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Supplement of

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 et al.

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1. Estimating county-level baseline mortality rates from CDC WONDER database

To get the baseline mortality rates for counties with suppressed data, we grouped counties with unsuppressed mortality in each state, calculated the total deaths, and then subtracted these unsuppressed deaths from the total deaths in that state. The resulting total deaths count for the suppressed counties were then divided by the total population in these suppressed counties to get the baseline mortality rates. In some instances, the state-level death counts were also suppressed (e.g. for IHD and STROKE, age-specific baseline mortality rates are required). When this happens, we calculated the national-level baseline mortality rates and assign the value to those suppressed states.

To get the baseline mortality rates for the counties with “missing” or “unreliable” data (hereafter, “unreliable counties”), we summed the total deaths from the unreliable counties in each state, divided the summed total deaths by the total population in these unreliable counties to calculate the average baseline mortality rate (y_0), and assigned this average y_0 to those counties; if the summed deaths from those counties in this state are still unreliable/missing, we aggregated unreliable counties at a regional level, dividing the US into four regions: Northeast, Midwest, South and West (BenMAP, 2017), recalculated the baseline mortality rates at the regional level, and then assign the regional y_0 to the counties; if there are still counties with no available data, we estimated and assigned the national-level y_0. By using the state/regional/national average to estimate the suppressed or unreliable mortality data, the accuracy of the local baseline mortality rates can be improved (Tiwari et al. 2014).

2. Sensitivity analysis for the O_3 mortality burden with simulated pre-industrial O_3 background concentration

We did a sensitivity analysis by using the “pre-industrial” O_3 concentration as the counterfactual, which evaluates the burden due to “anthropogenic” pollution, following previous studies (Anenberg et al. 2010; Fang et al. 2013; Lelieveld et al. 2013; Silva et al. 2013, 2016). We obtain the pre-industrial O_3 concentrations by using an ensemble of 14 global chemistry–climate models under the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al. 2013; Silva et al. 2013), in which pre-industrial air quality was simulated using anthropogenic emissions in 1850. The pre-industrial summertime (April to September) average of 1hr daily maximum O_3 simulated under the ACCMIP is higher in the southeast and intermountain areas of US (>30 ppbv), and lower in the northeast and central U.S, (>28 ppbv) (see Fig. S10), which are both higher than the counterfactual assumed by Fann et al. (2012) (30 ppb in the west and 22 ppb in the east). According to the HIF, higher background O_3 concentrations are expected to lead to lower O_3 mortality burden estimates in the current year. The summertime O_3 concentrations from the ensemble mean are lower than the average O_3 counterfactual (37.6 ppbv) we originally used in the O_3 mortality burden, which is expected to lead to higher O_3 mortality burden estimates.

References

Anderson RN, Miniño AM, Hoyert DL, Rosenberg HM. 2001. Comparability of cause of death between ICD-9 and ICD-10: preliminary estimates. Natl. Vital Stat. Rep. 49: 1–32.

Anderson RN, Rosenberg HM. 2003. 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. 22:1551–1570; doi:10.1002/sim.1511.

Anenberg SC, Horowitz LW, Tong DQ, West JJ. 2010. An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling. Environ. Health Perspect. 118:1189–1195; doi:10.1289/ehp.0901220.

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 LW, Mauzerall DL. 2013. 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.

Fann N, Lamson AD, Anenberg SC, Wesson K, Risley D, Hubbell BJ. 2012. Estimating the national public health burden associated with exposure to ambient $PM_{2.5}$ and ozone. Risk Anal. 32:81–95; doi:10.1111/j.1539-6924.2011.01630.x.

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, 179–206, https://doi.org/10.5194/gmd-6-179-2013, 2013 Lelieveld J, Barlas C, Giannadaki D, Pozzer A. 2013. Model calculated global, regional and megacity premature mortality due to air pollution. Atmos. Chem. Phys. 13:7023–7037; doi:10.5194/acp-13-7023-2013.

Silva RA, West JJ, Zhang Y, Anenberg SC, Lamarque J-F, Shindell DT, et al. 2013. Global premature mortality due to anthropogenic outdoor air pollution and the contribution of past climate change. Environ. Res. Lett. 8:34005; doi:10.1088/1748-9326/8/3/034005.

Silva RA, West JJ, Lamarque JF, Shindell DT, Collins WJ, Dalsoren S, et al. 2016. The effect of future ambient air pollution on human premature mortality to 2100 using output from the ACCMIP model ensemble. Atmos. Chem. Phys. 16:9847–9862; doi:10.5194/acp-16-9847-2016.

Tiwari C, Beyer K, Rushton G. 2014. The impact of data suppression on local mortality rates: The case of cdc wonder. Am. J. Public Health 104:1386–1388; doi:10.2105/AJPH.2014.301900.

Table S**1**. The list of International Classification of Diseases (ICD) codes for specific causes of death, with ICD9 implemented from 1990 to 1998, and ICD10 from 1999 to 2010.

	ICD9 code	**ICD10**
chronic respiratory diseases (RESP)	070.22-070.23, 070.32-070.33, 070.44-070.49, 070.54-070.59, 135, 456.0-456.2, 470-475, 477-478, 490-492.8, 493-496, 500-506, 508, 515-519, 571, 572.3-572.8, 573.9	B18, D86.0, D86.2, D86.9, I85, J30-J39, J40-J47, J60-J65, J66-J70(except J69), J82, J84, J92, J93.0, J93.1, J95, J98 (except J98.1, J98.2, J98.3, J98.9), K70, K71.7, K72.1-K72.9, K73-K74, K75.2-K75.9, K76.6-K76.7, K76.9
chronic obstructive pulmonary disease (COPD)	490-492.8, 494, 496	J40-J44, J47
ischemic heart disease (IHD)	410-414, 429.2	I20-I25
lung cancer (LC)	162-162.9, 231.1, 231.2, 231.8, 235.7	C33-C34, D02.1-D02.2, D38.1
cerebrovascular disease and ischemic stroke (STROKE)	430-435, 437.0-437.2, 437.5-437.8	I60-I63, I65-I67, I69.0-I69.3

Table S**2**. The comparability ratios for baseline mortality rates for specific causes of death between ICD9 and ICD10. The ratios for each disease were calculated as the total deaths categorized in ICD10 divided by the same total deaths categorized in ICD9 (Anderson et al., 2001; Anderson & Rosenberg, 2003). To eliminate the discontinuity, the health burden from 1990 to 1998 is multiplied by this ratio. For details please see methods.

Diseases	**Ratio**
RESP	1.046
COPD	1.056
IHD	0.999
LC	0.984
STROKE	1.059

Table S**3**. The regional mean of annual $PM_{2.5}$ and summertime average of 1hr daily maximum O_3 concentration in 1990, 2010, and decadal trends (units μg m^{-3} per decade for $PM_{2.5}$, and ppbv per decade for O_3). Bold values indicate trends that are significant with P-values for the standard Student-t test smaller than 0.05. Regions are defined as the US nine climate regions (see Fig. S3).

	Annual $PM_{2.5}$ (μg m^{-3})			**Summertime (Apr-Sep) average of 1hr daily maximum O_3 (ppbv)**		
	1990	2010	trends	1990	2010	trends
Northwest	4.2	3.6	-0.10	46	43	**-1.0**
West	4.9	3.3	0.16	55	52	**-1.1**
West N. Central	4.8	4.2	**-0.37**	50	47	-1.1
Southwest	3.6	3.1	-0.07	55	53	**-1.0**
South	9.9	7.9	**-1.00**	58	52	**-2.7**
East N. Central	11.9	7.7	**-2.06**	51	47	**-2.2**
Central	18.0	11.5	**-3.48**	61	53	**-5.1**
Southeast	14.0	9.6	**-1.53**	62	52	**-4.3**
Northeast	14.7	7.9	**-3.14**	57	50	**-4.4**

Table S**4**. The mortality burden associated with ambient $PM_{2.5}$ for specific causes of death (COPD, IHD, LC and STROKE), and with O_3 (for COPD and RESP) from 1990 to 2010, and their trends (deaths yr^{-1}). For O_3, COPD is a subset of RESP.

Year	**COPD**	**IHD**	**LC**	**STROKE**	**COPD-O_3**	**RESP-O_3**
1990	5941	96495	12469	8830	6875	10903
1991	6090	95053	12447	8392	7334	11545
1992	6074	93108	12392	8135	6627	10355
1993	6365	91860	11986	7614	7334	11226
1994	6638	92066	12495	8054	7691	11816
1995	6103	87079	11505	7303	8215	12651
1996	6381	86883	11756	7384	7435	11403
1997	6213	81730	11199	7083	7580	11590
1998	6386	79298	11323	6891	9175	13981
1999	6870	81395	11351	7153	9275	13499
2000	6846	79664	11728	7478	8475	12471
2001	6571	75530	11253	6859	8607	12732
2002	5742	68842	9835	5894	8296	12208
2003	5978	68317	10092	6053	7978	11760
2004	5529	62375	9635	5693	6651	9999
2005	5965	61577	9785	5357	8671	13039
2006	5363	58175	9183	4840	7651	11863
2007	5797	56745	9578	4956	8252	12716
2008	5771	54163	8677	4464	7797	11834
2009	4509	45319	7049	3580	6738	10379
2010	4465	43611	6963	3541	7942	12275
Trend	**-64**	**-2636**	**-251**	**-241**	**24**	**24**

Table S**5**. The trends (TRE) and relative changes (REL) for the spatial average, population-weighted average air quality concentration, total mortality burdens change and due to changes in each of three factors (concentration only, baseline mortality rates only and population only), and where the concentration change is excluded, for $PM_{2.5}$ and O_3 from 1990-2000, 2000-2010, and 1990 to 2010. The units for the trends in concentrations are µg m^{-3} $decade^{-1}$ for $PM_{2.5}$, and ppbv $decade^{-1}$ for O_3. The units for the trends of mortality burden are deaths yr^{-1}. The REL for the pollutants concentration changes are calculated as (Last year – First year)/First year, while for REL in mortality are used linear regression periods for each period to avoid inter-annual variability.

		1990-2000		**2000-2010**		**1990-2010**	
		TRE	REL	TRE	REL	TRE	REL
$PM_{2.5}$	Spatial average	-0.77	-7%	-1.4	-24%	-1.1	-29%
	Population-weighted average	-2.9	-14%	-3.8	-29%	-3.2	-39%
	Mortality burden	-2100	-15%	-4400	-45%	-3200	-54%
	Concentration change only	-1400	-9%	-2800	-30%	-2000	-36%
	MortalityRates change only	-2500	-20%	-3100	-31%	-2800	-45%
	Population change only	2200	18%	2800	19%	2400	40%
	Concentration change excluded	-700	-6%	-2300	-19%	-1600	-24%
O_3	Spatial average	-0.32	-2%	-4.4	-8%	-2.4	-9%
	Population-weighted average	-1.2	-4%	-4.0	-5%	-3.0	-9%
	Mortality burden	240	14%	-70	-2%	24	13%
	Concentration change only	-60	-11%	-210	-16%	-160	-25%
	MortalityRates change only	140	11%	110	8%	90	20%
	Population change only	170	15%	160	12%	160	30%
	Concentration change excluded	330	28%	310	21%	280	55%

Table S**6**. The standard deviation (STD) and coefficient of variation (CV) for the detrended annual mortality burden for $PM_{2.5}$ and O_3 for the periods 1990-2000, 2000-2010, and 1990 to 2010. In addition to the total mortality burdens, we show mortality burdens considering changes from 1990 to 2010 in each factor individually (concentration only, baseline mortality rates only and population only), and with the concentration change excluded. The units for the STD are µg m^{-3} and ppbv for $PM_{2.5}$ and O_3.

		1990-2000		**2000-2010**		**1990-2010**	
		STD	CV	STD	CV	STD	CV
$PM_{2.5}$	Mortality burden	4300	4%	4900	6%	4200	4%
	Concentration change only	2800	2%	4700	5%	3700	3%
	MortalityRates change only	1500	1%	1400	2%	1400	1%
	Population change only	600	0.4%	1300	0.8%	1000	0.7%
	Concentration change excluded	3100	3%	2800	3%	2800	3%
O_3	Mortality burden	1700	15%	1000	9%	1400	12%
	Concentration change only	680	7%	660	8%	670	7%
	MortalityRates change only	220	2%	290	2%	270	2%
	Population change only	60	0.5%	20	0.1%	40	0.4%
	Concentration change excluded	260	2%	380	2%	340	2%

Table S7. The relative (%) mortality burden changes for US 48 states and District of Columbia from 1990 to 2010, for the mortality burden calculated as the total, concentration change only, mortality rates change only and population change only. The relative changes are calculated as (2010-1990)/1990×100%.

	$PM_{2.5}$-related mortality				**O_3-related mortality**			
	Total	**Concentration**	**Mortality rates**	**Population**	**Total**	**Concentration**	**Mortality rates**	**Population**
AL	-45	-34	-36	36	-5	-43	34	24
AR	-33	-25	-32	32	20	-27	31	27
AZ	-65	-66	-48	102	73	-6	2	84
CA	-64	-50	-51	54	39	-3	6	40
CO	-84	-78	-55	80	39	-22	14	59
CT	-74	-59	-49	31	-15	-28	8	11
DC	-37	-29	-36	38	-23	-19	-21	21
DE	-46	-35	-43	53	34	-23	28	36
FL	-48	-40	-46	64	5	-38	17	47
GA	-38	-28	-48	75	14	-38	21	55
IA	-50	-30	-39	21	1	-29	26	13
ID	-31	-31	-48	96	54	-21	21	65
IL	-54	-28	-47	26	8	-10	9	15
IN	-46	-28	-41	33	25	-25	40	20
KS	-35	-11	-42	28	21	-24	37	17
KY	-43	-28	-38	35	10	-33	34	24
LA	-51	-34	-43	32	2	-30	25	15
MA	-68	-46	-49	27	-22	-37	7	14
MD	-41	-30	-39	46	1	-24	8	24
ME	-99	-98	-52	42	-46	-64	24	21
MI	-56	-40	-42	33	13	-22	33	12
MN	-57	-30	-55	42	18	-17	13	28
MO	-39	-24	-37	30	12	-27	28	21
MS	-55	-35	-44	28	5	-37	38	23
MT	-82	-80	-46	69	6	-31	20	28
NC	-41	-30	-47	68	19	-38	32	47

ND	-69	-58	-46	28	1	-38	39	9
NE	-52	-28	-47	29	9	-21	17	18
NH	-84	-74	-51	51	-34	-54	14	25
NJ	-59	-39	-48	34	8	-16	9	18
NM	-26	-38	-38	97	64	-15	33	44
NV	-97	-98	-52	175	76	-15	-10	140
NY	-62	-40	-47	25	-12	-15	-2	11
OH	-50	-30	-42	28	18	-21	34	12
OK	-29	-20	-33	33	45	-25	58	22
OR	-65	-45	-56	55	-27	-53	14	41
PA	-60	-37	-46	26	-11	-29	15	10
RI	-73	-55	-47	23	-10	-35	25	10
SC	-42	-34	-44	66	16	-42	41	41
SD	-51	-35	-44	29	17	-32	40	19
TN	-39	-34	-36	50	4	-40	31	35
TX	-36	-35	-46	73	34	-28	26	50
UT	-90	-88	-56	80	68	-11	9	76
VA	-43	-32	-43	56	10	-27	19	30
VT	-93	-90	-45	47	-53	-65	14	20
WA	-57	-45	-49	63	-27	-52	9	44
WI	-61	-42	-46	34	31	-14	23	26
WV	-54	-34	-40	22	-7	-38	38	11
WY	-65	-64	-26	132	51	-13	30	34

Table S**8**. The calculated $PM_{2.5}$- and O_3-related mortality burdens for the US from 1990 to 2010 with County-level baseline mortality rates and National-average baseline mortality rates. The absolute differences (Abs diff) are using the National-average method minus County-level method, and the relative differences (Rel diff) are calculated as (National-County)/County. Units are deaths yr^{-1}.

	$PM_{2.5}$-related mortality				O_3-related mortality			
Year	County	National	Abs diff	Rel diff	County	National	Abs diff	Rel diff
1990	123735	121587	-2148	-1.7%	10903	10930	27	0.2%
1991	121982	120015	-1967	-1.6%	11545	11604	59	0.5%
1992	119709	117734	-1975	-1.7%	10355	10350	-5	0.0%
1993	117825	115686	-2139	-1.8%	11226	11265	39	0.3%
1994	119253	116909	-2344	-2.0%	11816	11827	11	0.1%
1995	111990	109589	-2401	-2.1%	12651	12664	13	0.1%
1996	112404	110353	-2051	-1.8%	11403	11354	-49	-0.4%
1997	106225	104335	-1890	-1.8%	11590	11598	8	0.1%
1998	103898	102113	-1785	-1.7%	13981	13964	-17	-0.1%
1999	106770	104891	-1879	-1.8%	13499	13440	-59	-0.4%
2000	105717	103691	-2026	-1.9%	12471	12338	-133	-1.1%
2001	100213	98533	-1680	-1.7%	12732	12726	-6	0.0%
2002	90312	88907	-1405	-1.6%	12208	12193	-15	-0.1%
2003	90441	89082	-1359	-1.5%	11760	11751	-9	-0.1%
2004	83231	81862	-1369	-1.6%	9999	10049	50	0.5%
2005	82684	81200	-1484	-1.8%	13039	13132	93	0.7%
2006	77561	76208	-1353	-1.7%	11863	11913	50	0.4%
2007	77076	75865	-1211	-1.6%	12716	12832	116	0.9%
2008	73074	72031	-1043	-1.4%	11834	12070	236	2.0%
2009	60457	59642	-815	-1.3%	10379	10557	178	1.7%
2010	58580	57306	-1274	-2.2%	12275	12507	232	1.9%

Table S**9**. Comparisons between estimates of $PM_{2.5}$- related deaths in the US for the period 1990 to 2010 between this study and previous studies, corresponding to Figure 6 in the main paper.

Health impact study	Disease category	Air pollution metrics	Year of deaths	Attributable deaths (thousands)
This study[a]	COPD+IHD+LC+STROKE	Annual mean 24-h $PM_{2.5}$	1990	124 (71-178)
			1995	112 (62-164)
			2000	106 (57-157)
			2005	83 (42-128)
			2010	59 (25-99)
Fann et al., 2017[b]	Total all-cause deaths	Annual mean 24-h $PM_{2.5}$	1990	170 (110-220)
			2000	140 (98-190)
			2010	120 (83-160)
Cohen et al., 2017[a]	COPD+IHD+LC+STROKE + lower respiratory infections	Annual mean 24-h $PM_{2.5}$	1990	106 (84-129)
			1995	107 (85-133)
			2000	106 (84-133)
			2005	100 (78-127)
			2010	83 (64-108)
			2015	88 (67-115)
Punger & West, 2013[b]	Total all-cause deaths	Annual mean 24-h $PM_{2.5}$	2005	73 (43-93)
Fann et al., 2012[b]	Total all-cause deaths	Annual mean 24-h $PM_{2.5}$	2005	130 (51-200)
Giannadaki et al., 2017[b]	COPD+IHD+LC+STROKE + lower respiratory infections	Annual mean 24-h $PM_{2.5}$	2010	52 (25-76)

[a]The exposed population for these studies are for adults above 25-year old.
[b]The exposed population for these studies are for adults above 30-year old.

Table S**10**. Comparisons between estimates of O_3- related mortality burdens in the US for the period 1990 to 2010 between this study and previous studies, corresponding to Figure 6 in the main paper.

Health impact study	Disease category	Air pollution metrics	Year of deaths	Attributable deaths (thousands)
This study as reported in Table S4[a]	RESP	Summertime 1-hr daily maximum O_3	1990	6.9 (2.3-11.0)
			1995	8.2 (2.8-13.2)
			2000	8.5 (2.9-13.6)
			2005	8.7 (2.9-14.0)
			2010	7.9 (2.7-12.8)
This study as reported in Table S4[a]	COPD	Summertime 1-hr daily maximum O_3	1990	6.9 (2.3-11.0)
			1995	8.2 (2.8-13.2)
			2000	8.5 (2.9-13.6)
			2005	8.7 (2.0-14.0)
			2010	7.9 (2.7-12.8)
This study with pre-industrial background O_3[a]	RESP	Summertime 1-hr daily maximum O_3	1990	15.0 (5.1-23.8)
			1995	17.3 (5.9-27.5)
			2000	17.6 (6.0-28.0)
			2005	18.7 (6.4-29.8)
			2010	18.4 (6.3-29.6)
Cohen et al., 2017[a]	COPD	3-month of 1-hr daily maximum O_3	1990	7.5 (2.9-12.5)
			1995	9.2 (3.5-15.2)
			2000	10.6 (4.0-17.6)
			2005	11.1 (4.2-18.5)
			2010	11.2 (4.3-18.7)
			2015	11.7 (4.4-19.6)
Punger & West, 2013[b]	RESP	6-month of 1-hr daily maximum O_3	2005	24.0 (6.3-38.3)
Fann et al., 2012[b]	RESP	Summertime daily 8-hour maximum	2005	19.0 (7.6-29.0)

[a]The exposed population for these studies are for adults above 25-year old.
[b]The exposed population for these studies are for adults above 30-year old.

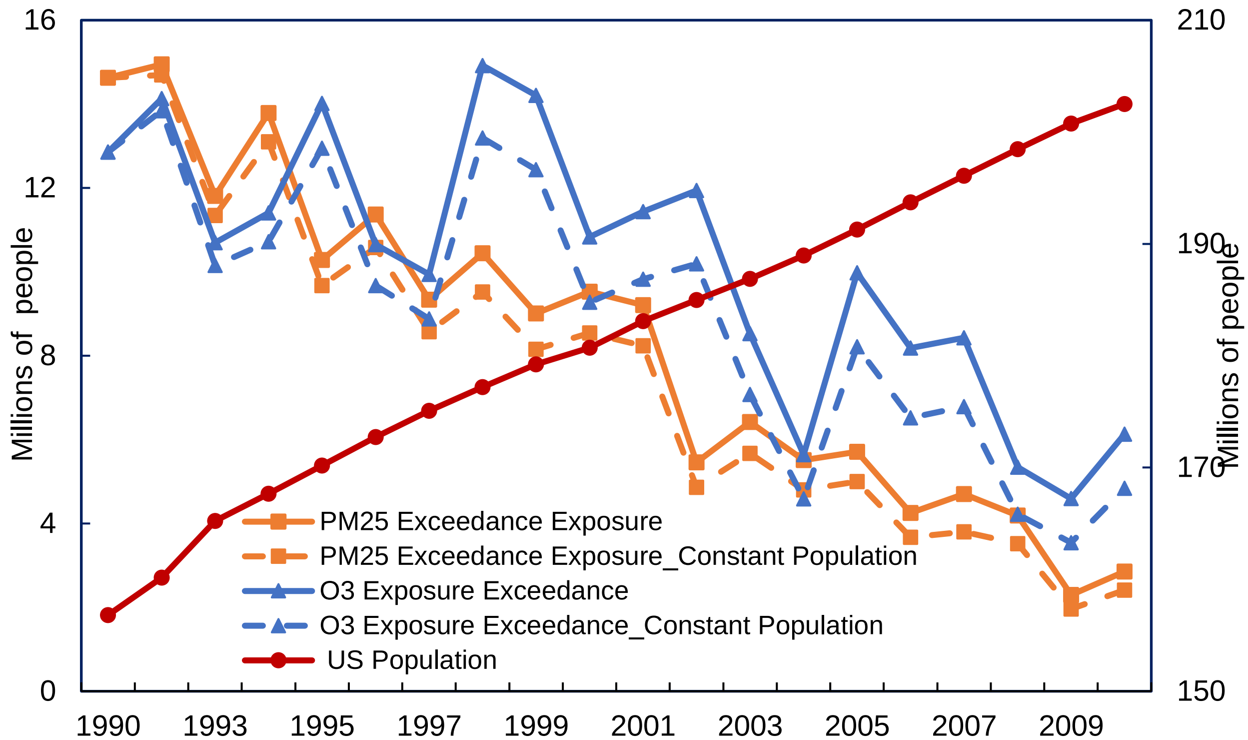

Figure S**1**. The population exposure exceedance using the adult population (> 25 yrs old) multiplied by the air quality exceedance days (million people-days, the number of days that exceed the daily $PM_{2.5}$ standard (35μg m^{-3}), and the daily MDA8 O_3 standard (70 ppbv)), for each year for $PM_{2.5}$ (orange lines) and O_3 (blue lines). The dashed lines are the population exposure exceedance in the case where the population stays constant at the 1990 level. The red line is the US total adult population > 25 yrs old from 1990 to 2010 with the y-axis on the right.

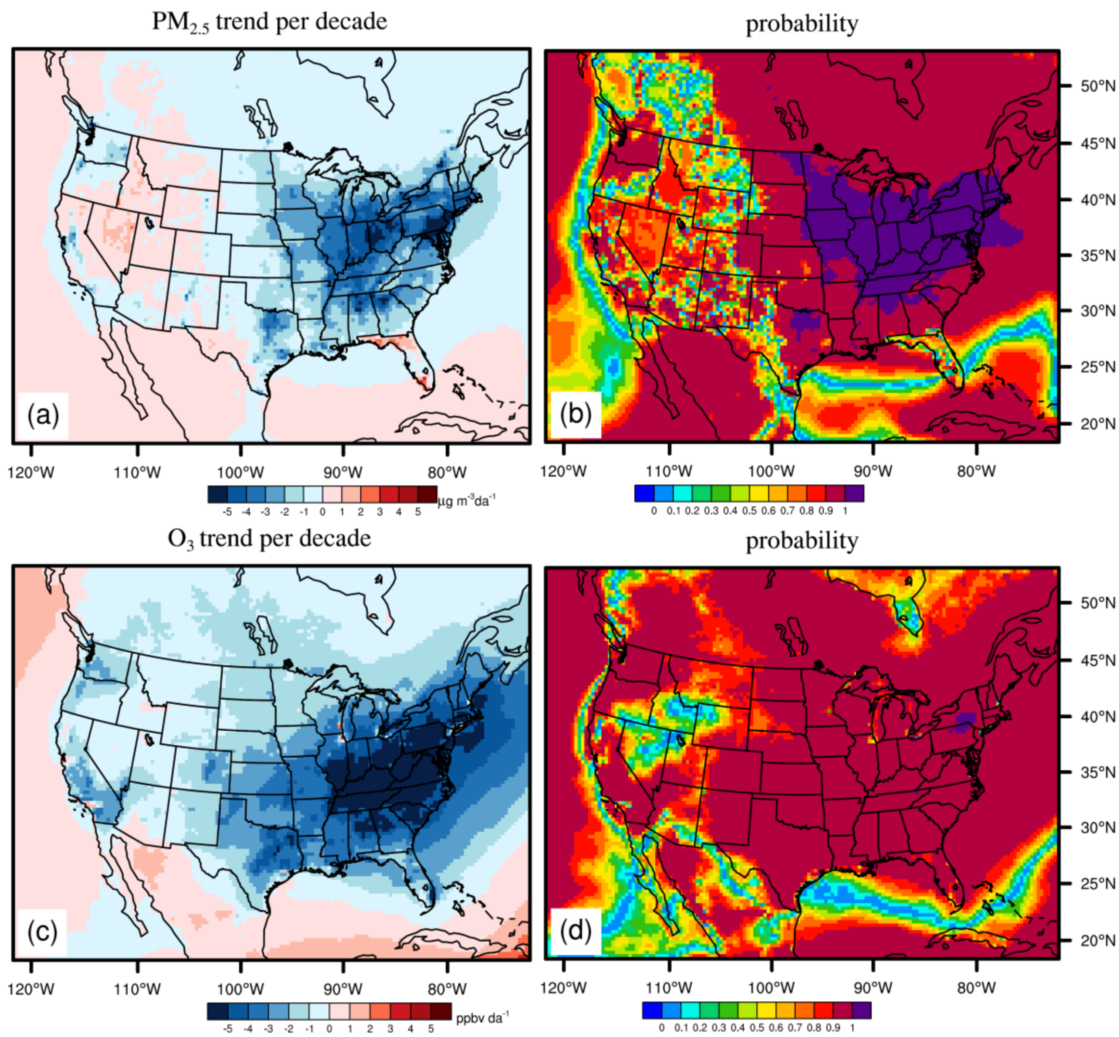

Figure S**2**. 21-yr air quality trends for annual mean $PM_{2.5}$ (a), and summertime (April to September) average of 1hr daily maximum O_3 (c) from 1990 to 2010, with the probability (the confidence estimated for the hypothesis that the trend is significant) for the trends (b,d). The purple color on the rightmost plot means the probability equals to 1.

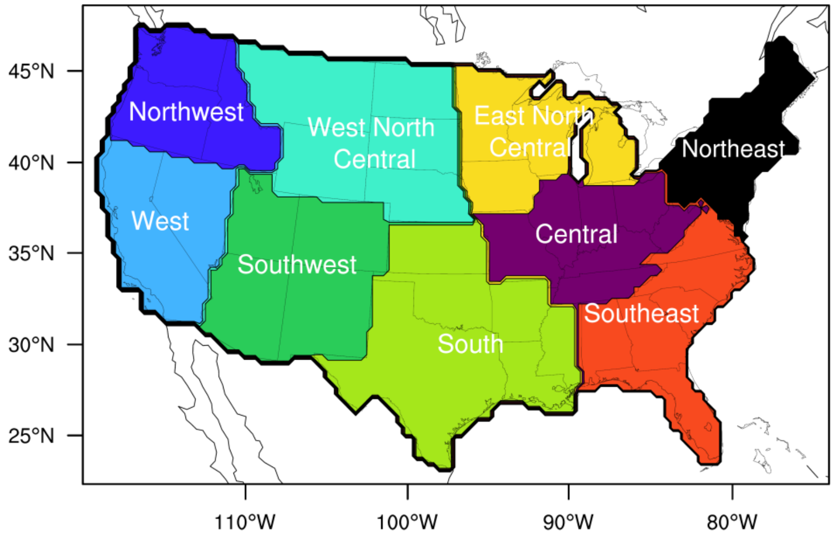

Figure S**3**. The nine US climate regions, following definitions from National Oceanic and Atmospheric Administration (http://www.ncdc.noaa.gov/monitoring-references/maps/us-climate-regions.php, accessed November 30 2017).

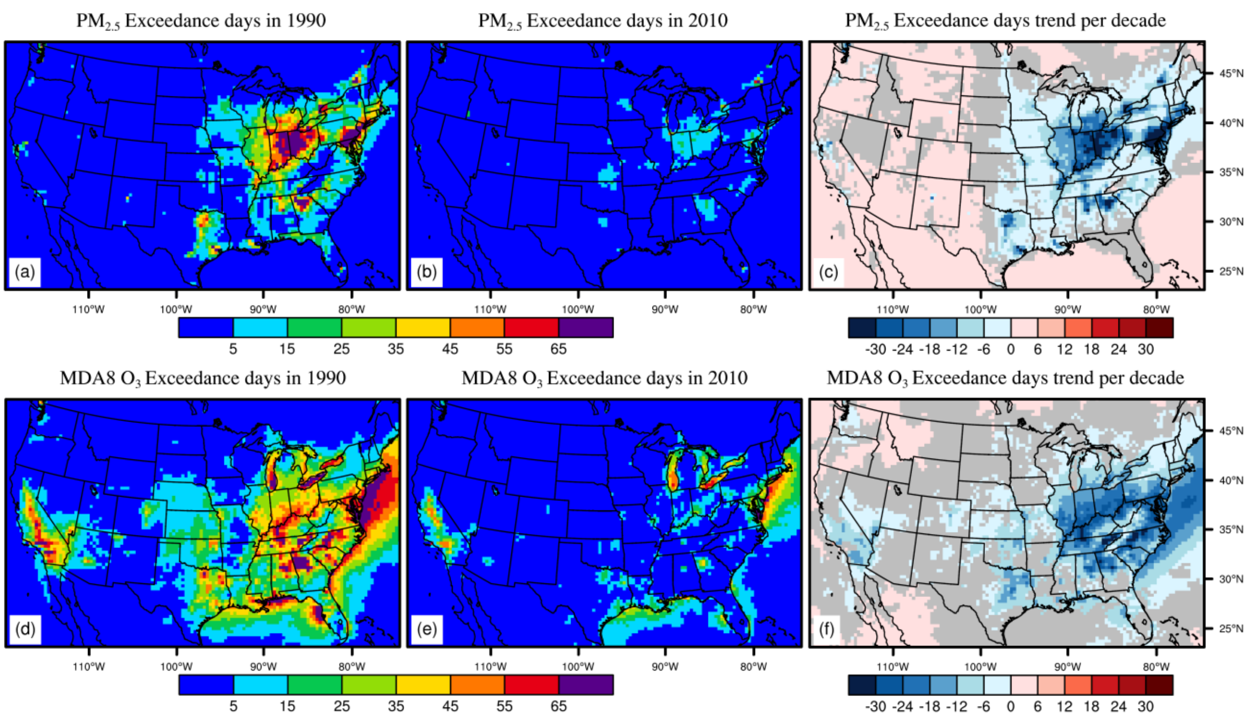

Figure S**4**. The total days of air quality exceedances for the year 1990 (a, d), 2010, (b, e) and the 21-yr trends (per decade, c,f), for daily $PM_{2.5}$ exceeding 35 μg m^{-3}(top), and for daily MDA8 O_3 exceeding 70 ppbv (bottom). The gray areas in (c,f) means the increase/decrease trends are insignificant with p-values for the standard Student-t test larger than 0.05.

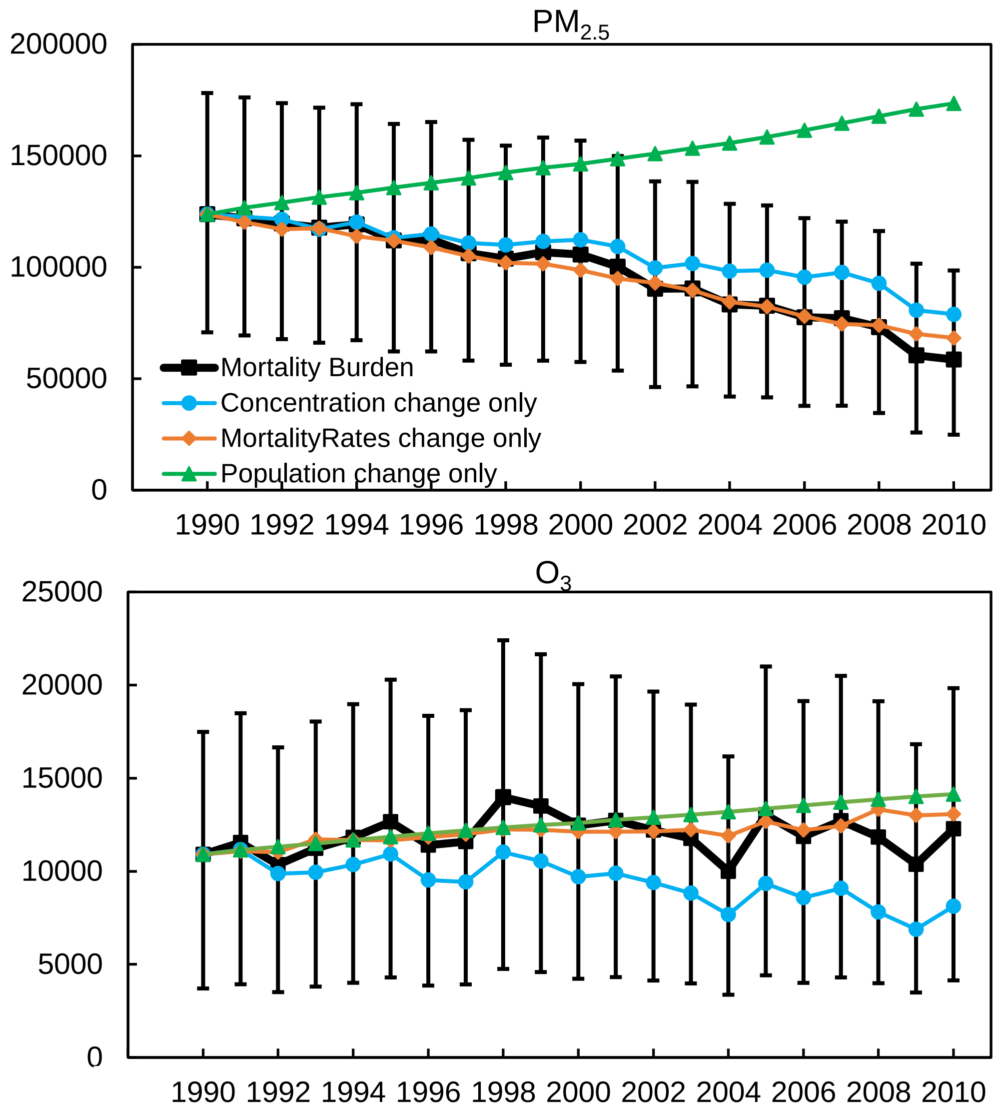

Figure S**5**: Total mortality burdens (black) attributed to $PM_{2.5}$ (top) and O_3 (bottom) considering all the three factors, mortality burden considering the air quality change only (blue), the baseline mortality rates change only (orange), and the population change only (green). Units are deaths yr^{-1}. The error bars are the 95% CI for the total mortality burdens (black).

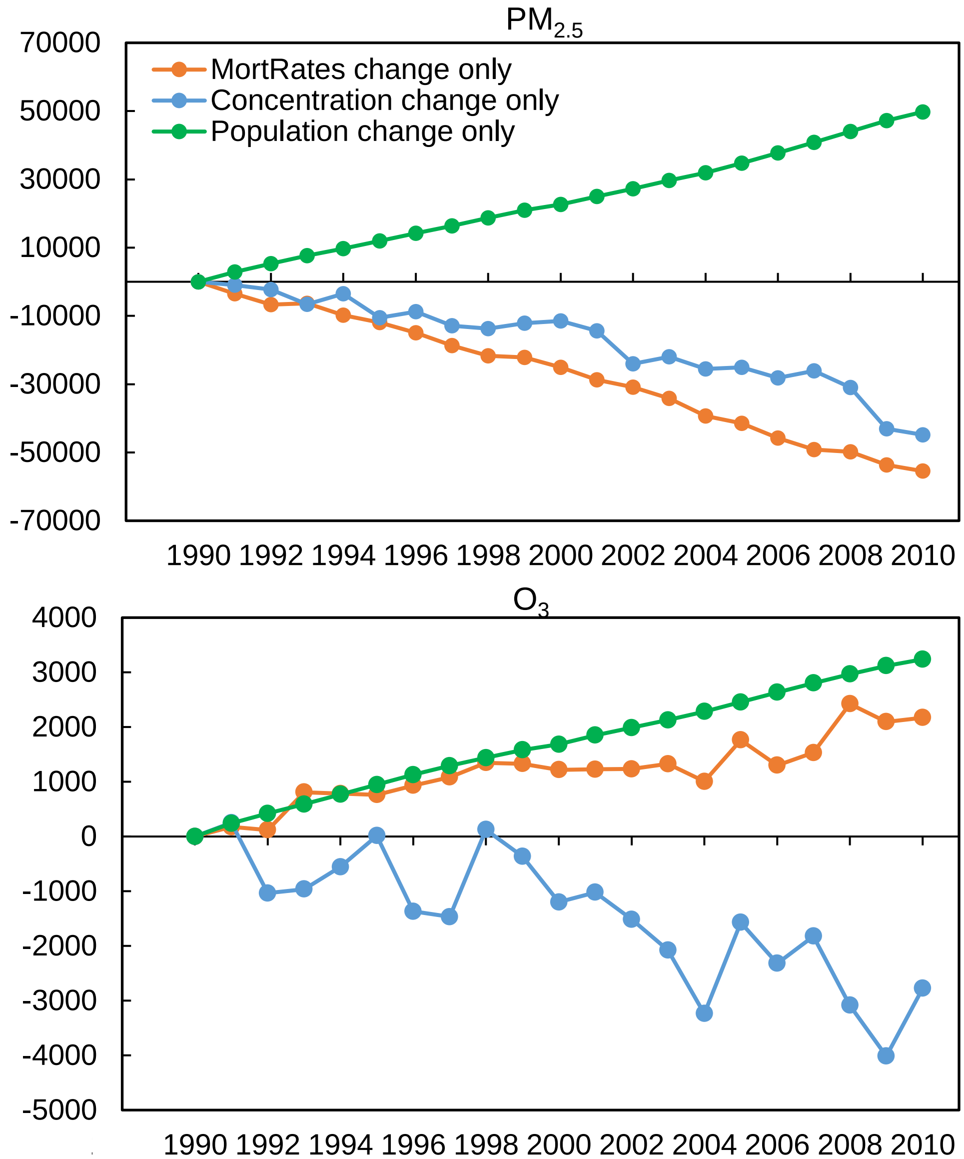

Figure S**6**: Trends for the absolute contribution of the three factors (baseline mortality rates, ambient air pollution concentration, and exposed population individually), in the net changes of the health burden changes for each year compared with 1990, for $PM_{2.5}$ (top) and O_3 (bottom). Units are deaths yr^{-1}.

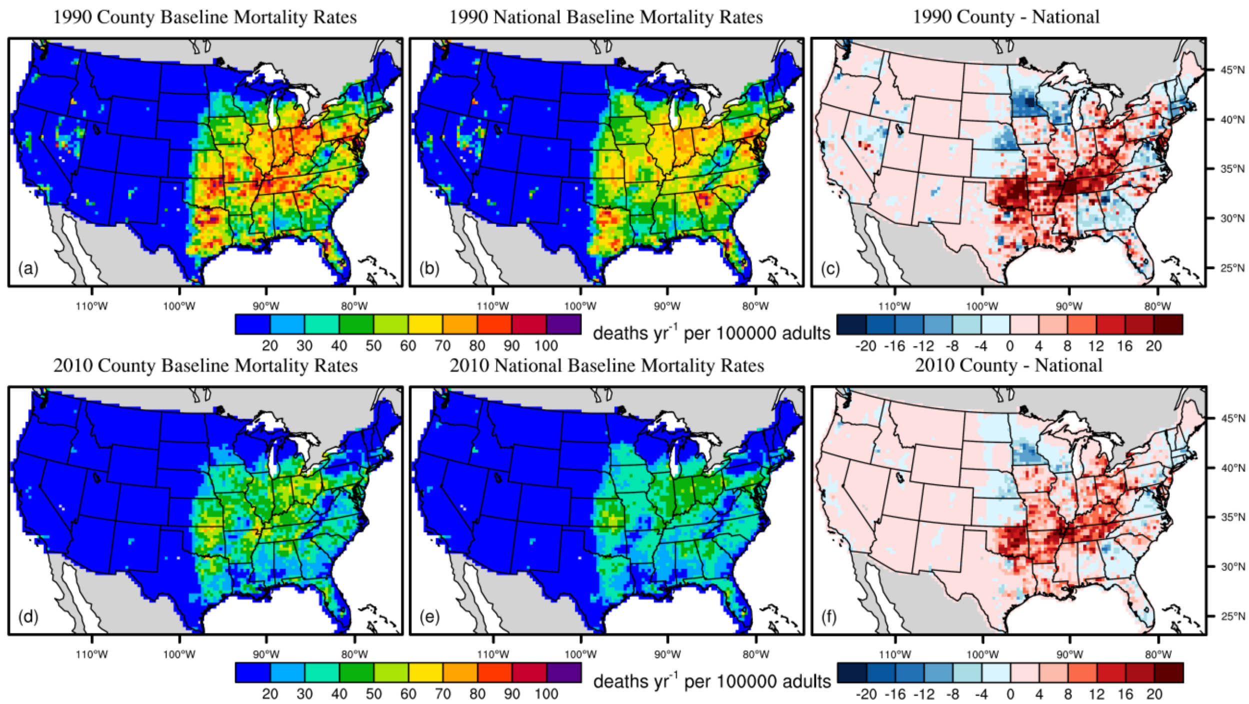

Figure S**7**. The mortality burden for $PM_{2.5}$ using county-level baseline mortality rates (a, d), national baseline mortality rates (b, e) and their differences (c, f) for the year 1990 (top) and 2010 (bottom). The units are deaths yr^{-1} per 100,000 adults.

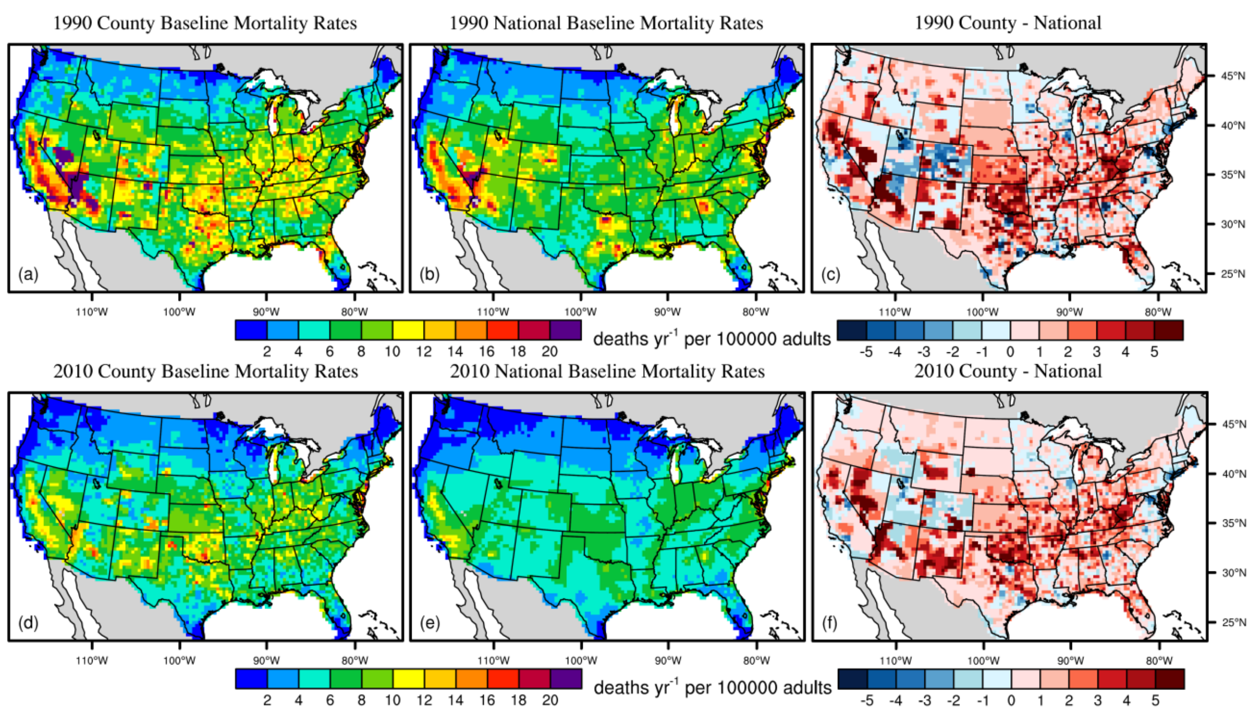

Figure S**8**. The same as fig. S7 but for the O_3-related mortality burden.

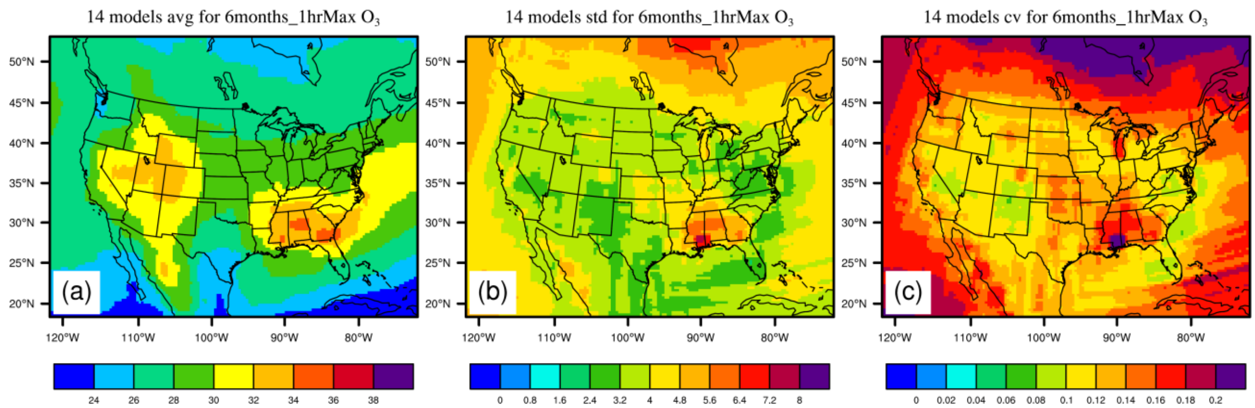

Figure S**9**. Spatial distribution of the summertime (April to September) average of 1hr daily maximum O_3 in 1850, for a) the 14 model average, b) standard deviation for the 14 models, and c) coefficient of variation. The results are regridded from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) 14 model means (Lamarque et al., 2013).