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Public health costs accounting of inorganic $PM_{2.5}$ pollution in metropolitan areas of the United States using a risk-based source-receptor model

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ABSTRACT

In order to design effective strategies to reduce the public health burden of ambient fine particulate matter ($PM_{2.5}$) imposed in an area, it is necessary to identify the emissions sources affecting that location and quantify their contributions. However, it is challenging because $PM_{2.5}$ travels long distances and most constituents are the result of complex chemical processes. We developed a reduced-form source-receptor model for estimating locations and magnitudes of downwind health costs from a source or, conversely, the upwind sources that contribute to health costs at a receptor location. Built upon outputs from a state-of-the-art air quality model, our model produces comprehensive risk-based source apportionment results with trivial computational costs. Using the model, we analyzed all the sources contributing to the inorganic $PM_{2.5}$ health burden in 14 metropolitan statistical areas (MSAs) in the United States. Our analysis for 12 source categories shows that 80–90% of the burden borne by these areas originates from emissions sources outside of the area and that emissions sources up to 800 km away need to be included to account for 80% of the burden. Conversely, 60–80% of the impacts of an MSA's emissions occurs outside of that MSA. The results demonstrate the importance of regionally coordinated measures to improve air quality in metropolitan areas.

1. Introduction

Air quality management is important for the protection of public health particularly in metropolitan areas, where large populations are exposed to a high level of air pollution. Controlling the ambient concentration of fine particulate matter ($PM_{2.5}$) is especially important because the effects of $PM_{2.5}$ on premature death (Krewski et al., 2009; Lepeule et al., 2012) typically account for a dominant fraction ($>$ 95%) of the monetized air pollution damages (National Research Council, 2010; U.S. EPA, 1999, 2011a). The effectiveness of air quality management strategies for metropolitan areas depends on the quality of information about source contributions, that is, how accurate we identify the emission sources of $PM_{2.5}$ and its gaseous precursors and quantify their contributions to public health burdens imposed at a target "receptor" location. However, estimation of source contributions, or better known as source apportionment, is an intrinsically challenging task because $PM_{2.5}$ and precursor pollutants are emitted by innumerable sources and travel long distances (i.e. hundreds of kilometers or more) while undergoing complex chemical reactions under changing meteorological conditions. Current tools for source apportionment are lacking in their ability to support air quality planning in terms of spatial, sectoral and temporal resolutions as well as ease-of-use.

One group of methods for source apportionment is called as receptor models (RMs), which estimate source contributions by relating physical and chemical characteristics of emissions sources with ambient measurements made at a downwind location or receptor (Hopke, 2016; Martin et al., 2011; McMurry et al., 2004; Watson et al., 2008). RMs have fundamental limitations in providing detailed source apportionment (Belis et al., 2013; Hopke and Cohen, 2011; Robinson et al., 2006; Watson et al., 2002, 2008). First, they cannot provide spatial or temporal distribution of source contributions. Although efforts have been made to combine RMs with dispersion models or back trajectory models, their estimations are still limited to qualitative description and not systematically evaluated yet (Watson et al., 2008). Second, relying on measurements, RMs are not suited for predicting source contributions from hypothetical scenarios. This limits their use for what-if policy analyses. Third, although secondary (or chemically produced) $PM_{2.5}$ species usually account for a dominant fraction of ambient $PM_{2.5}$, RMs basically assume that emissions are chemically stable and, therefore, have a limited ability to link secondary $PM_{2.5}$ species to their

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sources of gaseous precursors. Combining RMs with chemical aging model may partly address this limitation. Lastly, RMs cannot distinguish sources of similar chemical profiles, limiting the number of source types that can be analyzed.

The other group of methods is using a source-based model called as chemical transport models (CTMs), which simulate emissions, transport, chemical transformations, and removal processes of air pollutants in a three-dimensional grid. Unlike RMs, CTMs address complex photochemical processes explicitly. They can be used to inform hypothetical scenarios. However, because running CTMs is computationally expensive, simulating numerous emissions sources one by one in a "brute force manner" to quantify their contributions at a given receptor location is computationally very expensive. To overcome CTM's high computational burdens, CTM's 'add-on' sensitivity techniques were developed for emission tagging (e.g. the particulate source apportionment technology method) (Koo et al., 2009; Kwok et al., 2013; Wagstrom et al., 2008; Wang et al., 2009), source-oriented sensitivity (e.g. the decoupled direct method) (Dunker et al., 2002; Koo et al., 2007), and receptor-oriented sensitivity (e.g. the adjoint method) (Hakami et al., 2007; Henze et al., 2009). Although useful in their certain applications (e.g. Fann et al., 2012; Pappin et al., 2015; Pappin and Hakami, 2013; Wagstrom and Pandis, 2011), they can be too costly to explore a large set of policy options systematically (e.g. what combination of emissions controls over power plants in and out of state would be most cost-effective to improve air pollution in the New York metropolitan area?). In addition, they are not easy to use and thus not accessible to a wide range of users including policy practitioners and decision makers.

Furthermore, both RM and CTM methods require additional steps to analyze population risk from their outputs although the risk estimation is not usually as resource-intensive as air quality modeling. Ultimately, it is useful to have source apportionment tools that support risk-based assessments (National Research Council, 2004; Hubbell and Frey, 2011), which require going beyond source apportionment of ambient $PM_{2.5}$ concentrations to quantifying population exposed to ambient $PM_{2.5}$ and associated changes in mortality. Because RM's source profiles are not spatially resolved, such application is beyond their capability or intended scope. Although CTMs can support risk-based approaches in principle, their application could be limited by prohibitive technical and computational costs.

We developed a reduced-form model, the Air Pollution Social Cost Accounting (APSCA) model (available at http://barney.ce.cmu.edu/~jinhyok/apsca/), that estimates risk-based source-receptor relations for marginal changes in emissions of inorganic air pollutants with fine spatial, sectoral, and seasonal resolution. The ASPCA model is built on top of the Estimating Air pollution Impact Using Regression (EASIUR) model (Heo et al., 2016a, 2016b), which predicts mortality costs of emissions from anywhere in the United States at the accuracy similar to that of a CTM but with negligible computational requirements. By spatially disaggregating the mortality costs estimated by EASIUR to receptor locations, APSCA produces source contributions of public health costs borne by a receptor location, which are resolved in space (36 km × 36 km resolution), species (primary $PM_{2.5}$, SO_2, NO_x, and NH_3), sector (as resolved in emissions inventory), and season (winter, spring, summer, and fall). This paper describes and evaluates the method for building the APSCA model. We selected 14 metropolitan statistical areas in the United States to demonstrate the capability of APSCA as well as to explore the differences in the source contributions among the different areas across the nation. We discuss how the APSCA model would better assist decision-making for various air quality management and policy issues. Note that we often refer to mortality costs estimated by APSCA and EASIUR as "social costs" or "public health costs" in this paper because the mortality costs usually dominate other effects on morbidity and the natural environment as briefly mentioned in the beginning.

2. Method

The APSCA model is built based on the EASIUR model (Heo et al., 2016a, 2016b), a reduced-form model that quantifies the social costs of emissions. Derived from regressions on a large dataset generated by a CTM, the EASIUR model estimates the sum of social costs imposed at all downwind locations by emissions from a source location. The APSCA model estimates source contributions to receptor locations by spatially disaggregating EASIUR's estimates to receptor locations using empirically derived spatial distributions.

2.1. Summary of the EASIUR model

The EASIUR model is closely related to the APSCA model and, therefore, summarized here. For the complete description and evaluation of EASIUR, refer to Heo et al. (2016b). The EASIUR model estimates $PM_{2.5}$ mortality costs imposed at all downwind locations by one additional tonne of (or marginal) emissions of four inorganic $PM_{2.5}$ precursors: inert primary $PM_{2.5}$ (not including primary organic $PM_{2.5}$) and three inorganic $PM_{2.5}$ precursors (SO_2, NO_x, and NH_3). Extension to cover organic $PM_{2.5}$ is more challenging due to the complex and uncertain nature of secondary organic PM but is currently underway. The final product of EASIUR is a set of look-up tables or maps that provide marginal social cost estimates for each emitted species, for each of the four seasons, for three emissions elevations (i.e. ground-level, 150 m, and 300 m), and for every cell in a 36 km × 36 km grid covering the United States.

The EASIUR model follows a standard method for estimating mortality costs of emissions, which is an impact pathway analysis of estimating ambient $PM_{2.5}$ from precursor emissions, quantifying associated premature deaths based on epidemiological studies, and monetizing the mortality using the value of a statistical life. For simulating ambient $PM_{2.5}$, the model employed a state-of-the-art CTM, CAMx (ENVIRON, 2012), in the 148 × 112 grid of a 36 km × 36 km horizontal resolution, which is sufficiently acceptable for $PM_{2.5}$ public health analysis (Arunachalam et al., 2011; Gan et al., 2016; Punger and West, 2013; Thompson et al., 2014). EASIUR was developed using the $PM_{2.5}$ concentration-response relation from the most recent American Cancer Society cohort-based study (Krewski et al., 2009). EASIUR used $8.6 M (in 2010 USD) for the value of statistical life (VSL) as recommended by U.S. EPA (2010). Heo et al. (2016b) describes an easy method of adjusting EASIUR for different concentration-response relation and VSL.

The EASIUR model was developed in three steps. First, CAMx was run to calculate per-tonne social costs of the four species at a large number (100) of locations randomly selected based on population size. Second, the resulting per-tonne social costs were regressed as a function of covariates such as exposed population and key atmospheric variables, using a half of the sampled locations for the regressions and the other half for out-of-sample evaluations. Finally, using the regression models, per-tonne social costs were estimated for all the cells in the 36 km × 36 km grid covering the continental United States.

A central challenge for EASIUR development was to model exposed population in a simple but accurate way, accounting for highly inhomogeneous population densities and complex $PM_{2.5}$ impacts over hundreds of kilometers resulting from atmospheric chemistry, dispersion, and removal. In the EASIUR development, exposed population is expressed using 'average plumes.' The distribution of $PM_{2.5}$ concentrations formed by a marginal amount of emissions was estimated for 50 sample locations. An average plume is generated by averaging the 50 distributions of $PM_{2.5}$ concentrations after rotating them to align the average wind direction at each location to a common direction. The plume is divided by its sum and then used as spatial weighting factors. In the EASIUR model, exposed population was express by summing population grid cells weighted by a species-, season-, and height-specific average plume placed over a source location after aligning it for the average wind direction at the location. Fig. 1 shows wintertime

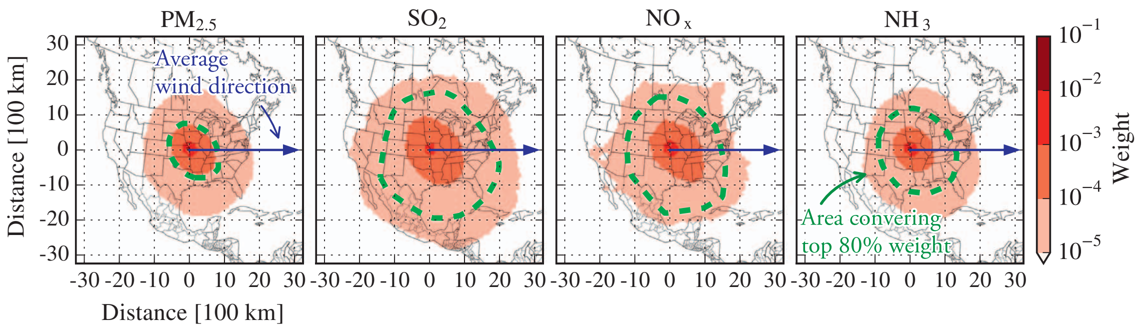

Fig. 1. Wintertime average plumes used in the APSCA model for ground-level emissions. $PM_{2.5}$ concentrations from marginal emissions at 50 sample locations estimated by a chemical transport model are averaged and normalized to produce distributions of spatial weighting factors (or average plumes). 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.

average plumes of four species emitted at the ground-level.

2.2. APSCA methods and development.

While the EASIUR model used the average plumes to estimate total downwind impacts, APSCA takes this a step further by using the average plumes to spatially disaggregate those downwind impacts. APSCA assumes that any given marginal tonne of emissions will have a plume of $PM_{2.5}$ impacts downwind very similar to a corresponding average plume, and our evaluation tests this assumption. More specifically, the APSCA model disaggregates EASIUR's estimates to downwind receptors by assigning damages in proportion to the product of the average plume weight (representing the concentration impact) and population. By doing this spatial disaggregation for all the emission sources, the source contribution or accounting of the social costs is estimated for each receptor location in the modeling grid.

As an illustration, Fig. 2a shows the wintertime average plume of primary $PM_{2.5}$ placed at Chattanooga in Tennessee. Fig. 2b depicts how the APSCA method estimates the spatial accounting of social costs caused by emissions at the location. Fig. 2c presents comparable results calculated using computationally expensive CAMx outputs. Figs. 2d and S2 corroborates the similarity between APSCA estimates and those calculated with CAMx outputs over a substantial fraction of receptors for the four APSCA species. In the APSCA model, we used average plumes (Fig. 1) that are four times larger (181 cells × 181 cells, or 6516 km × 6516 km) than those used in the EASIUR model (91 cells × 91 cells). Results from EASIUR suggest that these larger average plumes are required to capture the impacts of emissions, particularly from remote areas (e.g. Idaho or Montana).

2.3. Two evaluation methods for the APSCA model

We evaluated the APSCA model using two indirect methods: 1) comparison to ambient measurements; and 2) comparison to CTM-based estimates. We were not able to compare APSCA's estimates directly with other studies using receptor models or CTMs because they do not have comparable spatial (i.e. 36 km × 36 km) and sectoral (i.e. 12 source sectors) resolutions.

The first evaluation is to reconstruct annual average $PM_{2.5}$ concentrations using APSCA and to compare them to ambient measurements. EASIUR uses a standard method of calculating the social cost of emissions, S [\$]:

$$S = y^0 \cdot \left\{1 - exp\left(-\frac{ln\,R}{10} \cdot \Delta c\right)\right\} \cdot V \tag{1}$$

where y^0 is baseline mortality [number of deaths], R is the relative risk of $PM_{2.5}$, Δc is the changes in $PM_{2.5}$ concentrations [μg/m^3], and V is the value of a statistical life [\$/person]. Since S values from four inorganic pollutants in 2005 are known from EASIUR in all the grid cells, we back-calculated Δc for each inorganic pollutant in all the cells and compared to annual average concentrations of corresponding inorganic $PM_{2.5}$, which were obtained from U.S. EPA's Air Data (available at http://aqsdr1.epa.gov/aqsweb/aqstmp/airdata/download_files.html). We matched APSCA's concentrations using emissions of four species (elemental carbon, SO_2, NO_x, and NH_3) to elemental carbon, sulfate $PM_{2.5}$, nitrate $PM_{2.5}$, and ammonium $PM_{2.5}$ in the Air Data. Note that all the $PM_{2.5}$ species are assumed to have the same health effects in the regulations regardless of their chemical compositions due to insufficient epidemiological evidence (Industrial Economics, Incorporated, 2010; U.S. EPA, 2013).

There are two caveats in our comparisons to ambient measurements. First, EASIUR was designed to predict the social costs of "marginal" emissions. Therefore, ambient concentrations calculated using total emissions can be biased, considering that the inorganic $PM_{2.5}$ formation can depend on the size of emissions (Ansari and Pandis, 1998). Second, back-calculated concentrations of three inorganic gaseous species do not match directly to those of corresponding $PM_{2.5}$ species. This is because EASIUR's social costs, for example, from SO_2 emissions are the social costs of changes in all $PM_{2.5}$ species by the SO_2 emissions. However, the effects on other species (e.g. the effect of SO_2 emissions on nitrate and ammonium concentrations) were generally found to be minor (Heo et al., 2016b). Therefore, considering nonlinear relationships among inorganic species (Ansari and Pandis, 1998; West et al., 1999), the evaluation against measurements indicates only

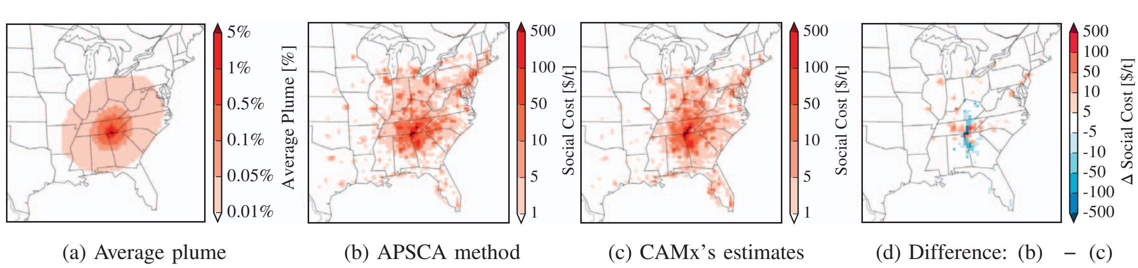

Fig. 2. An illustration of the spatial disaggregation method used in the APSCA model. (a) Shows a wintertime average plume of primary $PM_{2.5}$ placed at Chattanooga in Tennessee. (b) Shows the APSCA method of spatially disaggregating EASIUR's social cost estimates to receptor locations using the average plume weighted by underlying population. (c) Shows those estimated by a state-of-the-art chemical transport model, CAMx. (d) Shows the difference between the APSCA method (b) and CTM's estimates (c).

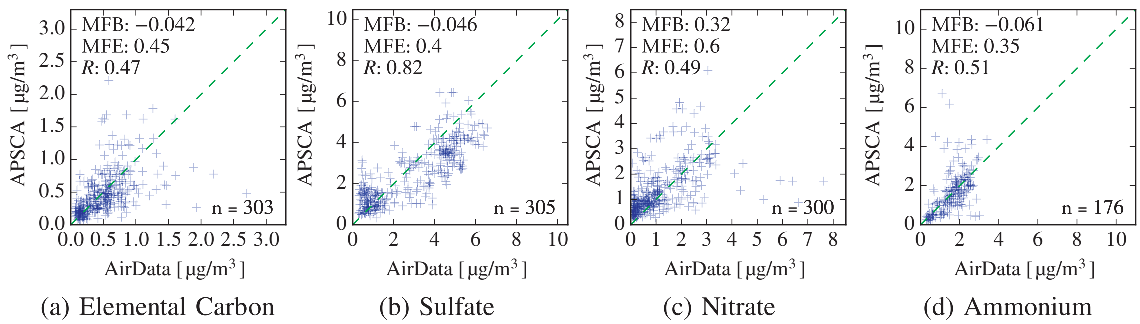

Fig. 3. Evaluation of APSCA's concentration estimates against ambient measurements. Spatial comparisons are presented in Fig. S3. MFB and MFE are defined in Eqs. (2) and (3), respectively, and *R* is the Pearson correlation coefficient.

whether APSCA estimates are within a reasonable order of magnitude.

We used mean fractional bias (MFB) and mean fractional error (MFE)—standard metrics to evaluate predictions of air quality models against observations (Boylan and Russell, 2006; Morris et al., 2005). MFB and MFE are defined in Eqs. (2) and (3), respectively.

$$\mathrm{MFB} = \frac{2}{n}\sum_{i=1}^{n}\frac{P_i - O_i}{P_i + O_i} \tag{2}$$

$$\mathrm{MFE} = \frac{2}{n}\sum_{i=1}^{n}\frac{|P_i - O_i|}{P_i + O_i} \tag{3}$$

where P_i is model-estimated concentrations at observation site i, O_i is observed concentrations, and n is the total number of sampled sites. MFB and MFE limit the maximum bias and errors within ± 200% and 200% respectively so that they are not severely skewed by outliers. Morris et al. (2005) defined the “Good” performance as having MBF ≤ ± 30% and MFE ≤50% and the “Average” as having MBF ≤ ± 60% and MFE ≤75%. Boylan and Russell (2006) suggested that meeting the “Good” criteria be “close to the best a model can be expected to achieve” and the “Average” be “acceptable for modeling applications.”

Secondly, we compared the APSCA's results to those estimated directly from CAMx outputs at 50 out-of-sample locations. For this evaluation, MFB and MFE were slightly adapted to prevent disproportionate biases caused by receptors far from source that account for a trivial fraction of social costs but tend to have large biases and errors. As Boylan and Russell (2006) suggested less stringent criteria for small values, we similarly defined metrics equivalent to MFB and MFE as follows to weight values at receptors relative to their fraction of social costs.

$$\mathrm{MFB_{eq}} = \frac{2}{m}\sum_{j=1}^{m}\frac{A_j - C_j}{A_j + C_j}\cdot\frac{C_j}{C_T} \tag{4}$$

$$\mathrm{MFE_{eq}} = \frac{2}{m}\sum_{j=1}^{m}\frac{|A_j - C_j|}{A_j + C_j}\cdot\frac{C_j}{C_T} \tag{5}$$

where C_j is social cost estimated by CAMx at receptor j, C_T is total social costs estimated by CAMx, A_j is social cost at j estimated by APSCA using C_T, and m is the total number of receptor locations. The $\mathrm{MFB_{eq}}$ and $\mathrm{MFE_{eq}}$ were calculated for ground-level emissions at the 50 out-of-sample locations. The averages of $\mathrm{MFB_{eq}}$ and $\mathrm{MFE_{eq}}$ were evaluated against the performance criteria from Morris et al. (2005).

2.4. Social cost accounting of inorganic $PM_{2.5}$ pollutions for 14 metropolitan areas

As an illustration of APSCA's application, we analyzed 14 metropolitan statistical areas across the United States (defined in Fig. S1) to explore the spatial variation in the social cost accounting of inorganic $PM_{2.5}$ pollution. Here, we are assuming APSCA's estimates stay linear over the entire range of emissions for a demonstration purpose. However, APSCA's strength and goal are to analyze marginal changes in emissions, which policy interventions are often concerned with. The accounting was estimated using the 2005 emissions inventory (U.S. EPA, 2011b), which was used in the EASIUR development, as follows. First, we prepared ground-level emissions of inert primary $PM_{2.5}$, SO_2, NO_x, and NH_3 by season for 12 source sectors (described below) at the APSCA's spatial resolution. For elevated point emissions, we aggregated emissions by stack height for every 10 m. Second, we estimated social costs for all the gridded emissions using EASIUR. For elevated emissions, EASIUR's three elevations (ground-level, 150 m, and 300 m) were interpolated linearly to match their elevations (e.g. for emissions at 50 m high, 2/3 of the EASIUR's ground-level estimate and 1/3 of EASIUR's 150 m estimate were summed.). Then, we spatially disaggregated the social costs using the APSCA method.

Finally, the social costs borne by each of the 14 metropolitan areas were prepared from APSCA's gridded estimates using high-resolution geospatial boundary information from U.S. Census Bureau (2013). For grid cells that partly overlap with the geospatial boundary of metropolitan areas, we allocated the fraction of grid estimates in proportion to the size of overlapped area. We analyzed the social cost accounting by spatial distribution, by sector, and by season. The 12 emissions sectors are listed here: EGU (electric generating units); non-EGU (non-EGU industrial sources); on-road (on-road gasoline and diesel vehicles); large marine vessels (category 3 commercial marine vessel), other transportation (locomotive and category 1 and category 2 commercial marine vessel); non-road (non-road engines); foreign (Mexico and Canada); area emissions (small sources that do not belong to EGU or non-EGU); fugitive dust; agriculture; fire (wild and prescribed fire); and biogenic sources. Heo et al. (2016a) describes the source categories in more detail.

3. Results

3.1. Evaluations of the APSCA model

The APSCA model shows good performance in the two evaluations. Fig. 3 compares APSCA's back-calculated concentrations with USEPA's Air Data measurements. Three species—EC, SO_2, and NH_3—meet the “Good” criteria suggested by Morris et al. (2005) while NO_x meets the “Average”, which is comparable to the evaluation of CTMs' predictions against measurements (Simon et al., 2012). Fig. S3 shows that areas with large population and/or large emissions (e.g. urban areas in the east) yield relatively smaller MFB and MFE than remote areas. This makes sense because a substantial fraction of public health effects from emissions originated from remote areas (e.g. Idaho or Montana) would occur relatively far from source locations, where APSCA's errors are expected to be larger. Since most applications of APSCA will tend to focus on areas where large emissions and large damages occur, APSCA's performance is expected to be better than that presented in Fig. 3. In addition, Fig. S3 shows that APSCA underpredicts sulfate, nitrate, and ammonium in the Southern California region by a factor of two or four.

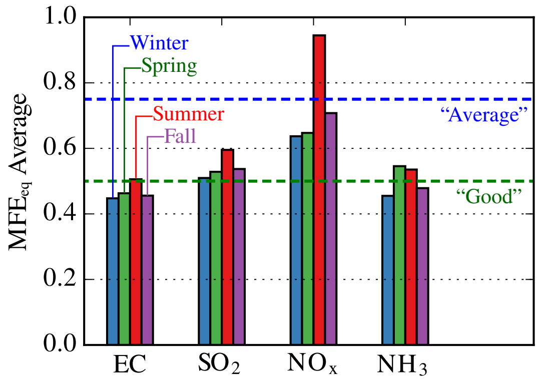

Fig. 4. Summary of APSCA evaluations against CTM-based estimates. "Average" and "Good" criteria are adopted from Morris et al. (2005). More detailed evaluations are presented in Figs. S4–S7. MFEeq is defined in Eq. (5).

Fig. 4 summarizes the comparisons between APSCA's spatial predictions and CTM's. Each bar corresponds to the average of MFE_{eq}'s of 50 source locations. APSCA's MFB_{eq} against CAMx is zero because the same amount of social costs are allocated to receptor areas and, therefore, not presented. APSCA's predictions generally meet or are close to "Good" criteria suggested by Morris et al. (2005). APSCA produced the least MFE_{eq} for EC and the largest for NO_x. This corresponds to the complexity of atmospheric processes associated with each species (Heo et al., 2016a). While EC is inert in the atmosphere and its impacts depend mainly on dispersion, other inorganic gases additionally undergo complicated photochemical reactions and gas-particle partitioning (Ansari and Pandis, 1998; West et al., 1999; Yu et al., 2005). Because the performance of summertime NO_x does not meet the less strict "Average" criteria, evaluations involving APSCA's summertime NO_x estimates should be interpreted carefully. Note also that marginal social costs of NO_x is generally smaller in summer (Heo et al., 2016a) and that summertime NO_x reduction may increase or decrease ozone concentrations depending on the ratio of volatile organic compounds to NO_x in the atmosphere (National Research Council, 1991). In Figs. S4–S7, we presented the spatial variation of APSCA's performance with respect to CAMx. As observed in the first evaluation, we found again that the performance is generally better in densely populated areas than in remote areas, and thus we expect APSCA's prediction to be better in areas where population and emissions are large.

3.2. Social cost accounting for 14 metropolitan statistical areas

The spatial accounting of emission sources responsible for the health costs in the 14 metropolitan statistical areas are presented in Fig. 5. The accounting maps show that a substantial fraction of the damages originated from areas far from receptor metropolitan areas. Emission sources affecting metropolitan areas in the west are geographically less spread out compared to those in the east. This is reasonable because they are far from sources in the east and emissions in the Rocky Mountains and the Great Plains are relatively small, limiting effects from long-range transport. Note that the social costs imposed in the Los Angeles metropolitan area would be underpredicted, considering the underprediction in the Southern California region shown in Fig. S3.

Fig. 6 provides the accounting by emission sector (top panel), by distance between emission source and each area's boundary (middle panel), and by species (bottom panel). Areas in the east are generally affected more by EGUs than in the west, reflecting the prevalence of coal-fired power plants in the east. Areas near Mexican or Canadian border have a substantial fraction from foreign sources. On-road and non-road mobile sources are important in all the areas. Areas near large ports have a sizeable fraction from marine vessels. Recall that the current APSCA does not include organic $PM_{2.5}$, which usually accounts for approximately half of ambient $PM_{2.5}$. When organic pollutants are included, we expect the fraction of on-road and non-road mobile sources, industrial sources, biogenic sources to be particularly much bigger than shown here. The middle panel in Fig. 6 presents the relative contributions of emission sources by their distance from receptor areas. Distance is calculated using the centroid of boundary cells of each area and that of emission source cells. Local (i.e. within the boundary of a metropolitan area) or relatively close (i.e. within 100 km) sources roughly account for 40% in the east and 60% in the west. Substantial portions of the health damages in any area originate from sources quite far from the area. For example, for the eastern areas, emission sources up to about 800 km away need to be included to account for 80% of the health burden; for the western areas, emissions sources up to about 400 km needs to be included. The bottom panel in Fig. 6 shows that primary $PM_{2.5}$ accounts for a bigger fraction in the western areas than in the eastern areas, indicating that regional secondary pollution affects eastern areas more.

Fig. 7 presents how much local (or within boundary) and external (or outside of boundary) emissions affect each area. In absolute terms, the magnitude of social costs varies substantially among the metropolitan areas, which is largely explained by the different size of population and the concentrations of ambient $PM_{2.5}$ in each area. The left panel shows that local emissions generally account for 10–20% of the total burden at each location while the remaining is attributable to external sources. The right panel shows that 60–80% of public health burden caused by local emissions is imposed outside. In addition, Fig. S8 presents seasonal accounting of the total burden, showing different seasonal variations among the areas. Figs. S9–S20 show seasonal accounting for each of the 12 source sectors. Certain sectors such as agriculture, fire, and biogenic show substantial seasonal variations.

4. Discussion

We have developed and evaluated the APSCA model, a reduced-form source-receptor model that estimates the contributions of emissions sources to the public health burden of inorganic $PM_{2.5}$ pollution experienced in downwind receptor locations. Using the average plume method derived from chemical transport model simulations to characterize the spatial distribution of exposed population and ambient $PM_{2.5}$ formations from precursor emissions, the APSCA model spatially disaggregates the social costs of emissions estimated by the EASIUR model. Estimating comprehensive source-receptor relations, the APSCA model generates detailed accounting of the public health burden from marginal changes in emissions at a given receptor. Our evaluations of APSCA against ambient measurements show that APSCA's performance can be considered "Good" for EC, SO_2, and NH_3 and "Average" for NO_x, which is comparable to a state-of-the-art CTM's performance. Compared to current receptor models and CTMs, APSCA can produce more comprehensive accounting information in terms of social, sectoral, and temporal resolutions and its ease-of-use would make it more accessible to practitioners and decision makers.

More specifically, the APSCA model has several merits for policy decision-making when compared to existing receptor models. First, it provides accounting information for any receptor location of interest while receptor models are limited to places where ambient measurements are available. Second, APSCA's outcomes are risk-based, accounting for population exposure and valuation, which is usually preferred in policy research such as cost-benefit analyses. Third, the APSCA model can be used to assess various hypothetical emissions scenarios, and hence is a powerful tool for what-if policy analysis, for which receptor models are not well-suited.

On the other hand, an APSCA model can be built only where input data for CTMs such as emissions inventory, meteorology, and other information are available. Hence, while receptor models can be used wherever ambient measurements exist, the APSCA model requires an infrastructure for chemical transport modeling, which is not commonly

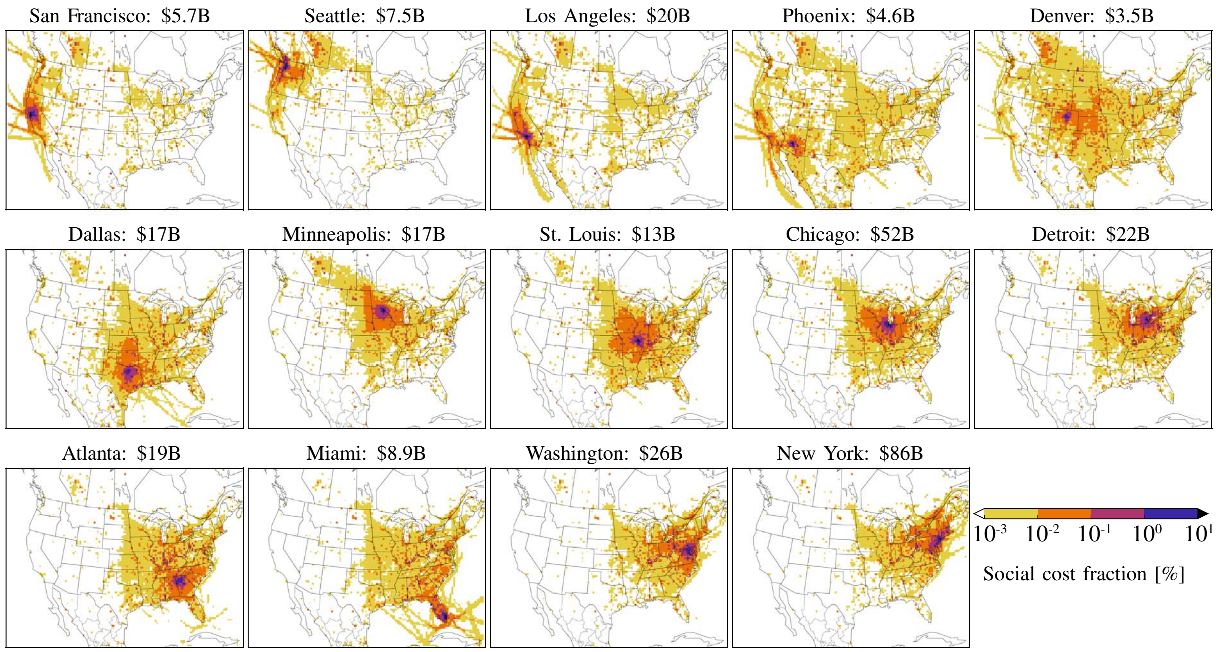

Fig. 5. Spatial accounting of emission sources responsible for the public health costs borne by the 14 metropolitan statistical areas. The value beside the name of each metropolitan area indicates the sum of public health costs borne by the metropolitan area in 2005. The colorbar shows the contribution fraction to the source locations. Definition of metropolitan statistical areas and their boundaries are presented in Fig. S1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

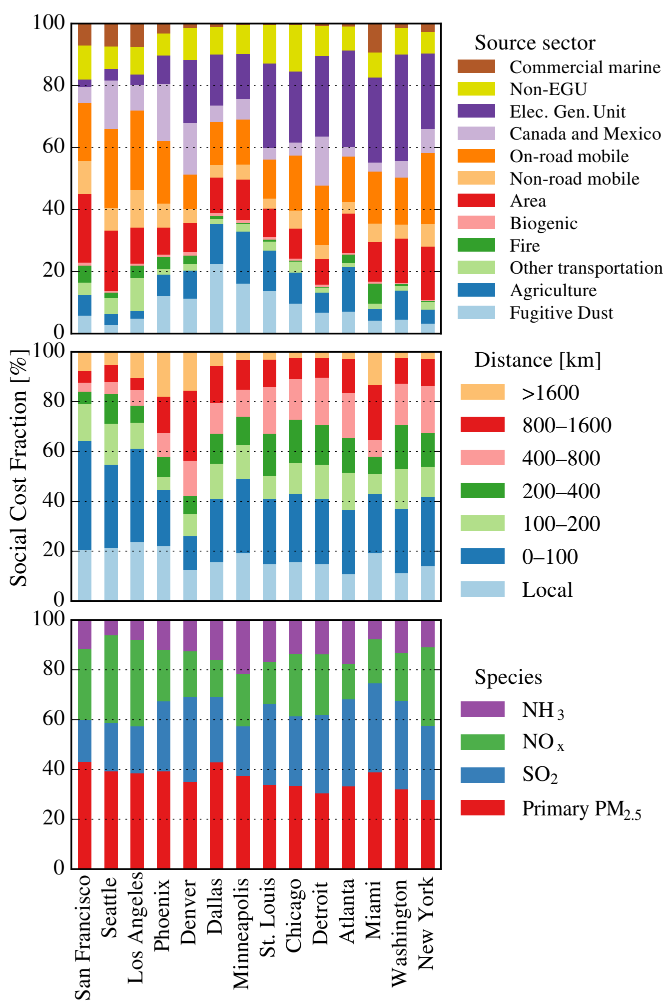

Fig. 6. Source contributions in terms of source sector, source distance, and air pollutant species.

available in many parts of the world. In addition, the APSCA model is built for long-term exposure to ambient $PM_{2.5}$ but not for episodic incidences because APSCA is relying on concentration-response relations derived from epidemiological studies on chronic exposure to ambient $PM_{2.5}$. Lastly, the APSCA model is yet to be developed for organic $PM_{2.5}$, for which gasoline and diesel engines, meat cooking, and prescribed fire are main anthropogenic sources. EASIUR did not address organic species due to recent advancement in the understanding of organic $PM_{2.5}$ and associated research gaps (Heo et al., 2016a, 2016b). However, an extension of EASIUR and APSCA to organic $PM_{2.5}$ is currently underway.

Our accounting analysis for the 14 metropolitan areas in the U.S. shows the importance of regionally based air quality management. For example, our results show that metropolitan areas need to consider emission sources as far as 400–800 km away to account for 80% of their health burden (See Fig. 6). Obligated by the "Good Neighbor" provision in the Clean Air Act, U.S. EPA has been paying attention to regionally based controls such as the NOx Budget Trading Program and the Cross-State Air Pollution Rule (U.S. EPA, 2011c, 2016). As "low-hanging fruits" in terms of local emissions reductions are becoming scarce, now it may make better sense to take more regional sources into account to achieve future clean air goals in metropolitan areas in a cost-effective manner. Our results support the EPA's ongoing efforts and can also help resource-starved local and state-level air quality managers with regulatory compliance.

Our results call for improved regional collaborations among local governments, state and federal agencies, and private sectors. The current level of such collaborations may not be sufficient. For example, the strategic plan of New York City (called Strategy 2011–2014, available at < http://www.nyc.gov/html/dep/html/about_dep/dep_strategic_plan.shtml >) includes four air quality related goals, all of which are about controlling *local* sources. In California, air quality is managed at a sub-state level; the state is divided into 15 air basins, each of which is managed by an air quality management district. However, our results suggest that there are potential benefits of collaborations among air basins. Since stakeholders at a regional level would have different interests and goals, it would be challenging to establish such regional collaborations, requiring a governance system that can bring all the

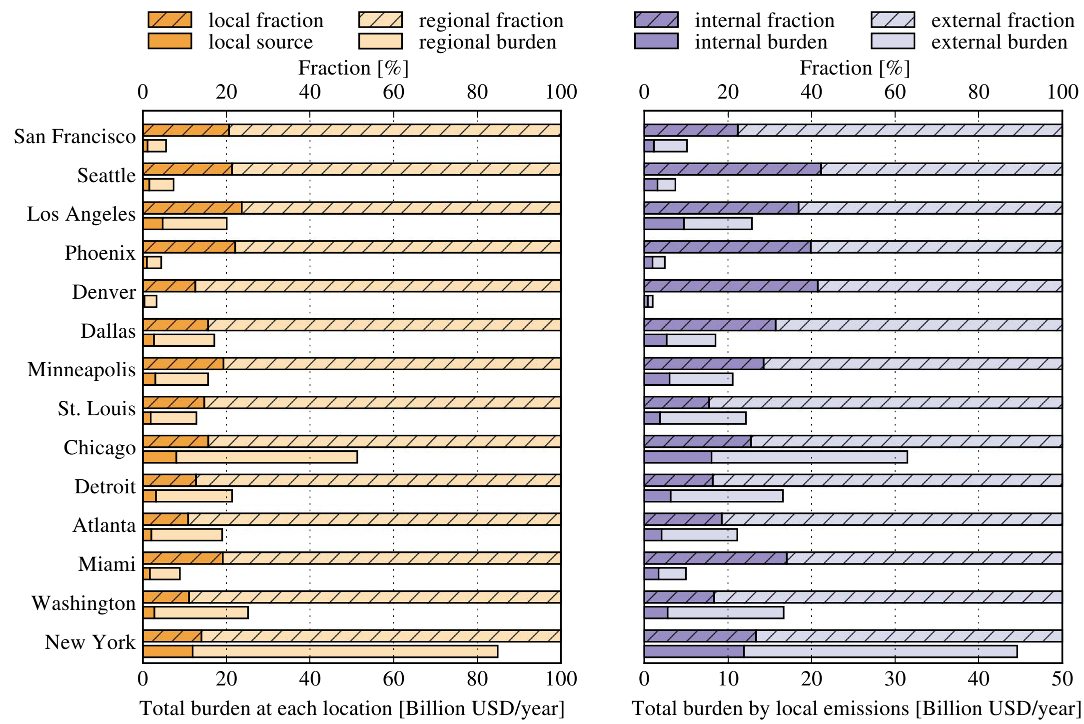

Fig. 7. Public health burden borne by or originated from metropolitan areas. Left-hand side panel shows health burden imposed on metropolitan areas and right-hand side panel health burden originated from emissions at metropolitan areas. Top dashed bars indicate fractions [%] and bottom solid bars social costs [Billion USD/year].

stakeholders together in the planning and decision-making processes. Detailed accounting analyses shown in this study could play an essential role in scoping and facilitating such collaborative process for air quality management at a proper regional scale. For example, Fig. 6 suggests that it would be important for eastern metropolitan areas and their states to make coordinated efforts to control upwind power plant emissions while western and midwestern areas may have to convince the agricultural sector for improving public health.

When used together with the EASIUR model, the APSCA model can be valuable for the design of optimal strategies in air quality management from a receptor location's point of view. The computational efficiency of the two models makes it possible to couple optimization methods, which can find the most cost-effective strategy among a large amount of policy options. At a local level, for example, New York City can identify emission sources inside and outside of its jurisdiction using APSCA and design policies that help maximize public health benefits within a given budget. Or, at the state or federal level, APSCA can assist state agencies such as New York State Department of Environmental Conservation (NYSDEC) to roll out a plan for regional collaborations to achieve policy goals such as maximizing public health benefits or equal distribution of the benefits in a cost effective manner. APSCA's detailed accounting of air pollution sources and receptors can facilitate various types of policy research associated with marginal changes in emissions such as cost-benefit analysis, climate mitigation co-benefit analysis, tax design, emission trading scheme, and environmental justice assessment.

The APSCA model has four major sources of uncertainty: CTM, reduced-form modeling, concentration-response relation, and VSL. Due to numerous inputs and parameters in CTMs, they are often evaluated "operationally" against ambient measurements (Boylan and Russell, 2006; Morris et al., 2005). A comprehensive evaluation of the CTM model and inputs showed the good performance of simulating inorganic pollutants (U.S. EPA, 2011d). Using a similar method, the reduced-form modeling of APSCA and EASIUR were evaluated against CTM's outputs in this study (see Fig. 4) and Heo et al. (2016a), respectively, which demonstrated that the associated uncertainties would be comparable to those of CTM's against ambient measurements, although it should be noted that APSCA shows large differences in the Southern California region and some other areas (see Section 3.1 and Fig. S3). Additionally, because the base year of APSCA and EASIUR is 2005, applying APSCA to different years may introduce more uncertainties because emissions have been decreasing substantially since then in the U.S and the changed emissions affect the formation of secondary inorganic $PM_{2.5}$ (Ansari and Pandis, 1998; Holt et al., 2015). The magnitude of the uncertainties would be modest overall within a moderate period of time (e.g. 10 years) as discussed in Heo et al. (2016a). Deriving EASIUR and APSCA with different baselines will clear the uncertainties. We plan to develop our models with more recent baseline inputs in the near future. According to standard procedures applied to regulatory assessments, the uncertainties from the other two sources, concentration-response relation and VSL, would range from −33% to +270% and from −90% to +160%, respectively (Heo et al., 2016a). It should also be noted that EASIUR and APSCA address only mortality, but no other morbidity and economic endpoints, which can be important in some cases. Although these uncertainties are considerable, the reduce-form modeling of APSCA and EASIUR is the only additional source of uncertainty when compared to those found in CTM-based major regulatory assessments.

Although it is desirable to compare APSCA results to those from other source apportionment studies, such comparisons are unfortunately not feasible yet. First of all, none of the existing studies have produced source apportionment that is risk-based and as detailed as what APSCA generates. Comparisons at an aggregate level are challenging as well because APSCA does not have organic $PM_{2.5}$ yet and the source sectors selected for APSCA from emissions inventory do not match easily measurement-based source profiles used in receptor models. As a result, comparisons to receptor models would have to be addressed in future research. An adjoint model (Hakami et al., 2007; Henze et al., 2007) or hybrid method (Hu et al., 2014) can also be used in future cross-evaluation of APSCA.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2017.06.006.

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