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Air pollution and early deaths in the United States. Part I: Quantifying the impact of major sectors in 2005

Fabio Caiazzo, Akshay Ashok, Ian A. Waitz, Steve H.L. Yim, Steven R.H. Barrett*

Laboratory for Aviation and the Environment, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, United States

HIGHLIGHTS

- Ozone and PM impacts of the major combustion sectors in the U.S. are modeled.
- Early deaths attributable to each sector are estimated.
- ~200,000 early deaths occur in the U.S. each year due to U.S. combustion emissions.
- The leading causes are road transportation and power generation.

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ABSTRACT

Combustion emissions adversely impact air quality and human health. A multiscale air quality model is applied to assess the health impacts of major emissions sectors in United States. Emissions are classified according to six different sources: electric power generation, industry, commercial and residential sources, road transportation, marine transportation and rail transportation. Epidemiological evidence is used to relate long-term population exposure to sector-induced changes in the concentrations of $PM_{2.5}$ and ozone to incidences of premature death. Total combustion emissions in the U.S. account for about 200,000 (90% CI: 90,000–362,000) premature deaths per year in the U.S. due to changes in $PM_{2.5}$ concentrations, and about 10,000 (90% CI: −1000 to 21,000) deaths due to changes in ozone concentrations. The largest contributors for both pollutant-related mortalities are road transportation, causing ~53,000 (90% CI: 24,000–95,000) $PM_{2.5}$-related deaths and ~5000 (90% CI: −900 to 11,000) ozone-related early deaths per year, and power generation, causing ~52,000 (90% CI: 23,000–94,000) $PM_{2.5}$-related and ~2000 (90% CI: −300 to 4000) ozone-related premature mortalities per year. Industrial emissions contribute to ~41,000 (90% CI: 18,000–74,000) early deaths from $PM_{2.5}$ and ~2000 (90% CI: 0 −4000) early deaths from ozone. The results are indicative of the extent to which policy measures could be undertaken in order to mitigate the impact of specific emissions from different sectors — in particular black carbon emissions from road transportation and sulfur dioxide emissions from power generation.
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1. Introduction

Air pollution adversely affects human health (U.S. EPA, 2011a; WHO, 2006; COMEAP, 2010). The emission of pollutants into the atmosphere is an inherent by-product of combustion processes. Recent research has found that ambient concentrations of fine particulate matter (smaller than 2.5 μm in aerodynamic diameter, $PM_{2.5}$) (Dockery et al., 1993; Pope et al., 2002; WHO, 2006) and ozone (Bell et al., 2004; Jerrett et al., 2009; WHO, 2008a) are associated with the incidence of premature mortality and morbidity outcomes. Although other anthropogenic air pollutants are recognized as causes of adverse health impacts, ground level $PM_{2.5}$ and ozone exposure is currently considered the most significant known cause of early deaths related to poor outdoor air quality (U.S. EPA, 2011a). The U.S. Environmental Protection Agency estimated that in 2010 there were ~160,000 premature deaths in the U.S. due to $PM_{2.5}$ exposure and ~4300 deaths related to ozone exposure. Fann et al. (2012) estimated between 130,000 and 340,000 $PM_{2.5}$-related early deaths in 2005, and between 4700 and 19,000 ozone-related early deaths.

In the U.S., air pollution is regulated by the Clean Air Act and its amendments (1970 through 1990), which enables the EPA to set

* Corresponding author. Tel.: +1 617 452 2550.
E-mail address: sbarrett@mit.edu (S.R.H. Barrett).

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national air quality standards for six criteria air pollutants including $PM_{2.5}$ and ozone (U.S. EPA, 2011a). The Environmental Protection Agency estimated that in 2012 about 74 million people in the U.S. are exposed to levels of $PM_{2.5}$ higher than the limit standard and that more than 131 million live in regions not compliant with maximum allowable ozone levels (U.S. EPA, 2012b). The EPA computed the costs for the implementation of the 1990 Clean Air Act to be about 65 billion dollars, with a potential benefit reaching 2 trillion dollars from 1990 to 2020, potentially avoiding ~230,000 premature deaths in 2020 (U.S. EPA, 2011a). Although the CAA90 policy-implementation costs are distributed among different source categories, the attribution of air quality-related premature mortalities to different sectors has not been quantified in the peer-reviewed literature. An assessment of the early deaths attributable to different sources would create the potential to drive specific policies with the aim of maximizing the health benefits related to emission reductions from a certain economic activity. In the U.S., anthropogenic combustion emissions represent the predominant source of ground level $PM_{2.5}$ and ozone concentrations (U.S. EPA, 2011a).

In the first part of the present study we evaluate premature deaths attributable to U.S. combustion emissions represented by the following sectors: electric power generation, industry, commercial/residential activities, road transport, marine transport and rail transport. The contribution of $PM_{2.5}$ and ozone-related mortalities is quantified to inform policy makers about opportunities to diversify regulations by taking into account the health impact caused by different types of human activities. The second part of the study (Part II) will focus on assessing future-year combustion emissions impacts from different sectors and on future possible mitigation strategies.

2. Data and methodology

The health impacts of combustion emissions from different sectors are evaluated through the derivation of a temporally, spatially and chemically resolved emissions inventory in the contiguous United States (CONUS), and parts of Canada and Mexico for the reference year 2005. Meteorology and air quality models are used to relate emissions to pollutant concentrations. A baseline simulation, including all emission sources, is performed to assess the model capability to predict meteorological fields, particulate matter and ozone concentrations. Sector emission scenarios are developed wherein combustion emissions from each of the six emission sectors defined above are removed in turn from the baseline inventory; differences in particulate matter and ozone concentrations between the baseline and sector scenario simulations are attributed to the contribution of that specific sector. Population exposure to sector-attributable $PM_{2.5}$ and ozone concentrations are used with concentration-response functions (CRFs) to estimate premature mortality impacts of each sector.

The calculated mortalities can be seen as potentially avoidable deaths in the reference year 2005 related to the instantaneous removal of combustion emissions from each specific sector. An extensive discussion about the use of number of premature deaths per year as a metric for anthropogenic health impact assessments is given by the UK Committee on the Medical Effects of Air Pollutants (COMEAP, 2010). The approach adopted in this study follows the methodology for the evaluation of "current" health burdens from air pollution described by COMEAP (2010). The remainder of this section details each of the steps previously described.

2.1. Meteorological modeling

The modeling domain is centered about the CONUS, including parts of Canada and Mexico. The horizontal resolution is 36 km (112 rows by 148 columns), with 34 sigma-pressure vertical layers. Meteorological fields for the year 2005 are derived using the Weather Research and Forecasting Model (WRF version 3.3.1; Skamarock et al., 2008), driven by four-dimensional data assimilation from the six-hourly NCEP Final Analyses (FNL) data at $1^\circ \times 1^\circ$ resolution. Meteorological simulations are validated against direct hourly temperature and wind observations from 1672 and 1619 stations, respectively. Observations are collected by the Meteorological Assimilation Data Ingest System (MADIS, 2010), developed by the National Oceanic and Atmospheric Administration (NOAA).

2.2. Emissions

Baseline emissions in the U.S., Canada and Mexico are derived from the 2005 EPA National Emissions Inventory (NEI; U.S. EPA, 2008a). This represents the most up to date emissions inventory at the time of this study. NEI 2005 emissions are compiled using data from numerous state and local agencies. The Sparse Matrix Operator Kernel Emissions program version 2.6 (SMOKE, 2010) is used to prepare emissions for the air quality model. SMOKE applies chemical speciation profiles (in case of PM, NO_x and Volatile Organic Compounds), temporal profiles and spatial surrogates for allocation of emissions into model grid-cells. The spatial surrogates are compiled by the EPA (SMOKE, 2010) to allocate area and line sources (which are often specified as county totals) to the CMAQ model grid cells. The emissions are distributed using area-weighting, and the emission allocation is done based on source classification codes (SCCs).

Pre-processed WRF meteorological fields are used to treat emissions from mobile sources for which emissions factors are significantly influenced by local temperature and relative humidity (Ashok, 2011) as well as to compute the plume rise of point-source emission sources and vertically allocate them into the model layers. Emissions scenarios are developed for six source categories ("sectors"): (a) electric power generation, (b) industry, (c) commercial/residential, (d) road transportation, (e) marine transportation, (f) rail transportation. Sectors are defined with differences relative to EPA source categories (U.S. EPA, 2008b) including that commercial and residential sources are merged together and transportation is divided into three separate sectors (discussed later). The division of the transportation sector is performed in order to capture contributions from different modes of transportation and assess modal emission mitigation strategies in future years in the second part of the study.

Sector emissions are taken out from each scenario by removing, in turn, the sources associated to the specific sector from the baseline NEI dataset. Aviation emissions are included in the baseline case, but aviation is not explicitly considered as a sector here since the premature mortalities related to this specific sector have been assessed in Yim et al. (2013). Sector-attributable emissions are considered only in the CONUS together with the U.S. maritime exclusive economic zone (200 nmi off the coastline, plus maritime boundaries with adjacent/opposite countries). Emissions from Canada and Mexico are kept in all the simulations at their original baseline values. We thus focus our investigation on the health impacts on U.S. population from sources located within the U.S. territory. The CONUS and maritime boundary specifications are taken from the National Atlas of the United States of America (2012) and from the Office of Coast Survey (OCS) of the NOAA (1998).

Totals for primary particulate matter, NO_x and SO_2 emissions for the reference year 2005 from each of the sectors are given in Table 1. Combustion emissions from the sectors considered account for 82% of the NO_x anthropogenic emissions in the continental U.S., and 98% of the sulfur dioxide emissions. Emissions from fugitive dust, agricultural activities, aviation and other non-combustion sources are not considered in the sector specifications.

Table 1
$PM_{2.5}$ (primary), NO_x and SO_2 emissions totals and percentages with respect to the baseline scenario (NEI, 2005 dataset, including all sources). Emissions are expressed in Tg year^{-1} for each sector considered in the study (data for 2005).

Sector	$PM_{2.5}$		NO_x		SO_x	
	Total	%	Total	%	Total	%
Electric power generation	0.46	11.7%	3.42	16.1%	9.46	70.4%
Industry	0.57	14.5%	2.75	13.0%	2.55	19.0%
Commercial/residential	0.69	17.6%	0.76	3.6%	0.49	3.6%
Road transportation	0.27	6.9%	8.17	38.5%	0.16	1.2%
Marine transportation	0.07	1.8%	1.30	6.1%	0.45	3.4%
Rail transportation	0.03	0.8%	1.01	4.8%	0.07	0.5%
Other	1.84	46.8%	3.81	18.0%	0.25	1.9%
Total	3.93	100.0%	21.22	100.0%	13.43	100.0%

It is possible to relate the totals found from the 2005 NEI to more recent estimates by using yearly total emissions trends for air pollutants in the U.S. (U.S. EPA, 2012a). The trends estimated by EPA indicate that with respect to 2005, in 2012 SO_2 emissions would be ~60% lower, NO_x emissions ~40% lower, and VOC emissions ~15% lower, while $PM_{2.5}$ and ammonia emissions are expected to increase by ~14% and ~5% respectively. We note that these figures are preliminary estimates and, particularly for SO_2 and NO_x, may be significantly revised.

2.3. Air quality modeling

Air quality simulations for the year 2005 are performed using the CMAQ (version 4.7.1) regional chemistry-transport model (Byun and Schere, 2006) at a spatial resolution of 36 km × 36 km. A two-week spin-up time is used to mitigate the influence of initial conditions. The initial and boundary conditions for the CMAQ simulations are provided by Barrett et al. (2012). Simulated $PM_{2.5}$ baseline concentrations are validated against 24-h averaged observations from 543 stations collected by the EPA Speciation Trends Network (STN). Ozone baseline concentrations are validated against hourly data from 538 stations from the U.S. EPA Air Quality System (AQS) (U.S. EPA, 2011b).

2.4. Health impacts

Epidemiological studies have quantified the relationship between adverse health effects and long-term exposure to $PM_{2.5}$ (U.S. EPA, 2011a; Lewtas, 2007; Krewski et al., 2009; Laden et al., 2006) and ozone (Bell et al., 2004; Jerrett et al., 2009). The quantitative association between premature mortality and ground-level concentrations of $PM_{2.5}$ and ozone is generally assessed through the derivation of relative risk (RR) factors and concentration-response functions (CRFs). An expert elicitation by the U.S. EPA reports a decrease of 1% (range 0.4%–1.8%) in annual all-cause deaths for a 1 μg m^{-3} decrease in the annual average $PM_{2.5}$ exposure in the United States (U.S. EPA, 2011a). Similar results are reported for Europe (Cooke et al., 2007). Jerrett et al. (2009) associated long-term ozone exposure with the risk of death from respiratory causes. In that study, the relative risk of early death from respiratory diseases as a consequence of an increase in ozone concentration of 10 ppb is estimated as 1.040 (95% confidence interval, 1.010–1.067).

$PM_{2.5}$ and ozone account for the majority of monetary losses related to the health impacts of air pollution (Ratliff et al., 2009), and as such long-term exposure to $PM_{2.5}$ and ozone form the focus of the present study. Premature deaths in the U.S. related to sector-attributable $PM_{2.5}$ are estimated using a linear CRF based on EPA assessments (U.S. EPA, 2011a) and described further in Barrett et al. (2012). The CRF associates long-term exposure to $PM_{2.5}$ with premature deaths from cardiopulmonary causes and lung cancer. For long-term exposure to ozone, a log-linear CRF derived from the results of Jerrett et al. (2009) is adopted, consistent with previous ozone health impact assessments in the U.S. (U.S. EPA, 2011a; Fann et al., 2012). The CRF evaluates the number of premature deaths Δy corresponding to a change in ozone concentration ΔO_3 (Abt Associates Inc. and U.S. EPA, 2012). Specifically,

$$\Delta y = y_0 \cdot \left(1 - \frac{1}{\exp(\beta \cdot \Delta O_3)}\right) \quad (1)$$

where y_0 is the baseline incidence rate of the health effect (death from respiratory diseases). The change in ozone concentration ΔO_3, specified in ppb, represents a change in the daily maximum ozone concentration averaged during the ozone season (April 1 – September 30), as described in Jerrett et al. (2009). The coefficient β takes on specific values for urban areas as well as region-specific values for rural areas based on the following geographical regions of the U.S.: Northeast, Industrial Midwest, Southeast, Upper Midwest, Northwest, Southwest, Southern California, as defined by the EPA (Krewski et al., 2000). Nominal values of β and standard error estimates used for uncertainty quantification are provided by the EPA (Abt Associates Inc. and U.S. EPA, 2012). For both $PM_{2.5}$ and ozone, mortalities are evaluated as single sector contributions for adults over 30 years old. Baseline incidences for pollutant-related mortalities (cardiopulmonary diseases and lung cancer for the $PM_{2.5}$ CRF, respiratory diseases for the ozone CRF) are taken from the WHO Global Burden of Disease (WHO, 2008b). Population density is retrieved from the Gridded Population of the World database (GPWv3, 2004).

2.5. Uncertainty assessment

The uncertainties inherent in the premature mortality calculations, including uncertainties from the CRF parameters as well as the air quality modeling, are quantified in this study. For $PM_{2.5}$ – related mortality calculations, the uncertainty in the CRF is accounted for with a triangular probability distribution of multiplicative factors with (low, nominal, high) values of (0.3, 1, 1.7) (U.S. EPA, 2006). The low, nominal and high values correspond to the vertices of the triangular distribution function. The distribution of CMAQ model normalized mean biases is used to account for the uncertainty in predicting PM concentrations, and it is modeled as a normal distribution of mean 7.55% and standard deviation of 28.1%. The minimum (−67.2%) and maximum (108.1%) normalized mean biases are adopted as limiting values to trim the tails of the normal distribution. The reciprocal of the biases distribution are used as multiplicative factors to correct CMAQ model predictions in the uncertainty calculations.

We note that the uncertainty related to different toxicities among $PM_{2.5}$ species as well as a ~10% probability of no causal link between $PM_{2.5}$ exposure and premature mortality (Roman et al., 2008) have not been accounted for quantitatively in this study. The assumption of equal toxicities is consistent with U.S. EPA expert elicitation studies (U.S. EPA, 2004), but represents an unquantified uncertainty (Levy et al., 2009). A similar approach is applied for the uncertainty assessment of ozone-related premature mortalities. For the ozone CRF shown in Equation (1) we consider a triangular probability distribution of multipliers with (low, nominal, high) values of ($\beta - 1.96\,\sigma_\beta$, β, $\beta + 1.96\,\sigma_\beta$), as tabulated in Abt Associates Inc. and U.S. EPA, 2012. The values σ_β of correspond to the standard errors for the health impact estimates performed by the CRF in different regions of the U.S. (Abt Associates Inc. and U.S. EPA, 2012). The β coefficients and their corresponding standard errors vary between each of the seven geographical regions of the U.S.

Table 2
Statistical model evaluation of WRF (wind speed and temperature) and CMAQ ($PM_{2.5}$ and ozone) against observations. Wind speed and temperature are evaluated on an hourly basis, $PM_{2.5}$ on a 24-h average, and ozone is evaluated as daily maximum values recorded during the ozone season (Apr–Sept). The units for each quantity are indicated in the table.

	Wind [m s^{-1}]	T [°C]	$PM_{2.5}$ [μg m^{-3}]	Ozone [ppb]
Model Mean	3.58	12.93	13.85	55.01
Model SD	2.14	11.76	9.39	15.74
Observed Mean	3.32	12.88	12.98	56.74
Observed SD	2.46	11.89	8.49	17.88
Index of Agreement	0.82	0.98	0.69	0.74
Correlation	0.68	0.97	0.49	0.57
Annual Mean Bias (%)	8.02	0.39	6.77	−3.04
Root-mean-square error	1.88	2.90	9.13	15.87
Mean Bias	0.22	0.05	0.88	−1.72
Mean Normalized Bias (%)	10.17	1.25	28.60	2.62
Normalized Mean Bias (%)	8.02	0.39	6.77	−3.04
Mean Fractional Bias (%)	30.24	10.42	1.90	−1.96
Mean Error	1.45	2.17	6.53	11.62
Normalized Mean Gross Error (%)	43.67	16.86	50.33	20.47
Mean Normalized Gross Error (%)	42.47	12.02	63.01	22.37
Mean Fractional Error (%)	65.47	−8.92	49.46	21.10
Data Availability (%)	74.74	76.94	73.73	98.12

described in Section 2.4. As such, the ozone CRF uncertainty bounds are computed individually for each of the regions. Region-specific uncertainty for the CMAQ ozone predictions is calculated using a normal distribution of normalized mean biases. Mean value, standard deviation and limits of the distributions are computed for each region following the same approach as for the $PM_{2.5}$-related model uncertainty evaluation.

3. Results

3.1. Model evaluation

Meteorological and air quality simulations are validated against observations using a set of statistical metrics recommended by the EPA (U.S. EPA, 2005). The definitions for each of the metrics can be found in Yim and Barrett (2012): in particular, an index of agreement (IA) of 1 indicates perfect agreement between the model and the available observations.

Overall the simulated meteorology and air quality statistics, shown in Table 2, are within the range or close to recent studies adopted for similar applications (Yim and Barrett, 2012; Gilliam and Pleim, 2010). Simulated wind speed (measured in m s^{-1}) exhibits an index of agreement of 0.82 and a normalized mean bias around 8% with respect to the available observations. Modeled temperature (measured in °C) shows an IA of 0.98 and a positive bias of 0.39%. The 24-h averaged fine particulate matter (in μg m^{-3}) computed by CMAQ has an index of agreement of 0.69. For ozone, daily maximum values (in ppb) during the ozone season (Apr–Sept) are computed, showing an index of agreement of 0.74. The model estimates the concentrations of $PM_{2.5}$ and ozone with a normalized mean bias of 6.77% and −3.04% respectively. The daily maximum evaluation of ozone during the ozone season yields a normalized mean gross error of 20.47%. Considering all the monitoring stations, the highest bias for the ozone seasonal daily maximum is 61%, the minimum is −42%. These values, computed in each of the seven U.S. regions that characterize the discrete application of the ozone CRF (1), are used as limits for the model uncertainty computations. The annual mean $PM_{2.5}$ modeling bias for all stations exhibits a maximum value of 108% and a minimum of −67%: as noted in

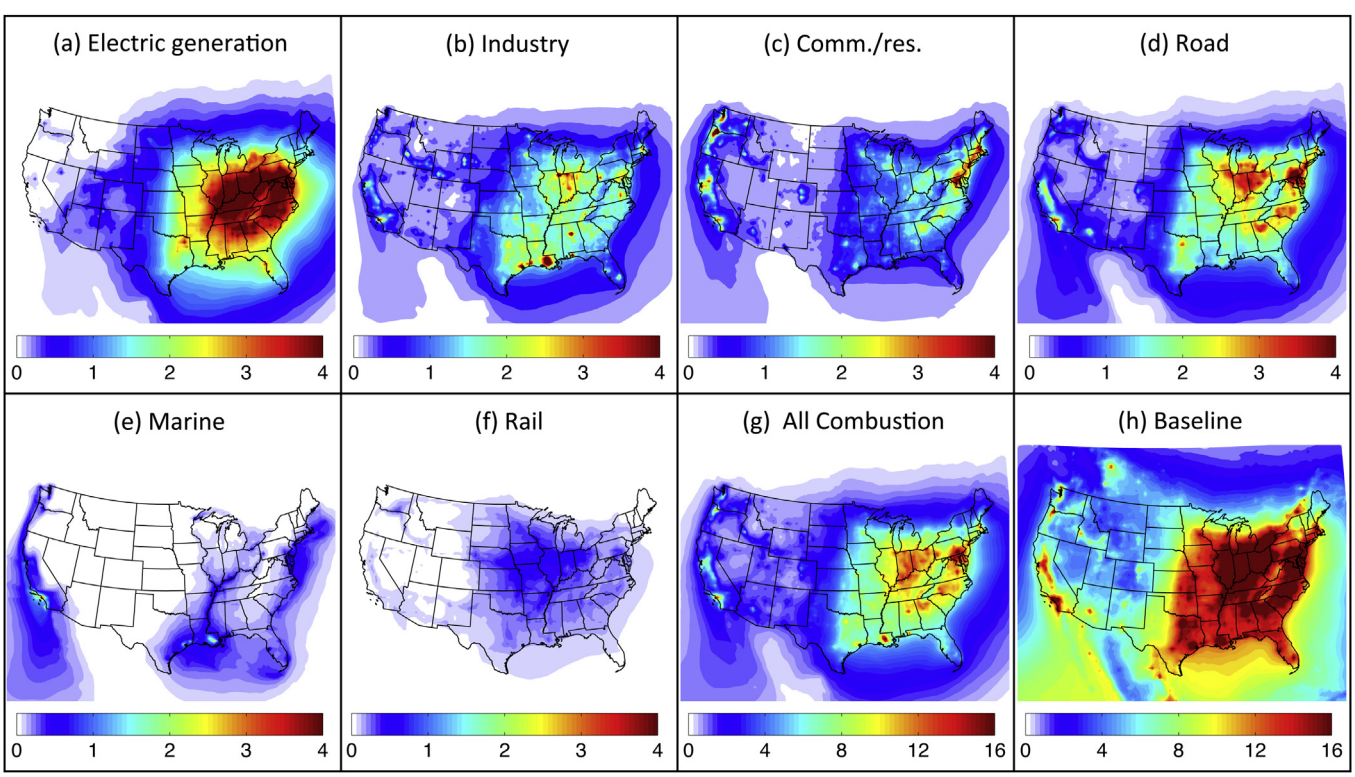

Fig. 1. Annual average ground-level $PM_{2.5}$ concentration (μg m^{-3}) from U.S. sources attributable to combustion emissions from (a) electric power generation; (b) industry; (c) commercial and residential sources; (d) road transportation; (e) marine transportation; (f) rail transportation; (g) sum of all combustion sources; (h) all sources (baseline case for this study). A different scale is adopted for (a–f) and (g–h).

Table 3
Population-weighted concentrations of $PM_{2.5}$ ($\mu g\ m^{-3}$) and ozone (ppb) attributable to combustion emissions from the six sectors considered in this study. $PM_{2.5}$ population-weighted annual mean concentration is speciated into six categories: sulfate (Sulf), nitrate (Nit), ammonium (Amm), black carbon (BC), organics (Org) and unspeciated (Uns). The total concentration of $PM_{2.5}$ is displayed in the second last column of the table. The PM concentrations are annually averaged while the ozone concentration is evaluated as daily maximum averaged over the ozone season (Apr–Sept).

Sector	$PM_{2.5}$							Ozone
	Sulf	Nit	Amm	BC	Org	Uns	Total $PM_{2.5}$	
Electric power generation	1.13	0.05	0.36	0.01	0.48	0.24	2.27	2.15
Industry	0.41	0.19	0.19	0.04	0.42	0.52	1.78	2.06
Commercial/residential	0.13	0.12	0.08	0.08	0.93	0.47	1.82	0.67
Road transportation	0.10	0.61	0.25	0.27	0.98	0.08	2.30	6.90
Marine transportation	0.11	0.03	0.04	0.06	0.09	0.03	0.36	0.39
Rail transportation	0.01	0.05	0.02	0.03	0.09	0.00	0.20	0.53
Total from combustion	1.89	1.05	0.94	0.49	2.99	1.34	8.73	12.70

section 2.4, these values are used as uncertainty ranges in the CMAQ $PM_{2.5}$ evaluation.

3.2. $PM_{2.5}$ impacts

Annual average ground-level $PM_{2.5}$ attributable to U.S. emissions from the different sectors considered in this study is shown in Fig. 1. The general distribution of particulate matter concentrations highlights the clustering of anthropogenic activities along the coastlines and in the Midwest regions of the U.S.

Table 3 shows the population-weighted annual mean concentrations of $PM_{2.5}$ (together with its composite species) and ozone attributable to the different sectors. Road transportation is responsible for a $PM_{2.5}$ population-weighted concentration of 2.30 $\mu g\ m^{-3}$ in U.S., representing the largest contributor to PM-related impacts. Most of the particulate matter attributable to road transport emissions is organic (0.98 $\mu g\ m^{-3}$) followed by nitrate aerosol (0.61 $\mu g\ m^{-3}$): this reflects the fact that onroad mobile emissions are the largest source of NO_x in the U.S., as shown in Table 1. Vehicle emissions are also the largest contribution to population-weighted black carbon concentrations (0.27 $\mu g\ m^{-3}$). The change in black carbon concentration attributable to road vehicles in the U.S. is shown in Fig. 2a. BC concentrations peak in major cities where the traffic is higher, in contrast to total $PM_{2.5}$ concentrations (Fig. 1d) which are more diffuse due to the inclusion of secondary particulate matter. For this reason, black carbon from road emissions has a relatively high adverse health impact with respect to other PM species.

Electric power generation is responsible for a population-weighted annual mean $PM_{2.5}$ concentration of 2.27 $\mu g\ m^{-3}$. Given the discrete distribution of power plants, the contribution of this sector is less ubiquitous with respect to road transportation (Fig. 1a), being less relevant on the western regions. Power plants account for 16% of NO_x emissions and 70% of SO_2 emissions in the U.S. (Table 1). Of the 9.46 million tons of sulfur dioxide emitted in 2005, about 95% comes from coal-fired power plants (NRDC, 2007) which represent the largest source of electricity in the U.S. (U.S. EIA, 2012).

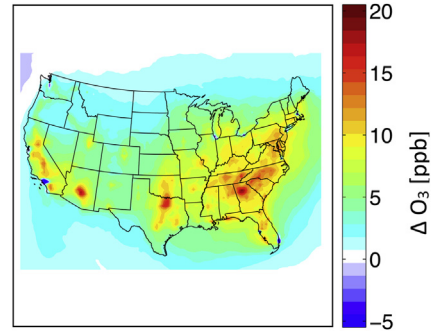

Fig. 3. Variation of mean (Apr–Sept) daily maximum ozone concentration (ppb) due to road transportation emissions in 2005.

Eastern power plants generally use coal with higher sulfur content than western power plants (U.S. EIA, 2002). This trend is shown in Fig. 2b, which displays the ground-level annual mean sulfate concentration attributable to electric generation. In the Midwest states, the sulfate concentration exhibits peaks of 3.5 $\mu g\ m^{-3}$, which account for the 1.13 $\mu g\ m^{-3}$ population-weighted concentration of sulfate due to the electric sector. Yim and Barrett (2012) reported a population-weighted mean annual sulfate concentration of about 0.25 $\mu g\ m^{-3}$ in the UK, showing a significantly smaller impact of the electric generation sector in this country with respect to what we found in the U.S. This is partially due to the fact that the largest power plants in the UK are generally located relatively far away as well as downwind from highly populated regions.

Combustion emissions from commercial and residential sources generate a mean annual population-weighted $PM_{2.5}$ concentration of 1.82 $\mu g\ m^{-3}$, mostly composed of organic particulate matter (0.93 $\mu g\ m^{-3}$). Due to the nature of these sources, the peaks in commercial/residential contributions occur in the most densely populated areas of the east and the west coast (Fig. 1c).

Fig. 1b shows mean $PM_{2.5}$ concentrations due to emissions from industrial activities, which account for a population-weighted annual concentration of 1.78 $\mu g\ m^{-3}$. The concentration distribution exhibits peaks in the Midwest industrial area between Chicago and Detroit, and in the regions around Philadelphia, Atlanta and Los Angeles. The largest contributions occur in the coastline of the U.S. Gulf Coast connecting Mobile (AL), New Orleans (LA) and Houston (TX). The high concentration of industry-attributable $PM_{2.5}$ in this

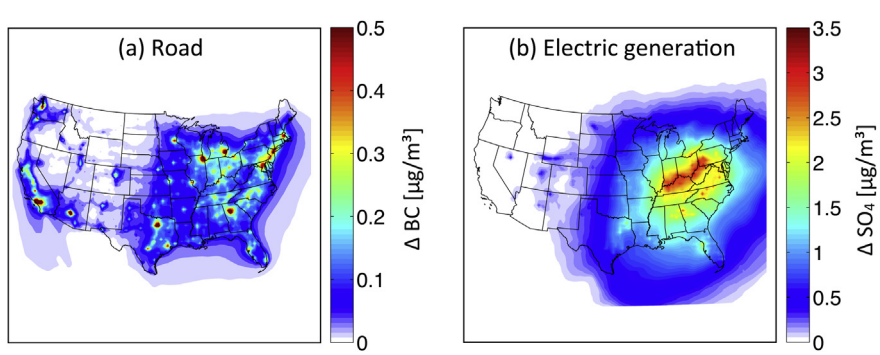

Fig. 2. Annual average ground-level concentration (in $\mu g\ m^{-3}$) in the U.S. of (a) black carbon (BC) due to road transportation; (b) SO_4 due to electric power generation.

Table 4
Premature deaths [90% confidence interval] in the U.S. in 2005 due to long-term exposure to $PM_{2.5}$ and ozone associated to combustion emissions from different sectors.

Sector	$PM_{2.5}$	O_3
Electric power generation	52,200 [23,400–94,300]	1700 [−250–3700]
Industry	40,800 [18,300–73,700]	1750 [−30–3500]
Commercial/residential	41,800 [18,700–75,500]	350 [−50–750]
Road transportation	52,800 [23,600–95,300]	5250 [−850–11,100]
Marine transportation	8300 [3700–15,000]	530 [−50–1100]
Rail transportation	4500 [2000–8100]	540 [−100–1200]
Aviation (Yim et al., 2013)[a]	1200 [550–2600]	155 [71–260]
Total from combustion[b]	200,400 [89,700–361,900]	10,100 [−1300–21,400]

[a] Refers to global full flight emission impact in the U.S., using the same CRFs described in Section 2.4.
[b] Excluding aviation.

region is related to the presence of the largest oil refineries in the United States (U.S. EIA, 2004).

Mean annual concentrations of particulate matter due to marine emissions are shown in Fig. 1e. Emission sources are considered only within the maritime exclusive economic zone (200 nmi off the coastline, plus maritime boundaries with adjacent/opposite countries), and Southern California exhibits their largest impact in terms of $PM_{2.5}$ concentration. Particulate matter forming in this region as a consequence of maritime emissions is then substantially advected to the southeast. Locally significant marine transportation-attributable $PM_{2.5}$ concentrations span along all the U.S. coastlines and along the navigable portions of the Mississippi and Ohio rivers. The population-weighted annual average concentration of total $PM_{2.5}$ is 0.38 $\mu g\ m^{-3}$, and is almost equally distributed between different PM species.

Finally, Fig. 1f shows the $PM_{2.5}$ concentration due to rail emissions: rail-attributable particulate matter spreads relatively

Table 5
Number of premature mortalities (NM) and mortality rate (MR) per year due to $PM_{2.5}$ concentrations attributable to different sectors in the 48 states of the CONUS (plus District of Columbia). Mortality rate (MR) corresponds to number of deaths per year per 100,000 people within the state.

State	Electric gen		Industry		Comm/Res		Road		Marine		Rail	
	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR
Alabama	1242	27.3	833	18.3	509	11.2	766	16.8	86	1.9	83	1.8
Arizona	127	2.5	269	5.3	386	7.6	616	12.1	41	0.8	37	0.7
Arkansas	630	23.7	410	15.4	219	8.2	411	15.4	56	2.1	72	2.7
California	468	1.3	4834	13.9	6459	18.6	5726	16.4	3484	10.0	280	0.8
Colorado	177	4.1	160	3.7	388	9.0	264	6.2	5	0.1	24	0.6
Connecticut	473	13.9	332	9.7	821	24.1	697	20.5	62	1.8	25	0.7
Delaware	248	31.4	162	20.5	179	22.7	230	29.2	35	4.4	12	1.6
DC	187	35.1	76	14.2	164	30.8	150	28.2	7	1.3	8	1.5
Florida	2402	15.1	1372	8.6	1045	6.6	1852	11.7	459	2.9	106	0.7
Georgia	2335	28.3	1232	15.0	1161	14.1	1809	22.0	103	1.2	141	1.7
Idaho	13	1.0	127	9.6	112	8.5	68	5.1	4	0.3	10	0.8
Illinois	3161	25.0	2840	22.5	1551	12.3	3135	24.8	176	1.4	437	3.5
Indiana	2032	32.8	1661	26.8	838	13.5	1639	26.5	100	1.6	209	3.4
Iowa	528	17.7	379	12.7	235	7.9	476	16.0	22	0.7	101	3.4
Kansas	448	16.2	365	13.2	211	7.6	396	14.3	15	0.5	99	3.6
Kentucky	1642	39.7	726	17.6	556	13.5	886	21.4	86	2.1	101	2.4
Louisiana	826	18.2	1133	24.9	319	7.0	568	12.5	314	6.9	74	1.6
Maine	98	7.5	81	6.2	192	14.7	105	8.1	14	1.1	3	0.3
Maryland	1885	34.9	987	18.3	1505	27.9	1558	28.8	104	1.9	96	1.8
Massachusetts	821	12.8	1211	18.8	1775	27.6	1368	21.3	131	2.0	42	0.7
Michigan	2289	22.3	1858	18.1	1050	10.2	2484	24.2	103	1.0	196	1.9
Minnesota	580	11.6	664	13.3	559	11.2	777	15.6	38	0.8	122	2.4
Mississippi	684	23.7	431	14.9	241	8.3	414	14.3	82	2.8	56	1.9
Missouri	1329	23.3	873	15.3	588	10.3	1048	18.4	82	1.4	196	3.4
Montana	8	0.8	24	2.7	26	2.8	18	1.9	1	0.1	4	0.5
Nebraska	227	13.1	168	9.7	92	5.3	193	11.1	6	0.3	57	3.3
Nevada	47	2.4	109	5.6	98	5.0	104	5.3	16	0.8	10	0.5
New Hampshire	137	10.9	176	14.0	279	22.2	185	14.7	12	1.0	6	0.5
New Jersey	1885	22.2	1260	14.8	2341	27.6	2420	28.5	328	3.9	78	0.9
New Mexico	63	3.4	79	4.4	85	4.7	97	5.3	5	0.3	14	0.8
New York	3744	19.8	2400	12.7	4442	23.5	4730	25.1	559	3.0	176	0.9
North Carolina	2570	32.0	1059	13.2	1196	14.9	1742	21.7	115	1.4	134	1.7
North Dakota	35	5.3	26	4.0	19	2.9	25	3.8	1	0.1	9	1.4
Ohio	4223	36.1	2024	17.3	1783	15.3	3054	26.1	204	1.7	328	2.8
Oklahoma	536	15.3	466	13.3	224	6.4	489	14.0	26	0.7	78	2.2
Oregon	35	1.0	238	6.8	1263	36.3	252	7.3	82	2.3	24	0.7
Pennsylvania	3864	31.1	2118	17.1	2431	19.6	3114	25.1	274	2.2	193	1.6
Rhode Island	145	14.1	128	12.5	237	23.1	178	17.3	20	2.0	6	0.6
South Carolina	1196	29.3	532	13.1	575	14.1	846	20.8	60	1.5	66	1.6
South Dakota	70	9.2	55	7.2	29	3.8	51	6.7	1	0.2	14	1.9
Tennessee	1787	31.1	928	16.2	641	11.2	1053	18.3	95	1.7	117	2.0
Texas	2835	13.4	3583	17.0	1869	8.8	3239	15.3	642	3.0	317	1.5
Utah	58	2.6	88	3.9	107	4.8	145	6.5	6	0.3	10	0.5
Vermont	57	9.2	36	5.8	69	11.2	56	9.1	3	0.5	3	0.5
Virginia	2433	33.8	1153	16.0	1416	19.7	1608	22.4	121	1.7	120	1.7
Washington	50	0.8	308	5.1	1625	26.9	554	9.2	149	2.5	38	0.6
West Virginia	683	36.5	269	14.4	243	13.0	307	16.4	23	1.2	31	1.6
Wisconsin	981	17.9	728	13.3	770	14.1	1083	19.8	52	1.0	130	2.4
Wyoming	15	3.0	23	4.7	9	1.8	10	2.1	1	0.1	3	0.6

Table 6
Number of premature mortalities (NM) and mortality rate (MR) per year due to $PM_{2.5}$ concentrations attributable to different sectors in the 20 most populous metropolitan areas (M) and cities (C) of the CONUS (2005 data). Mortality rate (MR) corresponds to number of deaths per year per 100,000 people within the state.

City/MA	Electric gen		Industry		Comm/Res		Road		Marine		Rail	
	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR
New York City (M)	2571	20.3	1713	13.5	3555	28.0	3615	28.5	483	3.8	103	0.8
Los Angeles (M)	137	1.5	1854	20.6	1891	21.1	2092	23.3	1505	16.8	90	1.0
Chicago (M)	1102	22.7	1378	28.4	716	14.8	1379	28.4	56	1.1	171	3.5
Detroit (M)	657	23.2	593	21.0	292	10.3	790	27.9	28	1.0	46	1.6
Philadelphia (M)	573	27.1	404	19.1	535	25.3	591	28.0	79	3.7	25	1.2
Boston (M)	242	12.4	546	28.0	682	35.0	540	27.7	47	2.4	13	0.7
Washington (M)	655	35.2	290	15.6	560	30.1	533	28.6	24	1.3	32	1.7
San Jose (M)	11	0.6	202	11.0	433	23.4	199	10.8	126	6.8	8	0.4
Houston (M)	255	14.1	506	27.9	258	14.2	304	16.8	158	8.7	25	1.4
San Diego (M)	56	3.4	143	8.7	339	20.7	288	17.5	201	12.3	12	0.7
Minn.-Saint Paul (M)	203	12.5	318	19.5	253	15.5	341	20.9	13	0.8	43	2.6
Dallas (M)	280	17.4	329	20.5	209	13.0	374	23.2	20	1.3	29	1.8
Baltimore (M)	475	34.7	368	26.9	441	32.2	430	31.4	35	2.6	25	1.8
Phoenix (C)	34	2.6	89	7.0	141	11.1	225	17.7	11	0.8	11	0.8
Cleveland (M)	466	36.8	222	17.6	222	17.5	384	30.3	32	2.5	37	2.9
Miami (C)	127	10.2	70	5.6	80	6.4	128	10.3	61	4.9	5	0.4
Denver (M)	53	4.4	50	4.2	128	10.7	103	8.6	1	0.1	7	0.6
Saint Louis (M)	280	26.8	204	19.5	141	13.5	235	22.5	22	2.1	31	2.9
Kansas City (C)	208	20.1	163	15.8	109	10.6	199	19.2	8	0.7	47	4.5

uniformly in the central-eastern part of the U.S., with a peak in the Midwest. Yearly averaged population-weighted concentration of rail-attributable $PM_{2.5}$ is 0.20 $\mu g\ m^{-3}$.

3.3. Ozone impacts

The impact on ozone concentrations is related to the atmospheric concentrations of VOC and NO_x. Fig. 3 shows the average daily maximum concentration of ozone attributable to road transportation emissions. Daily maximum ozone is temporally averaged only during the ozone season (Apr–Sep), consistent with EPA practice. Road mobile emissions induce a domain-wide increase in daily maximum seasonal ozone concentrations, except for some major urban areas (e. g. Miami), where the high background NO_x concentrations account for a decrease in the ozone concentrations due to the additional NO_x emitted by road vehicles.

Road transportation provides the most significant impact over ozone exposure among the combustions emission sources considered in this study. From Table 3, the population-weighted mean daily maximum ozone concentration due to vehicle emissions is 6.90 ppb, about three times larger than the population-weighted concentration change due to electric generation (2.15 ppb) and industry (2.06 ppb). Commercial/residential activities, as well as shipping and rail emissions, have an impact on the mean daily maximum ozone concentration below 1 ppb.

3.4. Health impacts

Premature deaths from cardiovascular diseases and lung cancer due to long-term exposures to $PM_{2.5}$ attributable to each sector are evaluated by applying the CRF described in Section 2.4, and are given in Table 4. Aggregated combustion emissions account for a total of about 200,000 (90% CI: 90,000–361,000) $PM_{2.5}$-related premature mortalities per year in the U.S. This result is comparable with total mortalities estimated by similar studies (U.S. EPA, 2011a; Fann et al., 2012). The distribution of early deaths among the different sectors reflects the population-weighted average $PM_{2.5}$ sector-attributable concentrations shown in Table 3.

The two largest contributors to $PM_{2.5}$-related premature deaths in the U.S. are road transport and power generation, accounting for 53,000 (90% CI: 24,000–95,000) and 52,000 (90% CI: 23,000–94,000) early deaths per year, respectively.

Commercial/residential sources and industry account for 42,000 (90% CI: 19,000–76,000) and 41,000 (90% CI: 18,000–74,000) early deaths, respectively. About 8000 (90% CI: 4000–15,000) deaths per year are attributable to marine transport and 4500 (90% CI: 2000–8000) to rail transport. Aviation mortalities are included in the table as estimated by Yim et al. (2013): a total of 1200 (90% CI: 550–2600) $PM_{2.5}$-related mortalities per year are attributable to full flight aviation emissions in North America.

Table 5 allocates the $PM_{2.5}$-related premature mortalities for each sector shown in Table 4 in the 48 states (and the District of Columbia) of the CONUS. This table displays for each state both the absolute number of premature deaths per year and the mortality rate, defined as number of early deaths per year per 100,000 people within the state.

CMAQ gridded results for each sector are attributed to each state using the code ArcGIS (ESRI, 2008). In terms of absolute impact of $PM_{2.5}$ combustion emissions, the most affected region is California, with about 21,000 early deaths per year. Of these, about 12,000 come from both commercial/residential sources and road transportation, and ~5000 from industry. About 3500 premature deaths per year in this state are attributable to marine transportation emissions, which exhibit a peak in Southern California (Fig. 1e).

The data in Table 5 show a large impact of electric generation emissions in the central-eastern U.S. and in the Midwest. This reflects the trend shown in Fig. 2b for power generation-related sulfate concentrations. In particular, with a mortality rate (MR) of about 40 premature deaths per year per 100,000 inhabitants in Kentucky, electric generation is the sector responsible for the highest mortality rate among the U.S. states.

Road transportation, consistent with its annual mean $PM_{2.5}$ concentration map (Fig. 1d), exhibits the most widespread distribution of sector-attributable premature deaths among the U.S. states. In terms of relative impacts, the state characterized by the highest relative mortality due to all the sectors is Maryland, with about 114 early deaths per year every 100,000 inhabitants.[1]

[1] It should be noted that the total number of early deaths given in Table 5 for each sector does not exactly coincide with the values of Table 4 for the whole U.S. This is due to slight inaccuracies in the allocation of the gridded population distribution within state boundaries, which yields an average error of 0.9% in the estimate of the cumulative number of deaths per each sector.

Table 7
Number of premature mortalities (NM) and mortality rate (MR) per year due to ozone concentrations attributable to different sectors in the 48 states of the CONUS (plus District of Columbia). Mortality rate (MR) corresponds to number of deaths per year per 100,000 people within the state.

State	Electric gen		Industry		Comm/Res		Road		Marine		Rail	
	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR
Alabama	97	2.13	69	1.51	14	0.31	240	5.27	22	0.49	24	0.52
Arizona	41	0.81	47	0.92	19	0.37	403	7.94	16	0.32	30	0.59
Arkansas	50	1.90	46	1.72	6	0.21	120	4.53	15	0.56	18	0.66
California	8	0.02	43	0.12	22	0.06	209	0.60	49	0.14	12	0.03
Colorado	27	0.62	23	0.54	3	0.08	57	1.33	1	0.03	7	0.17
Connecticut	−2	−0.06	−2	−0.07	−1	−0.04	−12	−0.35	−1	−0.02	0	−0.01
Delaware	−1	−0.08	−1	−0.07	0	−0.03	−3	−0.36	0	−0.03	0	−0.02
DC	0	0.00	0	0.00	0	0.00	0	0.00	0	0.00	0	0.00
Florida	175	1.10	97	0.61	82	0.52	191	1.20	9	0.06	22	0.14
Georgia	108	1.31	77	0.94	19	0.23	396	4.80	24	0.30	28	0.34
Idaho	2	0.15	6	0.43	1	0.07	16	1.20	1	0.07	2	0.17
Illinois	12	0.09	9	0.07	2	0.01	24	0.19	3	0.02	5	0.04
Indiana	−1	−0.01	0	−0.01	0	0.00	−3	−0.04	0	0.00	0	0.00
Iowa	46	1.56	36	1.20	6	0.20	97	3.24	5	0.18	19	0.64
Kansas	44	1.57	43	1.56	4	0.16	88	3.20	5	0.16	17	0.61
Kentucky	24	0.58	13	0.30	2	0.06	48	1.15	5	0.11	5	0.13
Louisiana	65	1.44	109	2.40	8	0.18	163	3.58	75	1.66	17	0.38
Maine	−1	−0.05	−1	−0.07	−1	−0.04	−5	−0.36	0	−0.04	0	−0.01
Maryland	−4	−0.07	−3	−0.06	−1	−0.02	−16	−0.29	−1	−0.02	−1	−0.02
Massachusetts	−3	−0.05	−2	−0.04	−2	−0.03	−4	−0.06	−1	−0.02	0	0.00
Michigan	−1	−0.01	−1	−0.01	0	0.00	−3	−0.03	0	0.00	0	0.00
Minnesota	54	1.08	42	0.84	9	0.18	119	2.39	6	0.12	21	0.42
Mississippi	51	1.76	50	1.73	6	0.22	135	4.68	26	0.91	16	0.55
Missouri	72	1.25	48	0.85	8	0.14	144	2.52	12	0.21	26	0.46
Montana	2	0.20	2	0.26	0	0.04	8	0.92	1	0.06	2	0.17
Nebraska	26	1.48	23	1.33	2	0.14	48	2.75	2	0.11	12	0.70
Nevada	2	0.12	4	0.19	1	0.08	20	1.05	1	0.07	2	0.11
New Hampshire	−1	−0.05	−1	−0.05	0	−0.03	−4	−0.28	0	−0.01	0	−0.01
New Jersey	−2	−0.03	−3	−0.04	−2	−0.02	−3	−0.04	1	0.01	0	−0.01
New Mexico	40	2.22	55	3.03	5	0.30	127	7.02	5	0.28	19	1.06
New York	−7	−0.04	−9	−0.05	−5	−0.03	−16	−0.09	2	0.01	−2	−0.01
North Carolina	150	1.86	98	1.22	31	0.38	489	6.08	32	0.40	33	0.41
North Dakota	8	1.16	5	0.79	1	0.11	12	1.78	0	0.07	3	0.52
Ohio	−2	−0.02	−1	−0.01	0	0.00	−6	−0.05	0	0.00	0	0.00
Oklahoma	72	2.06	95	2.71	9	0.25	222	6.33	13	0.37	25	0.71
Oregon	4	0.10	7	0.21	4	0.13	36	1.03	8	0.23	3	0.08
Pennsylvania	−10	−0.08	−7	−0.06	−3	−0.02	−37	−0.30	−1	−0.01	−2	−0.02
Rhode Island	−1	−0.07	−1	−0.06	0	−0.04	−4	−0.40	−1	−0.05	0	−0.01
South Carolina	73	1.79	53	1.30	15	0.36	260	6.38	20	0.50	18	0.43
South Dakota	12	1.58	10	1.30	1	0.14	21	2.75	1	0.11	6	0.73
Tennessee	101	1.76	67	1.17	13	0.23	277	4.82	23	0.39	27	0.48
Texas	252	1.19	495	2.34	43	0.20	1052	4.98	163	0.77	88	0.42
Utah	9	0.42	6	0.27	1	0.06	27	1.21	1	0.05	3	0.13
Vermont	0	−0.07	0	−0.07	0	−0.03	−2	−0.39	0	−0.02	0	−0.02
Virginia	39	0.54	22	0.31	7	0.09	69	0.95	−20	−0.28	7	0.10
Washington	3	0.05	5	0.08	4	0.06	29	0.48	3	0.05	2	0.04
West Virginia	−1	−0.03	0	−0.01	0	−0.01	−2	−0.08	0	0.00	0	0.00
Wisconsin	15	0.27	12	0.21	3	0.05	33	0.61	3	0.05	6	0.10
Wyoming	4	0.82	4	0.72	0	0.07	7	1.37	0	0.05	2	0.31

Table 6 shows the same results as Table 5 for the 20 most populous metropolitan areas in the U.S. Urban population data are retrieved from the National Atlas of the United States, 2005. As expected for all metropolitan areas, road transportation and commercial/residential sources have the largest and most uniformly distributed impact on all cities. The highest peaks of the $PM_{2.5}$-related health impacts due to vehicle emissions are found in the major East coast cities: New York (MR ∼ 28.5), Washington (MR ∼ 28.6) and Baltimore (MR ∼ 31.4). The city of Baltimore in particular is characterized by the highest total mortality rate from all combustion sources: about 130 early deaths attributable to $PM_{2.5}$ per year per 100,000 inhabitants. The highest absolute all-combustion sources impact is in New York, with about 12,000 total mortalities per year.

Of the set of 5695 cities considered, the highest $PM_{2.5}$-attributable all-combustion mortality rate (MR ∼144) has been found in Donaldsonville, LA. Here the presence of nine oil refineries within a 70-km radius, for a total production of ∼2.2 million barrels per day (NREL, 2012), accounts for a mortality rate by industrial sources of ∼81 early deaths per year per 100,000 people.

Table 4 also includes premature mortalities due to ozone concentrations attributable to the different sectors. Aggregated combustion emissions account for about 10,100 (90% CI: −1300 to 21,400) ozone-related premature deaths in the U.S. in 2005. As with $PM_{2.5}$, the aggregate ozone mortality estimate is consistent with previous national emissions assessments in the U.S. (U.S. EPA, 2011a; Fann et al., 2012). The negative lower bound is a consequence of the ozone depletion occurring in densely populated cities, due to NO_x emissions in NO_x-saturated environments.

The main contributor is road transportation, which is responsible for more than half of the ozone-related mortalities (∼5250). Both electric generation and industry account for about 1800 mortalities per year. Commercial/residential, marine and rail transport account for about 350, 530 and 540 ozone-related

mortalities annually, respectively. It is noted that, despite their relatively large contributions to $PM_{2.5}$ mortalities with respect to the other sectors, commercial and residential sources contribute only to a fraction of the total ozone-related early deaths. This can be explained by considering the NO_x emission attributions given in Table 1. Road transportation represents the single largest contributor to NO_x emissions (accounting for 38.5% of the total). Industry and electric generation both give a similar contribution to NO_x emissions. This trend is reflected in the national pattern of ozone-related mortalities shown in Table 4.

Similarly to the previous tables for $PM_{2.5}$, Table 7 and Table 8 provide the number of early deaths per year and the mortality rate due to ozone exposure as a consequence of emissions from the six sectors considered. Table 7 shows the data for each U.S. state, while Table 8 sorts the results for the 20 most populous metropolitan areas. The correlation between high ozone levels and high sunlight exposure, together with differences in emissions and background VOC and NO_x concentrations, account for the uneven distribution of ozone-related mortalities between northern and southern states.

More than 20% of the ozone-related mortalities from all sectors (~2100 early deaths) occur in Texas, mainly as a consequence of road transportation and industrial emissions. The second most affected state is North Carolina, with about 800 ozone-related early deaths per year, half of which attributable to vehicle emissions. Smaller states with high percentage of urban areas (e. g., Maryland, Connecticut) are characterized by an ozone-related mortality reduction due all-sectors emissions. In these regions, ozone is generally depleted by additional NO_x emissions. The same principle applies to many of the metropolitan areas considered in Table 8.

4. Discussion

The spatial distribution and speciation of $PM_{2.5}$ impacts per sector can be used to inform the design of sector-specific emission mitigation measures. Premature mortalities from sulfate attributable to power plants represent approximately half of the ~52,000 mortalities from the sector. These mortalities are mainly related to SO_x emissions from coal power plants, and could be reduced by promoting the purchase of low-sulfur content coal from the western deposits in the Powder River Basin in Wyoming and Montana (Stavins and Schmalensee, 2012), the introduction of lime scrubbers, or the adoption of alternative energy sources (e. g. natural gas, as forecasted by the U.S. EIA, 2012). Similarly, the mortalities related to marine combustion emissions (of which about one third is related to sulfate concentrations) could be reduced by enforcing limits to the sulfur content of bunker fuel used in ship engines. Regulations to this effect have recently been put in place by the International Maritime Organization (IMO, 2010). In 2010 a limit of 1% fuel sulfur content for the North America Emission Control Area (ECA) was established, to be lowered to 0.1% in 2015.

In using the results of this study to inform potential mitigation measures, it is important to note that premature mortality estimates are calculated assuming equal toxicity amongst the different types of particulate matter. Recent epidemiological studies (Lippmann and Chen, 2009; Levy et al., 2012) suggest that differential toxicity amongst PM species may be significant. In an extensive multi-site time-series analysis, Levy et al. (2012) showed differences in the correlations between changes in hospital admissions and concentrations of different types of $PM_{2.5}$, with black carbon showing the highest relative health impact. Furthermore, a recent ACS cohort analyses (Lippman, 2010) indicate that $PM_{2.5}$ correlations with premature mortality risk may vary with source category, with coal and traffic sources having the most significant associations. Despite these findings, no epidemiological study to date has provided a conclusive assessment of the relative toxicity of different $PM_{2.5}$ components, sufficient to develop CRFs accounting for those differences [as per Levy et al. (2012) and current EPA practice]. It is therefore possible that future CRFs will be able to describe particulate matter health impacts by weighting PM species. Table 3 of the present study provides data appropriate for such a calculation.

An assessment of the health impacts from $PM_{2.5}$ and ozone concentrations attributable to different source categories in the US has been performed in parallel with the present study by Fann et al. (2013), who adopt a source apportionment approach to allocate the concentrations of $PM_{2.5}$ and ozone among various different source categories. Their source categories follow the NEI source classification scheme, whereas we have reprocessed inventories to correspond to what may be termed "economic" sectors. For example, the "industrial" sources in this study are split between "industrial point sources" and "area sources" in Fann

Table 8
Number of premature mortalities (NM) and mortality rate (MR) per year due to ozone concentrations attributable to different sectors in the 20 most populous metropolitan areas (M) and cities (C) of the CONUS (2005 data). Mortality rate (MR) corresponds to number of deaths per year per 100,000 people within the state.

City/MA	Electric Gen		Industry		Comm/Res		Road		Marine		Rail	
	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR	NM	MR
New York City (M)	−2.22	−0.017	−4.66	−0.037	−2.67	−0.021	3.76	0.030	2.93	0.023	−0.53	−0.004
Los Angeles (M)	0.24	0.003	1.42	0.016	1.52	0.017	0.95	0.011	0.02	0.000	−0.17	−0.002
Chicago (M)	−0.13	−0.003	−0.12	−0.002	0.01	0.000	0.23	0.005	−0.01	0.000	0.06	0.001
Detroit (M)	−0.02	−0.001	−0.02	−0.001	−0.01	0.000	−0.02	−0.001	0.00	0.000	0.00	0.000
Philadelphia (M)	−0.16	−0.008	−0.15	−0.007	−0.07	−0.003	−0.75	−0.035	−0.06	−0.003	−0.03	−0.002
Boston (M)	−0.42	−0.021	0.10	0.005	−0.19	−0.010	8.96	0.459	0.19	0.010	0.21	0.011
Washington (M)	−0.77	−0.041	−0.67	−0.036	−0.28	−0.015	−3.57	−0.192	−0.11	−0.006	−0.21	−0.011
San Jose (M)	0.21	0.012	1.33	0.072	0.78	0.042	5.19	0.281	6.05	0.328	0.08	0.004
Houston (M)	9.17	0.505	22.37	1.233	3.24	0.179	47.30	2.607	11.25	0.620	2.78	0.153
San Diego (M)	0.02	0.001	0.28	0.017	0.11	0.007	0.13	0.008	−0.50	−0.031	0.05	0.003
Minn.-Saint Paul (M)	9.40	0.577	6.20	0.380	1.63	0.100	21.49	1.318	0.87	0.053	3.54	0.217
Dallas (M)	4.15	0.258	6.22	0.386	0.60	0.037	16.92	1.051	1.46	0.091	1.19	0.074
Baltimore (M)	0.00	0.000	0.00	0.000	0.00	0.000	0.00	0.000	0.00	0.000	0.00	0.000
Phoenix (C)	4.48	0.351	6.68	0.523	3.89	0.305	71.07	5.569	2.53	0.198	4.65	0.364
Cleveland (M)	−0.08	−0.006	−0.06	−0.005	−0.02	−0.002	−0.03	−0.002	0.03	0.003	0.01	0.001
Miami (C)	0.83	0.067	8.09	0.651	12.71	1.024	−94.1	−7.582	−13.11	−1.056	0.41	0.033
Denver (M)	3.28	0.275	2.77	0.231	0.68	0.057	11.07	0.926	0.18	0.015	0.93	0.078
Saint Louis (M)	−0.04	−0.004	−0.03	−0.003	−0.01	−0.001	−0.19	−0.018	−0.01	−0.001	−0.01	−0.001
Kansas City (C)	8.55	0.827	5.03	0.486	0.83	0.081	14.76	1.429	0.83	0.080	3.01	0.291

et al. (2013), where their area sources in turn also include part of the commercial/residential emissions considered in this study. Here we make a comparison for $PM_{2.5}$-related early deaths insofar as possible using Table 3 of the Fann et al. (2013) SI and assuming a nominal 12 life years lost per premature mortality for the purposes of this comparison. We note that these comparisons are not like-for-like due to the different inventory processing applied (as well as different meteorology and air quality models, and apportionment approach) and it is not clear the extent to which comparisons are appropriate. For power generation [Fann et al. (2013): electricity generating units] we estimate 52,200 early deaths per year, compared to their 51,700 using our conversion. For mobile sources [approximately our road transportation, marine transportation, rail transportation and aviation] we estimate 66,800 early deaths per year, cf. their estimate of 36,300. We note that our aircraft estimate includes cruise emissions, whereas theirs is based on a different inventory and only for landing and takeoff emissions. For industry [Fann et al. (2013): all industrial sub-categories except electricity generating units] we estimate 40,800 cf. their 22,400. However, our definition of industry includes some of their "area sources" so an upper bound on their early deaths would be 42,800. In total (excluding non-anthropogenic and transboundary pollution) Fann et al. (2013) estimates 148,000 early deaths per year, cf. our 200,000 early deaths per year. This implies that our estimates are broadly ~35% higher, although firm conclusions about individual sectors cannot be made. Additionally, we infer 16 life years lost per premature mortality for electricity generating units from their work which would expand the difference by ~30%, while our accounting for low $PM_{2.5}$ modeling biases in our probabilistic approach would serve to reduce the effective differences by ~25%. On a relative basis, we observe that in both assessments electric generation accounts for about 25% of the total $PM_{2.5}$ premature deaths. The relative importance of the aggregated transportation sectors (road, marine, rail and aviation) in the present study is higher (~33% versus ~20%) than the "mobile" sector considered in Fann et al. (2013).

5. Conclusions

Combustion emissions in the U.S. are found to be responsible for ~200,000 premature mortalities due to long-term exposure to increased $PM_{2.5}$ concentrations, and ~10,600 premature mortalities due to exposure to increased ozone concentrations. The totals computed do not consider non-linearities in the model response (e. g., in the formation of secondary $PM_{2.5}$). This effect is expected to be relatively small, potentially yielding an underestimation in total mortalities of the order of 6%, as found in a study using an analogous methodology in the United Kingdom (Yim and Barrett, 2012).

Among the different sectors considered in this study, road transportation accounts for the largest number of early mortalities, ~53,000 $PM_{2.5}$-related and ~5300 ozone-related. For comparison, we consider that in 2005 the number of fatalities related to car accidents in the U.S. was ~43,500 (U.S. DOT, 2012). This suggests that the air quality impact of road transportation in terms of premature deaths may likely exceed the number of fatal accidents by about 30%. It is documented (U.S. DOT, 2012) that about 40% of the fatal accidents involve people in the 0–44 years range, corresponding to a loss of about 35 life years per fatality. Emissions instead generally affect people at older ages, with an average loss of ~12 years per mortality (COMEAP, 2010), yielding a total of 0.70 million life years lost from both $PM_{2.5}$ and ozone exposure per year. This means that car accidents may still be the leading cause of loss of life years, despite the smaller number of fatalities. These issues related to the use of premature mortalities as a metric to assess the health burden related to air pollution are discussed in COMEAP (2010).

Considering concentrations of different types of $PM_{2.5}$, road vehicles account for a population-weighted concentration of black carbon larger than the sum of all the other sectors (Table 3).

Power generation emissions results in adverse health impacts similar to road transportation in terms of premature mortalities (Table 3). A large extent of this impact is related to sulfur dioxide emissions from coal-fired power plants. The population-weighted concentration of 1.13 $\mu g\ m^{-3}$ of sulfate due to electric generation is the highest among all the $PM_{2.5}$ species for all the sectors considered (Table 2). A reduction of sulfur dioxide emissions from power plants could therefore limit the adverse health impact of electric generation, and should be taken into account for future U.S. energy and air quality policies.

The extent of the impact on air quality by road transportation and electric power generation found in this assessment will drive the selection of future-year mitigation scenarios explored in Part II of the study.

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