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DOI
10.1038/s41370-020-0219-1

Publication date
2020

Document Version
Final published version

Published in
Journal of Exposure Science and Environmental Epidemiology

Journal of Exposure Science & Environmental Epidemiology (2021) 31:654–663
https://doi.org/10.1038/s41370-020-0219-1

ARTICLE

Comparisons of simple and complex methods for quantifying exposure to individual point source air pollution emissions

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Received: 14 June 2019 / Revised: 20 December 2019 / Accepted: 31 January 2020 / Published online: 17 March 2020

Abstract

Expanded use of reduced complexity approaches in epidemiology and environmental justice investigations motivates detailed evaluation of these modeling approaches. Chemical transport models (CTMs) remain the most complete representation of atmospheric processes but are limited in applications that require large numbers of runs, such as those that evaluate individual impacts from large numbers of sources. This limitation motivates comparisons between modern CTM-derived techniques and intentionally simpler alternatives. We model population-weighted $PM_{2.5}$ source impacts from each of greater than 1100 coal power plants operating in the United States in 2006 and 2011 using three approaches: (1) adjoint $PM_{2.5}$ sensitivities calculated by the GEOS-Chem CTM; (2) a wind field-based Lagrangian model called HyADS; and (3) a simple calculation based on emissions and inverse source-receptor distance. Annual individual power plants' nationwide population-weighted $PM_{2.5}$ source impacts calculated by HyADS and the inverse distance approach have normalized mean errors between 20 and 28% and root mean square error ranges between 0.0003 and 0.0005 μg m^{-3} compared with adjoint sensitivities. Reduced complexity approaches are most similar to the GEOS-Chem adjoint sensitivities nearby and downwind of sources, with degrading performance farther from and upwind of sources particularly when wind fields are not accounted for.

Keywords air pollution modeling · reduced complexity modeling · $PM_{2.5}$ · HyADS · exposure modeling

Introduction

New evidence of harm from exposure to air pollution even at the historically low levels seen in the United States motivates identification of the pollution impacts of individual sources [1, 2]. Many power plants in the United States continue to burn coal and emit harmful pollutants, and targeted air pollution regulations would benefit from evidence that accurately identifies which sources contribute disproportionately to the public exposure and health burden.

Recent public health research in epidemiology and environmental justice to these ends has employed chemical transport model (CTM) results (often combined with ground and/or satellite-based observations) to quantify exposure variability across populations on wide spatial scales [2–6]. Despite gains in computational power and algorithm efficiency, the complexity and increasing fidelity of CTMs impedes their application to research questions that would require many model runs. Recent air pollution exposure research has relied on reduced complexity approaches to

Supplementary information The online version of this article (https://doi.org/10.1038/s41370-020-0219-1) contains supplementary material, which is available to authorized users.

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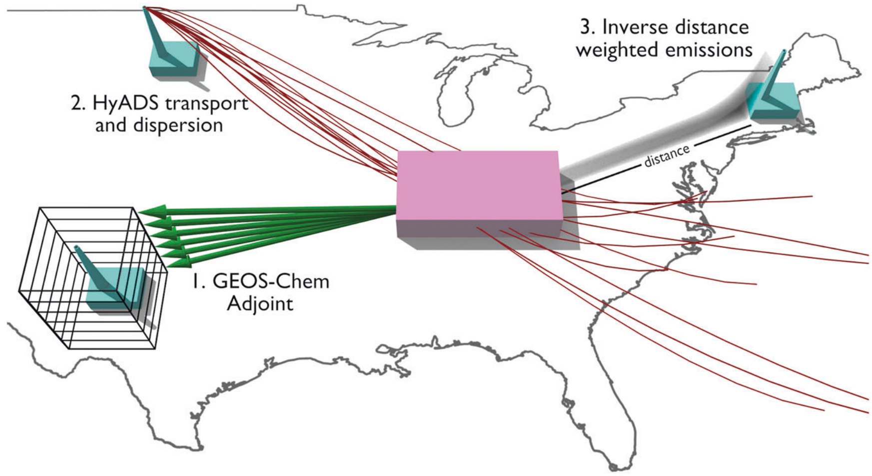

Fig. 1 Schematic of the three approaches for calculating $PM_{2.5}$ source impacts. Each of the three models calculate individual source impacts on given locations *P*, here represented by a pink rectangular prism.

quantify exposure variability in space and time [7–13]. These and other epidemiological and environmental justice applications quantify impacts from (many) individual sources over specific time periods and/or consider more refined spatial scales than commonly available in CTMs. In addition, these analyses are often conducted by researchers without the resources to effectively navigate the complexity of CTMs.

Increased use of reduced complexity approaches to quantify spatial-temporal impacts from individual sources motivates a need to evaluate their abilities to reproduce application-relevant features of CTM results [14]. This manuscript explores how two reduced-complexity characterizations of individual source exposure compare to annual GEOS-Chem adjoint sensitivities by their abilities to quantify source impact variability in time and space from each of greater than 1100 United States coal-powered electricity generating units. The vast effort required to produce the annual GEOS-Chem sensitivity results motivated this investigation. Reduced complexity models may be able to reproduce result characteristics important in exposure studies—namely, spatial-temporal variability in exposure metrics—and more readily extend them to other time and distance scales. Knowledge of the extent to which relatively simple exposure modeling approaches reproduce point source impacts modeled with a full-scale CTM will inform whether such simple calculations could propel applied research in directions currently limited by the computational burden of full-scale CTMs.

Methods

We present source impacts calculated in 2006 (2011) for each of 1256 (1177) electricity generating units comprising 503 (484) facilities with coal as their primary fuel operating in the United States. These are calculated using three unique modeling frameworks (Fig. 1)—the GEOS-Chem adjoint model employs a full suite of scientific atmospheric transport and chemistry on a 3D grid, the HYSPLIT Average Dispersion (HyADS) model employs the HYSPLIT Lagrangian wind field model, and the inverse distance weighted emissions (IDWE) model uses a simple calculation of distance and emissions. All three approaches employ coal power plant continuous emissions monitoring data in from the US Environmental Protection Agency's (US EPA) Air Markets Program [15].

A potential limitation of the two reduced complexity models—HyADS and IDWE—is that they do not, by default, estimate source impacts in terms of air pollution species or physically interpretable concentration units (e.g., μg m^{-3} or parts per million), and instead provide metrics only interpretable in terms of their relative source impacts. While such relative characterization has proven relevant in some health-outcomes and environmental justice studies [8, 11, 16], here we employ a quantitative post-processing procedure to improve the interpretability of the reduced complexity model exposure outputs. This procedure uses results from a second full-scale source impacts modeling platform—the Community Multiscale Air Quality (CMAQ)-Direct Decoupled Method (DDM) Hybrid—to build statistical models that convert raw HyADS and IDWE exposure metrics to $PM_{2.5}$ source impacts.

GEOS-chem adjoint sensitivities

The adjoint of the widely used global GEOS-Chem CTM enables the calculation of source-oriented sensitivities of model output (e.g., concentrations) to emissions perturbations [17]. We use the sensitivities described and calculated by Dedoussi et al. [18] for the nested North American domain for 2006 (using U.S. EPA's 2005 National

Emissions Inventory (NEI)) and 2011 (using the U.S EPA's 2011 NEI) [19]. The sensitivities have a horizontal resolution of 0.5° × 0.666° (~55 × ~55 km) (latitude × longitude), and a near-surface vertical resolution of ~130 m. This resolution has been shown to be adequate for application in health impacts assessments [20–22].

These adjoint sensitivities quantify state-level, annually averaged $PM_{2.5}$ population source impacts with respect to vertically-resolved emissions perturbations anywhere in the 3D domain. By multiplying the sensitivities with the corresponding emissions, we estimate population-weighted $PM_{2.5}$ source impacts ($PWSI^{Adjoint}$) of annual SO_2 emissions from each individual power plant (*i*) on each receptor location *P* (for the GEOS-Chem adjoint runs employed here, *P* will represent a state or the entire United States). While linearity is assumed with this multiplication, the individual source perturbations are estimated to be small enough for this to have negligible effects. Assessing annual impacts of thousands of individual emission sources using a conventional CTM approach is otherwise computationally (and likely numerically) impractical.

To distribute each coal unit's emissions into the horizontal grid, we assume that the emissions are contained in the grid column correspondent to the unit's coordinates. To test the hypothesis that various plume rise assumptions would affect the results, we compare results from five simple models: (a) the average sensitivity within ~650 m above surface (first five model layers; referred to hereafter as "Average"), (b) the average sensitivity between ~120 and ~650 m above surface (layers two–five; "Layers 2–5"), (c) the sensitivity at the layer of the stack height ("Stack Height"), (d) the average sensitivity of the stack height layer and the layer above it ("Stack Height +1"), and (e) the average sensitivity of the stack height layer and the two layers above it ("Stack Height +2"). Where the stack height was not available in the emissions data, we use the average of the first five layers. Dedoussi et al. included fully vertically-resolved plumes, modeled using the SMOKE model accompanying the NEI [23], when calculating the adjoint sensitivities [18].

Since observation-based ground truths for the source impacts attributable to individual coal units cannot be estimated using currently available methods, GEOS-Chem adjoint results are taken as the closest available estimate of actual impacts.

Reduced complexity approaches

As in the GEOS-Chem adjoint modeling, we employ the two reduced complexity models to calculate population-weighted source impacts for each coal-fired electricity generating unit. For these models, each unit's impact on a population in location *P* is:

$$PWSI^{m}_{P,j} = \frac{\sum_{i=p}^{N_P} PM^{m}_{2.5\,i,j} \times \text{population}_i}{\sum_{i=p}^{P} \text{population}_i}, \tag{1}$$

where $i = p \ldots N_P$ is the collection of 36 km grid centroids contained in location *P* and $PWSI^{m}_{P,j}$ is the population-weighted $PM_{2.5}$ source impact from source *j* on location *P* for reduced complexity model *m*. $PM^{m}_{2.5\ i,j}$, the $PM_{2.5}$ source impact from source *j* on receptor location *i*, is defined using one of the two reduced complexity models ($m \in$ [HyADS, IDWE]) described in the subsequent subsections. Annual Intercensal Population Estimates for Unites States Counties for 2006 and 2010 were retrieved using the *censusapi* R package (2010 was used as a proxy for 2011) and spatially aggregated to the same 36 km grid [24].

The HYSPLIT with average dispersion (HyADS) model

HyADS employs the HYSPLIT [25, 26] air parcel transport and dispersion model to identify exposure patterns from individual sources. To estimate these, HyADS initiates 100 emitted parcels at each stack location four times per day and tracks hourly locations of each parcel for 10 days using HYSPLIT—10 days approximates a conservative upper bound of sulfur's atmospheric residence time [27]. Any parcel trajectories that reach a height of zero are assumed to stop contributing to exposure thereafter. Monthly parcel locations are spatially aggregated to a 36 km grid. These are then weighted by each unit's monthly SO_2 emissions, resulting in a metric of unit-specific influence on a grid covering the United States. In a given time period, $\text{exposure}^{HyADS}_{i,j}$ is a linear combinations of emissions from each source *j* and the number of air parcels originating from that source over a grid cell *i*:

$$\text{exposure}^{HyADS}_{i,j} = \text{emissions}_j \times \text{parcels}_{i,j} \tag{2}$$

The exposure metric, defined as "HyADS emissions-weighted exposure," is interpreted as a relative metric (i.e., it does not correspond directly to individual air pollutants such as SO_2 or $PM_{2.5}$). In Henneman et al. [28], we describe the method in detail and evaluate HyADS's ability to capture annual exposure to coal-fired power plant emissions from all U.S. plants and change over time [28]. We found high annual correlation with observed sulfate and $PM_{2.5}$ coal source impacts modeled with the CMAQ-DDM Hybrid described below. Section 2.3 outlines our approach to convert the HyADS metric into $PM_{2.5}$ source impacts used for the calculation in Eq. (1).

Inverse distance weighted emissions (IDWE)

As an even more simplified approach with the most simplistic account of pollution transport, we use only distance and annual SO_2 emissions to estimate emissions exposure:

$$\text{exposure}_{i,j}^{\text{IDWE}} = \text{emissions}_j \times \text{distance}_{i,j}^{-1}, \tag{3}$$

where emmisions_j represents annual or monthly emissions from source j and $\text{distance}_{i,j}$ represents the distance between each source and centroids of a 36 km grid covering the continental United States. We refer to this approach as IDWE; Section 2.3 outlines our approach to convert this relative metric into $PM_{2.5}$ source impacts used for the calculation in Eq. (1).

Post-processing reduced complexity approaches to $PM_{2.5}$ source impacts

While raw exposure metrics estimated by HyADS and IDWE may be useful for some applications, these approaches are inherently limited by their nonphysical units and lack of interpretability relative to air quality observations and outputs of standard CTMs. It is useful, therefore, to relate the metrics to policy-relevant pollutant concentration in familiar units. By assuming that HyADS and IDWE exposure contributed to elevated $PM_{2.5}$ concentrations which is valid in particular because they are based on SO_2 emissions—atmospheric SO_2 oxidizes to sulfate, a $PM_{2.5}$ constituent—we employ an approach that adjusts these fields to $PM_{2.5}$ coal source impacts with units μg m^{-3}.

Calibrating the HyADS and IDWE fields to $PM_{2.5}$ coal impacts requires a spatially and temporally concurrent metric of coal source impacts measured in $PM_{2.5}$. As one such approximate gold standard, we employ results derived from the CMAQ model with the DDM calculated on a 36 km grid over the continental United States. The approach for creating these fields, called the CMAQ-DDM Hybrid, is detailed in full by Ivey et al. [29]. CMAQ-DDM Hybrid source impacts estimate the total $PM_{2.5}$ source impacts from all coal sources in the United States; coal power plants in our database represented 89% of total coal SO_2 emissions in 2005. While not available for a wide range of time periods, availability of CMAQ-DDM Hybrid estimates for 2005 and 2006 presents the opportunity to: (a) train the statistical calibrations described below on 1 year's worth of data from 2005, (b) evaluate the trained models using 2006 data (one of our model evaluation years) and (c) use the trained statistical model to predict HyADS and IDWE $PM_{2.5}$ source impacts in 2011, a year in which CMAQ-DDM is not available.

For HyADS and IDWE exposure fields to all emissions sources ($\sum_{j=1}^{J} \text{exposure}_j^m$), we projected raw exposure fields to match the CMAQ-DDM Hybrid grid and trained multiple models over the continental United States. The models took the form:

$$\text{PM}_{2.5}^{\text{CMAQ-DDM}} = \beta_0^m + \beta_{\text{exp}}^m \sum_{j=1}^{J} \text{exposure}_j^m + \beta_{\vec{X}}^m \vec{X} + \beta_{\text{exp},\vec{X}}^m \vec{X} \times \sum_{j=1}^{J} \text{exposure}_j^m + \epsilon^m, \tag{4}$$

where $\text{PM}_{2.5}^{\text{CMAQ-DDM}}$ is $PM_{2.5}$ coal source impacts from CMAQ-DDM Hybrid, $\vec{X}$ is the vector of meteorological variables and ϵ is assumed iid normal. We employed monthly and annual average temperature, accumulated precipitation, relative humidity, and x and y wind vectors for meteorological inputs from the North American Regional Reanalysis [30]. The raw meteorology values originally on a ~32 km grid were spatially projected to the same 36 km grid as the CMAQ-DDM Hybrid $PM_{2.5}$ source impacts.

CMAQ-DDM Hybrid results were available in 2005 and 2006. Model (4) was judged the best among four different model specification based on training in 2005 and using data from 2006 as a holdout sample to evaluate prediction error and bias (model variations and evaluation presented in Supplementary Section 1 [Supplementary Figs. 1–4]). Models for monthly impacts were obtained analogously using corresponding months in 2005 for training and 2006 as a holdout validation sample.

To calculate each unit's source impact on location i in terms of the recalibrated $PM_{2.5}$ metric, we evaluate the difference between two predictions using the trained version of Eq. (4) for HyADS and IDWE: (1) a prediction in which $\sum_{j=1}^{J} \text{exposure}_{i,j}^m = 0$ and (2) a prediction in which $\sum_{j=1}^{J} \text{exposure}_j^m = \text{exposure}_j^m$. We repeat this calculation for each source in each of 2006 and 2011, and we use the resulting outputs in Eq. (1) to calculate population-weighted source impacts on locations P ($\text{PWSI}_{P,j}^m$).

Method comparisons

We compare unit-level *PWSI*'s from HyADS and IDWE with GEOS-Chem adjoint *PWSI*'s. The GEOS-Chem adjoint model runs employed an objective function targeting annual average population-weighted $PM_{2.5}$ concentrations in individual states and on the entire United States. As such, we first compare the abilities of HyADS and IDWE to simulate annual population-weighted exposures attributable to emissions from all units on the entire country and on a group of states (Pennsylvania, Georgia, Kentucky, Wisconsin, Texas, Colorado, and California) selected to represent a range of proximities to and densities of nearby coal-fired power plants. We calculate the average distance of each state's population-weighted grid centroid from the emissions-weighted centroid

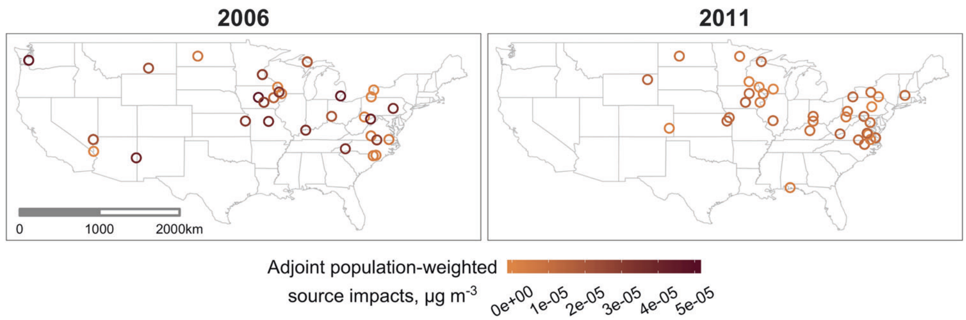

Fig. 2 50 top units in 2006 and 2011 by annual average population-weighted PM2.5 source impacts on the entire United States using the Average GEOS-Chem Adjoint results. Some colocated unites overlap in the plot.

of all power plants (termed population-emissions weighted distance; D^{pew}) as a representative distance for each location P from emissions sources:

$$D_P^{pew} = \frac{\sum_{j=1}^{J}\sum_{i=p}^{P} \text{distance}_{i,j} \times \text{population}_i \times \text{emissions}_j}{\sum_{j=1}^{J}\sum_{i=p}^{P} \text{population}_i \times \text{emissions}_j}, \tag{5}$$

where $j = 1, \ldots, J$ comprises all coal units. D^{pew} exhibits a minimum near Kentucky and a maximum in the western United States (Supplementary Fig. 5).

Evaluation metrics

We employ various evaluation metrics suggested by Emery et al. [31]: root mean square error (RMSE), mean bias (MB), normalized mean error (NME), normalized mean bias (NMB), and Pearson linear correlation (Pearson R). We add Spearman rank-order correlation (Spearman R) to quantify the ability to estimate relative importance of individual units.

Monthly source impacts

Subannual meteorology and emissions variability contributes to varying source impacts throughout the year. The GEOS-Chem adjoint sensitivities provided by Dedoussi et al. estimate annually averaged exposures with respect to emissions perturbations, preventing comparisons at subannual scales [18]. Nonetheless, we compare monthly unit-specific HyADS and IDWE population-weighted source impacts to evaluate a reduced complexity approach containing explicit information on pollutant transport (HyADS) against one rooted solely in distance and emissions levels.

Results and discussion

In this section, we compare the three exposure models by their abilities to quantify $PM_{2.5}$ source impacts from individual coal electricity generating units. The first subsection discusses annual impacts across all three metrics (GEOS-Chem adjoint, HyADS, IDWE) and the second discusses monthly impacts across HyADS and IDWE. The final sections explore limitations and implications.

Annual power plant impacts

The GEOS-Chem adjoint found the top-ranked units by population-weighted $PM_{2.5}$ source impacts $\text{PWSI}_{P=\text{US},j}^{\text{Adjoint}}$ on the entire United States (US) to be located across the eastern United States, with the densest collection of large-impact facilities located in the Ohio River Valley, consistent with recent findings that Ohio and Kentucky contribute the greatest cost per megawatt-hour on the entire country [32] (Fig. 2). Between 2006 and 2011, total population-weighted exposure on the entire United States from all power plants fell by 37%, mirroring emissions reductions from coal-fired power plants of 41%.

For the majority of the comparisons made in Fig. 3, correlations, bias, and error between GEOS-Chem adjoint and the two reduced complexity approaches were not affected by assumptions about the plume injection height in the GEOS-Chem adjoint model. Overall, annual average population-weighted $PM_{2.5}$ source impacts on the entire United States varied by less than 2.7% for our different plume height injection assumptions.

GEOS-Chem adjoint coal-fired power plant unit population-weighted $PM_{2.5}$ exposures on the entire United States are highly correlated with $\text{PWSI}_{P=\text{US},j}^{\text{HyADS}}$ and $\text{PWSI}_{P=\text{US},j}^{\text{IDWE}}$ population-weighted exposures, with Pearson correlations between 0.86 and 0.98 (Fig. 3 and Supplementary Fig. 6). NMB (RMSE) range between 2 and 23% (3.7×10^{-4} and 4.7×10^{-4} μg m^{-3}) for $\text{PWSI}_{P=\text{US},j}^{\text{HyADS}}$ and between -26 and -17% (3.0×10^{-4} and 3.0×10^{-4} μg m^{-3}) for $\text{PWSI}_{P=\text{US},j}^{\text{IDWE}}$. For individual source impacts on the entire United States, HyADS and IDWE tend to be positively and negatively biased, respectively, by similar amounts.

In Pennsylvania (PA), Kentucky (KY), and Georgia (GA)—states which are the easternmost and have the lowest characteristic population-emissions weighted distance D^{pew}—$\text{PWSI}_{P,j}^{\text{Adjoint}}$ correlations with $\text{PWSI}_{P,j}^{\text{HyADS}}$ and $\text{PWSI}_{P,j}^{\text{IDWE}}$ range from 0.62 and 0.98 in 2006 and 2011 (Fig. 3 and Supplementary Fig. 5). RMSE's range between 1.0×10^{-3}

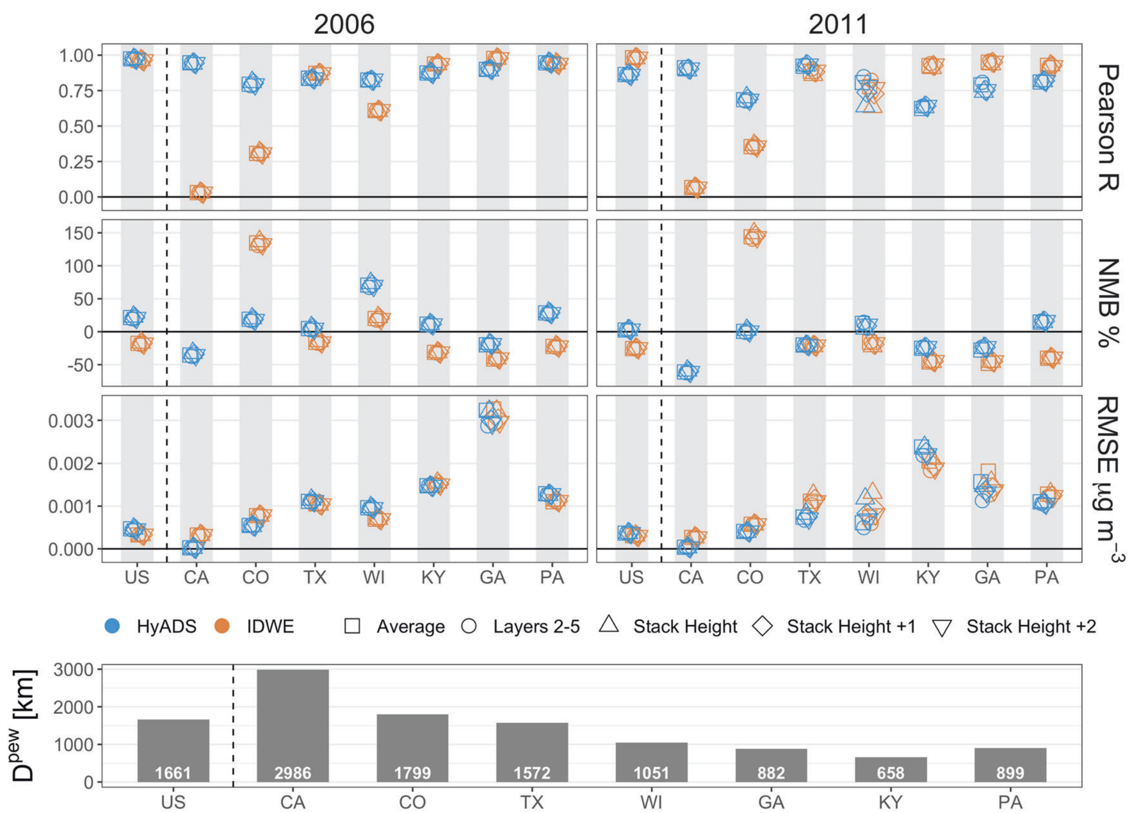

Fig. 3 Top: linear correlation (Pearson *R*), Normalized Mean Bias ($-100\% < \mathrm{NMB} < +\infty$) and root mean square error (RMSE) evaluations of $\mathrm{PWSI}_{P,j}^{\mathrm{HyADS}}$ and $\mathrm{PWSI}_{P,j}^{\mathrm{IDWE}}$ source impacts evaluated against $\mathrm{PWSI}_{P,j}^{\mathrm{Adjoint}}$ in individual states and entire United States (US). Bottom: Population and emissions weighted distance (Dpew, defined in Eq. (5)) for individual states and the entire United States. NMB for $\mathrm{PWSI}_{P,j}^{\mathrm{IDWE}}$ in CA and CO are removed because they are many times higher than the scale of the results in other states the removed values range from 1000 to 1800%.

and 3.2×10^{-3} μg m^{-3} for HyADS and between 1.1×10^{-3} and 3.3×10^{-3} μg m^{-3} for IDWE; NMB's range between −28 and 29% for HyADS and between −46 and −22% for IDWE. Overall, HyADS yields lower magnitude bias in these states, but both approaches yield similar error. This means that, on average, $\mathrm{PWSI}_{P,j}^{\mathrm{HyADS}}$ is closer to $\mathrm{PWSI}_{P,j}^{\mathrm{Adjoint}}$, but both $\mathrm{PWSI}_{P,j}^{\mathrm{HyADS}}$ and $\mathrm{PWSI}_{P,j}^{\mathrm{IDWE}}$ consistently differ from $\mathrm{PWSI}_{P,j}^{\mathrm{Adjoint}}$ by similar magnitudes. Evaluation metrics presented in the SI reinforce this narrative (Supplementary Fig. 7).

In states farther west (both farther from the largest concentrations of coal-fired power plants and more upwind on average), performance of IDWE degrades compared with HyADS. In California (CA) and Colorado (CO), $\mathrm{PWSI}_{P,j}^{\mathrm{IDWE}}$ yields very low correlation and very positive NMB relative to $\mathrm{PWSI}_{P,j}^{\mathrm{Adjoint}}$; the former is likely due to the increased importance of physical processes such as atmospheric transport and deposition in transporting pollution to these distant states, and the latter is attributable to low overall impacts in these states. Indeed, RMSE's in these states are low (below 1.0×10^{-3} μg m^{-3}) for both models and are lower for HyADS than IDWE. This trend of IDWE's decreasing performance in western states is consistent between 2006 and 2011 even with large emissions reductions from coal power plants between the years.

While the results show a relationship between population-emissions weighted distance (D^{pew}), this distance does not explain all of the variability in the evaluation metrics between GEOS-Chem adjoint results and the reduced complexity models. D^{pew} values in Pennsylvania, Georgia, Kentucky, Wisconsin, and Texas, for example, span a range of more than 1000 km, but correlations between source impacts estimated by HyADS and IDWE perform similarly across all of these states. In states further west, the benefits of the HyADS model over IDWE become clearer due to its relatively higher correlations and lower bias and error.

Monthly power plant impacts

Monthly unit Pearson NMB between HyADS and IDWE source impacts on the entire United States $\mathrm{PWSI}_{P=\mathrm{US},j}^{\mathrm{HyADS}}$ and $\mathrm{PWSI}_{P=\mathrm{US},j}^{\mathrm{IDWE}}$ range between 22 and 132% with peaks in the winter and summer. Correlations range between 0.90 and 0.99 and show little variability throughout 2006 and 2011

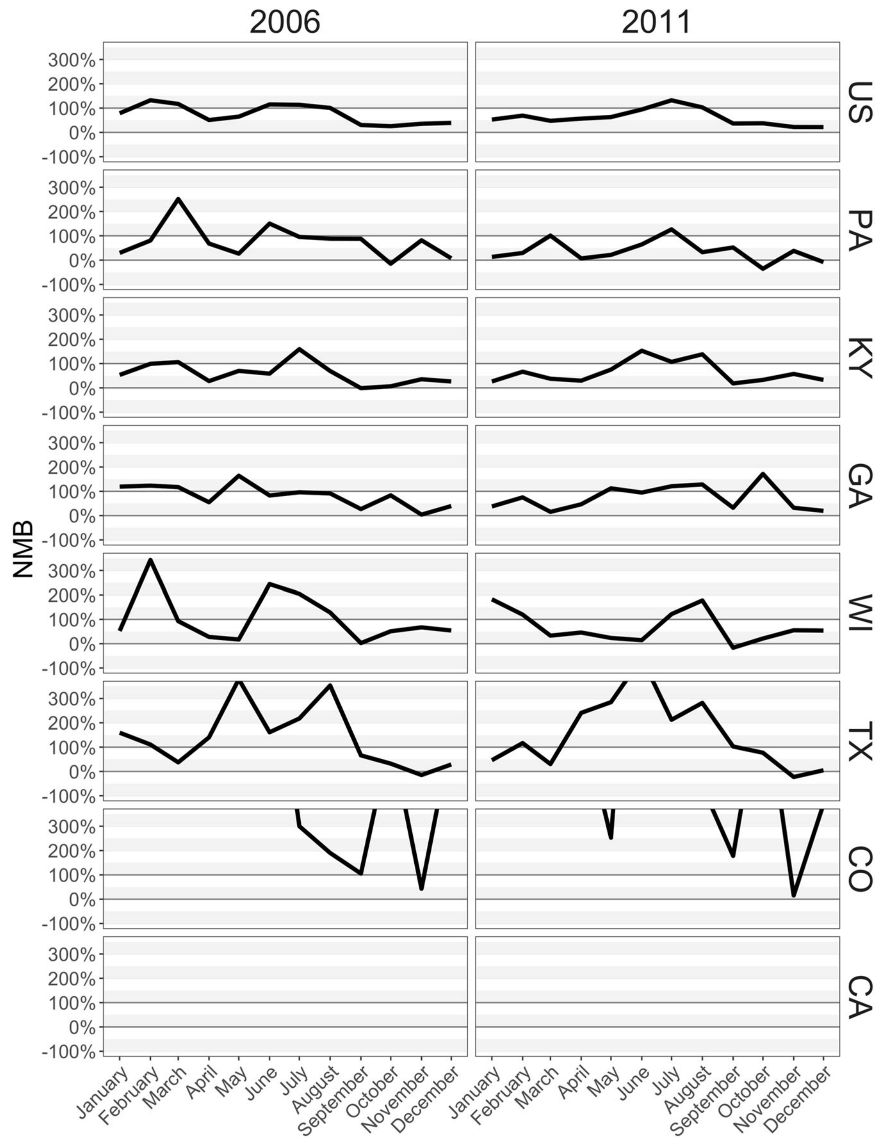

Fig. 4 Normalized mean bias ($-100\% < \mathrm{NMB} < +\infty$) of $\mathrm{PWSI}^{\mathrm{IDWE}}_{P,j}$ evaluated against $\mathrm{PWSI}^{\mathrm{HyADS}}_{P,j}$. The values in Colorado (CO) range up to 18,000% and in California range from 700% to greater than 2,000,000%.

(Supplementary Fig. 8). NMB's for the entire United States range between 22 and 132% and both NMB and NME are highest in the summertime for both years and winter 2006 (Fig. 4 and Supplementary Fig. 9), and MB shows a positive bias of about 0.01 μg m^{-3} in the summertime in both years (Supplementary Fig. 10).

Similar to the annual results, the three states near high concentrations of coal-fired power plants (Pennsylvania, Kentucky, and Georgia) exhibit similar bias, error, and correlations as source impacts on the entire United States, with poorer performance in the summer and winter than the fall and spring. Monthly NMB's average 58% in Pennsylvania, 62% in Kentucky, and 79% in Georgia (Fig. 4), with the evaluation statistics showing that IDWE is positively biased relative to HyADS in these states in most months (Supplementary Figs. 9–11).

Farther from large numbers of coal units in the east, bias, error, and correlations in Wisconsin, Texas, and Colorado between $\mathrm{PWSI}^{\mathrm{HyADS}}_{P,j}$ and $\mathrm{PWSI}^{\mathrm{IDWE}}_{P,j}$ are highly variable throughout 2006 and 2011 and generally reflect poorer agreement than in the eastern states. Monthly variability in performance increases even further west, with no monthly NMB's reaching above 79% in California. NMB and NME are extremely high in Colorado and California, though this is primarily a factor of low total impacts in these states.

Limitations

All three approaches employed here simulate point source emissions impacts using different methods that produce unique metrics. The two physical models (GEOS-Chem adjoint and HyADS) simulate different physical processes—

GEOS-Chem adjoint simulates the annual $PM_{2.5}$ source impacts attributable to SO_2 emissions perturbations using the full suite of physical and chemical processes in the atmosphere, while HyADS simulates monthly exposure to air influenced by such emissions. Atmospheric lifetimes of emitted species as dictated by deposition and chemical reactions in the atmosphere are most fully captured by the GEOS-Chem adjoint sensitivities; HyADS and IDWE largely ignore these processes, potentially explaining decreased performance of the reduced complexity models in western states.

The formulations that produce the HyADS and IDWE exposure metrics imply potentially differing policy implications but are generally limited by lack of interpretability in their raw form. In converting the raw exposure metrics to $PM_{2.5}$ source impacts, we alter the interpretation of the metrics and introduce potential bias in the conversion. The converted metrics $PWSI_{P,j}^{HyADS}$ and $PWSI_{P,j}^{IDWE}$ are interpretable as $PM_{2.5}$ source impacts—these are correlated, yet distinct, from the raw metrics. Individual source impacts calculated with the raw metrics correlate similarly with $PWSI_{P,j}^{Adjoint}$ as the converted metrics (Supplementary Fig. 12). While the raw exposure units produced by HyADS and IDWE are not directly comparable or applicable in health impact assessments that apply existing exposure-response functions, their variability across time and space may be useful in epidemiological and/or exposure variability studies [8, 11, 16].

The results here are not necessarily expected to extrapolate to other emitted species (e.g., NO_x) that could lead to elevated air pollution concentrations and adverse health impacts, and only the GEOS-Chem adjoint sensitivities approach account for chemical transformations and interactions of emitted species. More variability is expected in applying these approaches to other types of sources, such as ground-based area or mobile sources.

We chose to use a simplified plume approach for the vertical distribution of point source emissions in the GEOS-Chem adjoint sensitivities approach. While we originally hypothesized that various plume rise assumptions would have an important effect on the results, our sensitivity analysis (Fig. 3) showed that the plume rise assumption had little impact on annual correlations with the reduced complexity approaches.

Implications

We present comparisons between three approaches for estimating exposure attributable to large numbers of point sources. The results suggest that the HyADS and IDWE reduced complexity approaches are able to reproduce state-level $PM_{2.5}$ impacts from individual sources calculated by the GEOS-Chem adjoint sensitivity approach. The reduced complexity approaches perform similarly at annual time scales and for nearby, upwind sources. At longer distance scales, source impacts become more sensitive to atmospheric processes not captured by IDWE, the simplest approach. Recent evidence showing health impacts at even low $PM_{2.5}$ concentrations [1, 2] suggests that performance even in apparently clean areas has important implications on the models' potential to influence regulatory decision-making.

The monthly evaluation results highlight the importance of characterizing atmospheric transport on shorter time scales. Comparisons of the IDWE source impacts to HyADS show higher bias and error in summer and winter months than spring and fall. As in the annual evaluation, IDWE's performance degrades substantially in locations further from large groups of point sources. Dedoussi and Barrett showed the importance of monthly variability—sensitivity values during the summer months were a factor of ~4 times higher than the winter months in their analysis [33].

The GEOS-Chem CTM accounts for atmospheric processes including advective and diffusive transport, wet deposition, interaction with emissions from other sources, and background air constituents. The post-processing of the reduced complexity exposure metrics to $PM_{2.5}$ enabled us to develop, evaluate, and apply statistical parameterizations of these processes. One physical parameterization we tested—plume injection heights—turned out to have little impact on the eventual comparisons between HyADS and IDWE.

Overall, the results suggest three important factors in determining point source exposure patterns: emissions amount, source-receptor distance, and directionality relative to average transport patterns. These factors contribute to the relatively better performance of IDWE in states nearby large numbers of sources. Including information about transport—such as wind speed and direction—becomes more important at locations far from and upwind of sources. Adjusted to more complex model results that need only be run once, impacts from these reduced complexity models can be converted to physically interpretable units.

This work presents evidence that intentionally simpler alternatives to full-scale CTMs have potential to quantify population exposure to individual point source SO_2 emissions. Some processes (such as advective transport) were shown to be more important for identifying exposed areas than others (such as plume injection height), and the importance of invective transport, in particular, was shown to differ with distance and direction from source.

Data availability

Annual and monthly datasets of unit-level population-weighted $PM_{2.5}$ source impacts are available at https://github.com/lhenneman/simple_and_complex_AQ.

Code availability

We provide R code to reproduce the analyses and plots at https://github.com/lhenneman/simple_and_complex_AQ.

Acknowledgements This work was supported by research funding from NIHR01ES026217, NIHK99ES027023, EPA 83587201, and HEI 4953. Its contents are solely the responsibility of the grantee and do not necessarily represent the official views of the USEPA. Furthermore, USEPA does not endorse the purchase of any commercial products or services mentioned in the publication.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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References

1. Wei Y, Wang Y, Di Q, Choirat C, Wang Y, Koutrakis P, et al. Short term exposure to fine particulate matter and hospital admission risks and costs in the Medicare population: time stratified, case crossover study. BMJ. 2019;367:l6258.
2. Di Q, Wang Y, Zanobetti A, Wang Y, Koutrakis P, Choirat C, et al. Air pollution and mortality in the medicare population. N Engl J Med. 2017;376:2513–22.
3. Fann N, Baker KR, Fulcher CM. Characterizing the PM2.5-related health benefits of emission reductions for 17 industrial, area and mobile emission sectors across the U.S. Environ Int. 2012;49:141–51.
4. Fann N, Lamson AD, Anenberg SC, Wesson K, Risley D, Hubbell BJ. Estimating the national public health burden associated with exposure to ambient PM2.5 and ozone. Risk Anal. 2012;32:81–95.
5. Fann N, Coffman E, Timin B, Kelly JT. The estimated change in the level and distribution of PM2.5-attributable health impacts in the United States: 2005–2014. Environ Res. 2018;167:506–14.
6. Krall JR, Mulholland JA, Russell AG, Balachandran S, Winquist A, Tolbert PE, et al. Associations between source-specific fine particulate matter and emergency department visits for respiratory disease in four U.S. cities. Environ Health Perspect 2016. https://doi.org/10.1289/EHP271.
7. Abrams JY, Klein M, Henneman LRF, Sarnat SE, Chang HH, Strickland MJ, et al. Impact of air pollution control policies on cardiorespiratory emergency department visits, Atlanta, GA, 1999–2013. Environ Int. 2019;126:627–34.
8. Casey JA, Karasek D, Ogburn EL, Goin DE, Dang K, Braveman PA, et al. Coal and oil power plant retirements in California associated with reduced preterm birth among populations nearby. Am J Epidemiol. 2018. https://doi.org/10.1093/aje/kwy110/4996680.
9. Heo J, Adams PJ, Gao HO. Public health costs accounting of inorganic PM 2.5 pollution in metropolitan areas of the United States using a risk-based source-receptor model. Environ Int. 2017;106:119–26.
10. Levy JI, Baxter LK, Schwartz J. Uncertainty and variability in health-related damages from coal-fired power plants in the United States. Risk Anal. 2009;29:1000–14.
11. Mikati I, Benson AF, Luben TJ, Sacks JD, Richmond-Bryant J. Disparities in distribution of particulate matter emission sources by race and poverty status. Am J Public Health. 2018;108:480–5.
12. Strasert B, Teh SC, Cohan DS. Air quality and health benefits from potential coal power plant closures in Texas. J Air Waste Manag Assoc. 2019;69:333–50. https://doi.org/10.1080/10962247.2018.1537984.
13. Tessum CW, Apte JS, Goodkind AL, Muller NZ, Mullins KA, Paolella DA, et al. Inequity in consumption of goods and services adds to racial–ethnic disparities in air pollution exposure. Proc Natl Acad Sci. 2019;116:201818859.
14. Baker KR, Amend M, Penn S, Bankert J, Simon H, Chan E, et al. A database for evaluating the InMAP, APEEP, and EASIUR reduced complexity air-quality modeling tools. Data Br. 2020;28:104886.
15. U.S. EPA. Air Markets Program Data. 2016. https://ampd.epa.gov/ampd/.
16. Henneman LRF, Choirat C, Zigler CM. Decreases in negative health outcomes associated with coal emissions reductions between 2005 and 2012 in the United States. Epidemiology. 2019;30:477–85.
17. Henze DK, Hakami A, Seinfeld JH. Development of the adjoint of GEOS-Chem. Atmos Chem Phys 2007. https://doi.org/10.5194/acp-7-2413-2007.
18. Dedoussi IC, Eastham SD, Monier E, Barrett SRH. Premature mortality related to United States cross-state air pollution. Nature. 2020;578:261–5.
19. U.S. EPA. National Emissions Inventory. 2016. https://www.epa.gov/air-emissions-inventories/national-emissions-inventory.
20. Li Y, Henze DK, Jack D, Kinney PL. The influence of air quality model resolution on health impact assessment for fine particulate matter and its components. Air Qual Atmos Heal. 2016;9:51–68.
21. Thompson TM, Saari RK, Selin NE. Air quality resolution for health impact assessment: influence of regional characteristics. Atmos Chem Phys. 2014;14:969–78.
22. Punger EM, West JJ. 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 Heal. 2013;6:563–73.
23. U.S. EPA. SMOKE v3.5.1 User's Manual.
24. Recht H. censusapi: Retrieve Data from the Census APIs. 2019. https://cran.r-project.org/package=censusapi.
25. Draxler RR, Hess GD. An overview of the HYSPLIT_4 modelling system for trajectories, dispersion, and deposition. Aust Meteorol Mag. 1998;47:295–308.
26. Stein AF, Draxler RR, Rolph GD, Stunder BJB, Cohen MD, Ngan F. Noaa's hysplit atmospheric transport and dispersion modeling system. Bull Am Meteorol Soc. 2015;96:2059–77.
27. Barrie LA, Yi Y, Leaitch WR, Lohmann U, Kasibhatla P, Roelofs GJ, et al. A comparison of large-scale atmospheric sulphate aerosol models (COSAM): overview and highlights. Tellus, Ser B Chem Phys Meteorol. 2001;53:615–45.
28. Henneman LRF, Choirat C, Ivey CE, Cummiskey K, Zigler CM. Characterizing population exposure to coal emissions sources in the United States using the HyADS model. Atmos Environ. 2019;203:271–80.
29. Ivey CE, Holmes HA, Hu YT, Mulholland JA, Russell AG. Development of PM2.5 source impact spatial fields using a hybrid source apportionment air quality model. Geosci Model Dev. 2015;8:2153–65.
30. Kalnay E, Kanamitsu M, Kistler R, Collins W, Deaven D, Gandin L, et al. The NCEP/NCAR 40-year reanalysis project. Bull Am Meteorol Soc. 1996;77:437–71.
31. Emery C, Liu Z, Russell A, Talat Odman M, Yarwood G, Kumar N. Recommendations on statistics and benchmarks to assess photochemical model performance. J Air Waste Manag Assoc. 2016;67:582–98.

32. Dedoussi IC, Allroggen F, Flanagan R, Hansen T, Taylor B, Barrett SRH, et al. The co-pollutant cost of carbon emissions: an analysis of the US electric power generation sector. Environ Res Lett. 2019;14:094003.

33. Dedoussi IC, Barrett SRH. Air pollution and early deaths in the United States. Part II: attribution of PM2.5 exposure to emissions species, time, location and sector. Atmos Environ. 2014;99:610–7.