden of Anthropogenic Ozone and Fine Particulate Matter on Premature Human Mortality Using Atmospheric Modeling, Environ. Health Perspect., 118(9), 1189–1195, doi:10.1289/ehp.0901220, 2010.

Astitha, M., Luo, H., Rao, S. T., Hogrefe, C., Mathur, R. and Kumar, N.: Dynamic evaluation of two decades of WRF-CMAQ ozone simulations over the contiguous United States, Atmos. Environ., 164, 102–116, doi:10.1016/j.atmosenv.2017.05.020, 2017.

Brauer, M., Amann, M., Burnett, R. T., Cohen, A., Dentener, F., Ezzati, M., Henderson, S. B., Krzyzanowski, M., Martin, R. V, Van Dingenen, R., van Donkelaar, A. and Thurston, G. D.: Exposure assessment for estimation of the global burden of disease attributable to outdoor air pollution., Environ. Sci. Technol., 46(2), 652–60, doi:10.1021/es2025752, 2012.

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.

Butt, E. W., Turnock, S. T., Rigby, R., Reddington, C. L., Yoshioka, M., Johnson, J. S., Regayre, L. A., Pringle, K. J., Mann, G. W. and Spracklen, D. V: Global and regional trends in particulate air pollution and attributable health burden over the past 50 years, Environ. Res. Lett., 12(10), 104017, 2017.

Centers for Disease Control and Prevention (CDC) 2017. National Center for Health Statistics, https://wonder.cdc.gov/mortSQL.html, [accessed in 02/28/2017].

Chestnut, L. G. and Mills, D. M.: A fresh look at the benefits and costs of the US acid rain program, J. Environ. Manage., 77(3), 252–266, doi:10.1016/j.jenvman.2005.05.014, 2005.

Cohen, A. J., Brauer, M. and Burnett, R. T.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: An analysis of data from the Global burden of Diseases Study 2015, Lancet, 389(10082), 1907–1918, doi:10.1016/S0140-6736(17)30505-6, 2017.

Correia, A. W., Arden Pope, C., Dockery, D. W., Wang, Y., Ezzati, M. and Dominici, F.: Effect of air pollution control on life expectancy in the United States: An analysis of 545 U.S. Counties for the period from 2000 to 2007, Epidemiology, 24(1), 23–31, doi:10.1097/EDE.0b013e3182770237, 2013.

Dwyer-Lindgren, L., Bertozzi-Villa, A., Stubbs, R. W., Morozoff, C., Kutz, M. J., Huynh, C., Barber, R. M., Shackelford, K. A., Mackenbach, J. P., van Lenthe, F. J., Flaxman, A. D., Naghavi, M., Mokdad, A. H. and Murray, C. J. L.: US county-level trends in mortality rates for major causes of death, 1980-2014, JAMA, 316(22), 2385–2401, doi:10.1001/jama.2016.13645, 2016.

Dennison, P. E., Brewer, S. C., Arnold, J. D. and Moritz, M. A.: Large wildfire trends in the western United States, 1984–2011, Geophys. Res. Lett., 41(8), 2928–2933, doi:10.1002/2014GL059712.Received, 2014.

Environmental Benefits Mapping and Analysis Program – Community Edition, User's Manual, Appendices, 2017 (BenMAP 2017),https://www.epa.gov/sites/production/files/2017-04/documents/benmap_ce_um_appendices_april_2017.pdf, [accessed in 04/25/2017].

Fang, Y., Naik, V., Horowitz, L. W. and Mauzerall, D. L.: Air pollution and associated human mortality: The role of air pollutant emissions, climate change and methane concentration increases from the preindustrial period to present, Atmos. Chem. Phys., 13, 1377–1394, doi:10.5194/acp-13-1377-2013, 2013.

Fann, N., Lamson, A. D., Anenberg, S. C., Wesson, K., Risley, D. and Hubbell, B. J.: Estimating the national public health burden associated with exposure to ambient $PM_{2.5}$ and ozone., Risk Anal.,, 32(1), 81–95, doi:10.1111/j.1539-6924.2011.01630.x, 2012a.

Fann, N., Baker, K. R. and Fulcher, C. M.: Characterizing the $PM_{2.5}$-related health benefits of emission reductions for 17 industrial, area and mobile emission sectors across the U.S., Environ. Int., 49(2012), 141–151, doi:10.1016/j.envint.2012.08.017, 2012b.

Fann, N., Kim, S. Y., Olives, C. and Sheppard, L.: Estimated Changes in Life Expectancy and Adult Mortality Resulting from Declining PM2.5 Exposures in the Contiguous United States: 1980-2010, Environ. Health Perspect., 125(9), 097003, doi:10.1289/EHP507, 2017.

Gan, C. M., Pleim, J., Mathur, R., Hogrefe, C., Long, C. N., Xing, J., Wong, D., Gilliam, R. and Wei, C.: Assessment of long-term WRF-CMAQ simulations for understanding direct aerosol effects on radiation "brightening" in the United States, Atmos. Chem. Phys., 15(21), 12193–12209, doi:10.5194/acp-15-12193-2015, 2015.

Gan, C. M., Hogrefe, C., Mathur, R., Pleim, J., Xing, J., Wong, D., Gilliam, R., Pouliot, G. and Wei, C.: Assessment of the effects of horizontal grid resolution on long-term air quality trends using coupled WRF-CMAQ simulations, Atmos. Environ., 132, 207–216, doi:10.1016/j.atmosenv.2016.02.036, 2016.

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.

GBD 2013 Risk Factors Collaborators: Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks in 188 countries, 1990–2013: a systematic analysis

for the Global Burden of Disease Study 2013, Lancet., 386(10010), 2287–2323, https://doi.org/10.1016/s0140-6736(15)00128-2, 2015.

GBD 2015 Risk Factors Collaborators: Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 1990–2015: a systematic analysis for the Global Burden of Disease Study 2015, Lancet., 388(10053), 1659–1724, https://doi.org/10.1016/S0140-6736(16)31679-8, 2016.

GHDx (Global Health Data Exchange). 2013. Global Burden of Disease Study 2010 – Ambient Air Pollution Risk Model 1990–2010. http://ghdx.healthmetricsandevaluation.org/record/global-burden-disease-study-2010-gbd-2010-ambient-air-pollution-risk-model-1990-2010, (accessed 8 November 2016).

Giannadaki, D., Lelieveld, J. and Pozzer, A.: Implementing the US air quality standard for $PM_{2.5}$ worldwide can prevent millions of premature deaths per year, Environ. Heal.,, 15(1), 88, doi:10.1186/s12940-016-0170-8, 2016.

Hogrefe, C., Lynn, B., Goldberg, R., Rosenzweig, C., Zalewsky, E., Hao, W., Doraiswamy, P., Civerolo, K., Ku, J. Y., Sistla, G. and Kinney, P. L.: A combined model-observation approach to estimate historic gridded fields of $PM_{2.5}$ mass and species concentrations, Atmos. Environ., 43(16), 2561–2570, doi:10.1016/j.atmosenv.2009.02.031, 2009.

Jaffe, D., Chand, D., Hafner, W., Westerling, A. and Spracklen, D.: Influence of Fires on O_3 Concentrations in the Western U.S., Environ. Sci. Technol., 42(16), 5885–5891, doi:10.1021/es800084k, 2008.

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.

Jian, Y., Messer, L. C., Jagai, J. S., Rappazzo, K. M., Gray, C. L., Grabich, S. C. and Lobdell, D. T.: The Associations between Environmental Quality and Mortality in the Contiguous United States, 2000-2005., Environ. Health Perspect., 125(3), 2000–2005, doi:10.1289/EHP119, 2016.

Lamarque, J. F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A. and Zeng, G.: The atmospheric chemistry and climate model intercomparison Project (ACCMIP): Overview and description of models, simulations and climate diagnostics, Geosci. Model Dev., 6(1), 179–206, doi:10.5194/gmd-6-179-2013, 2013.

Lelieveld, J., Barlas, C., Giannadaki, D. and Pozzer, A.: Model calculated global, regional and megacity premature mortality due to air pollution, Atmos. Chem. Phys., 13(14), 7023–7037, doi:10.5194/acp-13-7023-2013, 2013.

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D. and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525(7569), 367–71, doi:10.1038/nature15371, 2015.

Li, Y., Henze, D. K., Jack, D., Henderson, B. H. and Kinney, P. L.: Assessing public health burden associated with exposure to ambient black carbon in the United States, Sci. Total Environ., 539(2016), 515–525, doi:10.1016/j.scitotenv.2015.08.129, 2016.

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., Tronstad Lund, M., 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(14), 10497–10520, doi:10.5194/acp-18-10497-2018, 2018.

Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., Amann, M., Anderson, H. R., Andrews, K. G., Aryee, M., Atkinson, C., Bacchus, L. J., Bahalim, A. N., Balakrishnan, K., Balmes, J., Barker-Collo, S., Baxter, A., Bell, M. L., Blore, J. D., Blyth, F., Bonner, C., Borges, G., Bourne, R., Boussinesq, M., Brauer, M., Brooks, P., Bruce, N. G., Brunekreef, B., Bryan-Hancock, C., Bucello, C., Buchbinder, R., Bull, F., Burnett, R. T., Byers, T. E., Calabria, B., Carapetis, J., Carnahan, E., Chafe, Z., Charlson, F., Chen, H., Chen, J. S., Cheng, A. T.-A., Child, J. C., Cohen, A., Colson, K. E., Cowie, B. C., Darby, S., Darling, S., Davis, A., Degenhardt, L., Dentener, F., Des Jarlais, D. C., Devries, K., Dherani, M., Ding, E. L., Dorsey, E. R., Driscoll, T., Edmond, K., Ali, S. E., Engell, R. E., Erwin, P. J., Fahimi, S., Falder, G., Farzadfar, F., Ferrari, A., Finucane, M. M., Flaxman, S., Fowkes, F. G. R., Freedman, G., Freeman, M. K., Gakidou, E., Ghosh, S., Giovannucci, E., Gmel, G., Graham, K., Grainger, R., Grant, B., Gunnell, D., Gutierrez, H. R., Hall, W., Hoek, H. W., Hogan, A., Hosgood, H. D., Hoy, D., Hu, H., Hubbell, B. J., Hutchings, S. J., Ibeanusi, S. E., Jacklyn, G. L., Jasrasaria, R., Jonas, J. B., Kan, H., Kanis, J. a, Kassebaum, N., Kawakami, N., Khang, Y.-H., Khatibzadeh, S., Khoo, J.-P., Kok, C., et al.,: A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010: a systematic analysis for the Global Burden of Disease Study 2010., Lancet, 380(9859), 2224–60, doi:10.1016/S0140-6736(12)61766-8, 2012.

Liu, M., Huang, Y., Ma, Z., Jin, Z., Liu, X., Wang, H., Liu, Y., Wang, J., Jantunen, M., Bi, J. and Kinney, P. L.: Spatial and temporal trends in the mortality burden of air pollution in China: 2004–2012, Environ. Int., 98, 75–81, doi:10.1016/j.envint.2016.10.003, 2017.

Malley, C. S., Henze, D. K., Kuylenstierna, J. C. I., Vallack, H. W., Davila, Y., Anenberg, S. C., Turner, M. C. and Ashmore, M. R.: Updated global estimates of respiratory mortality in adults ≥ 30 years of age attributable to long-term ozone exposure, Environ. Health Perspect., 125(8), 1–9, doi:10.1289/EHP1390, 2017.

Mathur, R., Xing, J., Gilliam, R., Sarwar, G., Hogrefe, C., Pleim, J., Pouliot, G., Roselle, S., Spero, T. L., Wong, D. C. and Young, J.: Extending the Community Multiscale Air Quality (CMAQ) modeling system to hemispheric scales: Overview of process considerations and initial applications, Atmos. Chem. Phys., 17(20), 12449–12474, doi:10.5194/acp-17-12449-2017, 2017.

Murphy, D. M., Chow, J. C., Leibensperger, E. M., Malm, W. C., Pitchford, M., Schichtel, B. A., Watson, J. G. and White, W. H.: Decreases in elemental carbon and fine particle mass in the United States, Atmos. Chem. Phys., 11(10), 4679–4686, doi:10.5194/acp-11-4679-2011, 2011.

Murray, C. J. L., Collaborators US Burden of Disease: The State of US Health, 1990-2010, JAMA, 310(6), 591, doi:10.1001/jama.2013.13805, 2013.

Pope, C. A., Ezzati, M. and Dockery, D. W.: Fine-Particulate Air Pollution and Life Expectancy in the United States, N. Engl. J. Med., 360(4), 376–386, doi:10.1056/NEJMsa0805646, 2009.

Porter, P. S., Rao, S. T., Hogrefe, C. and Mathur, R.: A reduced form model for ozone based on two decades of CMAQ simulations for the continental United States, Atmos. Pollut. Res., 8(2), 275–284, doi:10.1016/j.apr.2016.09.005, 2017.

Punger, E. M. and West, J. J.: The effect of grid resolution on estimates of the burden of ozone and fine particulate matter on premature mortality in the USA, Air Qual., Atmos. Heal.,, 6(3), 563–573, doi:10.1007/s11869-013-0197-8, 2013.

Silva, R. A., West, J. J., Zhang, Y., Anenberg, S. C., Lamarque, J.-F., Shindell, D. T., Collins, W. J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L. W., Nagashima, T., Naik, V., Rumbold, S., Skeie, R., Sudo, K., Takemura, T., Bergmann, D., Cameron-Smith, P., Cionni, I., Doherty, R. M., Eyring, V., Josse, B., MacKenzie, I. A, Plummer, D., Righi, M., Stevenson, D. S., Strode, S., Szopa, S. and Zeng, G.: Global premature mortality due to anthropogenic outdoor air pollution and the contribution of past climate change, Environ. Res. Lett., 8, 034005, doi:10.1088/1748-9326/8/3/034005, 2013.

Silva, R. A., West, J. J., Lamarque, J. F., Shindell, D. T., Collins, W. J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L. W., Nagashima, T., Naik, V., Rumbold, S. T., Sudo, K., Takemura, T., Bergmann, D., Cameron-Smith, P., Cionni, I., Doherty, R. M., Eyring, V., Josse, B., MacKenzie, I. A., Plummer, D., Righi, M., Stevenson, D. S., Strode, S., Szopa, S. and Zengast, G.: The effect of future ambient air pollution on human premature mortality to 2100 using output from the ACCMIP model ensemble, Atmos. Chem. Phys., 16(15), 9847–9862, doi:10.5194/acp-16-9847-2016, 2016a.

Silva, R. A., Adelman, Z., Fry, M. M. and West, J. J.: The impact of individual anthropogenic emissions sectors on the global burden of human mortality due to ambient air pollution, Environ. Health Perspect., 124(11), 1776–1784, doi:10.1289/EHP177, 2016b.

Spracklen, D. V., Logan, J. A., Mickley, L. J., Park, R. J., Yevich, R., Westerling, A. L. and Jaffe, D. A.: Wildfires drive interannual variability of organic carbon aerosol in the western U.S. in summer, Geophys. Res. Lett., 34(16), 2–5, doi:10.1029/2007GL030037, 2007.

Thompson, T. M., Saari, R. K. and Selin, N. E.: Air quality resolution for health impact assessment: influence of regional characteristics, Atmos. Chem. Phys., 14(2), 969–978, doi:10.5194/acp-14-969-2014, 2014.

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.

US Burden of Disease Collaborators: The state of us health, 1990-2010: Burden of diseases, injuries, and risk factors, J. Am. Med. Assoc., 310(6), 591–606, http://dx.doi.org/10.1001/jama.2013.13805, 2013.

US EPA. 2011. "Benefits and Costs of the Clean Air Act 1990-2020, the Second Prospective Study." https://www.epa.gov/clean-air-act-overview/benefits-andcosts-clean-air-act-1990-2020-second-prospective-study (accessed 24 Novermber 2017).

US EPA. 2014. "Control of Air Pollution from Motor Vehicles: Tier3 Motor Vehicle Emission and Fuel Standards Final Rule." https://www.epa.gov/regulations-emissions-vehicles-and-engines/final-rule-control-air-pollution-motor-vehicles-tier-3 (accessed 5 September 2018).

US EPA. 2017. " https://gispub.epa.gov/air/trendsreport/2017/" (accessed 2 August 2017).

van Donkelaar, A., Martin, R., Brauer, M. and Boys, B. L.: Use of Satellite Observations for Long-Term Exposure Assessment of Global Concentrations of Fine Particulate Matter Concentrations of Fine Particulate Matter, Environ. Health Perspect., 123(February), 135–143, doi:10.1289/ehp.1408646, 2015.

Wang, J., Xing, J., Mathur, R., Pleim, J. E., Wang, S., Hogrefe, C., Gan, C.-M., Wong, D. C. and Hao, J.: Historical Trends in $PM_{2.5}$-Related Premature Mortality during 1990-2010 across the Northern Hemisphere, Environ. Health Perspect., 125(3), 400–408, doi:10.1289/EHP298, 2017.

World Health Organization (WHO). 2016. Ambient air pollution: A global assessment of exposure and burden of disease, ISBN:9789241511353.

Wong, D. C., Pleim, J., Mathur, R., Binkowski, F., Otte, T., Gilliam, R., Pouliot, G., Xiu, A., Young, J. O. and Kang, D.: WRF-CMAQ two-way coupled system with aerosol feedback: Software development and preliminary results, Geosci. Model Dev., 5(2), 299–312, doi:10.5194/gmd-5-299-2012, 2012.

Xing, J., Pleim, J., Mathur, R., Pouliot, G., Hogrefe, C., Gan, C. M. and Wei, C.: Historical gaseous and primary aerosol emissions in the United States from 1990 to 2010, Atmos. Chem. Phys., 13(15), 7531–7549, doi:10.5194/acp-13-7531-2013, 2013.

Xing, J., Mathur, R., Pleim, J., Hogrefe, C., Gan, C. M., Wong, D. C., Wei, C., Gilliam, R. and Pouliot, G.: Observations and modeling of air quality trends over 1990-2010 across the Northern Hemisphere: China, the United States and Europe, Atmos. Chem. Phys., 15(5), 2723–2747, doi:10.5194/acp-15-2723-2015, 2015.

Xu, Y., Serre, M. L., Reyes, J. and Vizuete, W.: Bayesian Maximum Entropy Integration of Ozone Observations and Model Predictions: A National Application, Environ. Sci. Technol., 50(8), 4393–4400, doi:10.1021/acs.est.6b00096, 2016.

Zhang, Y., Bowden, J. H., Adelman, Z., Naik, V., Horowitz, L. W., Smith, S. J., and West, J. J.: Co-benefits of global and regional greenhouse gas mitigation for US air quality in 2050, Atmos. Chem. Phys., 16, 9533-9548, https://doi.org/10.5194/acp-16-9533-2016, 2016.

Tables and Figures

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 the (2010-1990)/1990. The mortality burdens in the US for $PM_{2.5}$ and O_3 in 1990 are 123,700 deaths yr^{-1} (70,800-178,100) and 10,900 deaths yr^{-1} (3,700-17,500).

		2010 (deaths yr^{-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%
	MortalityRates 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	8,100 (2,700-13,100)	-25%
	MortalityRates change only	13,100 (4,400-21,000)	20%
	Population change only	14,100 (4,800-22,700)	30%
	Concentration change excluded	16,900 (5,700-27,000)	55%

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 yr^{-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	13712	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

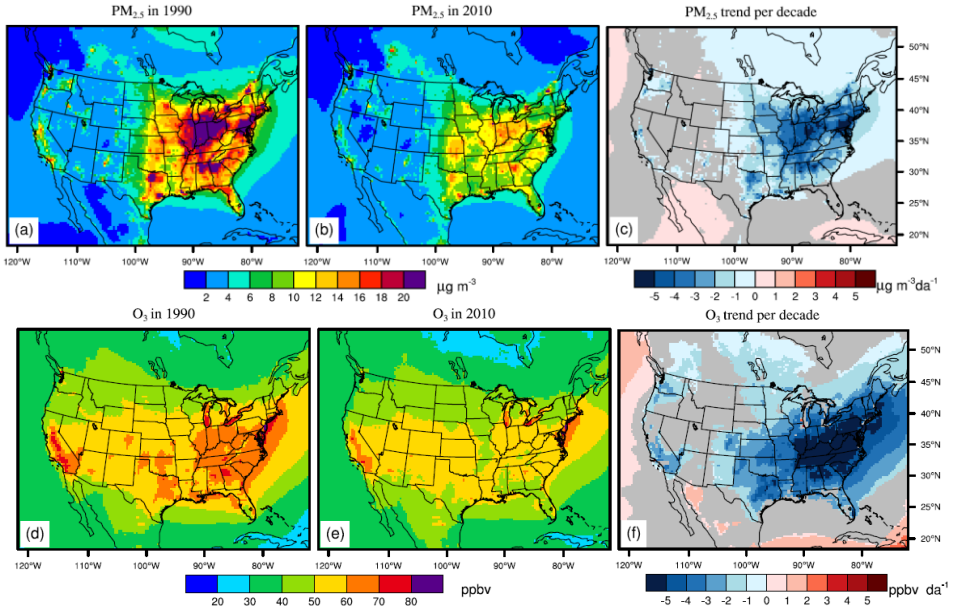


Figure 1: Annual mean $PM_{2.5}$ (μg m^{-3}) in 1990 (a), 2010 (b), and the 21-yr trends (c, μg m^{-3} da^{-1} (μg m^{-3} per decade)), and summertime average of 1hr daily maximum O_3 in 1990 (ppbv) (d), 2010 (e) and the trend (f, ppbv da^{-1} (ppbv per decade)). The grey shaded areas in panels c and f indicate trends that are insignificant with p-values for the standard Student-t test larger than 0.05.

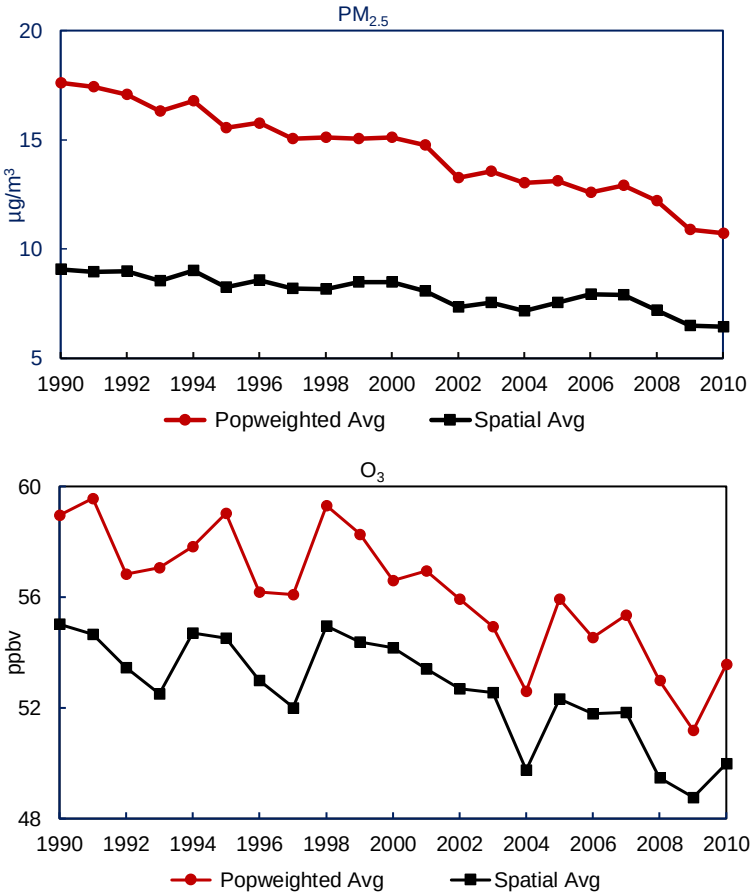


Figure 2: Population-weighted average (Popweighted-Avg) and spatial average over CONUS land areas of annual average $PM_{2.5}$ (top) and summertime average of 1hr daily maximum O_3 (bottom) concentration from 1990 to 2010. Population-weighted average concentrations are based on population in each year.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

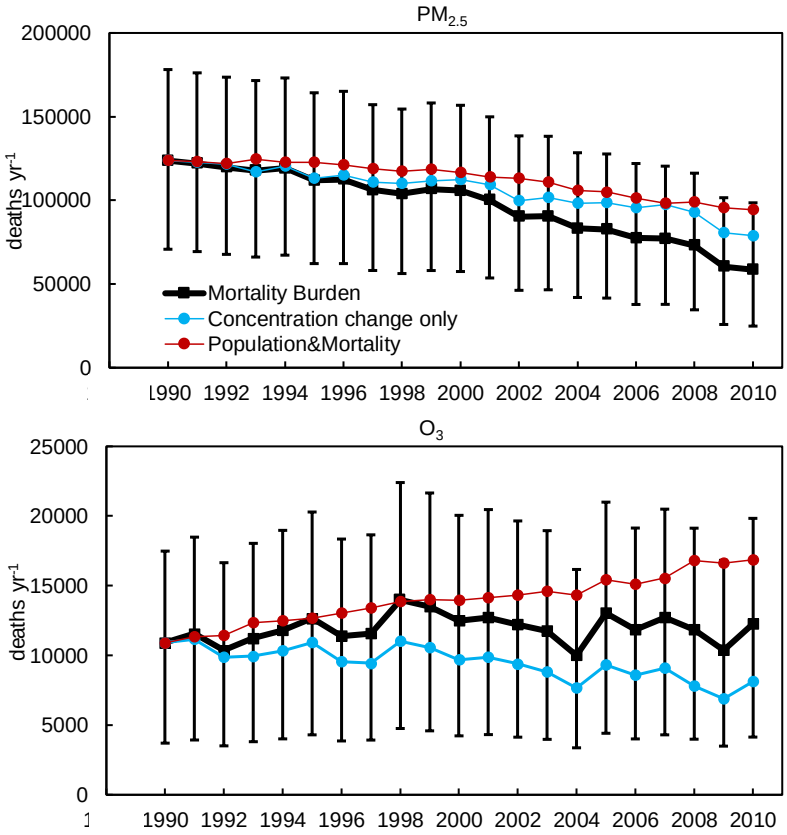


Figure 3: Trends in the total mortality burden (black) for $PM_{2.5}$ (top, as a total of ischemic heart disease (IHD) + stroke (STROKE) + chronic obstructive pulmonary disease (COPD) + lung cancer (LC)) and O_3 (bottom, chronic respiratory disease (RESP)), and mortality burdens considering the air quality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

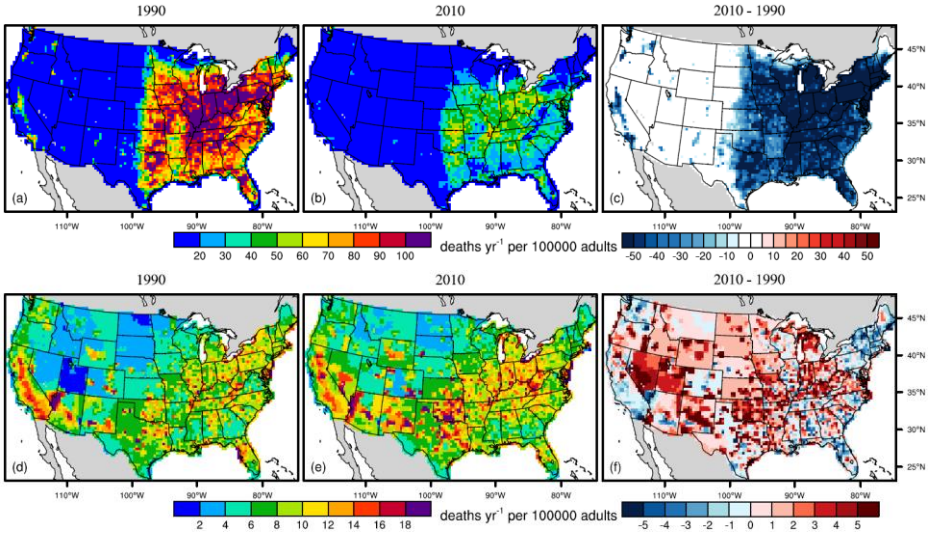


Figure 4: The mortality burdens associated with $PM_{2.5}$ (a, b), O_3 (d, e) in 1990 (a, c) and 2010 (b, d), and the differences (2010 minus 1990) (c, f) for each 36km×36km grid cell. Units are deaths yr^{-1} per 100,000 adults (above 25 yrs old).

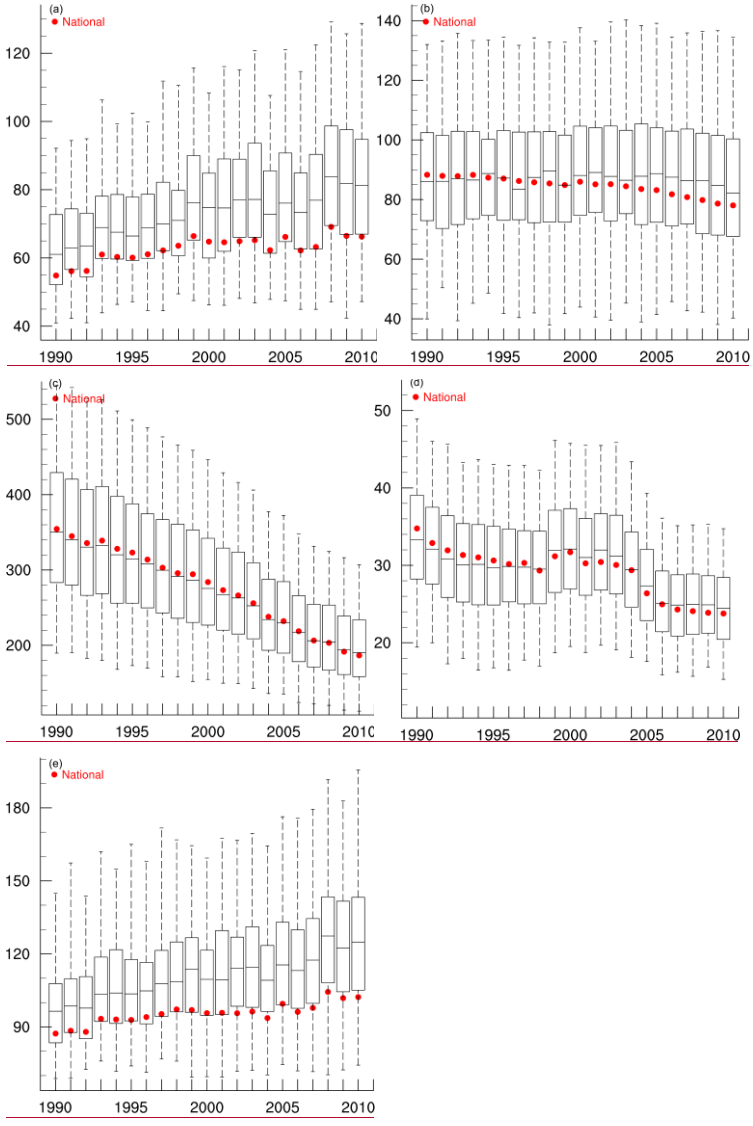

(a)
National
120
100
80
60
40
1990
1995
2000
2005
2010
(b)
National
140
120
100
80
60
40
1990
1995
2000
2005
2010
(c)
National
500
400
300
200
1990
1995
2000
2005
2010
(d)
National
50
40
30
20
1990
1995
2000
2005
2010
(e)
National
180
150
120
90
1990
1995
2000
2005
2010

Figure 5: The baseline mortality rates for specific causes of death related with $PM_{2.5}$, including chronic obstructive pulmonary disease (a), lung cancer (b), ischemic heart disease (c) and stroke (d), and respiratory diseases related with O_3 (e). The bottom whiskers, bottom border, middle line, top border and the top whiskers of the boxes, indicate the 5th, 25th, 50th, and 75th, 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 in y axis are per 100,000 people.

Formatted: English (United Kingdom)

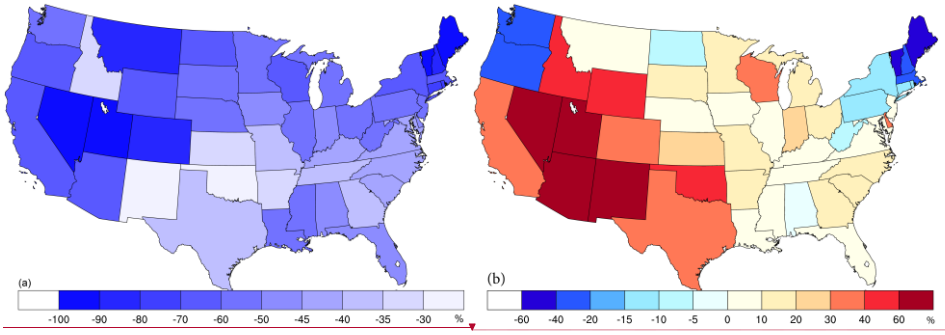
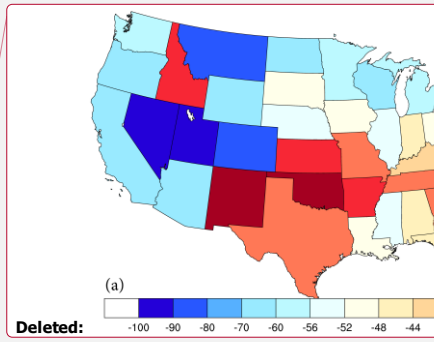


Figure 6: Relative mortality burden changes from 1990 to 2010 for the 48 states and the District of Columbia for $PM_{2.5}$ (a) and O_3 (b). The relative changes are calculated as (2010-1990)/1990×100%. Note the different color scales for the two plots. The values for the District of Columbia are -37%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



Deleted:

Deleted: 5

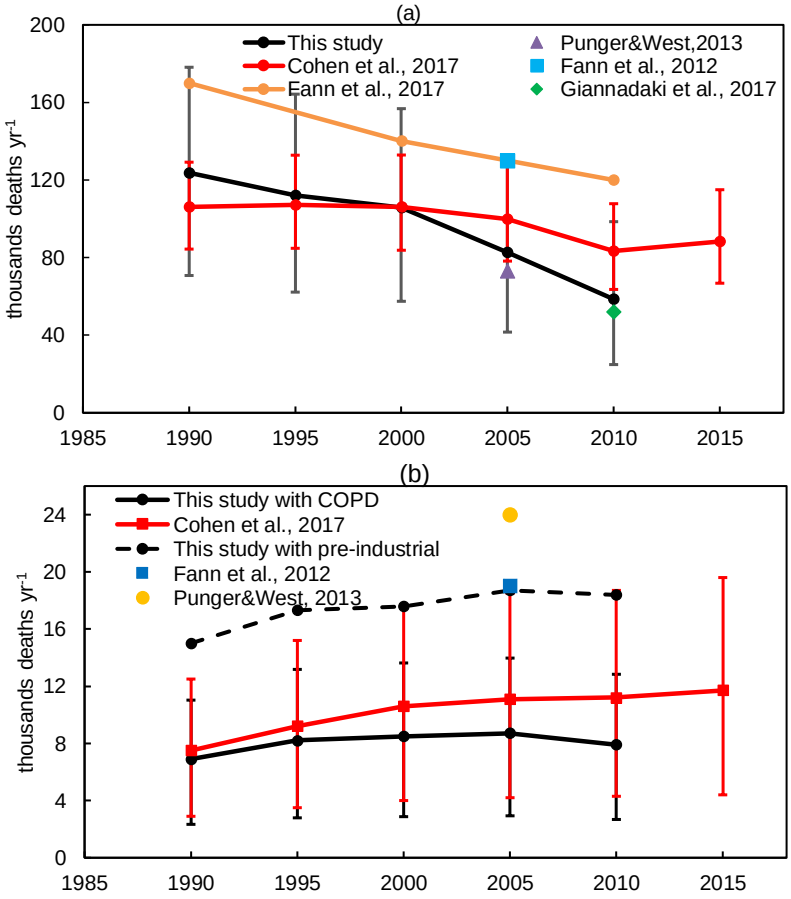


Figure 7: Comparisons of the U.S. mortality burdens attributed to $PM_{2.5}$ (a), and O_3 (b) in this study, with Cohen et al., (2017), Fann et al., (2017), Fann et al., (2012a), Punger and West (2013), and Giannadaki et al., (2017). The black line for O_3 is the recalculated O_3 mortality burden from 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).

Deleted: 6