# Air pollution and early deaths in the United States: Attribution of $PM_{2.5}$ exposure to emissions species, time, location and sector



by

Irene Constantina Dedoussi

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Submitted to the Department of Aeronautics and Astronautics
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## Abstract

Combustion emissions constitute the largest source of anthropogenic emissions in the US. They lead to the degradation of air quality and human health, by contributing to the formation of fine particulate matter ($PM_{2.5}$), which is harmful to human health. Previous work computed the population $PM_{2.5}$ exposure and number of early deaths caused by emissions from six major sectors: electric power generation, industry, commercial and residential activities, road transportation, marine transportation and rail transportation. In the present work we go beyond aggregate sectors and now attribute exposure and early deaths to sectors, emissions species, time of emission, and location of emission. This enables determination of the emissions reductions that would have the greatest benefit by sectors, species, time and location. We apply a long-term adjoint sensitivity analysis with population exposure to $PM_{2.5}$ in the contiguous US as the objective function, and calculate the four dimensional sensitivities (time and space) of $PM_{2.5}$ exposure with respect to each emissions species. Epidemiological evidence is used to relate increased population exposure to premature mortalities. This is the first regional application of the adjoint sensitivity analysis method to characterize long-term air pollution exposure. (A global scale application has been undertaken related to intercontinental pollution.) We find that for the electric power generation sector 75% of the attributable $PM_{2.5}$ exposure is due to $SO_2$ emissions, and 80% of the annual impacts are attributed to emissions from April to September. This suggests that burning of low sulfur coal has greatest benefit in the summer. In the road transportation sector, 29% of $PM_{2.5}$ exposure is due to $NO_x$ emissions and 33% from ammonia ($NH_3$), which is a result of emissions after-treatment technologies. We estimate that the benefit of reducing $NH_3$ emissions from road transportation is $\sim$20 times that of $NO_x$ per unit mass. 75% of the road transportation ammonia impacts occur during the months October to March. We rank the states based on their contribution to the overall combustion emissions-attributable $PM_{2.5}$ exposure in the US, and calculate that California contributes 12%, Pennsylvania 7% and Ohio 5.8%. We publicly

release the sensitivity matrices computed, noting their potential use as a rapid air quality policy assessment tool.

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# 1. Introduction

Air pollution and degraded air quality adversely impacts human health [1, 2]. Epidemiological studies link long-term exposure to particulate matter $PM_{2.5}$ (fine particulate matter, with an aerodynamic diameter less than 2.5 μm) to an increased risk of premature mortality [3–5]. Given it is the most significant known cause for early deaths associated with outdoor air pollution, $PM_{2.5}$ has been become the predominant metric to quantify air quality [1].

Fann et al. [6] estimated the overall $PM_{2.5}$ attributable premature mortalities in the US to be between 130,000 and 340,000 for the year of 2005, and a later study [7] provided an apportionment of these based on the emission source type. Within the US, combustion emissions are the predominant source of ground level $PM_{2.5}$ concentrations [1]. A study by Caiazzo et al. [8], which precedes this work, quantified the population $PM_{2.5}$ exposure and early deaths attributable to combustion emissions of the major sectors in the US in 2005. The major sectors were defined as electric power generation, industry, commercial and residential, and three modes of transportation: road, marine, and rail transportation. The number of $PM_{2.5}$–related premature mortalities attributable to combustion emissions was estimated to be 200,000 (90% CI: 90,000-362,000), ranking road transportation activities the largest contributor with ~53,000 (90% CI: 24,000-95,000) and electric power generation emissions the second largest with ~52,000 (90% CI: 23,000-94,000) premature mortalities. It was also shown that ozone–related premature mortalities were 5% of total (ozone and $PM_{2.5}$ attributable) mortalities. (In this work we only consider $PM_{2.5}$.)

Caiazzo et al. [8] quantified both the total health impacts and the spatial distribution of health impacts and $PM_{2.5}$ constituents in the US for each of the aforementioned sectors. While this is suggestive of mitigation measures at the level of prioritizing sectors, it does not provide information about which emission species, times and locations are responsible for the impacts, or in which sector there is the greatest marginal benefit for reductions. The aim of the present thesis is to determine these and specify emissions reductions (in terms of sector, species, location and time) that will result in the greatest health benefits.

In Caiazzo et al. [8] a forward chemistry-transport model subtraction method was used to quantify the $PM_{2.5}$ impacts. This method involved taking the difference of two forward atmospheric chemistry-transport simulations, one of the baseline case (including all combustion emissions) minus one for which one of the sector's emissions were eliminated. This approach has the benefit of being simple to implement and producing many disaggregated (spatially varying) outputs based on a few aggregated inputs. This method is matched to cases such as when the impact of a single emissions change on the different parts of the US is needed. In our case, however, the opposite is required. We are aiming to calculate the impact of multiple control parameters (species, time, and location) on population $PM_{2.5}$ exposure in the US. The influence of location, source and emission type on the estimates of human health benefits has previously reported by Fann et al. [9], using a reduced form (forward) air quality model, which allowed the assessment of 12 emission/source reduction scenarios, using the subtraction method. Performing forward chemistry-transport model simulations, by examining all of these parameters (sector, species, location, time) separately, would be computationally impractical.

In the present work an adjoint approach is employed to tackle the problem of quantifying the relationship between the emission characteristics (species, time and location) and the overall $PM_{2.5}$ exposure for the six major sectors in the US. The adjoint method traces sensitivities back to individual sources in a single simulation, and thus provides information about influences from individual sources on an aggregated output, which in

our case is the total population $PM_{2.5}$ exposure (or premature mortalities when multiplied by a concentration–response coefficient) in the US. A more detailed description of adjoint sensitivity methods in atmospheric modeling can be found in Henze et al. [10], Hakami et al. [11], and Sandu et al. [12].

Four dimensional (space and time) sensitivity matrices of population $PM_{2.5}$ exposure with respect to the various PM, and PM precursor emissions, are computed. These sensitivities can also used as a rapid policy assessment tool, to quickly quantify the impacts of emissions scenarios.

The adjoint sensitivity approach has previously been used in the context of intercontinental transport, as well as policy assessment. Koo et al. [13] used it to capture the intercontinental high-altitude pollution risks to human health. Gilmore et al. [14] used adjoint sensitivities to assess the temporal and spatial variability in $O_3$ production due to aviation $NO_x$ emissions. Mesbah et al. [15] took advantage of the rapid policy assessment benefits and estimated the regional impacts of electricity generating units combining it with an optimization tool to suggest mitigation measures. To our knowledge this is the first study to use an adjoint approach to compute long–term exposure and health impacts at a regional scale. [Work by Koo et al. [13] and forthcoming work by C.J. Lee et al. has applied an adjoint approach to compute health impacts in global scale modeling.]

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# 2. Methods

This section describes the modeling techniques applied to quantify the influence of the species, location, and time of combustion emissions on long-term total population $PM_{2.5}$ exposure and $PM_{2.5}$ exposure–attributable early deaths in the US. First, the combustion emissions dataset is described. The adjoint analysis that computes the sensitivity matrices of $PM_{2.5}$ exposure to all the species, locations and times is then presented. The multiplication of the sensitivity matrices with the emissions from each sector allows the assessment of the species, locations and times that have the most significant contribution to the annual $PM_{2.5}$ exposure increase on the US. Population exposure is then translated into premature mortalities using a concentration–response function.

## 2.1 Emissions dataset

The emissions dataset used in this study is based on the 2005 EPA National Emissions Inventory, developed by USA EPA [16], and processed by Caiazzo et al. [8], using SMOKE [17] and source characteristic data from Ashok et al. [18]. It includes combustion emissions from six different sectors: electric power generation, industry, commercial and residential, road transportation, marine transportation and rail transportation. The totals of the US $NO_x$, $SO_x$ and primary $PM_{2.5}$ emissions for each sector in Tg/year are summarized in Table 2.1. Their corresponding relative contribution to the total of emissions is also tabulated. Emissions of all other species are shown in Table A.1 in the Appendix.

These emissions totals have decreased between 2005 and the present time [19], and it

Table 2.1: Primary $PM_{2.5}$, $NO_x$ and $SO_x$ emissions totals and percentages with respect to the baseline scenario. Emissions are expressed in Tg/year for each sector considered in the study (data for 2005).

| Sector | Primary $PM_{2.5}$ | | $NO_x$ | | $SO_x$ | |
|---|---|---|---|---|---|---|
| | 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% |

remains part of future work to apply the same method the most recent emissions dataset and its projected values for the future years.

## 2.2 Air quality modeling

The GEOS-Chem adjoint model that is used to produce sensitivities of $PM_{2.5}$ exposure to emissions is described first, and the way these sensitivities were defined and applied is then presented.

### 2.2.1 GEOS-Chem adjoint

GEOS-Chem is a global tropospheric chemistry–transport model, originally developed by Bey et al. [20]. It performs transport, gas- and aerosol-phase chemistry, as well as wet and dry deposition calculations. In the GEOS-Chem version that was implemented in the present work the KPP chemical solver [21] and the RPMARES aerosol equilibrium model (an implementation of the MARS-A scheme of Binkowski and Roselle [22]) were used. A simplified linearized Ozone scheme (LINOZ) is used for the stratospheric chemistry

calculations [23]. GEOS-Chem takes as inputs emissions as well as GEOS5 meteorological data from the Global Modeling and Assimilation Office (GMAO) at the NASA Goddard Space Flight Centre.

The three dimensional grid focuses on the North American (NA) domain, 140 °W to 40 °W longitude, and 10 °N to 70 °N latitude. The resolution of the horizontal grid is $0.5\,^\circ \times 0.666\,^\circ$ (latitude $\times$ longitude), with 47 vertical layers up to 80 km. Boundary conditions for the NA domain are obtained by running GEOS-Chem with a global domain (at $4\,^\circ \times 5\,^\circ$ resolution). The adjoint simulations are run for a 15-month period, with the first 3 months of the (backward) simulation used as the adjoint spin-up time. This is time during which the model is run, but the outputs are not included in the analysis, to ensure that any initial conditions do not contribute significantly to the air quality impacts during the annual time period that is of interest.

The uncertainty of GEOS-Chem in predicting PM concentrations was quantified in terms of normalized mean biases in the same way as in Caiazzo et al. [8]. We note that total population $PM_{2.5}$ exposure as calculated by GEOS-Chem in the present work is 14% higher than as calculated by CMAQ in Part I. This additional correction is not accounted for so that differences in modeling are not obscured.

The adjoint model of GEOS-Chem provides a computationally efficient way of calculating sensitivities. It was developed by [10], and an aviation module was added and used by Koo et al. [13] and Gilmore et al. [14].

### 2.2.2 Sensitivities

The computed sensitivities are partial derivatives of a quantity of interest (objective function) with respect to various control parameters. The quantity of interest (objective function) for the adjoint sensitivity, $J$, is the annually averaged population exposure to $PM_{2.5}$ over a specific domain. In our case, the domain of interest is the contiguous US, including the 48 states and the District of Columbia (i.e. excluding Hawaii and Alaska). The

objective function $J$ is therefore defined as

$$J = \frac{1}{T} \cdot \sum_{i=1}^{N_{\text{lon}}} \sum_{j=1}^{N_{\text{lat}}} \sum_{t=1}^{T} \left[ \rho_{ij} \cdot \Delta\chi_{ijt} \right], \tag{2.1}$$

where $i$ and $j$ are indices for the longitude and latitude, respectively, and $N_{\text{lon}}$ and $N_{\text{lat}}$ the number of grid cells in the longitudinal and latitudinal sense, $t$ denotes the time step, $T$ the number of time steps in the simulation, $\Delta\chi_{ijt}$ is the concentration perturbation of $PM_{2.5}$ in $\mu g/m^3$ at time step $t$ in grid cell $(i, j)$ of the ground layer, and $\rho_{ij}$ is the population (over age 30 as this is consistent with the applicability of the concentration–response function applied) in grid cell $(i, j)$. Population data was processed from the GRUMP 2006 database [24]. The sensitivities were computed for the year of 2006.

The three-dimensional sensitivity matrices we obtain from the adjoint at every timestep $t$ are of the form

$$S^w_{ijkt} = \frac{\partial J}{\partial E^w_{ijkt}}$$

for emissions species $w$, where $J$ is the previously defined objective function and $E^w_{ijkt}$ are the emissions (in kg) of species $w$ in the three-dimensional grid at timestep $t$. These show the effect that a kg of emission of specie $w$ at grid location $(i, j, k)$ and time $t$, has on the annually averaged population exposure to $PM_{2.5}$ within the US.

We calculate the impact of each species, locations and time, on annual average population $PM_{2.5}$ exposure increase in the US by taking the inner (Forbenius) product of the sensitivity with the emissions from each of the sectors, i.e.

$$P^w_{ijkt} = S^w_{ijkt} : E^w_{ijkt},$$

where $P^w_{ijkt}$ is the total US annual average population $PM_{2.5}$ exposure (in people $\times$ $1\,\mu g/m^3$) caused by emission $E^w_{ijkt}$ (in kg) of species $w$ at location $(i, j, k)$ and time of emission (and not of exposure) $t$.

The adjoint method quantifies linearized relationship between emissions and $PM_{2.5}$ exposure. This makes it well suited to computing the impact of marginal emissions changes, e.g. at a particular location or time, or of marginal changes in emissions of a particular species and/or a particular sector. We note that in the context of attributing exposure to entire sectors, it could be argued that the non–linear response must be captured (as in Caiazzo et al. [8]), and so there is some degree of inaccuracy in using an adjoint for this purpose. On the other hand, it could be argued that a linear approximation is more meaningful as then the total impact equals the sum of the contributions, and attribution among strongly interacting sectors would have limited conceptual utility. As will be discussed in Chapter 3, the sector for which results imply a potentially significant degree of non–linearity is road transportation (dominated by $NO_x$ emissions), whereas, for example, power generation is dominated by relatively linear $SO_2$ to sulfate chemistry, and other sectors are relatively small.

## 2.3 Health impacts assessment

The health impacts in this work are quantified in terms of premature mortalities. Epidemiological studies have developed quantitative associations between increased human exposure to $PM_{2.5}$ and increased risk of premature death [1, 25–27], due mainly to cardiopulmonary diseases and lung cancer. We compute the expected number of early deaths using a concentration–response function (CRF) derived from an EPA study. Specifically, a 1% (range 0.4%-1.8%) increase in all-cause mortalities for every 1 $\mu g/m^3$ increase in the annually averaged $PM_{2.5}$ exposure in the US is applied for adults over the age of 30 years old [1]. The number of early deaths caused by emission $E^w_{ijkt}$ (in kg) of species $w$ at location $(i, j, k)$ and time of emission (and not of exposure) $t$ is then

$$D^w_{ijkt} = P^w_{ijkt} B_{ij} R,$$

where $B_{ij} = B$ is the baseline all-cause death rate (taken as the US average) and $R = 1\%$ is the risk coefficient.

Uncertainty is treated in the same way as in the previous work by Caiazzo et al. [8]. When quantifying the impact of emissions of a particular species, or emissions at a particular time, we term resulting premature mortalities as "equivalent". We define equivalent premature mortalities as a measure of the $PM_{2.5}$ exposure impact assuming that all $PM_{2.5}$ constituents are of equal toxicity and contribute equally towards the increased risk of death. We draw this distinction because the concentration–risk function applied was effectively derived for an urban mixture of PM species, and not for a specific PM species, so its application more narrowly than a mixture of PM types explicitly raises the differential toxicity question. As was noted by Levy et al. [28], it is likely that different PM species are not of equal toxicity, but there is a lack of quantitative basis for weighting PM species by toxicity. We also assume in our equivalent premature mortality metric that an exposure contribution at any time of the year has the same effect. In other words, any temporal weighting implicit in the derivation of the concentration–response function, and differences between sectors, are not accounted for. The equivalent premature mortality metric is more understandable than showing summed people $\times$ $1\,\mu g/m^3$, but we use the term "equivalent" to draw attention to the aforementioned uncertainties.

# 3. Results and Discussion

We first compare the GEOS-Chem adjoint method results with those from the CMAQ forward modeling of Caiazzo et al. [8] (section 3.1). The following three sections (3.2, 3.3, 3.4) then present the calculation of the impact of the species, time and location of each of the sectoral emissions.

Table 3.1: Comparison between Caiazzo et al. [8] (CMAQ) and present work (GEOS-Chem adjoint) excluding secondary organic aerosols (SOAs). The premature mortalities attributable to each sector are shown, as well as the percentage difference between our current GEOS-Chem adjoint model and the CMAQ model.

| Sector | Premature deaths | | |
|---|---|---|---|
| | Part I | Part II | % difference |
| Electric power generation | 41,570 [18,700–74,830] | 41,660 [18,750–75,000] | 0.2 % |
| Industry | 33,160 [14900–59,690] | 37,440[16,850–67,390] | 12.9% |
| Commercial/residential | 32,370 [14,570–58,270] | 35,790 [16,110–64,420] | 10.6% |
| Road transportation | 33,590[15,120–60,460] | 47,780 [21,500–86,000] | 42.2 % |
| Marine transportation | 6900 [3110–12420] | 5980 [2690–10,760] | -13.3 % |
| Rail transportation | 2630 [1180–4730] | 2500 [1130–4500] | -4.9% |
| Total | 150,220 [67,600–270,400] | 171,150 [77,000–308,100] | 13.9% |

## 3.1 Model intercomparison

We aggregate the temporal, spatial and speciated results to calculate the overall health impacts attributable to the combustion emissions for each of the six sectors and compare these with the forward CMAQ method from Caiazzo et al. [8]. Since the adjoint GEOS-Chem model does not account for secondary organic aerosols (SOAs) the results from Caiazzo et al. [8] were corrected to only include primary organic aerosol emissions.

The comparison is presented in Table 3.1 and it includes percentages differences of the GEOS-Chem adjoint model from the CMAQ model. Overall the GEOS-Chem adjoint approach results in a 13.9% higher estimate in early deaths attributable to US combustion emissions. All individual sector differences are lower than this (in magnitude), except road transportation (42.2%). Road transportation and power generation are the two largest sectors in terms of attributable early deaths, and are within $\sim$ 10% of each other. In the case of power generation, which as we will show is dominated by sulfate impacts, there is a 0.2% difference between the CMAQ and GEOS-Chem adjoint results. As $SO_2$ to sulfate chemistry is relatively linear compared to the $NO_x$ and $NH_3$ to nitrate chemistry that mediates the impact of road transportation, and the two sectors are of similar size in terms of their exposure impact, this suggests that non-linearity may be important in the case of road transportation (and is less important in other sectors).

Inter-model differences in transport, chemistry and aerosol thermodynamic equilibrium codes, as well as other components, may also play a role. The conclusion that can be drawn from this intercomparison is that CMAQ and the GEOS-Chem adjoint produce mortality estimates that are generally within 15%, with the exception of road transportation that is within 50%. We note that these differences are within both the uncertainty in the concentration-response function and the uncertainty in computed PM concentrations with either CMAQ or GEOS-Chem.

## 3.2 Speciation

We quantify how much each emissions species from each sector contributes towards the total annual average population $PM_{2.5}$ exposure in the US attributable to that sector. Figure 3-1 presents this finding by showing, for each sector, the percentage contribution of each emission species to overall $PM_{2.5}$ exposure attributable to that sector. (Note that this differs from Caiazzo et al. [8] in that the attribution is to the emission species, not the exposure

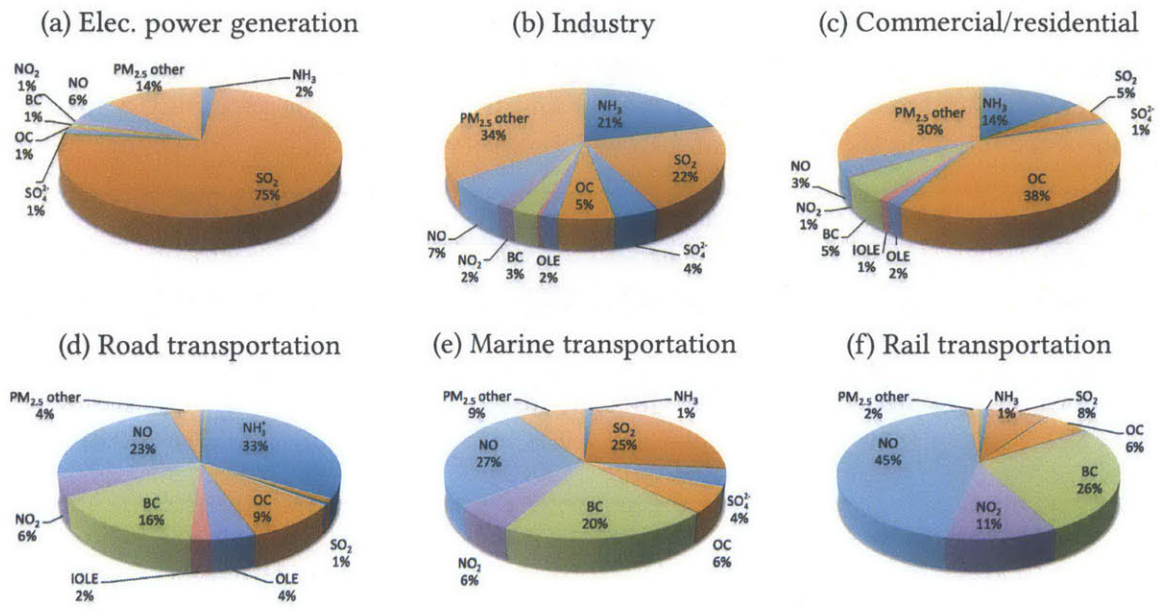



Figure 3-1: Speciated attribution of $PM_{2.5}$ exposure increase the US attributable to each of the sectors. OLE represents the olefin lumped group and IOLE the internal olefin (R-HC=CH-R). $PM_{2.5}$ other is primary PM other than BC or OC.

species.) Table 3.2 shows the benefits, expressed in terms of avoided equivalent premature mortalities per Tg of annual emissions reduction for each of the major species and all the sectors.

The majority of the mortalities (75%) from the electric power generation is attributable to the $SO_2$ emissions of this sector. $SO_2$ is a precursor for sulfate, which dominates the impacts of the sector.

For the industry sector, we estimate that the largest contributor (34%) to population exposure originates from primary $PM_{2.5}$ emissions. $SO_2$ and $NH_3$ follow with 22% and 21%, respectively. We find that the $NO_x$ emissions account for 9% of the population exposure attributable this sector.

The commercial and residential activities attributable $PM_{2.5}$ exposure impacts are 73% attributable to primary $PM_{2.5}$ emissions. This is consistent with the sector having the highest contribution of overall primary $PM_{2.5}$ emissions. Primary organic aerosols dominate this non-SOA $PM_{2.5}$ contribution.

Table 3.2: Equivalent mortalities reduction benefit expressed in avoided equivalent deaths/Tg of annual emissions. This metric quantifies how many mortalities are avoided for 1 Tg emissions reduction of each species and from each sector, assuming a spatially uniform reduction in the domain, and an equal toxicity for each of the $PM_{2.5}$ constituents.

| | Electric Power | Industry | Commercial | Road | Marine | Rail |
|---|---|---|---|---|---|---|
| CO | 2.17 | 2.80 | 3.3 | 3.14 | 3.71 | 3.27 |
| $NH_3$ | $1.79\times10^4$ | $2.93\times10^4$ | $6.53\times10^4$ | $5.58\times10^4$ | $8.98\times10^4$ | $3.16\times10^4$ |
| $SO_2$ | $1.66\times10^3$ | $1.60\times10^3$ | $1.72\times10^3$ | $1.59\times10^3$ | $1.72\times10^3$ | $1.48\times10^3$ |
| $NO_3^-$ | $1.66\times10^3$ | $5.27\times10^3$ | $7.98\times10^3$ | $10.3\times10^3$ | $11.6\times10^3$ | $6.30\times10^3$ |
| $SO_4^{2-}$ | $0.51\times10^4$ | $1.20\times10^4$ | $1.70\times10^4$ | $1.63\times10^4$ | $0.79\times10^4$ | $1.16\times10^4$ |
| $NO_x$ | $4.29\times10^2$ | $5.89\times10^2$ | $9.56\times10^2$ | $8.61\times10^2$ | $7.66\times10^2$ | $7.01\times10^2$ |
| Primary $PM_{2.5}$ | $0.80\times10^4$ | $1.60\times10^4$ | $1.95\times10^4$ | $2.67\times10^4$ | $2.06\times10^4$ | $1.62\times10^4$ |

For road transportation sector, the attributable $PM_{2.5}$ exposure impacts are distributed mostly between four emissions species. We estimate that BC and OC are responsible for 16% and 9% of sector–attributable $PM_{2.5}$ exposure , respectively. $NH_3$ is responsible for 33% of the exposure and equivalent mortalities, whereas $NO_x$ emissions account for 29%.

While road transportation is the highest contributor to the overall $NO_x$ emissions in the US (see Table 2.1) , the $NO_x$ contribution to the overall $PM_{2.5}$ exposure is lower than that of $NH_3$ for the sector. Efforts to reduce automobile $NO_x$ emissions by aftertreatment technologies such as catalytic reduction result in the sector's $NH_3$ emissions. In such catalysts typically ammonia or urea powder is added to the exhaust gas as a reductant, to enable the reaction of $4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O$, which removes the NO from the exhaust emissions. However, when incorrect amounts of ammonia are injected, or the optimal temperature for the reaction is not reached, there is ammonia leakage to the exhaust, commonly referred to as ammonia slip [29, 30]. Ammonia then leads to secondary particle formation through reaction with $HNO_3$ and sulfate (to form ammonium nitrate and ammoniated sulfate compounds). The extent of the contribution of ammonia emissions to the overall road transportation attributable population PM exposure suggests that attention must be paid to the potential compromise between the $NO_x$ emissions reduction benefits and the $NH_3$ leakage risk disbenefits. This compromise has been noted in literature before

[31–33], but has not been quantified in terms of exposure or equivalent mortalities to our knowledge.

The importance of $NH_3$ in the formation of $PM_{2.5}$ is evident in Table 3.2. The relative benefit of reducing 1 Tg of $NH_3$ emissions from the road transportation is ~20 times higher than a 1 Tg reduction of $NO_x$ emissions. Noting that the $NO_x$ and $NH_3$ attributable equivalent mortalities are now approximately equal for the sector, this result suggests that further reductions in $NO_x$ emissions at the expense of increased $NH_3$ emissions are only beneficial in terms of PM exposure if the ratio exceeds 20:1 on a mass basis.

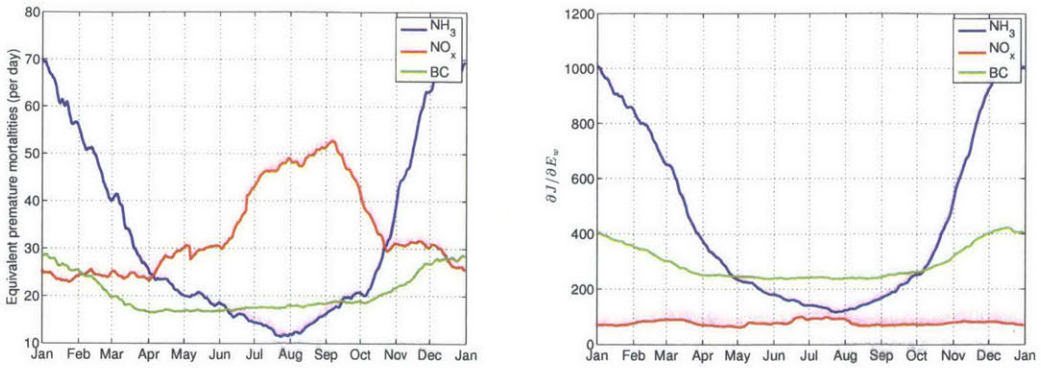
Roe et al. [30] compared the amount of $NH_3$ emissions from different sources in 2002 and estimated that in the US 70.9% of the $NH_3$ emissions comes from livestock, while 5.2% comes from on road mobile sources. However, when characterizing the $NH_3$ emissions in urban areas where they have the greatest potential to amplify PM concentations, this number increases. Roe et al. [30] found that ~50% of the ammonia contribution in the New York metropolitan area comes from on road mobile sources.

Marine transportation equivalent premature mortalities are 33% attributable to $NO_x$, 29% to $SO_x$ and 35% to primary $PM_{2.5}$ emissions. For rail transportation, 56% of the sector's impacts are attributable to $NO_x$ emissions.

## 3.3 Time of emission

The previous section quantified the annual impacts of the time averaged emissions for each of the sectors throughout the year. In this section we characterize the temporal variation of the attributable air quality impacts throughout the year for each sector.

Figure 3-2 shows the daily contribution to the annual equivalent premature mortalities for $NH_3$, $NO_x$, and BC emissions from the road transportation sector. These four species in total are responsible for 78% of the annual impacts of the sector. We observe a significant variation for $NH_3$ throughout the year, with the "winter" months (October




(a) Equivalent early deaths

(b) Sensitivities

Figure 3-2: Temporal variability of the health impacts attributable to the main road transportation emission species. Figure 3-2a shows the number of equivalent premature mortalities that the daily emissions of each species contributes towards the overall US equivalent deaths from the road sector. Figure 3-2b shows how the sensitivity of a kg/hr of emissions of each species to the annually averaged population exposure at $PM_{2.5}$ varies through the year. These sensitivities are for the ground level emissions, and are independent of sector. The temporal variation plots have been smoothed using a floating average.

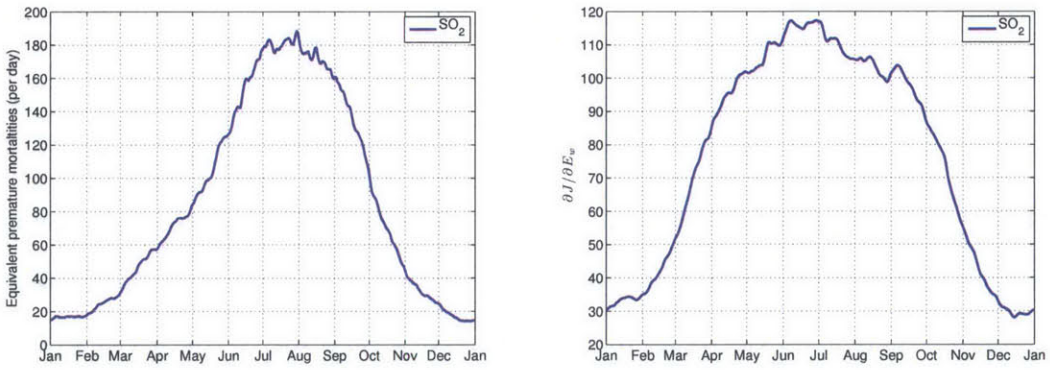
to March) $NH_3$ emissions contributing to the annual $PM_{2.5}$ impacts three times as much as the $NH_3$ emissions from the "summer" months (April to September). For $NO_x$ on the other hand, the impacts become more pronounced during the summer months, with the summer emissions contributing to ~65% of the annual impacts. For the road transportation sector these variations are driven by the sensitivity variations throughout the year. These are plotted in Figure 3-2b and are sector independent (except for the assumed altitude of emissions). The high winter impact of $NH_3$ directly scales with the higher winter sensitivity values.

Noting that the equivalent deaths caused by $NH_3$ emissions is highest in the winter, and the equivalent deaths caused by $NO_x$ emissions are greatest in the summer, this suggests that the relative benefit of catalytic reduction type emissions control is greatest in the summer.

In the electric power generation sector we also observe a significant temporal variability of the impacts, and this is shown in Figure 3-3. The electric power generation

emissions during the summer months contribute four times as much as the equivalent emissions during the winter months. This is driven by the temporal variability of the sensitivity of $PM_{2.5}$ exposure to $SO_2$ emissions, as shown in Figure 3-3b, where summer emissions of $SO_2$ cause four times as much annual average $PM_{2.5}$ exposure as in the winter. This suggests further $SO_2$ reduction in the summer (e.g. by burning of low sulfur coal) has four times as much benefit as in the winter. We also note that July $SO_2$ emissions from power generation cause an order of magnitude more equivalent deaths per day than emissions in January.

A similar trend regarding the $NO_x$ impacts to that of the road transportation sector is evident for the other sectors. The temporal variability plots for the rest of the sectors are provided in the Appendix (Figures B-1, B-2, B-4, B-3).




(a) Equivalent early deaths

(b) Sensitivity

Figure 3-3: Temporal variability of the health impacts attributable to the $SO_2$ emission from the electric power generation sector. Figure 3-3a shows the number of equivalent premature mortalities that the daily emissions of $SO_2$ contributes towards the overall US equivalent deaths from the electric power generation sector. Figure 3-3b shows how the sensitivity of $PM_{2.5}$ exposure to a kg/hr of $SO_2$ emissions to the annually averaged population exposure at $PM_{2.5}$ varies through the year. This sensitivity are independent of sector, and is averaged for altitudes between 0 and 1300m, where the electric power generation emissions occur (accounting for plume rise). The temporal variation plots have been smoothed using a floating average.

## 3.4 Location of emission

The extent to which each state contributes to overall US population PM exposure and early deaths was computed. Table 3.3 presents the number of premature mortalities and the relative contribution that each state has towards the total US early deaths from each sector. We note that California is the highest contributor to the US $PM_{2.5}$ in the industry (13.0%), the commercial and residential (14.9%), the road transportation (16.4%) and the marine transportation (45.4%) sectors. In the electric power generation sector Pennsylvania, Ohio, Indiana and Illinois are responsible for $\sim$35% of the impacts.

In terms of impacts from all the sectors together, the state of California contributes 12%. The states that follow are: Pennsylvania (7%), Ohio (5.8%), New York (5.8%) and Illinois (5.3%).

In Table 3.3 we also show a column

$$X = \frac{\%\text{ of deaths caused by the state's emissions}}{\%\text{ of US population in state}}.$$

When $X > 1$ it means that the state causes more early deaths in proportion to its population than average, while when $X < 1$ the state causes fewer early deaths in proportion to its population than average. Texas (19.7), Alabama (9.8) and Wisconsin (9.3) are the states with the three highest $X$ factor.

Table 3.3: $PM_{2.5}$ attributable premature mortalities (number of mortalities, NM) of the sectoral activity from each state. The percentage contribution of each state, as well as the sums per state, and sums per sector are also included. Note that the sum of the marine percentages does not add up to 100%. This is because marine emissions are included within 200 nmi off the coastline, some of which do not fall within the state boundaries.

| | Elec. generation | | Industry | | Comm/Res | | Road | | Marine | | Rail | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | NM | % | NM | % | NM | % | NM | % | NM | % | NM | % | sum/state | $X$ |
| AL | 1817 | 4.4 | 862 | 2.3 | 527 | 1.5 | 472 | 1.0 | 24 | 0.4 | 44 | 1.7 | 3700 | 9.8 |
| AR | 254 | 0.6 | 434 | 1.2 | 161 | 0.5 | 227 | 0.5 | 17 | 0.3 | 43 | 1.7 | 1100 | 0.4 |
| AZ | 186 | 0.4 | 253 | 0.7 | 290 | 0.8 | 547 | 1.1 | 0 | 0.0 | 20 | 0.8 | 1300 | 0.8 |
| CA | 342 | 0.8 | 4868 | 13.0 | 5348 | 14.9 | 7826 | 16.4 | 2096 | 35.1 | 253 | 10.1 | 20700 | 5.9 |
| CO | 190 | 0.5 | 126 | 0.3 | 266 | 0.7 | 241 | 0.5 | 0 | 0.0 | 18 | 0.7 | 800 | 0.0 |
| CT | 93 | 0.2 | 185 | 0.5 | 872 | 2.4 | 875 | 1.8 | 20 | 0.3 | 8 | 0.3 | 2100 | 0.8 |
| DE | 90 | 0.2 | 157 | 0.4 | 170 | 0.5 | 207 | 0.4 | 47 | 0.8 | 4 | 0.1 | 700 | 0.4 |
| FL | 1302 | 3.1 | 744 | 2.0 | 500 | 1.4 | 1059 | 2.2 | 214 | 3.6 | 19 | 0.8 | 3800 | 7.9 |
| GA | 1952 | 4.7 | 1030 | 2.8 | 972 | 2.7 | 1548 | 3.2 | 14 | 0.2 | 62 | 2.5 | 5600 | 0.5 |
| IA | 449 | 1.1 | 497 | 1.3 | 285 | 0.8 | 368 | 0.8 | 8 | 0.1 | 77 | 3.1 | 1700 | 0.3 |
| ID | 0 | 0.0 | 141 | 0.4 | 85 | 0.2 | 56 | 0.1 | 0 | 0.0 | 7 | 0.3 | 300 | 0.4 |
| IL | 2311 | 5.5 | 2307 | 6.2 | 1175 | 3.3 | 2812 | 5.9 | 78 | 1.3 | 282 | 11.3 | 9000 | 5.3 |
| IN | 3127 | 7.5 | 2007 | 5.4 | 912 | 2.5 | 1470 | 3.1 | 37 | 0.6 | 130 | 5.2 | 7700 | 9.3 |
| KS | 501 | 1.2 | 353 | 0.9 | 156 | 0.4 | 235 | 0.5 | 1 | 0.0 | 72 | 2.9 | 1300 | 0.2 |
| KY | 2529 | 6.1 | 654 | 1.7 | 580 | 1.6 | 578 | 1.2 | 64 | 1.1 | 48 | 1.9 | 4500 | 1.3 |
| LA | 347 | 0.8 | 1703 | 4.5 | 167 | 0.5 | 243 | 0.5 | 264 | 4.4 | 26 | 1.0 | 2700 | 1.7 |
| MA | 214 | 0.5 | 939 | 2.5 | 1398 | 3.9 | 1290 | 2.7 | 38 | 0.6 | 31 | 1.2 | 3900 | 1.6 |
| MD | 1041 | 2.5 | 747 | 2.0 | 1552 | 4.3 | 1656 | 3.5 | 56 | 0.9 | 58 | 2.3 | 5100 | 2.1 |
| MR | 22 | 0.1 | 71 | 0.2 | 193 | 0.5 | 84 | 0.2 | 9 | 0.2 | 0 | 0.0 | 400 | 0.1 |
| MI | 1632 | 3.9 | 1877 | 5.0 | 955 | 2.7 | 2513 | 5.3 | 60 | 1.0 | 58 | 2.3 | 7100 | 2.2 |
| MN | 442 | 1.1 | 656 | 1.8 | 611 | 1.7 | 775 | 1.6 | 35 | 0.6 | 69 | 2.7 | 2600 | 3.5 |
| MO | 819 | 2.0 | 655 | 1.7 | 512 | 1.4 | 640 | 1.3 | 25 | 0.4 | 100 | 4.0 | 2800 | 0.5 |
| MS | 237 | 0.6 | 381 | 1.0 | 192 | 0.5 | 261 | 0.5 | 42 | 0.7 | 32 | 1.3 | 1100 | 0.4 |
| MT | 67 | 0.2 | 154 | 0.4 | 27 | 0.1 | 39 | 0.1 | 0 | 0.0 | 20 | 0.8 | 300 | 0.1 |
| NC | 1465 | 3.5 | 701 | 1.9 | 1042 | 2.9 | 1288 | 2.7 | 37 | 0.6 | 55 | 2.2 | 4600 | 2.8 |
| ND | 442 | 1.1 | 85 | 0.2 | 33 | 0.1 | 51 | 0.1 | 0 | 0.0 | 32 | 1.3 | 600 | 1.1 |
| NE | 338 | 0.8 | 147 | 0.4 | 66 | 0.2 | 164 | 0.3 | 0 | 0.0 | 87 | 3.5 | 800 | 0.2 |
| NH | 140 | 0.3 | 193 | 0.5 | 369 | 1.0 | 232 | 0.5 | 2 | 0.0 | 2 | 0.1 | 900 | 2.5 |
| NJ | 609 | 1.5 | 1332 | 3.6 | 2820 | 7.9 | 3702 | 7.7 | 349 | 5.8 | 30 | 1.2 | 8800 | 8.8 |
| NM | 94 | 0.2 | 81 | 0.2 | 53 | 0.1 | 73 | 0.2 | 0 | 0.0 | 18 | 0.7 | 300 | 0.4 |
| NV | 74 | 0.2 | 82 | 0.2 | 42 | 0.1 | 51 | 0.1 | 0 | 0.0 | 7 | 0.3 | 300 | 0.1 |
| NY | 1190 | 2.9 | 1495 | 4.0 | 2895 | 8.1 | 3982 | 8.3 | 183 | 3.1 | 80 | 3.2 | 9800 | 8.9 |
| OH | 3996 | 9.6 | 1711 | 4.6 | 1879 | 5.2 | 2043 | 4.3 | 88 | 1.5 | 171 | 6.9 | 9900 | 7.0 |
| OK | 405 | 1.0 | 547 | 1.5 | 166 | 0.5 | 336 | 0.7 | 1 | 0.0 | 41 | 1.6 | 1500 | 0.1 |
| OR | 23 | 0.1 | 138 | 0.4 | 532 | 1.5 | 154 | 0.3 | 49 | 0.8 | 15 | 0.6 | 900 | 0.1 |
| PA | 4761 | 11.4 | 2124 | 5.7 | 2574 | 7.2 | 2827 | 5.9 | 170 | 2.8 | 98 | 3.9 | 12600 | 6.2 |
| RI | 78 | 0.2 | 69 | 0.2 | 152 | 0.4 | 132 | 0.3 | 12 | 0.2 | 3 | 0.1 | 400 | 0.2 |
| SC | 777 | 1.9 | 420 | 1.1 | 397 | 1.1 | 534 | 1.1 | 23 | 0.4 | 26 | 1.0 | 2200 | 0.3 |
| SD | 28 | 0.1 | 113 | 0.3 | 42 | 0.1 | 68 | 0.1 | 0 | 0.0 | 7 | 0.3 | 300 | 0.5 |
| TN | 1095 | 2.6 | 848 | 2.3 | 560 | 1.6 | 750 | 1.6 | 33 | 0.5 | 41 | 1.7 | 3300 | 1.3 |
| TX | 1950 | 4.7 | 2770 | 7.4 | 1231 | 3.4 | 2275 | 4.8 | 371 | 6.2 | 110 | 4.4 | 8700 | 19.7 |
| UT | 130 | 0.3 | 97 | 0.3 | 96 | 0.3 | 138 | 0.3 | 0 | 0.0 | 7 | 0.3 | 500 | 0.1 |
| VA | 1326 | 3.2 | 1230 | 3.3 | 1309 | 3.7 | 1378 | 2.9 | 69 | 1.2 | 74 | 3.0 | 5400 | 0.4 |
| VT | 2 | 0.0 | 34 | 0.1 | 109 | 0.3 | 58 | 0.1 | 0 | 0.0 | 1 | 0.0 | 200 | 0.1 |
| WA | 25 | 0.1 | 95 | 0.3 | 241 | 0.7 | 165 | 0.3 | 42 | 0.7 | 12 | 0.5 | 600 | 0.1 |
| WI | 705 | 1.7 | 641 | 1.7 | 870 | 2.4 | 990 | 2.1 | 22 | 0.4 | 58 | 2.3 | 3300 | 9.3 |
| WV | 1719 | 4.1 | 421 | 1.1 | 298 | 0.8 | 209 | 0.4 | 16 | 0.3 | 19 | 0.8 | 2700 | 0.7 |
| WY | 275 | 0.7 | 163 | 0.4 | 18 | 0.0 | 29 | 0.1 | 0 | 0.0 | 21 | 0.9 | 500 | 0.2 |
| Sum per sector | 41600 | 100 | 37300 | 100 | 35700 | 100 | 47700 | 100 | 4600 | 77 | 2500 | 100 | 169400 | |

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# 4. Conclusions

We apply the GEOS-Chem adjoint to compute four dimensional (space and time) sensitivity matrices for population exposure to $PM_{2.5}$ in the US. These sensitivities are used to quantify the speciated, temporal, and spatial variability in the early deaths attributable to emissions from major sectors. We characterize the impacts of power generation, industry, commercial/residential, road, marine and rail transportation. To our knowledge, this is the first time that an adjoint sensitivity analysis is used for long–term exposure and health impacts estimation on a regional scale.

We find that 75% of the electric power generation sector attributable health impacts originate from $SO_2$ emissions. Primary $PM_{2.5}$ emissions are responsible for 34% and 73% of the industry and commercial/residential attributable impacts, respectively. We quantify the road transportation impacts to be 33% attributable to $NH_3$, and 29% to $NO_x$ emissions. This indicates the relative importance of $NH_3$ leakage emissions from automobiles, which until now have been unregulated and demonstrates a tradeoff in emissions control. Marine transportation impacts are computed to be 25% attributable to $SO_2$, 35% to primary $PM_{2.5}$ and 33% to $NO_x$. Rail transportation impacts are 56% attributable to $NO_x$ emissions, and 35% to primary $PM_{2.5}$.

The temporal variability of the impacts is mostly driven by the temporal variability in the sensitivity values throughout the year, rather than variability in emissions. For the road transportation sector, we calculate that the winter months contribute three times as much as the summer months in terms of the $NH_3$ attributable $PM_{2.5}$ exposure. For

the electric power generation sector, we find that the summer months contribute four times as much as the winter months. These findings may pave the way for research into technologies, operation, or policies that account for the significant variations in the impact of emissions over the year. For example, using low sulfur coal in the summer would have four times as much benefit as in the winter.

In terms of spatial variability, we rank the contribution from each state and sector to the overall combustion emissions attributable health impacts in the US. We find that from the overall combustion activity in the US, 12% of the attributable health impacts originate from California, with Pennsylvania and Ohio following with 7% and 5.8% respectively.

The sensitivity matrices computed in this work can be used as a policy assessment tool, as they enable the assessment of premature mortalities in the US due to any emissions scenario in a very rapid manner. These matrices have been publicly released (with a wrapped code for ease of use) at http://lae.mit.edu.

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# A. Emissions totals

The emission totals in Tg/year for every sector and species is shown on Table A.1.

Table A.1: Speciated emissions totals for the year of 2005 in Tg/year for each sector.

| | Elec. Power | Industry | Commercial | Road | Marine | Rail |
|---|---|---|---|---|---|---|
| CO | 0.574 | 3.028 | 4.817 | 39.303 | 0.182 | 0.112 |
| $ALD_2$ | 0.000 | 0.002 | 0.010 | 0.012 | 0.000 | 0.002 |
| FORM | 0.006 | 0.208 | 0.077 | 0.082 | 0.003 | 0.006 |
| ETHA | 0.004 | 0.141 | 0.022 | 0.027 | 0.000 | 0.000 |
| NH3 | 0.023 | 0.134 | 0.038 | 0.142 | 0.000 | 0.000 |
| SO2 | 9.446 | 2.549 | 0.493 | 0.160 | 0.445 | 0.068 |
| $ALD_X$ | 0.000 | 0.030 | 0.115 | 0.071 | 0.000 | 0.008 |
| $SO_4$ | 0.061 | 0.068 | 0.011 | 0.007 | 0.017 | 0.000 |
| OC | 0.021 | 0.062 | 0.335 | 0.080 | 0.009 | 0.005 |
| OLE | 0.002 | 0.066 | 0.066 | 0.216 | 0.001 | 0.001 |
| IOLE | 0.000 | 0.008 | 0.021 | 0.046 | 0.000 | 0.000 |
| BC | 0.012 | 0.029 | 0.052 | 0.151 | 0.019 | 0.020 |
| $NO_2$ | 0.341 | 0.275 | 0.076 | 0.758 | 0.121 | 0.093 |
| NO | 2.004 | 1.614 | 0.447 | 4.834 | 0.770 | 0.596 |
| $PM_{2.5}$ other | 0.363 | 0.409 | 0.292 | 0.034 | 0.024 | 0.001 |

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# B. Temporal variation

The temporal variability of the health impacts attributable to the different emissions species from the different sectors, similar to the ones for the road transportation and the electric power generation sector that were presented on the main article, are shown here for the industry (B-1), commercial/residential (B-2), marine transportation (B-3), and rail transportation (B-4) sectors. The number of equivalent premature mortalities that the daily emissions of each species contribute towards the overall US equivalent deaths from each sector are shown. The temporal variation plots have been smoothed using a moving average.

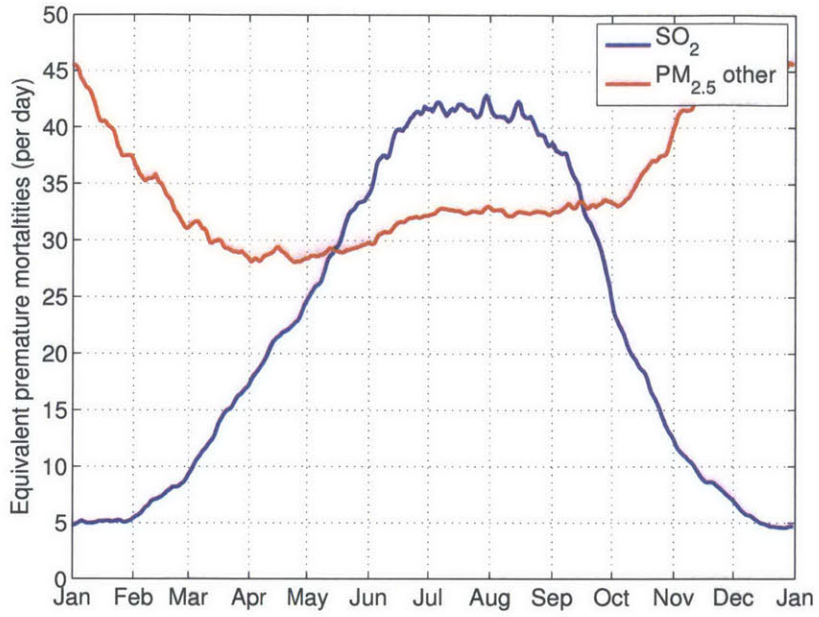


Figure B-1: Temporal variability of the health impacts attributable to the main industry sector emission species

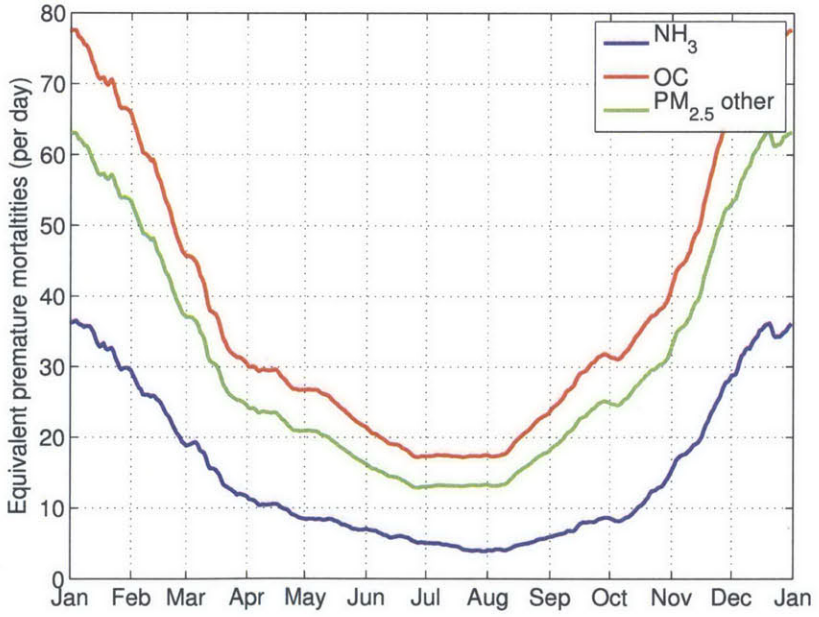


Figure B-2: Temporal variability of the health impacts attributable to the main commercial/residential emission species

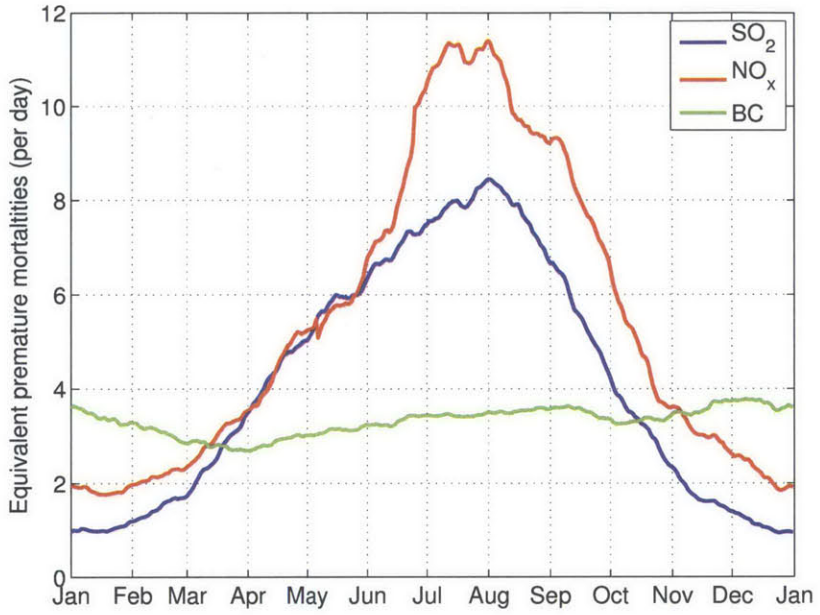


Figure B-3: Temporal variability of the health impacts attributable to the marine transportation emission species

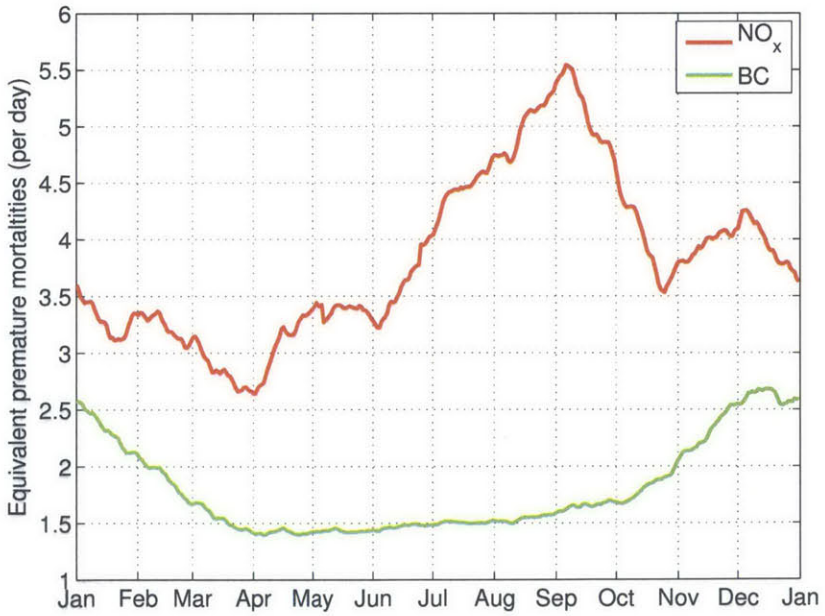


Figure B-4: Temporal variability of the health impacts attributable to the main rail transportation emission species