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# Public Health Costs of Primary $PM_{2.5}$ and Inorganic $PM_{2.5}$ Precursor Emissions in the United States

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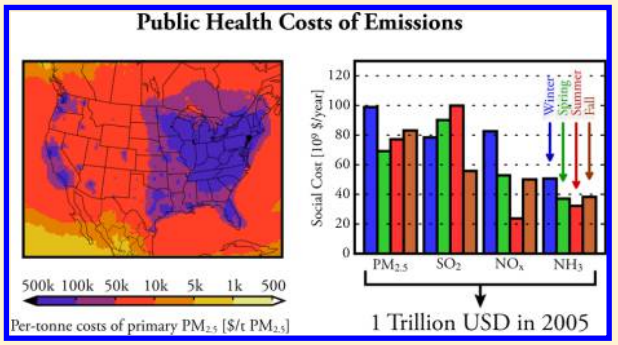
Supporting Information

**ABSTRACT:** Current methods of estimating the public health effects of emissions are computationally too expensive or do not fully address complex atmospheric processes, frequently limiting their applications to policy research. Using a reduced-form model derived from tagged chemical transport model (CTM) simulations, we present $PM_{2.5}$ mortality costs per tonne of inorganic air pollutants with the 36 km × 36 km spatial resolution of source location in the United States, providing the most comprehensive set of such estimates comparable to CTM-based estimates. Our estimates vary by 2 orders of magnitude. Emission-weighted seasonal averages were estimated at \$88,000−130,000/t $PM_{2.5}$ (inert primary), \$14,000−24,000/t $SO_2$, \$3,800−14,000/t $NO_x$, and \$23,000−66,000/t $NH_3$. The aggregate social costs for year 2005 emissions were estimated at \$1.0 trillion dollars. Compared to other studies, our estimates have similar magnitudes and spatial distributions for primary $PM_{2.5}$ but substantially different spatial patterns for precursor species where secondary chemistry is important. For example, differences of more than a factor of 10 were found in many areas of Texas, New Mexico, and New England states for $NO_x$ and of California, Texas, and Maine for $NH_3$. Our method allows for updates as emissions inventories and CTMs improve, enhancing the potential to link policy research to up-to-date atmospheric science.




## INTRODUCTION

Estimating the social costs of air pollution, i.e. the damages imposed on human health and the natural environment, plays an important role in policy research, such as benefit-cost analyses of air quality laws[1,2] and regulations;[3,4] externality analyses of energy and transportation systems;[5−9] and climate and energy technology assessments.[10−13] $PM_{2.5}$, particulate matter having a diameter of 2.5 $\mu$m or less, is especially important because $PM_{2.5}$ is strongly associated with premature mortality[14−17] and accounts for more than 90% of the monetized social costs.[1,2,18] $PM_{2.5}$ is a complex mixture of various chemical compounds, sizes, and shapes. There are active research efforts to understand where $PM_{2.5}$ toxicity comes from.[19−21] Nevertheless, $PM_{2.5}$ is currently regulated on a mass concentration basis by the U.S. EPA[22] because other metrics do not have sufficient epidemiological and toxicological evidence to support regulatory rule-making.[23]

A standard method of estimating the social cost of emissions is using an impact pathway analysis.[1,24,25] First, an air quality model estimates changes in $PM_{2.5}$ concentrations by comparing a simulation with baseline emissions and another with perturbed emissions. Next, changes in mortality rate are estimated using concentration−response relations reported by epidemiological studies. Then, the number of premature deaths is estimated by applying the changed mortality rates to exposed population. U.S. EPA usually publishes two separate mortality estimates using two landmark series of epidemiological studies, one from the American Cancer Society (ACS) studies[14] and the other from the Harvard Six Cities (H6C) studies[15] due to pros and cons of the two series.[1,26] Finally, the premature deaths are monetized with the value of statistical life (VSL), or people's willingness-to-pay to avoid the mortality risk. U.S. EPA recommends[25] using 8 M USD (in 2010 USD) for VSL after adjusting it for income growth.

Marginal social cost (\$/t), the social cost per tonne of air pollutant emitted, is a useful metric because policy research often deals with "marginal" changes in emissions and the social cost of emissions can be conveniently estimated by multiplying the amount of emissions by the marginal social cost. However, one should be cautious when applying such estimates outside of designed marginal ranges because nonlinear chemistry may result in substantial errors.[27] Intake fraction (ppm) is another impact metric,[28] defined as the mass fraction of the air pollutant or its precursors emitted that are inhaled by an exposed

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**Table 1. Tools for Estimating the Social Costs of Air Pollutant Emissions**

| | | reduced-form models derived from | | |
|---|---|---|---|---|
| | chemical transport models | dispersion models (e.g., CRDM) | chemical transport models | EASIUR |
| air quality modeling | state-of-the-art | simplified | similar to state-of-the-art | similar to state-of-the-art |
| computational cost | very high | low | low | low |
| spatial resolution | detailed, flexible (typically 4−36 km) | county-level | subject to underlying CTM simulations; typically, limited | 36 km resolution; similar to or finer than county-level |
| temporal resolution | detailed, flexible (typically ≤15 min) | annual | subject to underlying CTM simulations; typically, annual | seasonal |
| user interface | FORTRAN/UNIX environment, additional work needed for exposure/valuation analysis | graphical user interface, county-based lookup tables, regression equations | regression equations, lookup tables | spatial lookup tables |
| examples | CAMx (ENVIRON, 2012), CMAQ (Byun and Schere, 2006), WRF/Chem (Grell et al., 2005), GATOR-GCMOM (Jacobson, 2001) | COBRA (U.S. EPA, 2011), APEEP/AP2 (Muller and Mendelsohn, 2007; Muller, 2011), Levy et al. (2009) | RSM (U.S. EPA, 2006), Fann et al. (2009), Fann et al. (2012), Buonocore et al. (2014) | |

One of the major challenges in estimating social costs is to address properly the complex atmospheric processes of chemical reactions, transport, and removal associated with $PM_{2.5}$ and precursor gases, which determine population exposures over hundreds or more from an emissions source. Primary $PM_{2.5}$, species directly emitted as inert particulate matter such as elemental carbon (EC) and fugitive dust, poses a substantial public health burden. However, secondary $PM_{2.5}$, species produced through photochemical processes in the atmosphere from gaseous precursors, usually accounts for a dominant (>80%) fraction of ambient $PM_{2.5}$.[29,30] Inorganic gases—sulfur dioxide ($SO_2$), nitrogen oxides ($NO_x$), and ammonia ($NH_3$)—are important precursors that form $PM_{2.5}$ by well-understood but nonlinear thermodynamic interactions. For example, even when sulfate ($SO_4^{2-}$) is a major component of $PM_{2.5}$, $SO_2$ reductions may have little impact on $PM_{2.5}$ due to a sulfate-nitrate substitution phenomenon, a scenario common enough to have regulatory significance.[27,31,32] Certain volatile organic compounds (VOCs) are also major precursors of organic $PM_{2.5}$, the understanding of which is relatively poor but has substantially advanced in recent years.[30,33]

Current tools for linking changes in emissions to ambient $PM_{2.5}$ levels and, therefore, health effects often find limited utility in policy research because they are either computationally too expensive, too simplified, or too specific. Table 1 summarizes current methods. One is to use a state-of-the-science chemical transport model (CTM),[34−37] which divides the atmosphere into a three-dimensional grid and calculates complex atmospheric processes in each box at a high time resolution (typically, ∼15 min). Built in order to advance the understanding of atmospheric science as well as to inform air quality policy-making, CTMs are the most sophisticated tools to simulate air quality. Once CTMs estimate pollutant concentrations, software tools such as BenMAP[38] assist with population exposure estimation and valuation. However, running CTMs is computationally too expensive for many applications. Therefore, CTMs are usually run only for limited numbers of emissions scenarios and remain virtually out of reach to a large research community that wants to explore many policy and technological scenarios, perform rigorous uncertainty analyses (e.g., Monte Carlo), and so on.

To overcome the limitations of CTMs, reduced form models[5,39−41] have been built using dispersion models such as the Climatological Regional Dispersion Model (CRDM).[42] CRDM translates emissions from one source county to $PM_{2.5}$ concentrations at each downwind (or receptor) county using annual and seasonal average meteorology and emissions. Using CRDM, the Air Pollution Emissions Experiments and Policy (APEEP) model[41] and an updated version AP2[43] provide per-ton social costs of six major pollutants for all (about 3100) U.S. counties. It would require about 6000 CPU-y to generate comparable spatial detail with a CTM using brute force approaches (assuming 500 min per a simulation day[34]). However, CRDM's Gaussian dispersion approach[44] may have limitations for predicting the dispersion of long-range transport pollutants like $PM_{2.5}$ and precursors because it assumes conditions at the point of emissions are held for all downwind locations. CRDM also relies on simple assumptions for secondary $PM_{2.5}$ formations, which is complex and sensitive to meteorological conditions.[27,31] For example, in order to estimate the formation of ammonium nitrate $PM_{2.5}$ ($NH_4NO_3$), it calculates potential maximum amounts with a stoichiometric mass balance among related inorganic species and then simply divides the amounts by four assuming it does not form in warm periods.[42] Moreover, CRDM's prediction[45] of organic $PM_{2.5}$ is out-of-date as recent research has shown that there is much more secondary organic $PM_{2.5}$ than previously thought and that there are neglected-but-highly potent organic $PM_{2.5}$ precursors.[33,46] Moreover, because the size of county varies substantially across the nation, CDRM's county-based spatial resolution may be spatially inconsistent and too coarse in some cases.

Another group is reduced form models[47−49] built using CTMs. Although their predictions should be comparable to their parent CTMs, their parent CTM's high computational costs have typically limited the results in terms of resolution in source sectors and source locations. For example, U.S. EPA's Response Surface Model (RSM)[48] is such a model built using a multidimensional kriging method based on CMAQ[35] outputs. The estimates derived from RSM[8] are limited to 9 urban areas and 1 nationwide average and to 12 emission sectors. Another set of per-ton damages estimated by U.S. EPA[50] using a CTM with tagging is limited to national averages for 17 emission sectors.

In order to overcome the limitations of current reduced-form models, we recently developed a method that provides county-scale impact metrics similar to CRDM or APEEP but derived from a state-of-the-art CTM. Built from tagged CTM simulations and generalized via regressions, this method, called the Estimating Air quality Social Impacts Using Regression (EASIUR) model,[51] predicts marginal social costs and intake fractions accurately like a CTM but at trivial computational


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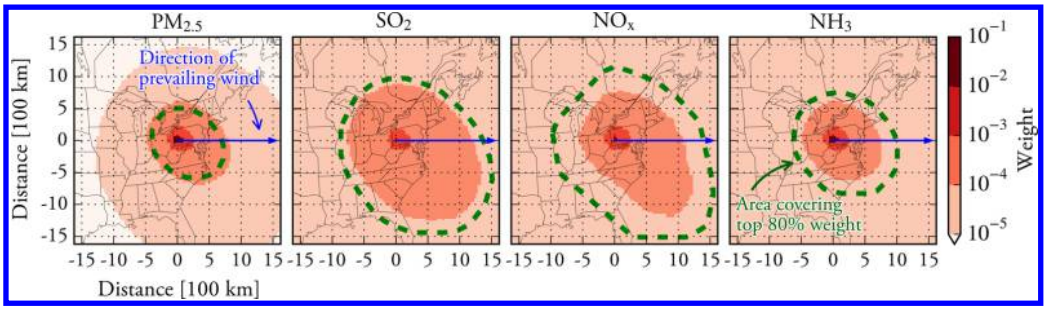


**Figure 1.** Average plume weights of the four species. Each average plume was averaged from four seasons. Dashed lines indicate the region encompassing 80% of the weights. To illustrate a sense of scale, they are placed on Pittsburgh, Pennsylvania. All the plumes are skewed to the right from the aligned wind direction, which is caused by the Coriolis effect from the rotation of the Earth.

costs. In this paper, we present marginal and aggregate public health costs estimated by EASIUR, compare to other estimates, and discuss uncertainties and their implications.

## METHOD

**Reduced-Form Model Built From Tagged CTM Simulation and Regression.** The EASIUR model is summarized here; a full description can be found elsewhere.[51] First, we select two sets of 50 random locations, one for model building and the other for out-of-sample evaluations, in a 148 × 112 grid covering the contiguous United States. Next, we run a CTM, Comprehensive Air Quality Model with extensions (CAMx) version 5.41,[34] with 2005 emissions and meteorological inputs developed and evaluated for a regulatory impact analysis.[52,53] Then, marginal social costs and intake fractions for emissions at the 100 source locations were calculated by a standard method used by U.S. EPA,[25,54] using a concentration−response relation from a recent ACS study,[14] $8.6 M (in 2010 USD) for VSL (U.S. EPA's recommended value[25] adjusted to 2005 income level), and population and baseline mortality in 2005 derived from BenMAP.[55] Although we based the chosen concentration−response relation, VSL, and population, we provide an easy method[51] to adjust our estimates for these factors over reasonable ranges. Finally, we derived regression models with one set of 50 locations and did out-of-sample evaluations against the other set of 50 locations. The models were built for three different emission heights: ground-level, 150 m, and 300 m. Regression models parametrize marginal social cost or intake fraction with population and atmospheric variables such as temperature and atmospheric pressure.

We developed an "average plume method" to address the challenge of describing population exposure to $PM_{2.5}$ occurring in downwind areas over hundreds of kilometers. An average plume is a spatial distribution of downwind impacts empirically derived by normalizing averaged $PM_{2.5}$ concentrations predicted by CAMx for our 50 training locations. Exposed population for any given emissions species and source location is expressed in regressions by convoluting the surrounding map of population with the spatial distribution of this average plume, essentially producing a population estimate weighted by proximity to the pollution source.

**Marginal and Aggregate Public Health Effects of Primary $PM_{2.5}$, $SO_2$, $NO_x$, and $NH_3$.** We estimated marginal social costs and intake fractions at every cell of the 148 × 112 grid using the EASIUR models. In our analysis, primary $PM_{2.5}$ indicates only inert primary species (mainly, elemental carbon and fugitive dust) but not primary organic $PM_{2.5}$, which is not inert although it has been treated inert in most CTMs until recently.[33]

In order to explore the magnitude of air quality public health burden of the United States, we estimated social costs for 11 emissions categories, using 2005 emissions inventory[53] (Emissions and detailed definitions of sectors are summarized in Table S1): EGU (electric generating units); non-EGU (non-EGU point sources and aircraft emissions); on-road (on-road gasoline and diesel vehicles); other transportation (locomotive, and marine vessel); nonroad (nonroad engines); foreign (Mexico and Canada); area emissions (relatively small sources that are not categorized as EGU or non-EGU); fugitive dust; agriculture; fire (wild and prescribed fire); and biogenic emissions. Height-specific marginal costs, which were interpolated from EASIUR's three elevations, were used for elevated point emissions. Note that the size of emissions used in estimating aggregate social costs is beyond the range where we tested to find our marginal social costs stay constant[51] and, therefore, the estimates may result in some bias for secondary species. However, they provide useful first-order estimates and the associated uncertainties would be less than a factor of 2 (see the Discussion).

## RESULTS

**Average Plumes.** Here we present the average plumes that represent a generic distribution of downwind impacts. Although these are primarily an intermediate step in the EASIUR method for estimating social costs, it is informative for users to be able to visualize these distributions; many users are surprised at the long-range impacts of $PM_{2.5}$ emissions and precursor gases. These plumes describe the expected spatial features of an air pollutant's dispersion and chemical conversion. To provide a sense of scale, the average plumes are plotted in Figure 1 on a map as if the emissions were originating from Pittsburgh and prevailing winds were west to east. The weights indicate the relative amount of the ambient $PM_{2.5}$ mass created by emissions in downwind grid cells; the sum across all grid cells is one. The impacts of all four species are highly regional, easily spanning more than 1000 km; the dashed lines encompass the region that accounts for 80% of the resulting $PM_{2.5}$. $SO_2$ and $NO_x$ emissions lead to more widely distributed impacts than primary $PM_{2.5}$ because they must be oxidized before forming $PM_{2.5}$, a process with a time scale on the order of hours or days. $NH_3$ emissions also require chemical transformation to form $PM_{2.5}$ but do so faster than $SO_2$ or $NO_x$; therefore, their impacts are distributed only somewhat more broadly than primary $PM_{2.5}$. Consistent with findings from other studies,[56,57] these spatial distributions provide partial justification for EASIUR's 36 km ×


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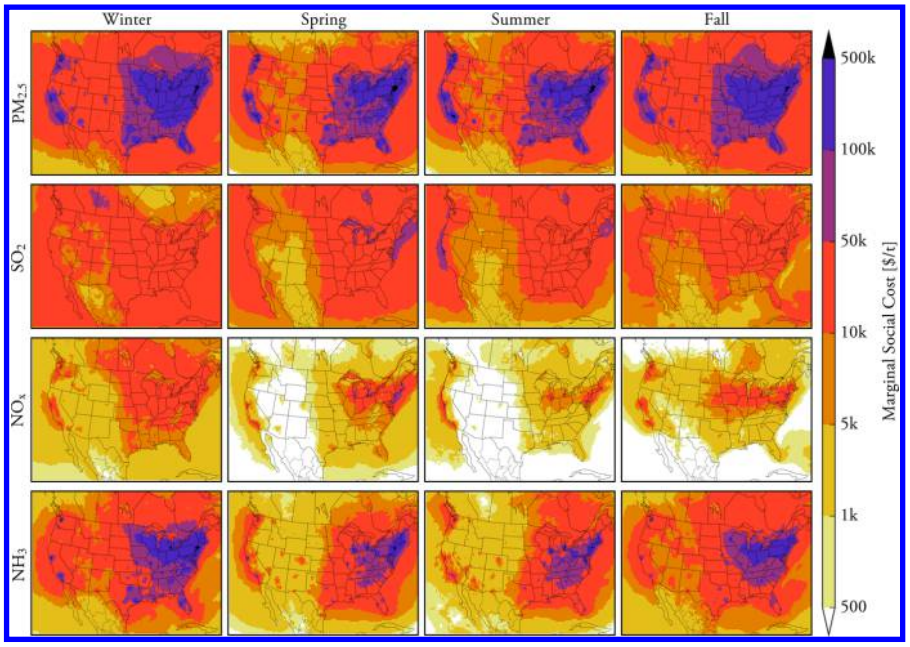


**Figure 2.** Marginal social costs for ground-level emissions of $PM_{2.5}$, $NO_x$, $SO_2$, and $NH_3$. $PM_{2.5}$ represents inert primary $PM_{2.5}$ species. Each point presents the sum of public health costs that would be imposed at all downwind areas if marginal emissions were made at the point. Marginal social costs are based on the relative risk of $PM_{2.5}$ from the work of Krewski et al.,[14] which is a 6% mortality increase per a 10 $\mu g/m^3$ increase in $PM_{2.5}$, and \$8.6 M (in 2010 USD) for the value of a statistical life. Winter is defined as January−March, spring, as April−June, summer, as July−September, and fall, as October−December.

36 km spatial resolution because a substantial fraction of created ambient $PM_{2.5}$ travels long distances in all cases. This is also consistent with observations showing that long transport secondary $PM_{2.5}$ usually dominates air quality burden in urban areas.[29,30]

However, an average plume does not directly translate into a public health burden because it does not account for population exposed to the plume, which highly depends on the location of emissions source. A product of an average plume placed at a certain location and population under the plume would describe a distribution of relative public health burden. For example, when such population-weighted average plumes are placed in New York City, the source grid cell accounts for 65% and 39% for primary $PM_{2.5}$ and $SO_2$, respectively, indicating the potential value of increasing spatial resolution for highly populated areas.

**Marginal Social Costs and Intake Fractions.** Marginal social costs and intake fractions estimated by the EASIUR models are presented in Figures 2 and S1, respectively. Seasonal-average estimates, weighted by the amount of emissions in the 2005 emissions inventory, are presented in Figures 3a and S2a, which are \$88,000−130,000/t $PM_{2.5}$, \$14,000−24,000/t $SO_2$, \$3,800−14,000/t $NO_x$, and \$23,000−66,000/t $NH_3$. The average intake fractions are 1.2−1.7 ppm for $PM_{2.5}$, 0.18−0.32 ppm for $SO_2$, 0.05−0.17 ppm for $NO_x$, and 0.31−0.85 ppm for $NH_3$.

The maps show that spatial variability largely depends on the population of the surrounding region. The effects of $SO_2$ and $NO_x$ emissions show less spatial variability than primary $PM_{2.5}$ and $NH_3$ because, having impacts that span broader regions, they effectively average out more of the variability in population. $NH_3$ and primary $PM_{2.5}$ impacts, in contrast, more closely reflect smaller-scale variations in population density. Seasonal variability is also distinct species by species. Wintertime emissions generally have higher values because air pollutants are less diluted vertically due to cold temperature. Wintertime marginal effects are especially large for $NO_x$ and $NH_3$ because $NO_x$ and $NH_3$ form ammonium nitrate $PM_{2.5}$ more readily under cold temperatures. Though $SO_2$ also shows higher values in cold seasons in some areas, it generally shows higher values in summer, since oxidation of $SO_2$ to sulfate $PM_{2.5}$ is controlled by sunlight-driven photochemistry.

The rank ordering of the social costs and intake fractions directly corresponds to the efficiency with which an emitted species forms $PM_{2.5}$. Recall that current epidemiology and valuation do not distinguish some $PM_{2.5}$ components as more toxic than others. Primary $PM_{2.5}$ emissions, by definition, directly enhance $PM_{2.5}$ concentrations and, therefore, have the highest social costs. In contrast, $SO_2$, $NO_x$, and $NH_3$ are all $PM_{2.5}$ precursors and must undergo one or more steps before forming $PM_{2.5}$. Therefore, the efficiency with which they form $PM_{2.5}$ depends on the fraction of $SO_2$ and $NO_x$ that oxidizes to sulfuric acid and nitric acid, respectively. For $NO_x$ and $NH_3$, thermodynamics also plays a role as one is generally limiting for ammonium nitrate formation. Lastly, because ammonia has a low molecular weight and is often the limiting factor for ammonium nitrate formation,[27,31,58,59] the social cost expressed on a per ton basis is high.

Figure S3 presents the comparisons among three emissions heights: ground-level, 150 m high, and 300 m high. Generally, if emissions are released at a higher elevation nearby densely populated areas, marginal social costs become lower, which indicates that air pollutants impact nearby population areas less strongly. However, marginal social costs are not much lower in sparsely populated areas; in some remote locations, elevated emissions have higher social costs than ground-level when elevation enhances the transport of the pollutant to distant urban areas. Because the lifetime of $PM_{2.5}$ (several days) is longer than the vertical mixing time scale for the planetary boundary layer (~1 day), vertical differences would generally


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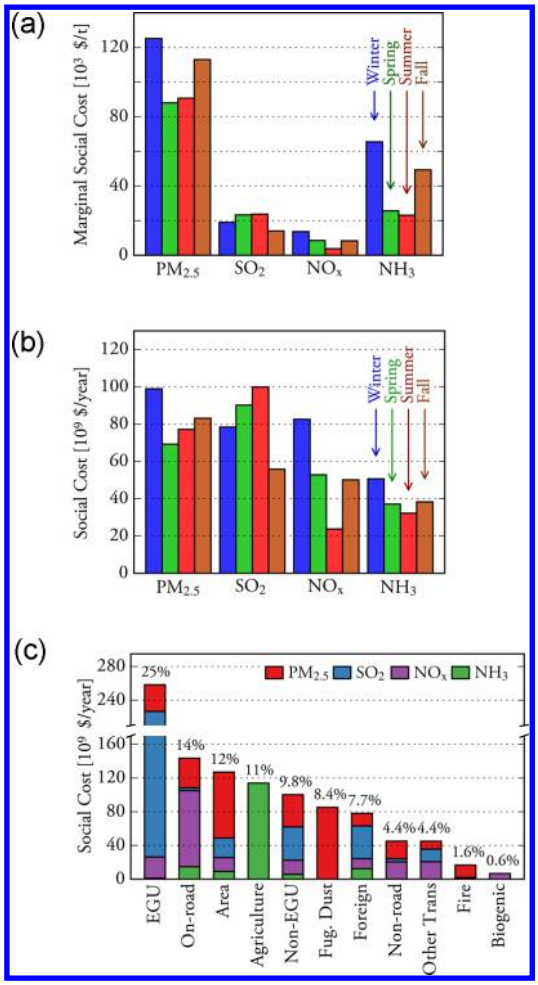


**Figure 3.** Marginal and aggregate social costs of inorganic air pollutants in 2005. $PM_{2.5}$ consists of inert primary $PM_{2.5}$ species (mainly, fugitive dust and elemental carbon), which does not include organic species.

smooth out relatively quickly on the regional scales under consideration, resulting in social costs, especially for $SO_2$ and $NO_x$, that are not very strong functions of emissions height.

**National Air Quality Burden.** Seasonal aggregate social costs by species are presented in Figure 3b. Aggregate social costs were $330 billion for primary $PM_{2.5}$ (not including primary organic PM), $320 billion for $SO_2$, $210 billion for $NO_x$, and $160 billion for $NH_3$. Intake fractions are presented in Figure S2b. Although wintertime $NH_3$ emissions are much smaller than in other seasons (agriculture is the dominant source of $NH_3$), wintertime social costs are the largest due to higher wintertime sensitivity of $PM_{2.5}$. The sum of the social costs of the four species is $1.0 trillion.

Sectoral social costs are presented in Figure 3c and broken down by species. The top 81% comes from six sectors: 25% from EGU, 14% from on-road, 12% from area sources, 11% from agricultural ammonia, 9.8% from non-EGU, and 8.4% from fugitive dust. Emissions from Mexico and Canada represent 7.7% of the total.

**Comparison to Other Studies.** EASIUR estimates are compared to those from three other studies in Figure 4 (numeric values are presented in Tables S3–S5). In order to isolate differences to air quality modeling, social cost estimates were harmonized to the best of our knowledge to account for differences in factors such as VSL, concentration–response relation, emissions unit (metric ton), population year, and $PM_{2.5}$ lag effects using methods described elsewhere.[51] Intake fractions are compared in Figure S4 with recommended values from Humbert et al.[60]

In Figures 4a and b, EASIUR is compared with AP2,[43] an updated version of APEEP,[40,61] county-by-county for ground-level emissions. We chose AP2 reported using 2005 emissions and population. Averaged over many locations, there is little net bias between EASIUR and AP2 for $SO_2$ and $NO_x$, but we frequently predict higher impacts stemming from primary $PM_{2.5}$ and ammonia. The high Pearson correlation coefficient (0.81) for primary $PM_{2.5}$ suggests that CRDM on which AP2 is based predicts net exposures that are comparable to a full CTM when averaged over long time periods and all downwind locations. However, comparisons for secondary species show much lower correlations, when the subsequent atmospheric chemistry is more complex. For $NO_x$, which is affected by both gas-phase oxidation and thermodynamic phase partitioning, the correlation between EASIUR and AP2 results is quite low, only 0.085, indicating very little agreement between the two models as to which counties emissions tend to result in higher impacts. Spatial comparisons presented in Figure 4b show that the differences between EASIUR and AP2 are not randomly distributed but have differing patterns in different regions. For example, AP2 estimates social costs for $NO_x$ emissions that are an order of magnitude lower than EASIUR in throughout the New England states.

Figure 4c presents similar comparisons between EASIUR and the work of Fann et al.[50] Running CAMx with the same emissions and meteorology as EASIUR, they reported per-ton estimates for 17 sectoral emissions for three species (primary $PM_{2.5}$, $SO_2$, and $NO_x$). Due to limited sectoral resolution in our emissions inventory, we regrouped their 17 sectors into 7 sectors: 5 sectors have the same definitions; our "area sources" include residential wood combustion additionally; and "other sources" include 11 industrial sectors. Emissions-weighted averages are compared in Figure 4c, showing our 11 sectoral-average estimates are mostly within a factor of 2. EASIUR's estimates were about 20% smaller for $PM_{2.5}$. For $SO_2$, EASIUR produced 30% smaller on average but less variability among sectors. For $NO_x$, the most difficult species, EASIUR is generally about 2 times larger but up to 3 times for nonroad mobile sources.

Lastly, in Figure 4d, EASIUR is compared to marginal social costs for one nationwide and nine urban areas from U.S. EPA's RSM.[48,62] Although RSM was derived running CMAQ with 2015 emissions projected from the 2001 National Emissions Inventory and 2001 meteorology, the two methods show similar national average estimates except for $PM_{2.5}$: for $PM_{2.5}$, RSM's estimate is 2–3 times larger than EASIUR's average; for $SO_2$, −10% to 90%; for $NO_x$, −48% to −20%; and for NH3, −52% to 24%. However, individual urban areas generally show large differences for all species. RSM-based estimates for some specific urban areas seem counterintuitive. For example, the RSM estimate is 4.5 times larger for area source carbon and 11 times for industrial $SO_2$ in Phoenix than in the combined New York and Philadelphia (NY/Phi) area, although NY/Phi has 6 times larger population than Phoenix. This may be related with the limitation of RSM design that cannot completely separate the effects of emissions from one area on others.[48]

**Uncertainties in EASIUR Estimates.** There are three major sources of uncertainty associated with EASIUR: air


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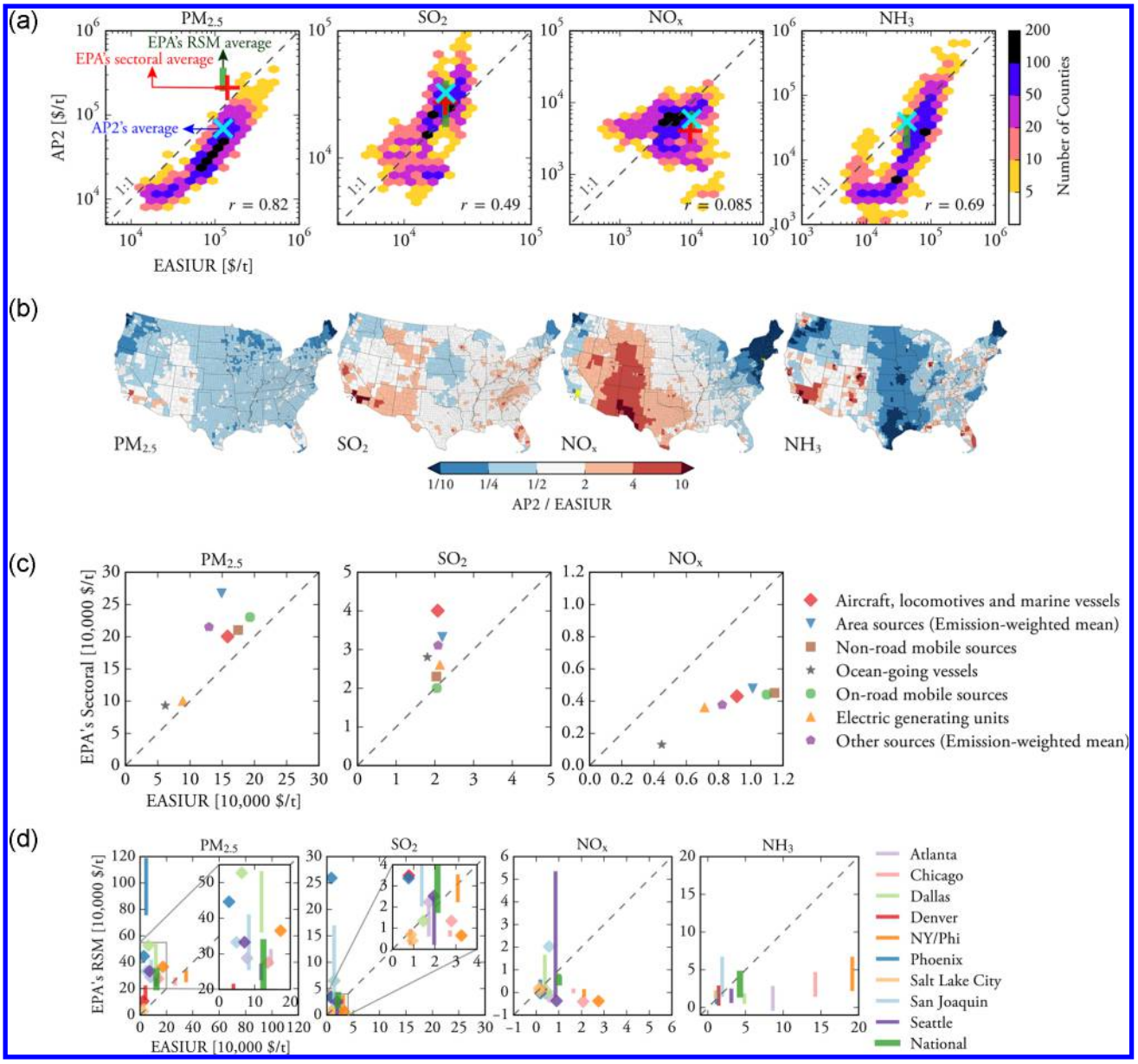


**Figure 4.** Comparisons of EASIUR to other studies. Several factors such as VSL, concentration−response relation, and emissions unit (metric ton), population year, and $PM_{2.5}$ lagged effects were adjusted for all estimates the same as EASIUR to isolate the role of air quality modeling. (a) Comparison of EASIUR to AP2,[43] U.S. EPA's sectoral estimates,[50] and U.S. EPA's Response Surface Model (RSM).[62] Since AP2 provides county-based estimates, EASIUR and AP2 are paired for every county for ground-level emissions. EPA's RSM estimates (ranges cover area and mobile sources), EPA's sectoral averages, and AP2's averages are compared to EASIUR's averages (weighted by emissions in U.S. land area). EPA's sectoral averages and AP2's averages are emissions-weighted averages. The Pearson correlation coefficients ($r$) between EASIUR and AP2 are presented on the bottom right. (b) Spatial comparison to AP2.[43] Estimates for ground-level emissions are compared. Yellow areas indicate where AP2 reported negative values. (c) Comparisons to estimates for industrial, area, and mobile sectors derived from U.S. EPA's tagged chemical transport model simulations.[50] (d) Comparisons to estimates for one national and nine urban areas derived using U.S. EPA's RSM.[62] EASIUR's ground-level estimates were compared to area and mobile sources and EASIUR's 150 m elevation estimates to EGU and non-EGU sources. Ranges are used if Fann et al.[62] provided two estimates for area and mobile sources, and diamond markers are used if they provide one estimate. EASIUR's emissions-weighted national averages are compared to RSM's national estimates.

quality modeling, concentration−response relations, and VSL. We present the uncertainties in Table S6, showing that air quality modeling introduces generally similar or smaller uncertainties than the other two factors, but it is larger for some cases. The uncertainties are quantified and discussed as follows.

Air quality modeling requires a detailed discussion about two sources of uncertainty: errors in CAMx compared to reality and errors in the regressions compared to CAMx. Although CTM's uncertainties are largely associated with key inputs such as emissions and meteorology and less significantly with atmospheric algorithms (at least for inorganic $PM_{2.5}$),[63,64] it is not feasible[65] to quantify formally uncertainties surrounding those factors due to measurement coverage and the sheer number of parameters in CTMs. Instead, CTMs are generally evaluated against ambient measurements, which are also uncertain due to factors associated with measurement accuracy and frequency. U.S. EPA's evaluation[52] reports fractional biases


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and fractional errors, common evaluation metrics for CTMs,[65,66] that are averaged over regional areas (of 5−10 states) on a seasonal basis. CTMs may have less biases and errors for predicting $PM_{2.5}$ changes from "additional" emissions than predicting total $PM_{2.5}$ concentrations from all emissions.[67,68]

Turning to the statistical regressions used by EASIUR, we report both confidence intervals (CIs) and prediction intervals (PIs) from the fitting procedures.[51] Since CIs quantify errors in the social cost when averaged over many locations, they are appropriate for applications that are regional to national in scale (e.g., covering 5−10 states). On the other hand, some applications will require an uncertainty estimate for damages from emissions in one specific location (or cluster of nearby locations such as a metropolitan statistical area), which are better represented by PIs.

Lacking a formal method, we suggested the uncertainties in Table S6 based on our best judgment. Assuming the two sources of uncertainty are independent, we combined them. For regional-scale applications (covering emissions sources in four or more states, or in areas covering 500 000 $km^2$ or larger), we averaged the absolute values of fractional biases (weighted by the number of observations) reported in the CAMx evaluation[52] to represent the uncertainty of CAMx as percent error. For the uncertainty introduced by regressions, we calculated percent errors using the average upper bounds of the 95th CIs relative to mean estimates in EASIUR regressions.[51] We combined both errors as if they are combined as standard deviations and used the combined errors to express upper bounds of uncertainty. For lower bounds, we calculated by taking the same fraction relative to 100%; for example, a +25% upper bound produces a lower bound of −20%. Similarly, for subregional analyses (covering less than 500 000 $km^2$), we used the average of fractional errors (weighted by the number of observations) and the average upper bounds of the 95th PIs relative to mean estimates. For primary $PM_{2.5}$, we used only 50% of fractional biases and fractional errors for elemental carbon, our proxy species for all inert primary $PM_{2.5}$, because the biases and errors are considered to be overpredicted due to errors in emissions inventory[69] rather than CTM itself. Although this method is our judgment rather than formal quantification, we believe that suggested values would sufficiently represent the uncertainty.

For concentration−response relation, we suggest the range of uncertainty that covers the 95% CIs of $PM_{2.5}$ relative risks from two land-mark epidemiological studies: the ACS studies[14] and the H6C studies H6C[15] due to pros and cons of population samples in two studies.[1,26] Therefore, alternatively, EASIUR's estimates can be explored separately for each study, using the log-linear concentration−response relation as described elsewhere.[51] Lastly, the 95% CIs of the Weibull distribution derived based on 26 VSL studies are suggested[25] for the uncertainty surrounding VSL.

## DISCUSSION

We presented marginal social costs and intake fractions for four major inorganic air pollutants in the United States: inert primary $PM_{2.5}$, $SO_2$, $NO_x$, and $NH_3$. These two metrics were estimated using the EASIUR model, which were built using regression on a data set generated by a CTM. The EASIUR estimates are the most comprehensive set of marginal health costs and intake fractions, having a spatial resolution of 36 km × 36 km for source location, a temporal resolution of four seasons, and three emission elevations (ground-level, 150 m high, and 300 m high). Because the EASIUR model can be rederived as CTMs and input data change in the future, it offers a streamlined approach for incorporating up-to-date air quality science into policy research.

Because EASIUR estimates are derived based on emissions and meteorology in 2005, there are caveats to consider when EASIUR is used for different years. First, the emissions baseline matters to the marginal costs of $SO_2$, $NO_x$, and $NH_3$ because the availability of coreactants influences the formation of secondary inorganic $PM_{2.5}$.[27,31] In this context, it is important to note that $SO_2$ and $NO_x$ emissions have been rapidly decreasing in recent years (e.g., from 2005 to 2014, $SO_2$ emissions decreased by 66% and $NO_x$ by 39%)[70] and are expected to further decrease in coming years. Holt et al.[59] showed that changed emissions from 2005 to 2012 would result in 23−42% larger sensitivity for $SO_2$ (due to more active aqueous oxidation), 37−200% larger for $NO_x$ depending on location (due to more nitrate PM formation in winter), and 28−38% smaller for $NH_3$ (due to less acidic atmosphere). Pinder et al.[71] showed $NH_3$ sensitivity would decrease by up to 30% in winter over the period of 2001−2020. A quantitative assessment can also be inferred from the work of Fann et al.,[50] which reported per-ton benefits of emissions reductions for 17 sectors for 2 periods, 2005 and 2016. After adjusting differences in population and income level between two periods, changed emissions from 2005 to 2016 resulted in 7% increase of marginal benefits for $SO_2$ and 26% increase for $NO_x$ on emissions-weighted average. These studies suggest that social cost estimates may change by up to a factor of 2, but usually less, even for fairly large changes in baseline emissions. Considering other uncertainties, the effect of changing emissions baseline is not prohibitive but is worth noting.

Interannual variability in meteorology should have a minor influence on these results, affecting seasonal or annual marginal health effects generally less than 10%.[72−74] Studies[11,75−77] suggest that the influence of climate change on $PM_{2.5}$ concentrations is negligible in short-term assessments and relatively small even in the long term.

An important group of species, primary and secondary organic $PM_{2.5}$, which roughly account for a half of ambient $PM_{2.5}$, is not modeled for EASIUR due to "structural" uncertainty of current CTMs. Although primary organic $PM_{2.5}$ has been treated inert in most CTMs so far,[78,79] one needs to be careful when applying our primary $PM_{2.5}$ estimates to primary organic $PM_{2.5}$ because most primary organic species were found to evaporate and undergo chemical reactions.[33] Current CTMs also underpredict secondary $PM_{2.5}$ by an order of magnitude,[78] suggesting a possibility of large biases in social cost estimates of VOCs (e.g., those from RSM[62] and APEEP/AP2[43,61]). Recent advancements in the understanding of organic $PM_{2.5}$ are being introduced to CTMs,[33,80−82] and we plan to do a separate study dedicated to modeling EASIUR for organic species.

Though current reduced-form models including EASIUR showed relatively good agreement for aggregate estimates (e.g., national or sectoral averages), our comparisons in Figure 4 show that they can produce substantial differences in assessments dealing with secondary species like $NO_x$ and $NH_3$ and/or with a small spatial domain. Such assessments would require a careful sensitivity analysis using multiple models as a "conservative" way of exploring uncertainty for air quality modeling beyond suggested EASIUR uncertainties. Our


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systematic comparisons in Figure 4 would be able to show in what areas the community has large discrepancies.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b06125.

Additional tables and figures (PDF)

The EASIUR estimates are available via the Internet at http://barney.ce.cmu.edu/~jinhyok/easiur/.

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### Notes

The authors declare no competing financial interest.

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## ■ REFERENCES


(1) *The Benefits and Costs of the Clean Air Act from 1990 to 2020*; U.S. Environmental Protection Agency. Office of Air and Radiation: Washington, DC, 2011.

(2) *The Benefits and Costs of the Clean Air Act, 1990 to 2010, EPA report to Congress*, EPA-410-R-99-001; U.S. Environmental Protection Agency. Office of Air and Radiation. Office of Policy: Washington, DC, 1999.

(3) *Regulatory Impact Analysis for the Final Mercury and Air Toxics Standards*, EPA-452/R-11-011; U.S. Environmental Protection Agency, Office of Air and Radiation: Washington, DC, 2011.

(4) *Regulatory Impact Analysis for the Federal Implementation Plans to Reduce Interstate Transport of Fine Particulate Matter and Ozone in 27 States; Correction of SIP Approvals for 22 States*, EPA-HQ-OAR-2009-0491; U.S. Environmental Protection Agency, Office of Air and Radiation: Washington, DC, 2011.

(5) Levy, J. I.; Baxter, L. K.; Schwartz, J. Uncertainty and Variability in Health-Related Damages from Coal-Fired Power Plants in the United States. *Risk Anal.* **2009**, *29* (7), 1000−1014.

(6) Levy, J. I.; Buonocore, J. J.; von Stackelberg, K. Evaluation of the public health impacts of traffic congestion: a health risk assessment. *Environ. Health* **2010**, *9* (1), 65.

(7) Mashayekh, Y.; Jaramillo, P.; Chester, M.; Hendrickson, C.; Weber, C. Costs of Automobile Air Emissions in U.S. Metropolitan Areas. *Transp. Res. Rec.* **2011**, *2233* (1), 120−127.

(8) Buonocore, J. J.; Dong, X.; Spengler, J. D.; Fu, J. S.; Levy, J. I. Using the Community Multiscale Air Quality (CMAQ) model to estimate public health impacts of $PM_{2.5}$ from individual power plants. *Environ. Int.* **2014**, *68*, 200−208.

(9) Gilmore, E. A.; Lave, L. B.; Adams, P. J. The Costs, Air Quality, and Human Health Effects of Meeting Peak Electricity Demand with Installed Backup Generators. *Environ. Sci. Technol.* **2006**, *40* (22), 6887−6893.

(10) Siler-Evans, K.; Azevedo, I. L.; Morgan, M. G.; Apt, J. Regional variations in the health, environmental, and climate benefits of wind and solar generation. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110* (29), 11768−11773.

(11) Heo, J.; McCoy, S. T.; Adams, P. J. Implications of Ammonia Emissions from Post-Combustion Carbon Capture for Airborne Particulate Matter. *Environ. Sci. Technol.* **2015**, *49* (8), 5142−5150.

(12) Michalek, J. J.; Chester, M.; Jaramillo, P.; Samaras, C.; Shiau, C.-S. N.; Lave, L. B. Valuation of plug-in vehicle life-cycle air emissions and oil displacement benefits. *Proc. Natl. Acad. Sci. U. S. A.* **2011**, *108*, 16554.

(13) Garcia-Menendez, F.; Saari, R. K.; Monier, E.; Selin, N. E. U.S. air quality and health benefits from avoided climate change under greenhouse gas mitigation. *Environ. Sci. Technol.* **2015**, *49*, 7580.

(14) Krewski, D.; Jerrett, M.; Burnett, R. T.; Ma, R.; Hughes, E.; Shi, Y.; Turner, M. C.; Pope, C. A., III; Thurston, G.; Calle, E. E.; et al. *Extended follow-up and spatial analysis of the American Cancer Society study linking particulate air pollution and mortality*, Research Report 140; Health Effects Institute: Boston, MA, 2009.

(15) Lepeule, J.; Laden, F.; Dockery, D.; Schwartz, J. Chronic Exposure to Fine Particles and Mortality: An Extended Follow-up of the Harvard Six Cities Study from 1974 to 2009. *Environ. Health Perspect.* **2012**, *120* (7), 965−970.

(16) Beelen, R.; Raaschou-Nielsen, O.; Stafoggia, M.; Andersen, Z. J.; Weinmayr, G.; Hoffmann, B.; Wolf, K.; Samoli, E.; Fischer, P.; Nieuwenhuijsen, M.; et al. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. *Lancet* **2014**, *383* (9919), 785−795.

(17) Crouse, D. L.; Peters, P. A.; van Donkelaar, A.; Goldberg, M. S.; Villeneuve, P. J.; Brion, O.; Khan, S.; Atari, D. O.; Jerrett, M.; Pope, C. A.; et al. Risk of Nonaccidental and Cardiovascular Mortality in Relation to Long-term Exposure to Low Concentrations of Fine Particulate Matter: A Canadian National-Level Cohort Study. *Environ. Health Perspect.* **2012**, *120* (5), 708−714.

(18) National Research Council. *Hidden Costs of Energy: Unpriced Consequences of Energy Production and Use*; The National Academies Press: Washington, DC, 2010.

(19) Dominici, F.; Wang, Y.; Correia, A. W.; Ezzati, M.; Pope, C. A.; Dockery, D. W. Chemical Composition of Fine Particulate Matter and Life Expectancy: In 95 US Counties Between 2002 and 2007. *Epidemiology* **2015**, *26*, 556.

(20) Franklin, M.; Koutrakis, P.; Schwartz, J. The Role of Particle Composition on the Association Between $PM_{2.5}$ and Mortality. *Epidemiol. Camb. Mass* **2008**, *19* (5), 680−689.

(21) Bell, M. L. *Assessment of the health impacts of particulate matter characteristics*, Research Report 161; Health Effects Institute: Boston, MA, 2012; pp 5−38.

(22) U.S. EPA. National Ambient Air Quality Standards for Particulate Matter; Final Rule. *Fed. Regist.* **2013**, *78* (10), 3085−3287.

(23) Industrial Economics, Incorporated. *Uncertainty Analyses to Support the Second Section 812 Benefit-Cost Analysis of the Clean Air Act*; Prepared for Office of Air and Radiation, U.S. Environmental Protection Agency: Washington DC.; Cambridge, MA, 2010.

(24) Rabl, A.; Spadaro, J. V. Public Health Impact of Air Pollution and Implications for the Energy System. *Annu. Rev. Energy Environ.* **2000**, *25* (1), 601−627.

(25) *Guidelines for Preparing Economic Analyses*; National Center for Environmental Economics, Office of Policy, U.S. Environmental Protection Agency, 2010.

(26) Krewski, D.; Burnett, R. T.; Goldberg, M. S.; Hoover, B. K.; Siemiatycki, J.; Jerrett, M.; Abrahamowicz, M.; White, W. H. Overview of the reanalysis of the Harvard Six Cities Study and American Cancer Society Study of Particulate Air Pollution and Mortality. *J. Toxicol. Environ. Health, Part A* **2003**, *66* (16−19), 1507−1551.

(27) Ansari, A. S.; Pandis, S. N. Response of Inorganic PM to Precursor Concentrations. *Environ. Sci. Technol.* **1998**, *32* (18), 2706−2714.

(28) Bennett, D. H.; McKone, T. E.; Evans, J. S.; Nazaroff, W. W.; Margni, M. D.; Jolliet, O.; Smith, K. R. Defining Intake Fraction. *Environ. Sci. Technol.* **2002**, *36* (9), 206A−211A.




DOI: 10.1021/acs.est.5b06125
*Environ. Sci. Technol.* 2016, 50, 6061−6070

(29) Hand, J. L.; Schichtel, B. A.; Pitchford, M.; Malm, W. C.; Frank, N. H. Seasonal composition of remote and urban fine particulate matter in the United States. *J. Geophys. Res.* **2012**, *117*, D05209.
(30) Zhang, Q.; Jimenez, J. L.; Canagaratna, M. R.; Allan, J. D.; Coe, H.; Ulbrich, I.; Alfarra, M. R.; Takami, A.; Middlebrook, A. M.; Sun, Y. L.; et al. Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes. *Geophys. Res. Lett.* **2007**, *34*, L13801.
(31) West, J. J.; Ansari, A. S.; Pandis, S. N. Marginal $PM_{2.5}$: Nonlinear aerosol mass response to sulfate reductions in the eastern United States. *J. Air Waste Manage. Assoc.* **1999**, *49* (12), 1415−1424.
(32) Blanchard, C. L.; Roth, P. M.; Tanenbaum, S. J.; Ziman, S. D.; Seinfeld, J. H. The Use of Ambient Measurements To Identify which Precursor Species Limit Aerosol Nitrate Formation. *J. Air Waste Manage. Assoc.* **2000**, *50* (12), 2073−2084.
(33) Robinson, A. L.; Donahue, N. M.; Shrivastava, M. K.; Weitkamp, E. A.; Sage, A. M.; Grieshop, A. P.; Lane, T. E.; Pierce, J. R.; Pandis, S. N. Rethinking Organic Aerosols: Semivolatile Emissions and Photochemical Aging. *Science* **2007**, *315* (5816), 1259−1262.
(34) ENVIRON. *CAMx User's Guide Version 5.41*; Environ International Corporation: Novato, CA, 2012.
(35) Byun, D.; Schere, K. L. Review of the Governing Equations, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. *Appl. Mech. Rev.* **2006**, *59* (2), 51−77.
(36) Grell, G. A.; Peckham, S. E.; Schmitz, R.; McKeen, S. A.; Frost, G.; Skamarock, W. C.; Eder, B. Fully coupled "online" chemistry within the WRF model. *Atmos. Environ.* **2005**, *39* (37), 6957−6975.
(37) Jacobson, M. Z. GATOR-GCMM: A global- through urban-scale air pollution and weather forecast model: 1. Model design and treatment of subgrid soil, vegetation, roads, rooftops, water, sea ice, and snow. *J. Geophys. Res.* **2001**, *106*, 5385−5401.
(38) *Environmental Benefits Mapping and Analysis Program − Community Edition: User's Manual*; U.S. Environmental Protection Agency: Washington, DC, 2015.
(39) *User's Manual for the Co-Benefits Risk Assessment (COBRA) Screening Model Version: 2.61*; U.S. Environmental Protection Agency: Washington, DC, 2013.
(40) Muller, N. Z. Linking Policy to Statistical Uncertainty in Air Pollution Damages. *BE J. Econ. Anal. Policy* **2011**, *11* (1), 1935−1682.
(41) Muller, N. Z.; Mendelsohn, R.; Nordhaus, W. Environmental accounting for pollution in the United States economy. *Am. Econ. Rev.* **2011**, *101* (5), 1649−1675.
(42) Latimer, D. A. *Particulate Matter Source-Receptor Relationships Between All Point and Area Sources in the United States and PSD Class I Area Receptors*, VI-A-2 A-95-83; Prepared for U.S. EPA, OAQPS: Research Triangle Park, NC, 1996.
(43) Muller, N. Z. Boosting GDP growth by accounting for the environment. *Science* **2014**, *345* (6199), 873−874.
(44) Turner, D. B. *Workbook of Atmospheric Dispersion Estimates*, A-26; U.S. Environmental Protection Agency, Office of Air Programs: Research Triangle Park, NC, 1970.
(45) Grosjean, D.; Seinfeld, J. H. Parameterization of the formation potential of secondary organic aerosols. *Atmos. Environ.* **1989**, *23* (8), 1733−1747.
(46) Jathar, S. H.; Gordon, T. D.; Hennigan, C. J.; Pye, H. O. T.; Pouliot, G.; Adams, P. J.; Donahue, N. M.; Robinson, A. L. Unspeciated organic emissions from combustion sources and their influence on the secondary organic aerosol budget in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 10473.
(47) Buonocore, J. J.; Lee, H. J.; Levy, J. I. The influence of traffic on air quality in an urban neighborhood: a community-university partnership. *Am. J. Public Health* **2009**, *99* (Suppl 3), S629−635.
(48) *Technical Support Document for the Proposed PM NAAQS Rule, Response Surface Modeling*; Office of Air Quality Planning and Standards: Research Triangle Park, NC, 2006.
(49) Baker, K. R.; Foley, K. M. A nonlinear regression model estimating single source concentrations of primary and secondarily formed $PM_{2.5}$. *Atmos. Environ.* **2011**, *45* (22), 3758−3767.
(50) Fann, N.; Baker, K. R.; Fulcher, C. M. Characterizing the $PM_{2.5}$-related health benefits of emission reductions for 17 industrial, area and mobile emission sectors across the U.S. *Environ. Int.* **2012**, *49*, 141−151.
(51) Heo, J.; Adams, P. J.; Gao, H. O. Reduced-form modeling of public health impacts of inorganic $PM_{2.5}$ and precursor emissions. *Atmos. Environ.* **2016**, *137*, 80−89.
(52) *Air Quality Modeling Final Rule Technical Support Document*; Office of Air Quality Planning and Standards, Air Quality Assessment Division, U.S. Environmental Protection Agency, 2011.
(53) *Emissions Inventory Final Rule TSD*, EPA-HQ-OAR-2009-0491; Office of Air Quality Planning and Standards. Air Quality Assessment Division, U.S. Environmental Protection Agency, 2011.
(54) Hubbell, B.; Fann, N.; Levy, J. I. Methodological considerations in developing local-scale health impact assessments: balancing national, regional, and local data. *Air Qual., Atmos. Health* **2009**, *2* (2), 99−110.
(55) Abt Associates, Inc. *Environmental Benefits Mapping and Analysis Program (BenMAP)*, Version 4.0.35; Prepared for Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency: Research Triangle Park, NC, 2010.
(56) Thompson, T. M.; Saari, R. K.; Selin, N. E. Air quality resolution for health impact assessment: influence of regional characteristics. *Atmos. Chem. Phys.* **2014**, *14* (2), 969−978.
(57) Punger, E. M.; West, J. J. The effect of grid resolution on estimates of the burden of ozone and fine particulate matter on premature mortality in the USA. *Air Qual., Atmos. Health* **2013**, *6* (3), 563−573.
(58) Pinder, R. W.; Adams, P. J.; Pandis, S. N. Ammonia Emission Controls as a Cost-Effective Strategy for Reducing Atmospheric Particulate Matter in the Eastern United States. *Environ. Sci. Technol.* **2007**, *41* (2), 380−386.
(59) Holt, J.; Selin, N. E.; Solomon, S. Changes in Inorganic Fine Particulate Matter Sensitivities to Precursors Due to Large-Scale US Emissions Reductions. *Environ. Sci. Technol.* **2015**, *49* (8), 4834−4841.
(60) Humbert, S.; Marshall, J. D.; Shaked, S.; Spadaro, J. V.; Nishioka, Y.; Preiss, P.; McKone, T. E.; Horvath, A.; Jolliet, O. Intake Fraction for Particulate Matter: Recommendations for Life Cycle Impact Assessment. *Environ. Sci. Technol.* **2011**, *45* (11), 4808−4816.
(61) Muller, N. Z.; Mendelsohn, R. Measuring the damages of air pollution in the United States. *J. Environ. Econ. Manag.* **2007**, *54* (1), 1−14.
(62) Fann, N.; Fulcher, C. M.; Hubbell, B. J. The influence of location, source, and emission type in estimates of the human health benefits of reducing a ton of air pollution. *Air Qual., Atmos. Health* **2009**, *2* (3), 169−176.
(63) Russell, A.; Dennis, R. NARSTO critical review of photochemical models and modeling. *Atmos. Environ.* **2000**, *34* (12−14), 2283−2324.
(64) Fine, J.; Vuilleumier, L.; Reynolds, S.; Roth, P.; Brown, N. Evaluating Uncertainties in Regional Photochemical Air Quality Modeling. *Annu. Rev. Environ. Resour.* **2003**, *28* (1), 59−106.
(65) Boylan, J. W.; Russell, A. G. PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. *Atmos. Environ.* **2006**, *40* (26), 4946−4959.
(66) Morris, R. E.; McNally, D. E.; Tesche, T. W.; Tonnesen, G.; Boylan, J. W.; Brewer, P. Preliminary evaluation of the Community Multiscale Air Quality model for 2002 over the southeastern United States. *J. Air Waste Manage. Assoc.* **2005**, *55* (11), 1694−1708.
(67) Committee to Evaluate Measures of Health Benefits for Environmental, Health, and Safety Regulation. *Valuing Health for Regulatory Cost-Effectiveness Analysis*, 1 ed.; Miller, W., Robinson, L. A., Lawrence, R. S., Eds.; National Academies Press: Washington, DC, 2006.
(68) Hogrefe, C.; Civerolo, K. L.; Hao, W.; Ku, J.-Y.; Zalewsky, E. E.; Sistla, G. Rethinking the Assessment of Photochemical Modeling Systems in Air Quality Planning Applications. *J. Air Waste Manage. Assoc.* **2008**, *58* (8), 1086−1099.


DOI: 10.1021/acs.est.5b06125
*Environ. Sci. Technol.* 2016, 50, 6061−6070

(69) Chow, J. C.; Watson, J. G.; Lowenthal, D. H.; Antony Chen, L.-W.; Motallebi, N. $PM_{2.5}$ source profiles for black and organic carbon emission inventories. *Atmos. Environ.* **2011**, *45* (31), 5407−5414.
(70) U.S. EPA. National Emissions Inventory (NEI) Air Pollutant Emissions Trends Data. http://www.epa.gov/ttnchie1/trends/ (accessed Jun 25, 2015).
(71) Pinder, R. W.; Adams, P. J.; Pandis, S. N.; Gilliland, A. B. Temporally resolved ammonia emission inventories: Current estimates, evaluation tools, and measurement needs. *J. Geophys. Res.* **2006**, *111*, D16310.
(72) Vukovich, F. M.; Sherwell, J. Comparison of Fine Particles and the Relationship between Particle Variations and Meteorology at an Urban Site and a Remote Site in the Eastern United States. *J. Air Waste Manage. Assoc.* **2002**, *52* (5), 573−584.
(73) Aw, J.; Kleeman, M. J. Evaluating the first-order effect of intraannual temperature variability on urban air pollution. *J. Geophys. Res.* **2003**, *108* (D12), 4365.
(74) Tai, A. P. K.; Mickley, L. J.; Jacob, D. J. Correlations between fine particulate matter ($PM_{2.5}$) and meteorological variables in the United States: Implications for the sensitivity of $PM_{2.5}$ to climate change. *Atmos. Environ.* **2010**, *44* (32), 3976−3984.
(75) Tai, A. P. K.; Mickley, L. J.; Jacob, D. J. Impact of 2000−2050 climate change on fine particulate matter ($PM_{2.5}$) air quality inferred from a multi-model analysis of meteorological modes. *Atmos. Chem. Phys.* **2012**, *12* (23), 11329−11337.
(76) Pye, H. O. T.; Liao, H.; Wu, S.; Mickley, L. J.; Jacob, D. J.; Henze, D. K.; Seinfeld, J. H. Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. *J. Geophys. Res.* **2009**, *114*, D01205.
(77) Dawson, J. P.; Racherla, P. N.; Lynn, B. H.; Adams, P. J.; Pandis, S. N. Impacts of climate change on regional and urban air quality in the eastern United States: Role of meteorology. *J. Geophys. Res.* **2009**, *114*, D05308.
(78) Goldstein, A. H.; Galbally, I. E. Known and Unexplored Organic Constituents in the Earth's Atmosphere. *Environ. Sci. Technol.* **2007**, *41* (5), 1514−1521.
(79) Kanakidou, M.; Seinfeld, J. H.; Pandis, S. N.; Barnes, I.; Dentener, F. J.; Facchini, M. C.; Van Dingenen, R.; Ervens, B.; Nenes, A.; Nielsen, C. J.; et al. Organic aerosol and global climate modelling: a review. *Atmos. Chem. Phys.* **2005**, *5* (4), 1053−1123.
(80) Shrivastava, M. K.; Lane, T. E.; Donahue, N. M.; Pandis, S. N.; Robinson, A. L. Effects of gas particle partitioning and aging of primary emissions on urban and regional organic aerosol concentrations. *J. Geophys. Res.* **2008**, *113*, D18301.
(81) Murphy, B. N.; Pandis, S. N. Exploring summertime organic aerosol formation in the eastern United States using a regional-scale budget approach and ambient measurements. *J. Geophys. Res.* **2010**, *115*, D24216.
(82) Koo, B.; Knipping, E.; Yarwood, G. 1.5-Dimensional volatility basis set approach for modeling organic aerosol in CAMx and CMAQ. *Atmos. Environ.* **2014**, *95*, 158−164.


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