

Estimated Long-Term (1981–2016) Concentrations of Ambient Fine Particulate Matter across North America from Chemical Transport Modeling, Satellite Remote Sensing, and Ground-Based Measurements

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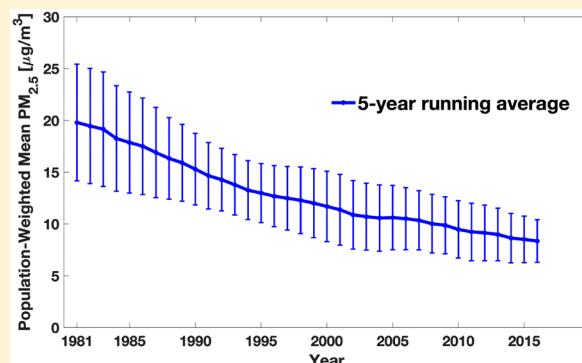
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Supporting Information

ABSTRACT: Accurate data concerning historical fine particulate matter ($PM_{2.5}$) concentrations are needed to assess long-term changes in exposure and associated health risks. We estimated historical $PM_{2.5}$ concentrations over North America from 1981 to 2016 for the first time by combining chemical transport modeling, satellite remote sensing, and ground-based measurements. We constrained and evaluated our estimates with direct ground-based $PM_{2.5}$ measurements when available and otherwise with historical estimates of $PM_{2.5}$ from PM_{10} measurements or total suspended particle (TSP) measurements. The estimated $PM_{2.5}$ concentrations were generally consistent with direct ground-based $PM_{2.5}$ measurements over their duration from 1988 onward ($R^2 = 0.6$ to 0.85) and to a lesser extent with $PM_{2.5}$ inferred from PM_{10} measurements from 1985 to 1998 ($R^2 = 0.5$ to 0.6). The collocated comparison of the trends of population-weighted annual average $PM_{2.5}$ from our estimates and ground-based measurements was highly consistent ($RMSD = 0.66 \mu\text{g m}^{-3}$). The population-weighted annual average $PM_{2.5}$ over North America decreased from $22 \pm 6.4 \mu\text{g m}^{-3}$ in 1981, to $12 \pm 3.2 \mu\text{g m}^{-3}$ in 1998, and to $7.9 \pm 2.1 \mu\text{g m}^{-3}$ in 2016, with an overall trend of $-0.33 \mu\text{g m}^{-3} \text{ yr}^{-1}$ (95% CI: -0.35 , -0.31).



1. INTRODUCTION

Ambient fine particulate matter with aerodynamic diameter less than $2.5 \mu\text{m}$ ($PM_{2.5}$) is recognized as the leading environmental risk factor for the global burden of disease, with an estimated 4.1 million (3.6 to 4.6 million) attributable deaths in 2016.¹ Long-term exposure to high $PM_{2.5}$ adversely affects human health.^{2–8} Several epidemiological studies reported adverse effects from long-term exposure at levels of $PM_{2.5}$ concentrations^{9–12} below the World Health Organization (WHO) guideline ($10 \mu\text{g m}^{-3}$ annual average), the United States standard ($12 \mu\text{g m}^{-3}$ annual average), and the Canadian standard ($10 \mu\text{g m}^{-3}$ annual average, to be reduced to $8.8 \mu\text{g m}^{-3}$ in 2020). However, the shape of the concentration–response function at these low $PM_{2.5}$ concentrations remains uncertain. Information about historical $PM_{2.5}$ concentrations across Canada and the United States is needed

to understand long-term changes in exposure and their implications for health effects research.

Understanding historical long-term exposure is complicated by the paucity of $PM_{2.5}$ monitoring sites across North America before the late 1990s and by the spatial variation of monitoring sites over time. Ground-based monitoring provides historical time series at specific points for $PM_{2.5}$, PM_{10} , and total suspended particles (TSP). Several cohort studies have attempted to infer historical PM estimates using monitoring data for urban areas in later years.^{4,13,14} A recent study by Kim et al.¹⁵ demonstrated that historical measurements of PM_{10} and TSP offer valuable information for the prediction of

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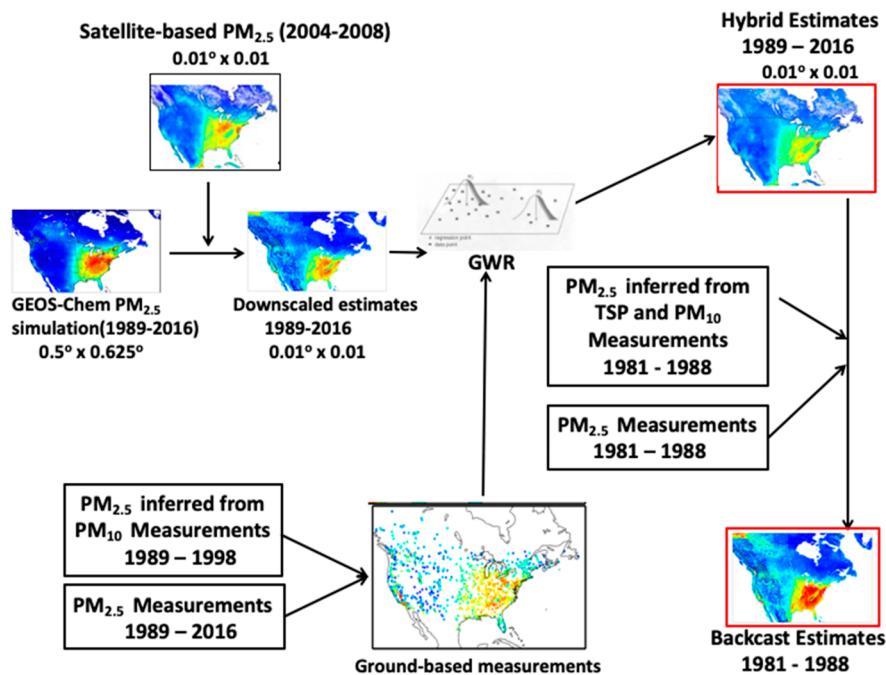


Figure 1. Overview of the estimation method.

historical PM_{2.5} concentrations across the continental United States.

Additional sources of data are available to inform estimates of historical PM_{2.5} spatial and temporal variations to improve the overall representativeness. Chemical transport modeling offers additional valuable information about historical PM_{2.5} concentrations through the representation of atmospheric processes with historical emission inventories.^{16–18} Satellite remote sensing offers a powerful additional constraint on PM_{2.5} spatial distributions,^{19,20} especially after 2002, when both the Terra and Aqua satellites were in orbit. Some studies^{21,22} have developed prediction models to estimate historical PM_{2.5} by backcasting using the ratio between PM_{2.5} and PM₁₀ or TSP observations. Other studies^{19,23–25} use land-use regression, which includes predictor variables derived from geographic information systems or combines information from other PM measurements or satellite data. However, those studies focused on either smaller regions^{21,25} or shorter durations.¹⁹

In this Article, we present historical estimates of PM_{2.5} across North America by combining information from chemical transport modeling, satellite-derived PM_{2.5} estimates, and ground-based monitoring from 1981 to 2016. These estimates can be used to assess long-term health impacts associated with low levels of PM_{2.5} throughout North America.

2. MATERIALS AND METHODS

Figure 1 provides an overview of our method to develop estimates of historical PM_{2.5} concentrations across North America by incorporating information from ground-based monitoring, chemical transport modeling, and satellite-derived PM_{2.5}. We started with a fine-resolution chemical transport model (GEOS-Chem) simulation across North America for 1989–2016. We downscaled the simulation to 0.01° × 0.01° using a satellite-derived PM_{2.5} data set.²⁰ We applied geographically weighted regression (GWR) to the downscaled simulation to incorporate information from ground-based measurements into the estimates. For 1981–1988, we relied

on information on interannual variation from ground-based measurements to backcast the gridded PM_{2.5} concentrations. Each step is described further below.

2.1. Historical Particulate Matter Monitoring Data.

We collected ground-based measurements for 1981–2016 across Canada and the United States. Canadian PM data were obtained from the National Air Pollutant Surveillance (NAPS) (<http://maps-cartes.ec.gc.ca/rnspa-naps/data.aspx?lang=en>). This database includes continuous PM measurement data, dichotomous sampler (dichot, PM₁₀, and PM_{2.5}) data, and TSP data. Instrument-specific calibrations were applied as recommended by the Canadian Council of Ministers of the Environment (CCME).²⁶ Daily PM data for the United States were obtained from the United States Air Quality System Data Mart for PM₁₀ and PM_{2.5} (https://aqs.epa.gov/aqswb/airdata/download_files.html). In addition, data from the inhalable particle network (IPN), which consisted of PM_{2.5} measurements in the early 1980s, were included. Table S1 summarizes the available monitoring data by measurement type in the selected years (1981–2016). In Canada, dichot PM_{2.5} and PM₁₀ sampling began in the mid-1980s, followed by continuous PM_{2.5} monitoring in the late 1990s. In the United States, most PM₁₀ sampling began in the late 1980s, followed by widespread PM_{2.5} monitoring in 1999. Limited PM_{2.5} measurements were available prior to 1999. Separate predictive models based on the uniform method were created for Canadian and United States monitoring data because the larger number of monitoring stations in the United States would overwhelm the Canadian dataset. Detailed information about the predictive models of inferring monthly PM_{2.5} concentrations from the historical PM₁₀ and TSP measurements is provided in the Supporting Information SI.1

2.2. Estimated Historical Gridded PM_{2.5} Data.

2.2.1. GEOS-Chem Chemical Transport Model. We use the GEOS-Chem chemical transport model (version 11-01, <http://www.geos-chem.org>), with updated historical emissions inventories and meteorological data, to consistently simulate

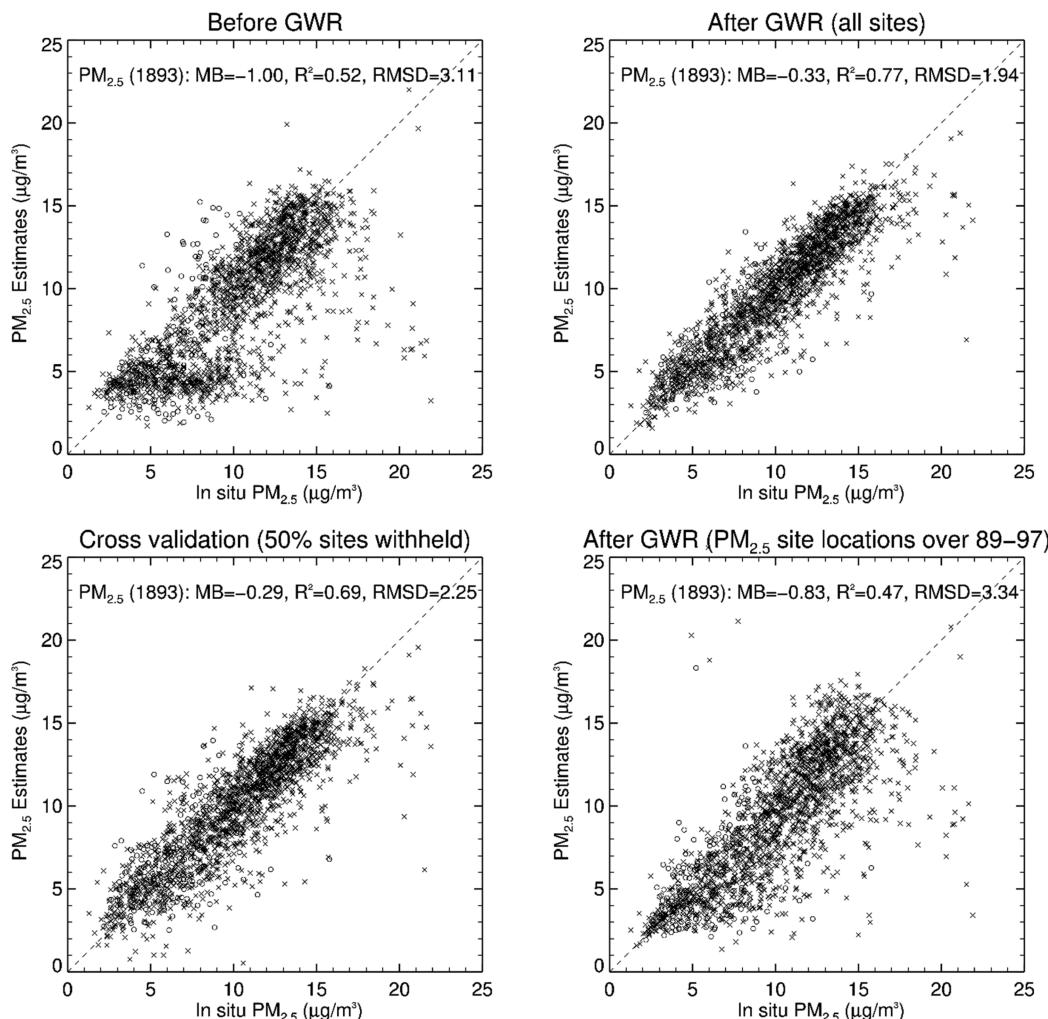


Figure 2. Comparison over 2004–2008 of mean $\text{PM}_{2.5}$ estimates with in situ measurements before (top left) and after GWR adjustment using all sites (top right), using cross-validation sites using 50% random holdout (bottom left), and using $\text{PM}_{2.5}$ sites present over 1989–1997 (bottom right). Open circles are Canadian sites, and crosses are United States sites. The number of sites is shown in brackets. Statistics shown are mean bias (MB, in $\mu\text{g m}^{-3}$), coefficient of determination (R^2), and root-mean-square difference (RMSD, in $\mu\text{g m}^{-3}$).

$\text{PM}_{2.5}$ concentrations across North America for 1989–2016. GEOS-Chem includes detailed aerosol–oxidant chemistry.^{27,28} The simulation of concentrations of $\text{PM}_{2.5}$ components includes the sulfate–nitrate–ammonium (SNA) aerosol system,^{28,29} mineral dust,³⁰ sea salt,³¹ and carbonaceous aerosol,³² with updates to black carbon³³ and secondary organic aerosol (SOA),^{34,35} including an aqueous-phase mechanism for SOA from isoprene.³⁵ Our simulation used a relative-humidity-dependent and composition-dependent fixed size distribution following Martin et al.,³⁶ with updates to organics³⁷ and mineral dust.³⁸ We drove the simulation using MERRA-2 meteorological data from NASA's Global Modeling and Assimilation Office (GMAO) with a nested resolution at $0.5^\circ \times 0.625^\circ$ over North America for 1989–2016 for which updated historical emissions were available. Anthropogenic emissions over North America were from the 2011 National Emissions Inventory (NEI2011, <http://www.epa.gov/air-emissions-inventories>) for the United States and the Criteria Air Contaminants (CAC, <http://www.ec.gc.ca/inrp-npri/>) for Canada, with historical scale factors applied to each simulating year. Black carbon (BC) and organic carbon (OC) emissions were calculated by applying sector-specific OC and BC to $\text{PM}_{2.5}$ emission ratios.^{18,39,40} Open fire emissions were from

GFED4⁴¹ for 1997–2016 and from the RETRO fire emission inventory⁴² for earlier years.

2.2.2. Creation of Historical Gridded $\text{PM}_{2.5}$ Data Set. Given our objective of a consistent data set over the entire 1989–2016 period and the lack of satellite aerosol optical depth (AOD) for the entire period, we used the 5-year average from near the middle of the period (2004–2008) of geophysical satellite-based $\text{PM}_{2.5}$ estimates (referred to as PM_{sat}),²⁰ derived from both the Terra and Aqua satellites, to downscale the GEOS-Chem model simulation (1989–2016) to a resolution relevant for exposure at $0.01^\circ \times 0.01^\circ$ following Li et al.¹⁸ We calculated the ratio between PM_{sat} and the 5 yr average (2004–2008) of GEOS-Chem simulations. Then, we used this ratio to downscale simulations in all years from 1989 to 2016. The downscaling process does not change the simulated relative temporal variation of $\text{PM}_{2.5}$ because the same scale factor was applied to all years. This downscaled estimate (referred to as PM_{scl}) contained fine-scale spatial information from satellite-derived $\text{PM}_{2.5}$ estimates (PM_{sat}) and long-term temporal information from the GEOS-Chem simulation. We evaluate the approach by excluding the satellite-based estimates.

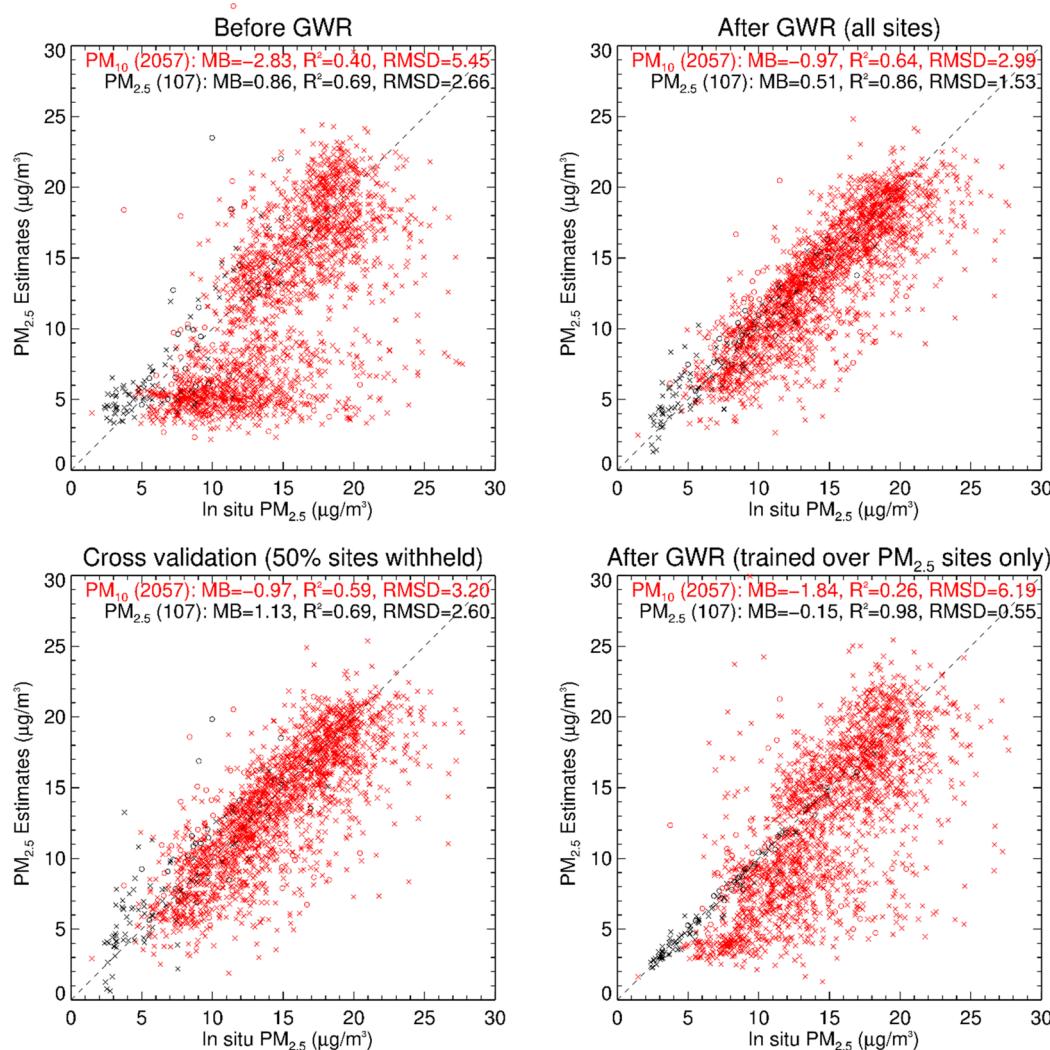


Figure 3. Comparison over 1992–1996 of mean PM_{2.5} estimates with in situ measurements before (top left) and after GWR adjustment using all sites (top right), using cross validation using 50% random holdout (bottom left), and using only PM_{2.5} sites (bottom right). Open circles are Canadian sites, and crosses are United States sites. Number of sites is shown in brackets. Comparison of PM_{2.5} (black) and PM₁₀ (red) sites is shown separately. Statistics shown are mean bias (MB, in µg m⁻³), coefficient of determination (R²), and root-mean-square difference (RMSD, in µg m⁻³).

Ground-based monitoring offers reliable information on PM_{2.5} when and where available. We used this information to constrain our estimates. We included monitor information across both the United States and Canada to produce a continuous surface for North America. Following van Donkelaar et al.,²⁰ we applied GWR to PM_{scl} over 1989–2016 using available PM_{2.5} observations and PM_{2.5} concentrations inferred from PM₁₀ observations. GWR⁴³ is a multiple regression, an extension of least-squares regression, to allow predictor coefficients to vary by choosing different spatial weighting function at several geographic locations according to their inverse-squared distance from individual observation sites. We used GWR to regress the spatial relationship between multiple predictors and the bias between PM_{2.5} estimates and PM_{2.5} measurements. Our predictors in GWR include urban land cover (ULC), subgrid elevation difference (SED), and aerosol chemical composition from GEOS-Chem simulation. We fit the GWR model at the same resolution (0.01° × 0.01°) as the downscaled PM_{2.5} estimates, which was scaled by satellite-driven PM_{2.5} following eq 1

$$(measured PM_{2.5} - estimated PM_{2.5})$$

$$= \alpha_1 ULC + \alpha_2 SED + \alpha_3 SUL + \alpha_4 NIT + \alpha_5 PrC \\ + \alpha_6 SOA + \alpha_7 DST + \varepsilon \quad (1)$$

where α_1 to α_7 represented the spatial weighted predictor coefficients for each predictor and ε is the error. ULC is the percent of urban land cover from the 500 m spatial resolution MODIS land cover type product.⁴⁴ The SED is the difference between the site elevations, which are from the ETOPO1 Global Relief Model of the National Geophysical Data Center,⁴⁵ and the annual mean elevation of the GEOS-Chem grid cell. SUL, NIT, PrC, SOA, and DST are sulfate, nitrate, primary carbon, secondary carbon, and dust, respectively, as simulated with GEOS-Chem. We conducted sensitivity tests by changing the weight of PM₁₀ observations in the GWR regression and found that a reduction by 75% of the weight of PM₁₀ best represented its uncertainty compared with direct PM_{2.5} measurements from ground-based measurements, GEOS-Chem transport model simulations, and satellite remote sensing.

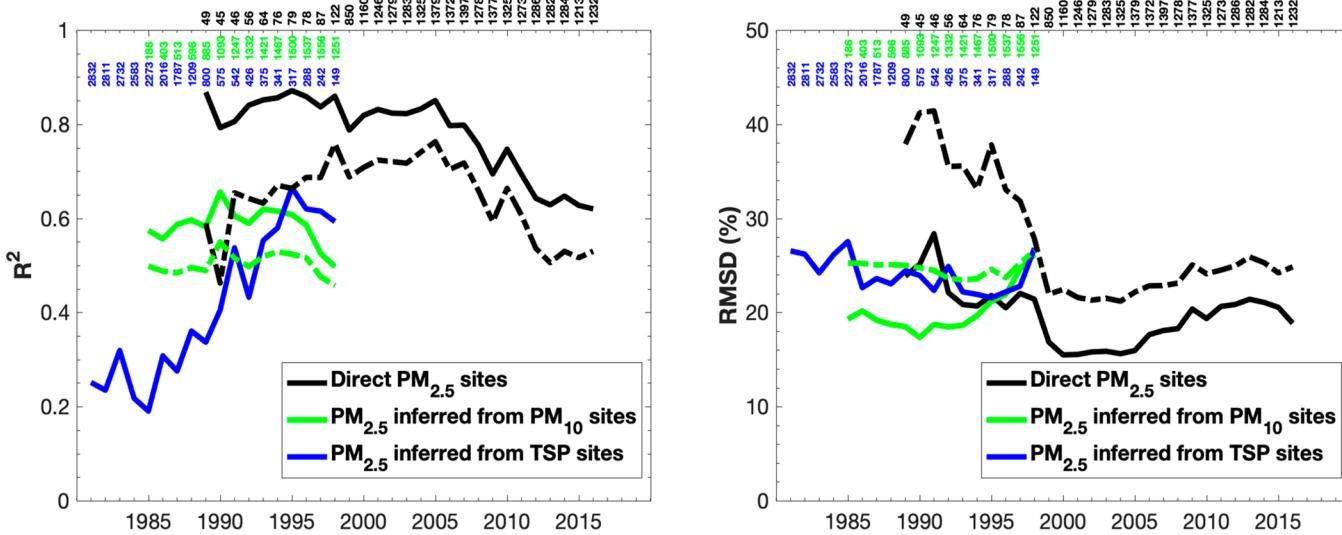


Figure 4. Statistics (R^2 and RMSD) of estimated $\text{PM}_{2.5}$ against ground-based measurements from 1981 to 2016. Solid lines indicate the performance of base estimates. Dashed lines indicate the performance of sensitivity estimates that exclude satellite remote sensing information (no blue dashed line). Numbers at the top of each panel indicate the number of monitors of direct $\text{PM}_{2.5}$ (black), $\text{PM}_{2.5}$ inferred from PM_{10} (green), and $\text{PM}_{2.5}$ inferred from TSP (blue).

For 1981–1988, reliable emission inventories were not available for the GEOS-Chem simulation. Instead, we used the information on interannual variation from ground-based measurements to backcast the gridded $\text{PM}_{2.5}$ concentrations following previous studies.^{21,22} Ground-based measurements include TSP measurements, PM_{10} measurements, and $\text{PM}_{2.5}$ measurements. Ground-based $\text{PM}_{2.5}$ concentrations inferred from TSP measurements were included for this time period because fewer than 200 PM_{10} sites existed before 1986, and even fewer $\text{PM}_{2.5}$ monitoring sites existed. For each year (e.g., 1988), we calculated the ratio between the annual mean $\text{PM}_{2.5}$ of this year and the following 3-year mean $\text{PM}_{2.5}$ (e.g., 1989–1991) for each ground-based monitoring site. We used the ratios from TSP sites as the basis, which were overwritten by the ratios from PM_{10} sites and then by the ratios from $\text{PM}_{2.5}$ sites. This ratio field from ground-based measurements was then interpolated to other grids using distance-weighted interpolation. Finally, we applied this gridded ratio field to the following 3-year mean $\text{PM}_{2.5}$ estimates to get the estimated $\text{PM}_{2.5}$ for each year. The process is described by eq 2

$$Y(t) = \gamma[Y(t+1) + Y(t+2) + Y(t+3)]/3 \quad (2)$$

where $Y(t)$ represents the $\text{PM}_{2.5}$ estimates in year t and γ is the gridded ratio field.

We evaluated the backcasting method by repeating the procedure for 2001–2008 using measurements from 2001 to 2011 for comparison with our estimates from 2001 to 2008 (Table S5).

We calculated the overall root-mean-square difference (RMSD) between the estimates and measurements for each year over 1981–2016 as a measure of uncertainty.

3. RESULTS AND DISCUSSION

We first evaluated the approach in the years when only $\text{PM}_{2.5}$ stations were used for GWR adjustment to statistically incorporate information from ground-based observations into the downscaled model results. Figure 2 shows scatter plots of 2004–2008 mean $\text{PM}_{2.5}$ from the downscaled simulation (PM_{scl}) before and after GWR adjustment versus in situ $\text{PM}_{2.5}$.

As found by van Donkelaar et al.,²⁰ the GWR model significantly reduces the mean bias (MB) and RMSD over both Canada and the United States Out-of-sample cross validation using 50% of randomly selected sites to train the GWR model exhibits significantly improved performance ($R^2 = 0.69$; $\text{RMSD} = 2.3 \mu\text{g m}^{-3}$) (bottom left panel) compared with the base case ($R^2 = 0.52$; $\text{RMSD} = 3.1 \mu\text{g m}^{-3}$). In such a holdback analysis, GWR parameter coefficients are trained using only 50% of available ground-based monitors. The withheld sites provide an independent data set with which to evaluate the quality of fused $\text{PM}_{2.5}$ estimates in areas without ground-based observation. The improvement at these independent sites suggests improvement in the GWR-adjusted surface, even at locations away from ground-based observation. The bottom right panel of Figure 2 shows the 2004–2008 mean performance of GWR-adjusted values made using only the $\text{PM}_{2.5}$ sites that were also available before 1998 (<70 sites in total), consisting mostly of remote and rural United-States-based sites. Limiting the GWR-based adjustment to only these earlier available $\text{PM}_{2.5}$ sites provided no improvement in agreement compared with the initial estimates without GWR. The negative MB in PM_{scl} ($-1.00 \mu\text{g m}^{-3}$) (top left panel) is not corrected in the adjusted estimates ($-0.83 \mu\text{g m}^{-3}$) (bottom right panel) due to a lack of representative urban and Eastern sites, which generally have higher $\text{PM}_{2.5}$ levels. Complementary information from PM_{10} sites that are representative of urban environments is necessary for early years.

Figure 3 shows scatter plots for the 1992–1996 time period to evaluate the performance of $\text{PM}_{2.5}$ inferred from PM_{10} . The top panels show that the performance of the scaled geophysical estimate is promising, with an R^2 versus $\text{PM}_{2.5}$ monitors of 0.69 that increases to 0.86 after GWR adjustment. The RMSD decreases from 2.7 to $1.5 \mu\text{g m}^{-3}$ over ~100 $\text{PM}_{2.5}$ sites in the adjusted estimates. For ~2000 PM_{10} sites, significantly improved agreement is also found after GWR adjustment. Cross validation with 50% out-of-sample sites (bottom left) further confirms the overall robustness of the approach. As found in the 2004–2008 period, using only $\text{PM}_{2.5}$ sites for

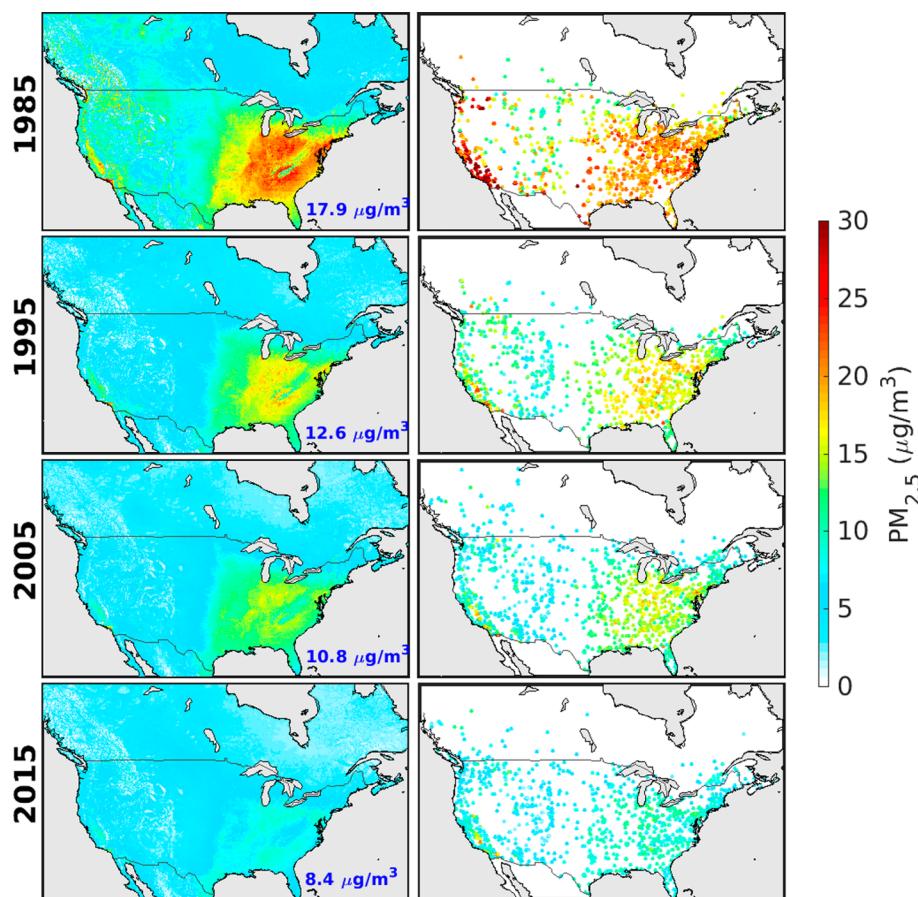


Figure 5. Estimated fine particulate matter annual means in 1985, 1995, 2005, and 2015 over North America. Left panels are estimated PM_{2.5}. Inset values in the left panel are the population-weighted average PM_{2.5} mass. Right panels indicate PM_{2.5} derived from ground-based measurements of PM_{2.5}, PM₁₀, and TSP.

GWR modeling does not improve the overall representation of the estimates, especially for PM₁₀ sites in urban areas.

Figure 4 shows the R^2 and RMSD for each year (1981–2016) of the estimates versus ground-based measurements to provide an overall assessment of uncertainty. Only PM_{2.5} data are used over 1999–2016 because sufficient PM_{2.5} measurements are available after 1999. Because the number of PM₁₀ sites significantly decreases prior to 1989 (~1000 in 1989, ~600 in 1988, ~400 sites in 1986, and <50 sites in 1984), the backcasting from 1985 to 1981 is primarily based on the trend information from TSP-based estimates and is expected to be more uncertain. The R^2 increases with the increase in PM₁₀ sites for 1985–1990. The R^2 is ~0.8 for 1989–2005 compared with PM_{2.5} sites. The relative RMSD at only PM_{2.5} sites drops from 30% in the early 1990s to <20% prior to 1999, when the PM_{2.5} measurements became more widespread. The decrease in R^2 after 2008 reflects weaker spatial PM_{2.5} gradients in recent years as PM_{2.5} levels decrease across North America. Higher RMSD errors are expected before 1999 due to more uncertainties in emission inventories as well as larger uncertainties in the monitor data used in GWR adjustments. Overall, the GWR-adjusted PM_{2.5} estimates yield an estimated error of <20% since 1999 and <30% from 1981 to 1998.

We tested how the satellite-derived PM_{2.5} data used for downscaling affected the performance of the estimated data set. Supporting Information SI.3 describes sensitivity estimates of PM_{2.5} data without satellite remote sensing. The R^2 values of these sensitivity estimates are between 0.1 and 0.2 lower than

our base estimate across all years, with larger differences in years preceding 1999 when fewer PM_{2.5} measurements were available. The relative RMSDs of the sensitivity estimates at direct observed PM_{2.5} sites are higher than the base estimates by 10 to 20%. This analysis indicates the significance of the constraints on PM_{2.5} spatial distributions offered by satellite remote sensing.

Figure 5 shows the distribution of PM_{2.5} estimates and ground-based measurements for 1985, 1995, 2005, and 2015 from this study. Enhancements in both the GWR-adjusted estimates and ground-based measurements are apparent across the Eastern United States and California. The estimated PM_{2.5} is generally consistent with ground-based measurements (Figure 4), especially with the direct PM_{2.5} measurements. PM_{2.5} concentrations decreased dramatically during the last three decades, especially in the Eastern United States.

Figure 6 shows the time series of population-weighted annual average PM_{2.5} concentrations across North America. We used gridded population estimates from the Socio-economic Data and Applications Center^{46,47} for calculating the population-weighted average (Supporting Information SI.3). The population-weighted annual average PM_{2.5} over North America decreased from $22 \pm 6.4 \mu\text{g m}^{-3}$ in 1981 to $7.9 \pm 2.1 \mu\text{g m}^{-3}$ in 2016. The linear tendency over this period is $-0.33 \pm 0.2 \mu\text{g m}^{-3} \text{ yr}^{-1}$. Both time series of the in situ measurements and estimates of population-weighted annual mean PM_{2.5} exhibit minor peaks in 2005 and 2007. The collocated comparison of the trends of population-weighted

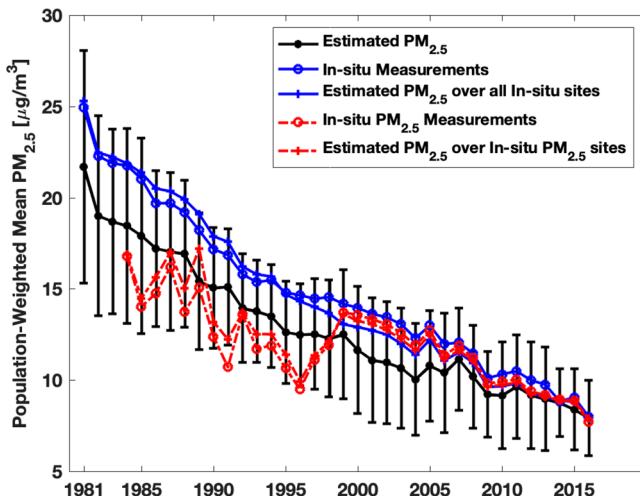


Figure 6. Time series of population-weighted average annual $\text{PM}_{2.5}$ concentrations across North America. Error bars are included for population-weighted annual mean estimated $\text{PM}_{2.5}$ concentrations.

annual average $\text{PM}_{2.5}$ from our estimates and ground-based measurements is highly consistent ($\text{RMSD} = 0.66 \mu\text{g m}^{-3}$) over 1985–1995. The population-weighted annual average $\text{PM}_{2.5}$ calculated from direct $\text{PM}_{2.5}$ sites is 20% lower than that calculated from all in situ sites, illustrating the effects of changes in monitor placement over time when assessing long-term changes in ambient $\text{PM}_{2.5}$ and the value of spatiotemporally continuous $\text{PM}_{2.5}$ estimates from this work. Larger error bars prior to 1990 reflect greater uncertainty in the TSP data set.

Figure S5 shows regional time series of the population-weighted annual average $\text{PM}_{2.5}$. Figure S6 shows regional time series of the relative percentage change of population-weighted annual average $\text{PM}_{2.5}$ concentrations using 2016 as the reference year. Northwestern North America has the most dramatic decrease for population-weighted average $\text{PM}_{2.5}$ concentrations with a factor of 2.7 decrease over 1981–2016, followed by Southeastern and Northeastern North America, with a factor of 2.4 decrease over 1981–2016. The relative changes in North Central, South Central, and Southwestern North America are similar, with a factor of 1.6 to 2.0 decrease in population-weighted $\text{PM}_{2.5}$ over 1981–2016. Overall, the spatially resolved historical $\text{PM}_{2.5}$ data set across North America reveals a factor of 1.7 decrease in population-weighted $\text{PM}_{2.5}$ over 1981–2016.

The comparison with previous estimates of historical $\text{PM}_{2.5}$ concentrations is instructive. Our estimated historical $\text{PM}_{2.5}$ concentrations from 1982–1991 in the Southeastern United States indicate a decrease of $3.9 \mu\text{g m}^{-3}$, similar to the reported decrease of $3\text{--}5 \mu\text{g m}^{-3}$ found by Parkhurst et al.²¹ We find similar large-scale reductions in historical $\text{PM}_{2.5}$ concentrations from 1981–2000, as found by Lall et al.,²² albeit with smoother temporal trends in the present study that are more consistent with Kim et al.¹⁵ The primary difference with our prior historical $\text{PM}_{2.5}$ estimates^{48–50} is that our current study spans a time period (1981–2016) that is about twice as long as our prior work by including more trend information from our GEOS-Chem simulation and includes historical ground-based measurements prior to 1999. Nonetheless, the population-weighted trends from our current data set remain within $0.03 \mu\text{g m}^{-3} \text{ yr}^{-1}$ of our prior work, indicating overall consistency, as further discussed in the Supporting Information SI.4.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: [10.1021/acs.est.8b06875](https://doi.org/10.1021/acs.est.8b06875).

Detailed description of prediction of monitoring historical $\text{PM}_{2.5}$ from measured PM_{10} and TSP, sensitivity test of estimated $\text{PM}_{2.5}$ data set without satellite remote sensing information, population data and further discussion of population-weighted $\text{PM}_{2.5}$ trends, and supporting figures for Section 3 (PDF)

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Notes

The authors declare no competing financial interest.

The annual mean estimated $\text{PM}_{2.5}$ for 1981–2016 across the North America data set has been deposited in the Zenodo Digital Repository (DOI: [10.5281/zenodo.2616769](https://doi.org/10.5281/zenodo.2616769)).⁵¹

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