# Public Health Impacts of Secondary Particulate Formation from Aromatic Hydrocarbons in Gasoline

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**Abstract**

**Background**

Aromatic hydrocarbons emitted from gasoline-powered vehicles contribute to the formation of secondary organic aerosol (SOA), which increases the atmospheric concentration of fine particles ($PM_{2.5}$). Here we estimate the public health burden associated with exposures to the subset of $PM_{2.5}$ that originates from vehicle emissions of aromatics under business as usual conditions.

**Methods**

The $PM_{2.5}$ contribution from gasoline aromatics is estimated using the Community Multiscale Air Quality (CMAQ) modeling system and the results are compared to ambient measurements from the literature. Marginal $PM_{2.5}$ annualized concentration changes are used to calculate premature mortalities using concentration-response functions, with a value of mortality reduction approach used to monetize the social cost of mortality impacts. Morbidity impacts are qualitatively discussed.

**Results**

Modeled aromatic SOA concentrations from CMAQ fall short of ambient measurements by approximately a factor of two nationwide, with strong regional differences. After accounting for this model bias, the estimated public health impacts from exposure to $PM_{2.5}$ originating from aromatic hydrocarbons in gasoline lead to a best estimate of approximately 3800 predicted premature mortalities nationwide, with best estimates ranging from 1800 to over 4700 depending on the specific concentration-response function used. These impacts are associated with total social costs of $28.2B, and range from $13.6B to $34.9B in 2006$. Assuming that the contribution of SOA precursors originating from aromatic hydrocarbons in gasoline is higher in urban areas increases these estimates to 5100 predicted premature mortalities nationwide, with best estimates ranging from over 2400 to 6300, associated with total social costs of $37.9B, ranging from $18.2B to $46.8B in 2006$.

**Conclusions**

This preliminary quantitative estimate indicates particulates from vehicular emissions of aromatics demonstrate a sizeable public health burden.

**Keywords**: Aromatic hydrocarbons, secondary organic aerosol (SOA), secondary particulate, social cost, gasoline

## Background

Field studies suggest 10% - 60% of fine particulate matter ($PM_{2.5}$) is comprised of organic compounds [1](Seinfeld *et al*. 1998). This material may be directly emitted to the atmosphere (primary) or formed from the gas-phase oxidation of hydrocarbon molecules and subsequent absorption into the condensed phase (secondary). The latter portion, referred to as secondary organic aerosol (SOA), is a major contributor to the $PM_{2.5}$ burden in both urban and rural atmospheres [2-5] (Zhang *et al*. 2007; Yu *et al.* 2007; Castro *et al.* 1999; Brown *et al.* 2002; Lim and Turpin 2002), which contributes to a range of adverse health effects [6-8] (Pope *et al.* 1995; Donaldson *et al.* 1998; Pope 2000), visibility reduction [9-10] (Eldering and Cass 1996; Kleeman *et al.* 2001), and global climate change [11-13] (Pilinis *et al.* 1995; Kanakidou *et al.* 2004; Maria *et al.* 2004).

In the atmosphere, SOA can originate from both anthropogenic (e.g., solvent use, mobile sources) and biogenic (e.g., forests) sources. Of the anthropogenic precursors, evidence is growing that aromatic hydrocarbons are among the most efficient at forming SOA [14-15] (Odum *et al.* 1997; de Gouw *et al.* 2008). Table 1 lists several empirical studies that estimated the contribution of SOA precursors to observed $PM_{2.5}$ concentrations. These studies show that aromatics typically contribute between 0.08 and 0.2 μg $C/m^3$ to observed $PM_{2.5}$ concentrations [16-19] (Kleindienst *et al.* 2010; Lewandowski *et al.* 2008; Offenberg *et al.* 2011; Stone *et al.* 2009).

A series of sunlight-irradiated, smog-chamber experiments have confirmed that the $PM_{2.5}$ formation potential of whole gasoline vapor can be accounted for solely in terms of the aromatic fraction of the fuel (Odum *et al.* 1997). More recent chamber studies show that SOA yields measured under low-nitrate conditions greatly exceed formation under high-nitrate conditions, and that SOA yields under high-nitrate conditions are greater than were observed previously (Ng *et al.* 2007). Evidence is growing that aromatics in gasoline exhaust are among the most efficient secondary organic matter precursors (de Gouw *et al.* 2008). In general, air quality models do not adequately capture these increased yields or potential interactions (Docherty *et al.* 2008) although improvements have been made (Carlton *et al.* 2010a).

In the United States, gasoline-powered vehicles are the largest source of aromatic hydrocarbons to the atmosphere (Simon *et al.* 2010). Most gasoline formulations consist of approximately 20% aromatic hydrocarbons (EPA 2012), which are used in place of lead to boost octane. Therefore, it has been suggested that removal of aromatics could reduce SOA concentrations and yield a substantial public health benefit (Gray and Varcoe 2005). The issue is complicated by the fact that any change to fuel composition will affect vehicular emissions of various pollutants (e.g., hydrocarbons, carbon monoxide, oxides of nitrogen, primary $PM_{2.5}$) which, in turn, will react in the atmosphere to produce a different mix of pollutants that may have adverse effects (e.g., Cook *et al.* 2011).

The purpose of this study is to estimate the public health impacts and social costs associated with exposure to SOA from vehicular emissions of aromatic hydrocarbons. This analysis provides a baseline case to explore the magnitude of the issue and against which to evaluate the cost and impacts of potential substitutes for aromatics. The next section describes the methods for the analysis, followed by results and a concluding discussion.

## METHODS

Predicted secondary $PM_{2.5}$ concentrations attributable to single-ringed aromatic hydrocarbons are estimated for a baseline year (2006) using the Community Multiscale Air Quality model version 5.0 (CMAQv5.0). Given that regulatory models are known to underestimate anthropogenic SOA formation (Volkamer *et al.* 2006; de Gouw *et al.* 2008; Docherty *et al.* 2008), these results are compared to available data to estimate scaling factors to adjust the model results. Adjusted $PM_{2.5}$ concentrations are then used in the US EPA Benefits and Mapping Program v4.0 (BenMAP) model to estimate morbidity health and mortality outcomes associated with exposure to these concentrations across the lower 48 states (Abt Associates, Inc. 2011).

### Exposure Concentrations

The CMAQ model is among the most widely used air quality models, with 3000+ registered users in 100 different countries (www.cmaq-model.org). Federal and State regulatory agencies use CMAQ for policy analyses and for routine air quality forecasting (Foley *et al.* 2010). The model provides a means for quantitatively evaluating the impact of air quality management policies prior to implementation. This analysis relied on CMAQv5.0 with the Carbon Bond 2005 (CB05) chemical mechanism, which includes a fairly comprehensive list of precursors that lead to SOA formation via both gas- and aqueous-phase oxidation processes, as well as particle-phase reactions (Carlton *et al.* 2010a).

Air quality model simulations based on CMAQv5.0 are used to estimate the total concentration of SOA from all single-ring aromatic compounds (e.g., benzene, toluene, xylenes) in 12km grid cells for many urban areas and 36km grid cells for the remaining areas covering the lower 48 states for a baseline year (2006).

**Potential Underestimates in Predictions of SOA Formation**

Although CMAQv5.0 contains updated algorithms and processes for predicting SOA formation, evidence suggests that the model may still underestimate secondary $PM_{2.5}$ concentrations (Carlton *et al.* 2010a; Docherty *et al.* 2008; Zhang and Ying 2011), particularly during the summer (Foley *et al.* 2010). Experiments conducted at Carnegie Mellon University to study SOA formation from the photooxidation of toluene show significantly larger SOA production than parameterizations employed in current air-quality models (Hildebrandt *et al.* 2010).

Using an organic tracer-based source apportionment approach, independently conducted research over the last five years provides increasing evidence that aromatic hydrocarbons in gasoline contribute, depending on the specific region, approximately 0.1 to 0.45 $\mu g/m^3$ of PM (Lewandowski *et al.* 2008; Offenberg *et al.* 2011; Kleindienst *et al.* 2010; Stone *et al.* 2009).

Given our objective to estimate the public health impact of aromatic SOA, CMAQv5.0 model results must be adjusted to reflect any biases in this $PM_{2.5}$ component. Monthly-averaged model results are compared against empirical estimates of aromatic SOA concentrations derived from ambient measurements of 2,3-dihydroxy-4-oxopentanoic acid collected at twelve locations across the U.S. (Lewandowski *et al.* 2008; Offenberg *et al.* 2011; Kleindienst *et al.* 2010; Kleindienst *et al.* unpublished data). We develop region-specific regression relationships between predicted CMAQ and measured concentrations in μg of carbon per $m^3$ and use these to adjust the model results prior to estimating health effects. We develop a mixed model with a random slope for each region, as there is some indication that slopes should vary by region. For example, Hildebrandt *et al.* (2010) report elevated SOA yields from toluene under high UV intensity, low-$NO_x$ conditions, and lower temperatures, relative to the parameters used typically in models. Therefore, the slope might be low in CA where there is a lot of $NO_x$ and high in the Midwest and East where ambient temperatures remain relatively low. The overall fixed effect and region-specific random effects models are developed using REML in R (http://www.r-project.org/) based on the following equation:

Formula: SOA ~ CMAQv5.0 + (CMAQv5.0 | region) (Eq. 1)

**SOA Formation from Aromatic Hydrocarbons in Gasoline**

SPECIATE, a US EPA database, provides a repository of volatile organic compound (VOC) speciation profiles of air pollution sources. We use these source profiles in conjunction with the 2005 National Emissions Inventory for VOCs to estimate the proportion of aromatic SOA formation attributable to emissions from gasoline vehicles. We rank order all sources of aromatic VOCs to quantify the contribution to total emissions specifically from gasoline-based sources.

**Health and Mortality Impacts**

The BenMAP model was used to estimate resulting health impacts associated with exposures to the change in $PM_{2.5}$ concentrations attributable to aromatic hydrocarbons from gasoline vehicles predicted by the process described above. The BenMAP model is widely used by regulatory agencies to quantify and monetize potential heath impacts associated with changes in air quality, and contains concentration-response functions for various pollutants, including $PM_{2.5}$, census data and population projections, and baseline mortality and morbidity rates for the lower 48 United States. Concentration response functions incorporated in BenMAP are based on published studies incorporating different assumptions regarding potential thresholds and observed slopes between concentrations and responses.

Four studies are included in this analysis (Krewski *et al.* 2009; Laden *et al.* 2006; Pope *et al.* 2002; Industrial Economics, Inc. 2006). Two major cohort studies are generally thought to provide estimates regarded as most robust and applicable to the general population, with the Harvard Six Cities Study publications reporting central estimates of an approximate 1.2-1.6% increase in all-cause mortality per $\mu g/m^3$ increase in annual average $PM_{2.5}$ (Laden *et al.* 2006) and the American Cancer Society studies reporting estimates of approximately 0.4-0.6% (Pope *et al.* 2002), with higher estimates when exposure characterization was more spatially refined (Krewski *et al.* 2009). Within the expert elicitation study (Industrial Economics, Inc. 2006) the median concentration-response function across experts was approximately 1%, midway between these cohort estimates, with a median 5th percentile of 0.3% and a median 95th percentile of 2.0%. The EPA external Advisory Committee on Clean Air Act Compliance Analysis recommended developing a distribution with the Pope and Laden studies at the 25$^{th}$ and 75$^{th}$ percentiles, respectively, leading to a mean of the new distribution close to the mean of the central estimates of both Pope and Laden. This generally will be consistent with the distribution identified in the expert elicitation (US EPA, SAB, HES, 2010). BenMAP applies these functions to the baseline mortality rate and the number of people potentially exposed by census tract. BenMAP provides distributions of premature mortality estimates based on the uncertainty in the concentration-response functions. That is, the 5$^{th}$ and 95$^{th}$ percentiles in the results are based on the distributions for concentration-response functions only.

**Monetized Estimates of Premature Mortality**

Monetized estimates of premature mortality are based on regulatory estimates of the value of mortality risk as defined by the U.S. EPA (http://yosemite.epa.gov/ee/epa/eed.nsf/pages/MortalityRiskValuation.html). This estimate is based on research in which people are asked how much they would pay for consumer products (such as water filters) that reduce risk or alternatively, that examine how much more employers have to pay employees (adjusting for age, education, experience, etc.) to compensate for taking an increased risk of accidental death. Hence this estimate is not a price on a life, but a price of risk reduction. For convenience it is converted into what was referred to as a value of a statistical life and is now referred to as the value of mortality risk. The implication is if people are willing to pay $X for a reduction in risk of 1 in 10,000, than reducing risk in enough people to produce, on average, one fewer death would be worth 10,000 X dollars. The U.S. EPA recommends a value of $7.4M in 2006 dollars (USEPA 2010) based on over 30 labor market and contingent valuation studies.

## RESULTS

### CMAQv5.0 Modeling Results Compared to Measurements

Table S1 compiles measurement-based estimates of aromatic SOA collected at twelve locations between 2004 and 2010. Concentrations reach as high as 0.41 μgC/m$^3$ during the summer in Cincinnati, with a median value of 0.14 μgC/m$^3$ across all 77 samples. In contrast, the CMAQv5.0 model results from the corresponding 12km grid cells and averaged over the appropriate month in 2006 show a maximum value of 0.13 and a median of 0.052 μgC/m$^3$ (see Table S1). This systematic bias in the model results warrants some adjustment of the CMAQv5.0 output before it is used in the BenMAP calculations. The mixed model obtained by regressing observations against the CMAQv5.0 results are shown in Table 2. The slopes do differ by region, with the highest slopes observed in the East and Midwest. Aggregated up to the national level, unadjusted CMAQ results predict a nationwide average concentration of 0.0448 μg/m$^3$, which increases to 0.17 following the adjustment, a factor of approximately two, consistent with initial estimates ranging from factors of two to five.

### Predicted $PM_{2.5}$ Concentrations from Aromatic Hydrocarbons in Gasoline

Source-specific speciation of total VOC in the National Emissions Inventory reveals that the U.S. emissions of aromatic hydrocarbons are 3.6 million tons per year, of which 69% are from gasoline-powered vehicles (Simon et al., 2010) as shown in Table 3. A source-by-source breakdown of all aromatic hydrocarbon emissions is provided in Table S2. To subtract the contribution of other emission sources (e.g., solvent usage, diesel exhaust) from our calculations, the adjusted aromatic SOA concentrations from CMAQv5.0 are multiplied by 0.69.

Spatial patterns of aromatic emissions are similar across sources. After gasoline, the next highest source of aromatics is solvent usage, and Reff et al. (2009) show that the spatial pattern of solvent usage is similar to gasoline, that is, occurs predominantly in urban areas. In addition, most major refineries are also in close proximity to urban areas.

**Adjusted CMAQv5.0 Results**

Figure 1 shows the final nationwide distribution of annual average $PM_{2.5}$ concentrations attributable to aromatic hydrocarbons emitted from gasoline vehicles, after applying all the adjustments to the CMAQv5.0 output described above. The nationwide average predicted $PM_{2.5}$ concentration based on the average predicted value for each state is approximately 0.124 $\mu g/m^3$ (standard deviation = 0.059 $\mu g/m^3$; minimum = 0.025 $\mu g/m^3$, maximum = 0.227 $\mu g/m^3$) and ranges from 0.013 to greater than 0.257 $\mu g/m^3$ at the county level. On a statewide basis, Table 4 shows the rank ordered concentrations by state, with Connecticut, Rhode Island, Ohio, New York, New Jersey and Indiana predicted statewide concentrations at 0.20 $\mu g/m^3$ or higher.

**BenMAP Modeling Results**

Figure 2 presents a nationwide map of predicted premature mortalities attributable to aromatic hydrocarbons in gasoline associated with the expert elicitation concentration-response function. Table 5 and Figure 3 provide a summary of predicted premature mortality and monetized estimates of social cost based on all four different concentration-response functions. Predicted premature mortalities range from nearly 1,850 to more than 4,700 cases, depending on which concentration-response function is used, which correspond with approximately \$13.6B to \$34.9B in total social costs. The 5th and 95th percentiles from each study are included in the parentheses, and represent the effect of uncertainty in the concentration-response functions only (e.g., there are many potential sources of uncertainty, but only those associated with the concentration-response functions are captured in BenMap). Our recommended best estimate is approximately 3,800 premature mortalities based on the mean of the expert elicitation concentration-response function. Based on the central estimates from the Krewski and Laden studies, respectively, results in a confidence interval of 1,850 to 4,700 for a central estimate.

The results in columns 3-4 (a) in Table 5 are adjusted by 0.69 to account for the fraction of aromatic emissions attributable to gasoline sources based on the National Emissions Inventory. However, it is still possible that the fraction of aromatic emissions from gasoline could be higher in urban areas (although, as noted previously, Reff et al. 2009 have shown that spatial patterns of emissions from other sources of aromatics such as solvent usage are similar to gasoline). To explore this assumption, we adjust only those counties designated as rural counties (CDC 2012) by 0.69 and assume that 100% of emissions in urban areas are derived from gasoline sources. The results are shown in the final two columns of Table 5 and in Figure 3. Predicted premature mortality increases to a little over 5,000, and based on the concentration-response function used, ranges from 2,400 to over 6,300.

Table 6 provides predicted premature mortalities and associated social costs for each of the four concentration-response functions, adjusted by 0.69. Figure 4 provides the results for each state, sorted from highest to lowest predicted impacts, using MetaDataViewer available from the National Toxicology Program (Boyles *et al.* 2011) for the best estimate represented by the expert elicitation slope (the remaining results are proportional based on the results presented in Table 6; results not shown graphically). New York, with a predicted average $PM_{2.5}$ concentration of 0.21 μg/m$^3$, shows the highest predicted impacts based on the number of exposed individuals. Ohio and Pennsylvania follow, with approximately 260 predicted premature mortalities each (based on the midpoint of the combined expert elicitation concentration-response function). The two states with the highest population,, Texas and California, are ranked eighth and tenth, respectively, for premature mortalities at approximately 160 and 130 expected cases, respectively.

## DISCUSSION

The best estimate of potential impacts is based on the expert elicitation concentration-response function recently endorsed by a US EPA Science Advisory Board Panel together with the regression-based adjustment factors to the CMAQv5.0 predictions resulting in 3,800 predicted premature mortalities. This compares to a recent nationwide estimate of approximately 130,000 overall premature mortalities (for 2005) associated with all $PM_{2.5}$ exposures recently discussed by Fann *et al.* (2012) and based on the Krewski *et al.* (2009) concentration-response function. The results presented in Fann *et al.* (2012) were based on CMAQv4.7 together with additional monitoring data to estimate premature mortalities attributable to exposure to $PM_{2.5}$ concentrations from all sources. The incremental contribution from exposure to aromatic hydrocarbons in gasoline using the adjusted results presented here and the Krewski concentration-response function represents approximately 1.4% of the total 130,000 estimated by Fann *et al.* (2012). While this may seem a small fraction of total PM-attributed mortality, these results are substantial as compared to many public health measures, and with the Cross State Air Pollution Rule implementation in the next five years, are likely to constitute a higher portion of PM-related deaths in the future. Under this rule, if $SO_2$ emissions decrease by an expected 50%, sulfate will become a smaller fraction of $PM_{2.5}$; therefore, other sources will become more important, particularly since SOA from aromatic hydrocarbon precursors are not expected to decrease and could represent an increasingly larger fraction of exposures.

In addition to premature mortality, which dominates monetized estimates of total social cost, exposures to SOA from aromatics in gasoline are associated with other health outcomes, including exacerbation of asthma, upper respiratory symptoms, lost work days, and hospital emergency room visits.

A recent study evaluated the public health impacts associated with exposure to direct emissions of $PM_{2.5}$ attributable to congested traffic conditions (Levy et al. 2010) and estimated a total social cost of $31B (in 2007$), comparable to the central estimate of $28.2B developed here. US EPA's Heavy-Duty Highway Diesel Final Rule estimates an

8,300 reduction in premature mortalities (US EPA 2006), a little more than twice the number of premature mortalities from this analysis.

In some areas, particularly urban areas, anthropogenic precursors to SOA, and specifically emissions from gasoline vehicles, are associated with contributions to $PM_{2.5}$ ranging from less than 5% to nearly 50% (Bahreini *et al.* 2012, Cabada *et al.* 2004; Wyche *et al.* 2009; Docherty *et al.* 2008) . A recent study in Los Angeles (Bahreini *et al.* 2012) found that gasoline emissions dominated SOA formation, accounting for nearly 90% of total aerosol formation, and the ratio of SOA to primary organic aerosol was approximately a factor of three. Across most areas in the U.S., SOA represents some 30%-40% of organic carbon concentrations (Yu *et al.* 2007; Pachon *et al.* 2010; Cabada *et al.* 2004; Zhang *et al.* 2009).

Many factors contribute to variability in SOA formation that are not well understood, including spatial and chemical variability in emissions, the amount of time needed for PM formation, and varying ambient conditions at different scales. CMAQ model performance of SOA formation has improved substantially with each version of the model, but likely doesn't capture every process, given that SOA formation depends on varying atmospheric physical and chemical conditions which are simulated at coarser scales in the CMAQ model relative to the (unknown) scales at which they occur in the environment.

The error in aromatic SOA formation is estimated at approximately ±33% (Kleindienst et al. 2007); therefore, measurements are somewhat better understood than the specific processes and conditions leading to those observations. A strength of this analysis is the combination of modeling corroborated by empirical studies to provide a baseline estimate of predicted premature mortality associated with secondary particulate formation. We provide an indication of the magnitude of the resulting public health burden that is more likely to underestimate rather than overestimate potential impacts.The results show that exposure to aromatic hydrocarbons in gasoline and resulting secondary organic aerosol formation demonstrates a non-trivial potential public health impact. As alternatives to aromatics in gasoline are contemplated, it will be important to consider the potential public health impacts associated with different transportation, fuel, and infrastructure design options (see, for example, Cook et al. 2011, who developed a life-cycle assessment approach to evaluate the impacts of increased use of ethanol under several scenarios).

## Competing Interests

The authors declare no competing financial interests. Funding for KvS, JB, and JS was provided by the Harvard Center for Risk Analysis. PVB participated as part of employment with the US EPA.

## Authors' Contributions

KvS wrote the manuscript with oversight from JS, JB conducted the BenMAP modeling using CMAQ results provided by PVB. All authors edited the final manuscript.

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Figure 1: Annual Average $PM_{2.5}$ Concentrations Attributed to Aromatic Emissions from Gasoline Vehicles

Figure 2: Estimated Cases of Premature Mortality Based on the Consensus Expert Elicitation Concentration-Response Function

Figure 3: Incidence and Total Social Cost Associated with Exposure to Aromatic SOA from Gasoline Emissions

Figure 4: Total Social Costs by State Based on Expert Elicitation Concentration-Response Function

**Table 1: Studies Evaluating the Contribution of Aromatic Hydrocarbons to SOA**

| Reference | Description | Source Apportionment | Concentrations (µg/$m^3$) |
|---|---|---|---|
| Kleindienst et al. 2010 | Contribution of primary and secondary sources of OC to $PM_{2.5}$ in Southeastern Aerosol Research and Characterization (SEARCH) network samples | Toluene used as a chemical tracer (2,3-hydroxy-4-oxopentanoic acid) | 0.10 to 0.45 across 4 sampling locations |
| Lewandowski et al. 2008 | Contribution of primary and secondary sources of OC to $PM_{2.5}$ in five midwestern United States cities throughout 2004: East St. Louis, IL Detroit, MI Cincinnati, OH Bondville, IL and Northbrook, IL | Toluene used as a chemical tracer (2,3-hydroxy-4-oxopentanoic acid) | Bondville: 0.09 - 0.25; Northbrook: 0.06 - 0.21; Cincinnati: 0.02 - 0.29; Detroit: 0.07 - 0.33; East St. Louis: 0.06 - 0.26 |
| Offenberg et al. 2011 | Contribution of primary and secondary sources of OC to $PM_{2.5}$ in 2006 in Research Triangle Park, NC over the course of a year | Toluene used as a chemical tracer (2,3-hydroxy-4-oxopentanoic acid) | average = 0.1, stdev = 0.09, min = 0.02, max = 0.36, n=33 |
| Williams et al. 2010 | Positive matrix factorization to determine primary and secondary components of organic aerosol | SOA from motor vehicles contribute 11% of total organic aerosols | concentrations not provided |
| Stone et al. 2009 | Contribution of primary and secondary sources of OC to $PM_{2.5}$ in July-August 2007 in Cleveland, OH, Detroit, MI and LA, CA | Toluene used as a chemical tracer (2,3-hydroxy-4-oxopentanoic acid) | 0.05 - 1.1 in the midwest; 0.95 - 1.61 in CA |

**Table 2: Regression Relationships Developed by Region to Adjust CMAQv5.0 Results Based on Data in Table S1**

| **Overall Estimate and Slope** | | | | |
|---|---|---|---|---|
| | Value | Standard Error | t-value | p-value |
| Intercept | 0.01875 | 0.16 | -0.69 | 0.49 |
| CMAQv5.0 | 1.896 | 2.34 | 1.99 | 0.05 |
| | | | | |
| **Random Effects by Region** | | | | |
| Region | Intercept | CMAQv5.0 | | |
| Midwest/East | 0 | 1.12 | | |
| South | 0 | -0.269 | | |
| West | 0 | -0.856 | | |
| | | | | |
| **Final Equations Used to Adjust Original CMAQv5.0 Results** | | | | |
| Midwest/East | SOA = 0.01875 + 3.016*CMAQv5.0 | | | |
| South | SOA = 0.01875 + 1.627*CMAQv5.0 | | | |
| West | SOA = 0.01875 + 1.04*CMAQv5.0 | | | |
| | | | | |
| **Model based on Eq.1:** | | | | |
| Linear mixed model fit by REML | | | | |
| Formula: SOA ~ CMAQv5.0 + (CMAQv5.0 \| region) | | | | |

0

**Table 3: National Emissions Inventory of Aromatic Hydrocarbons**

| Source | Aromatic VOC (ton/yr) | % of Total |
|---|---|---|
| **Gasoline** | **2,491,313** | **69%** |
| Solvent Usage | 518,334 | 14% |
| Diesel | 25,436 | 1% |
| Other | 573,679 | 16% |
| Total | 3,608,762 | 100% |

Note:
This information was obtained by combining VOC emissions from the National Emissions Inventory with speciation profiles from the SPECIATE database.
See Table S2.

**Table 4: State-wide Annual Average Estimates of $PM_{2.5}$ Attributed to Aromatic SOA from Gasoline Emissions**

| State | Predicted $PM_{2.5}$ Concentration (µg/m$^3$) |
|---|---|
| CT | 0.23 |
| RI | 0.23 |
| OH | 0.21 |
| NY | 0.21 |
| NJ | 0.20 |
| IN | 0.20 |
| MA | 0.19 |
| NH | 0.18 |
| IL | 0.17 |
| PA | 0.17 |
| MO | 0.17 |
| MI | 0.16 |
| SC | 0.16 |
| NC | 0.16 |
| GA | 0.16 |
| VT | 0.15 |
| IA | 0.15 |
| WI | 0.15 |
| ME | 0.14 |
| KY | 0.14 |
| DE | 0.14 |
| TN | 0.14 |
| AL | 0.14 |
| WV | 0.13 |
| VA | 0.13 |
| MS | 0.13 |
| KS | 0.12 |
| DC | 0.12 |
| MD | 0.12 |
| AR | 0.11 |
| MN | 0.11 |

| | |
|---|---|
| NE | 0.11 |
| OK | 0.09 |
| LA | 0.09 |
| SD | 0.09 |
| TX | 0.08 |
| ND | 0.08 |
| FL | 0.08 |
| NV | 0.05 |
| AZ | 0.05 |
| CA | 0.04 |
| ID | 0.04 |
| MT | 0.03 |
| UT | 0.03 |
| WY | 0.03 |
| OR | 0.03 |
| WA | 0.03 |
| NM | 0.03 |
| CO | 0.03 |

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**Table 5: Premature Mortality and Total Social Cost for Health Impacts Associated with Exposure to SOA from Aromatic Hydrocarbons in Gasoline in the Lower 48 States**

| Reference | Beta | Premature Mortality (cases) | Value of Mortality Reduction ($M) |
|---|---|---|---|
| Laden *et al.* 2006 | 0.015 | 4714 (2533, 6897) | $34.9B ($18.7B, $51.0B) |
| Pope *et al.* 2002 | 0.006 | 1833 (717, 2951) | $13.6B ($5.3B, $21.8B) |
| Krewski *et al.* 2009 | 0.006 | 1833 (1335, 2332) | $13.6B ($9.9B, $17.2B) |
| Expert Elicitation | 0.011 | 3816 (886, 6814) | $28.2B ($6.6B, $50.4B) |

Notes:

Value of mortality reduction = $7.4M per case in 2006$

**Table 6: Predicted Premature Mortalities and Associated Social Costs by State (Baseline Year = 2006)**

| State | Population (2006) | Premature Mortality based on Expert Elicitation (cases) | Total Social Cost based on Expert Elicitation ($M) | Premature Mortality based on Krewski et al. 2009 (cases) | Total Social Cost based on Krewski et al. 2009 ($M) | Premature Mortality based on Pope et al. 2002 (cases) | Total Social Cost based on Pope et al. 2002 ($M) | Premature Mortality based on Laden et al. 2006 (cases) | Total Social Cost based on Laden et al. 2006 ($M) |
|---|---|---|---|---|---|---|---|---|---|
| NY | 11,721,250 | 359 | $ 2,659 | 173 | $ 1,277 | 173 | $ 1,277 | 443 | $ 3,278 |
| OH | 7,027,236 | 266 | $ 1,972 | 128 | $ 947 | 128 | $ 947 | 329 | $ 2,433 |
| PA | 7,856,478 | 263 | $ 1,943 | 126 | $ 933 | 126 | $ 933 | 324 | $ 2,395 |
| IL | 7,826,777 | 215 | $ 1,592 | 103 | $ 765 | 103 | $ 765 | 266 | $ 1,966 |
| NJ | 6,003,804 | 189 | $ 1,402 | 91 | $ 673 | 91 | $ 673 | 234 | $ 1,730 |
| FL | 12,353,717 | 173 | $ 1,279 | 83 | $ 615 | 83 | $ 615 | 214 | $ 1,581 |
| MI | 6,269,921 | 169 | $ 1,254 | 81 | $ 602 | 81 | $ 602 | 209 | $ 1,549 |
| TX | 13,969,855 | 166 | $ 1,231 | 80 | $ 592 | 80 | $ 592 | 206 | $ 1,526 |
| NC | 5,523,143 | 147 | $ 1,090 | 71 | $ 524 | 71 | $ 524 | 182 | $ 1,347 |
| CA | 22,483,409 | 133 | $ 988 | 64 | $ 475 | 64 | $ 475 | 165 | $ 1,221 |
| GA | 5,572,237 | 133 | $ 986 | 64 | $ 474 | 64 | $ 474 | 165 | $ 1,221 |
| IN | 3,915,380 | 131 | $ 968 | 63 | $ 465 | 63 | $ 465 | 162 | $ 1,196 |
| MA | 4,049,798 | 125 | $ 922 | 60 | $ 443 | 60 | $ 443 | 153 | $ 1,136 |
| MO | 3,608,441 | 106 | $ 787 | 51 | $ 378 | 51 | $ 378 | 131 | $ 972 |
| VA | 4,873,441 | 102 | $ 754 | 49 | $ 362 | 49 | $ 362 | 126 | $ 931 |
| TN | 3,822,406 | 99 | $ 729 | 47 | $ 350 | 47 | $ 350 | 122 | $ 902 |

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| WI | 3,619,422 | 84 | $ 624 | 40 | $ 300 | 40 | $ 300 | 104 | $ 769 |
| CT | 2,253,322 | 83 | $ 617 | 40 | $ 296 | 40 | $ 296 | 103 | $ 761 |
| SC | 2,772,416 | 82 | $ 605 | 39 | $ 291 | 39 | $ 291 | 101 | $ 749 |
| AL | 2,927,474 | 81 | $ 599 | 39 | $ 288 | 39 | $ 288 | 100 | $ 742 |
| KY | 2,675,868 | 69 | $ 507 | 33 | $ 244 | 33 | $ 244 | 85 | $ 628 |
| MD | 3,715,953 | 67 | $ 493 | 32 | $ 237 | 32 | $ 237 | 82 | $ 610 |
| MN | 3,314,038 | 54 | $ 397 | 26 | $ 191 | 26 | $ 191 | 66 | $ 490 |
| IA | 1,906,272 | 48 | $ 353 | 23 | $ 170 | 23 | $ 170 | 59 | $ 435 |
| MS | 1,846,049 | 45 | $ 335 | 22 | $ 161 | 22 | $ 161 | 56 | $ 416 |
| LA | 2,630,768 | 43 | $ 315 | 20 | $ 151 | 20 | $ 151 | 53 | $ 391 |
| AR | 1,803,802 | 40 | $ 296 | 19 | $ 142 | 19 | $ 142 | 49 | $ 366 |
| OK | 2,233,442 | 38 | $ 283 | 18 | $ 136 | 18 | $ 136 | 47 | $ 350 |
| KS | 1,680,031 | 35 | $ 257 | 17 | $ 123 | 17 | $ 123 | 43 | $ 317 |
| WV | 1,187,545 | 33 | $ 242 | 16 | $ 116 | 16 | $ 116 | 40 | $ 298 |
| AZ | 3,911,781 | 29 | $ 217 | 14 | $ 104 | 14 | $ 104 | 36 | $ 269 |
| NH | 952,282 | 26 | $ 190 | 12 | $ 91 | 12 | $ 91 | 32 | $ 234 |
| RI | 653,356 | 25 | $ 187 | 12 | $ 90 | 12 | $ 90 | 31 | $ 230 |
| ME | 904,612 | 23 | $ 169 | 11 | $ 81 | 11 | $ 81 | 28 | $ 208 |
| NE | 1,058,917 | 19 | $ 140 | 9 | $ 67 | 9 | $ 67 | 23 | $ 172 |
| WA | 4,138,920 | 18 | $ 135 | 9 | $ 65 | 9 | $ 65 | 22 | $ 166 |
| NV | 1,603,777 | 14 | $ 101 | 7 | $ 49 | 7 | $ 49 | 17 | $ 125 |
| DE | 522,705 | 13 | $ 95 | 6 | $ 45 | 6 | $ 45 | 16 | $ 117 |
| OR | 2,383,414 | 12 | $ 90 | 6 | $ 43 | 6 | $ 43 | 15 | $ 111 |
| VT | 436,489 | 10 | $ 77 | 5 | $ 37 | 5 | $ 37 | 13 | $ 95 |
| CO | 2,974,597 | 9 | $ 70 | 5 | $ 34 | 5 | $ 34 | 12 | $ 87 |
| SD | 473,989 | 7 | $ 53 | 3 | $ 25 | 3 | $ 25 | 9 | $ 65 |

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| NM | 1,300,700 | 6 | $ 47 | 3 | $ 22 | 3 | $ 22 | 8 | $ 58 |
| UT | 1,399,252 | 6 | $ 45 | 3 | $ 22 | 3 | $ 22 | 8 | $ 56 |
| ND | 413,558 | 5 | $ 41 | 3 | $ 19 | 3 | $ 19 | 7 | $ 50 |
| ID | 907,667 | 5 | $ 38 | 2 | $ 18 | 2 | $ 18 | 6 | $ 47 |
| MT | 639,955 | 4 | $ 29 | 2 | $ 14 | 2 | $ 14 | 5 | $ 35 |
| DC | 217,088 | 4 | $ 28 | 2 | $ 14 | 2 | $ 14 | 5 | $ 35 |
| WY | 347,896 | 2 | $ 13 | 1 | $ 6 | 1 | $ 6 | 2 | $ 16 |

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Table S1: Comparison of Observed SOA Measurements and Unadjusted CMAQv5.0 Predictions

Table S2: US EPA's SPECIATE Database Used to Determine the Fraction of Anthropogenic SOA from Aromatic Hydrocarbons in Gasoline