Comparison of multiple PM$_{2.5}$ exposure products for estimating health benefits of emission controls over New York State, USA

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Abstract

Ambient exposure to fine particulate matter (PM$_{2.5}$) is one of the top global health concerns. We estimate the PM$_{2.5}$-related health benefits of emission reduction over New York State (NYS) from 2002 to 2012 using seven publicly available PM$_{2.5}$ products that include information from ground-based observations, remote sensing and chemical transport models. While these PM$_{2.5}$ products differ in spatial patterns, they show consistent decreases in PM$_{2.5}$ by 28%–37% from 2002 to 2012. We evaluate these products using two sets of independent ground-based observations from the New York City Community Air Quality Survey (NYCCAS) Program for an urban area, and the Saint Regis Mohawk Tribe Air Quality Program for a remote area. Inclusion of satellite remote sensing improves the representativeness of surface PM$_{2.5}$ in the remote area. Of the satellite-based products, only the statistical land use regression approach captures some of the spatial variability across New York City measured by NYCCAS. We estimate the PM$_{2.5}$-related mortality burden by applying an integrated exposure-response function to the different PM$_{2.5}$ products. The multi-product mean PM$_{2.5}$-related mortality burden over NYS decreased by 5660 deaths (67%) from 8410 (95% confidence interval (CI): 4570–12 400) deaths in 2002 to 2750 (CI: 700–5790) deaths in 2012. We estimate a 28% uncertainty in the state-level PM$_{2.5}$ mortality burden due to the choice of PM$_{2.5}$ products, but such uncertainty is much smaller than the uncertainty (130%) associated with the exposure-response function.

1. Introduction

Ambient exposure to fine particulate matter (defined as particles with less than 2.5 $\mu$m in aerodynamic diameter) is associated with mortality (Dockery et al 1993, Di et al 2017), cardiovascular (Gauderman et al 2004, Pope et al 2002, 2004, 2014), respiratory (Peng et al 2009), and other diseases (Pope and Dockery 2012). In the past several decades, efforts have been made to reduce the emissions from stationary and mobile sources in the United States (US) under federal and state regulations (US EPA 2018a). Between 2000 and 2017, the total anthropogenic emissions over the US have declined by 83%, 52%, 47%, 27%, and 7%
for SO$_2$, NO$_x$, CO, primary PM$_{2.5}$ and non-methane volatile organic compounds respectively (US EPA 2018a), which led to a 42% decrease in the national annual average PM$_{2.5}$ (US EPA 2018b). The reduction in PM$_{2.5}$ is associated with longer life expectancy (Correia et al. 2013, Fann et al. 2017), and decrease in mortality burden over recent decades (Butt et al. 2017, Wang et al. 2017, Zhang et al. 2018).

To quantify the health benefits of emission reduction, an important step is to determine the ambient concentration of ground-level PM$_{2.5}$. In general, ambient PM$_{2.5}$ is estimated using information from at least one of the following three categories: ground-based observations, atmospheric chemical transport model (CTM) simulations, and remote sensing observations. Early studies (e.g. Pope et al. 2004, Jerrett et al. 2005) relied on ground-based monitors to estimate PM$_{2.5}$ exposure. For regions without monitors, PM$_{2.5}$ distributions can be filled spatially using geostatistical interpolation techniques such as kriging (Jerrett et al. 2005, Fann et al. 2017) and inverse distance weighting (IDW, Lipsiet et al. 2011). Another approach is to build relationships between in situ observed PM$_{2.5}$ and land use, meteorological, and geospatial information using statistical methods (Henderson et al. 2007, Paciorek and Liu 2009, Beckerman et al. 2013, Wang et al. 2014, Yanosky et al. 2014), which can resolve the fine-scale PM$_{2.5}$ spatial gradient, but their skill depends on the availability of ground-based monitors (Lee et al. 2012). CTMs simulate PM$_{2.5}$ concentrations by solving the mass continuity equations for each PM component given emissions, meteorology, and topography. CTMs have been used to estimate PM$_{2.5}$ exposure and its historical or future trends nationwide (Wang et al. 2017, Zhang et al. 2018) and globally (Anenberg et al. 2010, Silva et al. 2013, Butt et al. 2017), and are especially valuable for regions where long-term ground-based measurements are sparse. However, CTMs generally have coarse spatial resolution (> 12 km), limiting their ability to characterize air pollution at local scales (Wang et al. 2016), and are subject to uncertain emissions, meteorology and chemical processes.

Space-based remote sensing products offer global coverage and more than two decades of continuous observations (Kaufman et al. 1997, King et al. 1999, Kaufman et al. 2002). Satellite retrieved aerosol optical depth (AOD), which is a measure of total light extinction by aerosol, is correlated with the column mass of aerosols (Wang and Christopher 2003, Koelmeijer et al. 2006). Satellite-derived AOD is generally incorporated into estimates of PM$_{2.5}$ in surface air in two ways: (1) forward geophysical approaches that rely on CTMs to simulate the relationship between PM$_{2.5}$ and AOD (e.g. Liu et al. 2004, van Donkelaar et al. 2006, 2014, 2016); (2) statistical approaches that either directly build a relationship between AOD and PM$_{2.5}$ (e.g. Gupta et al. 2006, Al-Hamdan et al. 2009, 2014), or add AOD as a predictor along with other land use, meteorological variables in regression models (e.g. Kloog et al. 2014, Ma et al. 2014, Just et al. 2015). Satellite-derived PM$_{2.5}$ is valuable for filling the spatial gaps over regions with sparse monitors (van Donkelaar et al. 2014, 2016), providing observational constraints to models (Anenberg et al. 2017, Lacey et al. 2017), and improving the predictive power of statistical models (Beckerman et al. 2013). However, using satellite AOD to predict PM$_{2.5}$, especially at shorter time scales, is challenging due to retrieval uncertainties (Martin 2008, van Donkelaar et al. 2012, Jin et al. 2019), missing data due to the inability to retrieve over cloud and snow (Gupta and Christopher 2008, Levy et al. 2009), and the dependence of PM$_{2.5}$-AOD relationship on aerosol speciation, vertical distributions, and aerosol optical properties (Chin et al. 2002, Gupta et al. 2006, Jin et al. 2019).

Over the US, several PM$_{2.5}$ products have become publicly available, owing to the increasing availability of observations, both in situ and space-based, and ever-growing computing capacity. However, most epidemiological studies, for practical purposes, rely on a single exposure estimate (e.g. Correia et al. 2013, Girgious et al. 2017, Al-Hamdan et al. 2018, Zhang et al. 2018). Jerrett et al. (2017) find a robust association of PM$_{2.5}$ with cardiovascular diseases using multiple PM$_{2.5}$ products, but the derived relative risk varies. A comparative study by McGuinn et al. (2017) over North Carolina finds the urban-rural difference in the relative risk varies with exposure assessment methods. However, objective assessment of the exposure models has long been challenging, mostly due to the lack of externally valid observations (Jerrett et al. 2017). To address this gap, we use independent ground-based observations to evaluate seven publicly accessible PM$_{2.5}$ products for both urban and rural environments over New York State (NYS). These products include information from ground-based observations, atmospheric models and satellite remote sensing, which cover the most commonly used and up-to-date exposure assessment methods. We then estimate decadal changes in the NYS mortality burden attributable to PM$_{2.5}$ exposure using these PM$_{2.5}$ products, and assess the extent to which health impact analyses are sensitive to the choice of exposure datasets for NYS.

2. Data and methods

2.1. PM$_{2.5}$ products

We collected seven publicly accessible PM$_{2.5}$ exposure products for NYS. These products cover the commonly used approaches to estimate PM$_{2.5}$ exposure, and most of them have been applied to health studies (table 1). Table 1 provides short names for each PM$_{2.5}$ product, along with their spatial and temporal coverage, resolution, and the data sources used to derive PM$_{2.5}$. All products span multiple years from 2002 to 2012, except the CDC WONDER product, which is only available between 2003 and 2011. We compare differences in PM$_{2.5}$ by calculating spatial, temporal and population weighted spatial root mean squared
Table 1. Summary of PM$_{2.5}$ products and ground-based observations used in this study. The spatial and temporal coverage is based on the coverage of the original dataset.

| Dataset | Short name | Spatial coverage | Temporal coverage | Spatial resolution | Temporal Resolution | Reference | Data source | Example applications in health studies |
|---------|------------|------------------|-------------------|-------------------|---------------------|-----------|-------------|--------------------------------------|
| Global Geophysical Satellite-Based PM$_{2.5}$ | Dalhousie/GL$^a$ (PM$_{2.5}$ Dal_GL) | Global | 1998–2016 | 0.01° × 0.01° | Annual | van Donkelaar et al. (2016) | US EPA AQS$^b$, MODIS$^c$, MISR$^d$ and SeaWiFS$^e$ AOD | GEOS-Chem (v9-01-03), Crouse et al. (2012), Cohen et al. (2017) |
| North America Geophysical Satellite-Based PM$_{2.5}$ | Dalhousie_NA$^b$ (PM$_{2.5}$ Dal_NA) | North America | 2000–2016 | 0.01° × 0.01° | Monthly | van Donkelaar et al. (2019) | US EPA AQS, MODIS$^c$, MISR$^d$ and SeaWiFS$^e$ AOD | GEOS-Chem (v9-01-03), None |
| Statistical Satellite-Based PM$_{2.5}$ | Emory$^d$ (PM$_{2.5}$ Emory) | NYS | 2002–2012 | 1 × 1 km$^2$ | Daily | Bi et al. (2019) | US EPA AQS, MODIS (MAIAC$^f$ AOD) | None, Gergis et al. (2017) |
| CMAQ Simulation | CMAQ (PM$_{2.5}$ CMAQ) | USA | 2002–2012 | 12 × 12 km$^2$ | Daily or Hourly | Byun and Schere (2006) | None, None | CMAQ (v4.7), Zhang et al. (2018) |
| Fused Air Quality Surface using Downscaling | FAQSD (PM$_{2.5}$ FAQSD) | USA | 2002–2012 | 12 × 12 km$^2$ | Daily | Berrocal et al. (2010, 2011) | US EPA AQS | None, Breitner et al. (2016), Hao et al. (2016), Bravo et al. (2017) |
| AQS and Remote Sensing Merged PM$_{2.5}$ | CDC WONDER$^g$ (PM$_{2.5}$ CDC) | USA | 2003–2011 | 10 × 10 km$^2$ | Daily | Al-Hamdan et al. (2014) | US EPA AQS, MODIS AOD | None, McClure et al. (2017), Al-Hamdan et al. (2017, 2018), Loop et al. (2018) |
| Inverse distance weighed AQS PM$_{2.5}$ | IDW (PM$_{2.5}$ IDW) | NYS | 1999–present | 0.1° × 0.1° | Daily | US EPA (2018c) | US EPA AQS | None, None, Lipsett et al. (2011) |
| US EPA Air Quality System | AQS (PM$_{2.5}$ AQS) | USA | 1999–present | Point observation | Daily (24 h average) | US EPA (2018c) | US EPA AQS | None, None, Lipsett et al. (2011) |
| St. Regis Mohawk Tribe Air Quality Program | SRMT (PM$_{2.5}$ SRMT) | Northern NYS New York City | 2002–2012 (with gaps) | Point observation | Daily | Benedict (2011) | US EPA AQS | None, None, Lipsett et al. (2011) |
| NYC Community Air Quality Survey | NYCCAS (PM$_{2.5}$ CAS) | NYS New York City | 2009–2016 | Point observation | 2-week average | Matte et al. (2013) | None, None, Lipsett et al. (2011) |

$^a$ The short names are mostly given as the institution of the data developers.
$^b$ The annual ground-based PM$_{2.5}$ from the global burden disease (GBD) database is used for the development of global PM$_{2.5}$. Over the US, the GBD ground-based PM$_{2.5}$ data are from the US EPA AQS network.
$^c$ MODIS: MODerate resolution imaging spectroradiometer.
$^d$ MISR: Multi-angle imaging spectroradiometer.
$^e$ SeaWiFS: sea-viewing wide field-of-view sensor.
$^f$ MAIAC: MODIS multi-angle implementation of atmospheric correction.
$^g$ The official dataset included in the Center for Disease Control and Prevention Wide-ranging ONline Data for Epidemiologic Research (CDCWONDER) database.
differences (RMSD, equations (S1))–(S3) are available online at stacks.iop.org/ERL/14/084023/mmedia), and the spatial and temporal correlation coefficients (Rt and Rτ, equations (S4) and (S5)). We define two metrics to characterize the variations in PM2.5 across multiple products: the normalized range (equation (S6)) and the uncertainty (σ0,δ, calculated from the 95% confidence interval (CI) assuming at statistical distribution; equation (S9)). Detailed methods are described in the supplementary material.

Satellite retrieved AOD products are used in four datasets, including the two Dalhousie products (Dalhousie_GL; V4.GL.02 and Dalhousie_NA; V4.NA.03), Emory and CDC WONDER, but the methods used to build the PM2.5-AOD relationship differ. The Dalhousie products use a global CTM (GEOS-Chem) to explicitly simulate the PM2.5-AOD relationship (van Donkelaar et al 2016). Although the Dalhousie products are designed for regional domains or larger, we evaluate their performance at the smaller spatial scale of a single state. The Emory product incorporates satellite AOD as a predictor along with other land use and meteorological variables to a machine learning model (random forest) (Bi et al 2019). The CDC WONDER product builds a linear regression model between satellite AOD and ground-based PM2.5, and then merges satellite-derived PM2.5 with spatially interpolated ground-based PM2.5 (Al-Hamdan et al 2014). Each of these approaches uses different AOD products (table 1). Four products include simulated PM2.5 from global or regional atmospheric chemistry models. The Dalhousie products use GEOS-Chem (v9-01-03) to simulate global distributions of PM2.5 and AOD (van Donkelaar et al 2012, Boys et al 2014, Philip et al 2014). The CMAQ simulation of PM2.5 was accessed from the US EPA Remote Sensing Information Gateway (RSIG) (US EPA, RSIG 2016). The FAQSD product fuses this CMAQ PM2.5 with AQS observations using a space-time downsampling model (Berrocal et al 2010, 2011). All products except the CMAQ simulation have been calibrated or merged with ground-based observations of 24 h average PM2.5 from the EPA Air Quality System (AQS). To assess the added value of satellite remote sensing and model, we construct another dataset that spatially interpolates the daily AQS observations within NYS using IDW.

2.2. Independent ground-based PM2.5 observations
We use ground-based observations from the NYC Community Air Quality Survey (NYCCAS) Program to evaluate these PM2.5 products over urban NYC. NYCCAS collected integrated samples for every 2-week period in each season from 2009 to 2016 at 150 distributed sites (figure S1) over NYC, which are chosen to represent a range of land use, traffic intensity and other characteristics (Matte et al 2013). While NYCCAS and filter-based AQS data are sampled with different instruments, Matte et al (2013) found that the two-week integrated PM2.5_CAS mirrors PM2.5_AQS (R2 = 0.96, slope = 1.0).

Over a remote area of upstate NY, we use ground-based measurements collected by the Saint Regis Mohawk Tribe (SRMT) Air Quality Program (Benedict et al 2011). SRMT is located in northern NYS, situated in the northwest corner of Franklin County, bordered by St. Lawrence County (figure S1). There are two SRMT sites that collect hourly PM2.5 samples continuously with a tapered element oscillating microbalance monitor during our study period of 2002–2012: one located in Saint Lawrence County (hereafter St. Lawrence Site, Latitude: 44.93 °N Longitude: 74.85 °W, AQS code: 360897001), providing data before August 2004; the other located in Franklin County (hereafter Franklin Site, Latitude: 44.98 °N Longitude: 74.69 °W, AQS code: 360337003), providing data since March 2009. Observations from these two sites are not included in the 24 h PM2.5 AQS data. The St. Lawrence Site is 37 km away from the nearest 24 h AQS monitor (code: 360893001), but this AQS monitor was discontinued in 2009. Thus, there is no operational AQS site near Franklin Site after 2010, and the evaluation at the Franklin Site represents areas far from monitors (figure S1).

2.3. Calculation of the mortality burden due to PM2.5 exposure
We estimate the mortality burden for PM2.5 products by resampling them to a common grid of 0.01° × 0.01°. We acquire the administrative boundary shapefiles from the Database of Global Administrative Areas (GADM), extract the shapefiles for NYS, and rasterize them to the 0.01° grid, so that each grid cell belongs to one county. The excess mortality attributable to ambient exposure to PM2.5 (ΔMort) is estimated using the health impact function (Zhang et al 2018):

\[ \Delta \text{Mort} = y_0 \times AF \times \text{Pop}, \]

where \( y_0 \) is the baseline mortality rate for specific diseases; Pop is exposed population age 25 years and older; AF is the attributable fraction, which is a function of the relative risk (RR):

\[ \text{AF} = 1 - 1/RR. \]

We use the RR factors from the GBD Study 2010, based on an integrated exposure-response model of Burnett et al (2014) developed from a meta-analysis:

\[ \text{For } C > C_0: \text{RR}(C) = 1 + \alpha (1 - \exp(\gamma (C - C_0)^{\delta})), \]

\[ \text{For } C < C_0: \text{RR}(C) = 1, \]

where \( C \) is the annual average ambient concentration of PM2.5; \( C_0 \) is the counter-factual level below which no additional risk is assumed; \( \alpha, \gamma, \) and \( \delta \) are fitting parameters. We acquired the RRs along with their 95% CIs for four causes of diseases, including chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), lung cancer (LC), and cerebrovascular
and ischemic stroke (STROKE) from the Global Burden of Disease Collaborative Network (2013). We use the county-level baseline mortality rate from the National Center for Health Statistics (CDC 2017) from 2002 to 2012 for each specific disease, following the definition of the GBD study (Lim et al 2012, Zhang et al 2018). We assign the annual county-level baseline-mortality to grid cells falling in the county. County-level population data for age ≥ 25 years are acquired from the CDC WONDER database. Since the population density varies spatially within a county, we distribute the county-level population data for each county by applying the spatial patterns acquired from the Gridded Population of the World (GPW, version 4) data from the Socioeconomic Data and Applications Center (SEDAC). We acquire GPW data for 2000, 2005, and 2010, and linearly interpolate them for each year from 2002 to 2012.

3. Results

3.1. Comparison across PM$_{2.5}$ products at multiple scales

Figure 1 compares the spatial distribution of annual average PM$_{2.5}$ from multiple products in 2002 and 2012 (2003 and 2011 for CDC WONDER) over New York State, with zoom-in maps for the New York City and surrounding area (upper-left of each panel). PM$_{2.5}$ products labeled in red (left half) include information from satellite remote sensing, and those without remote sensing (right half) are labeled in blue. Annual average PM$_{2.5}$ from the AQS and SRMT sites are shown as circles and triangles respectively. The boxplot shows the range of variation of annual average PM$_{2.5}$ in 2002 and 2012 (2003 and 2011 for CDC WONDER) over New York State. The box shows the inter-quartile (IQR), and the whiskers extend to show the rest of the distribution. Outliers (defined as values either 1.5 × IQR or more above the third quartile or below the first quartile) are shown as single points. The red triangles show the spatial median PM$_{2.5}$.

![Figure 1. Annual average PM$_{2.5}$ estimated from seven PM$_{2.5}$ products in 2002 and 2012 (2003 and 2011 for CDC WONDER) over New York State, with zoom-in maps for the New York City and surrounding area (upper-left of each panel). PM$_{2.5}$ products labeled in red (left half) include information from satellite remote sensing, and those without remote sensing (right half) are labeled in blue. Annual average PM$_{2.5}$ from the AQS and SRMT sites are shown as circles and triangles respectively. The boxplot shows the range of variation of annual average PM$_{2.5}$ in 2002 and 2012 (2003 and 2011 for CDC WONDER) over New York State. The box shows the inter-quartile (IQR), and the whiskers extend to show the rest of the distribution. Outliers (defined as values either 1.5 × IQR or more above the third quartile or below the first quartile) are shown as single points. The red triangles show the spatial median PM$_{2.5}$.](image-url)
data to accurately capture the temporal variability on shorter time scales. At the monthly scale, the temporal variabilities of statewide average PM$_{2.5}$, Emory, PM$_{2.5}$ IDW, and PM$_{2.5}$ FAQSD are almost identical ($R_T > 0.9$, table 2), all closely matching the variability of PM$_{2.5}$ AQS ($R_T > 0.97$). PM$_{2.5}$ Dal NA and PM$_{2.5}$ FAQSD show weaker correlations with PM$_{2.5}$, Emory, PM$_{2.5}$ IDW, and PM$_{2.5}$ FAQSD-PM$_{2.5}$, FAQSD-PM$_{2.5}$, CDC however, shows weak to no correlation with all of the other products ($R_T < 0.55$). We attribute this difference to the seasonal cycle of PM$_{2.5}$, FAQSD which differs from other products (figure 2(c)). At daily scales, PM$_{2.5}$, Emory, PM$_{2.5}$ IDW, PM$_{2.5}$ FAQSD and PM$_{2.5}$, CDC closely match ($R_T > 0.8$, figure 2(d)). Over NYC, where ground-based monitors are densely distributed, we find consistency across all products except for PM$_{2.5}$, CMAQ at all scales, with $\delta_{PM} = 10\%$ for annual average PM$_{2.5}$ after excluding PM$_{2.5}$, CMAQ (table S1).

3.2. Evaluation with independent ground-based observations

The intensive NYCCAS measurements are ideal for evaluating whether the PM$_{2.5}$ products capture the spatial patterns of PM$_{2.5}$ at the intra-urban scale. Only
six pixels cover NYC with the ~10 km resolution of PM$_{2.5,CMAQ}$, PM$_{2.5,FAQSD}$, PM$_{2.5,IDW}$ and PM$_{2.5,CDC}$ data, but they show moderate spatial correlation with NYCCAS data with $R_s$ ranging from 0.31 to 0.58 (table 2). The Emory product has a finer spatial resolution at 1 km, but it only shows slightly better spatial correlation with PM$_{2.5,CAS}$ ($R_s = 0.62$). The Dalhouse products show weak (PM$_{2.5,Dal,GL}$: $R_s = 0.33$) to no spatial correlation (PM$_{2.5,Dal,CL}$: $R_s = 0.1$) with PM$_{2.5,CAS}$, suggesting limited capability to capture the detailed spatial variability within cities, as expected by the coarser resolution inputs to those datasets. Averaging across all monitors, all products except PM$_{2.5,CMAQ}$ show strong monthly temporal correlation with PM$_{2.5,CAS}$ ($R_T > 0.8$, table 2). PM$_{2.5,CMAQ}$ is overall biased high, and shows an opposite seasonal cycle to PM$_{2.5,CAS}$ (figure S4).

To evaluate the performance of these PM$_{2.5}$ products over upstate NY, where the ground-based monitors are sparse, we use the PM$_{2.5}$ measurements from two SRMT sites (hereafter PM$_{2.5,SRMT}$). All products correlate more strongly with PM$_{2.5,SRMT}$ at the St. Lawrence site than the Franklin site. At the St. Lawrence site, PM$_{2.5,Emory}$ correlates best with the observed PM$_{2.5,SRMT}$ ($R_T = 0.89$, table 2), while PM$_{2.5,CDC}$ has the smallest RMSD$_T$ (1.52 μg m$^{-3}$, figure S2(c)). At the monthly scale, PM$_{2.5,IDW}$ and PM$_{2.5,Emory}$ are more consistent with PM$_{2.5,SRMT}$ in the cold season (November to March), and PM$_{2.5,FAQSD}$ is more consistent with PM$_{2.5,SRMT}$ from May to September, but overestimates PM$_{2.5}$ in winter by 33%. PM$_{2.5,Dal,NA}$ overestimates PM$_{2.5}$ in winter, and underestimates in the warm season (figure S4), though it captures the seasonal cycle and the temporal variability ($R_T = 0.81$). At the Franklin site, which is far from the AQS monitors, we find PM$_{2.5,Dal,GL}$ best captures the observed temporal variability ($R_T = 0.72$), though it is overall biased high by 40%. PM$_{2.5,Emory}$ agrees well with PM$_{2.5,SRMT}$ in summer, but is biased high in winter. PM$_{2.5,CMAQ}$ shows an opposite seasonal cycle that peaks in January, leading to the lowest $R_T$ value and highest RMSD$_T$ with PM$_{2.5,SRMT}$ among all products (figure S4).

### 3.3. Decadal changes in PM$_{2.5}$ and the associated mortality burden

Despite the differences in spatial resolution and PM$_{2.5}$ derivation methods, all products (excluding the PM$_{2.5,CDC}$) show significant decreases in statewide average PM$_{2.5}$, by 28% (PM$_{2.5,FAQSD}$) to 37% (PM$_{2.5,CMAQ}$) from 2002 to 2012 (figure 1). The ensemble average PM$_{2.5}$ over NYS decreased by 33% from 10.5 in 2002 to 7.0 μg m$^{-3}$ in 2012. The decreasing trend is widespread across all counties with 28%–40% decreases in the ensemble mean of county-level PM$_{2.5}$ (figure S5). The decrease in PM$_{2.5}$ is largely driven by the decrease in secondary inorganic aerosols (Boys et al 2014) attributed to anthropogenic emission reductions (US EPA, 2018a, 2018b). The annual average PM$_{2.5}$ shows larger decreases before 2009, and then levels off (figure 2(a)). The stabilization is partly due to the inter-annual variability in meteorology: the near-surface air temperature, which correlates with PM$_{2.5}$ over NYS (Porter et al 2015), is overall warmer in 2010 to 2012 than other years over NYS. Squizzato et al (2018) suggest PM$_{2.5}$ started to decline again over NYS since 2013.

The consistent decreasing trend provides evidence that PM$_{2.5}$-related air quality has improved significantly over NYS, which should decrease the PM$_{2.5}$-related mortality burden. We apply the integrated exposure-response function of Burnett et al (2014) to seven long-term PM$_{2.5}$ products. We estimate a 67% decline in the ensemble mean PM$_{2.5}$-related mortality burden (all causes combined) from 8410 (rounded to three significant figures; 95% CI due to uncertainty in relative risk factor, 4570–12 400) deaths in 2002 to 2753 (CI: 700–5790) deaths in 2012. Depending on the choice of PM$_{2.5}$ products, the estimated annual mortality burden varies from 6860 (PM$_{2.5,IDW}$, CI: 3630–10 200) to 9990 (PM$_{2.5,CMAQ}$, CI: 5780–14 300) deaths in 2002, and
1740 (PM2.5_IDW, CI: 162–4520) to 4270 (PM2.5_CMAQ, CI: 2080–7010) deaths in 2012. All products show consistent decreases in the mortality burden (figure 3). Using PM2.5_Emory yields the largest absolute decrease in mortality burden, by 5990 (CI: 4050–6860) deaths from 2002 to 2012, while using PM2.5_IDW yields the smallest decrease, by 5130 (CI: 3460–5685) deaths. In terms of relative change, using PM2.5_Emory, PM2.5_IDW, or PM2.5_DalNA yields the largest decrease in mortality burden (all three at 74%), while using PM2.5_CMAQ gives the smallest decrease (57%). The decrease in mortality burden combines decreases in PM2.5 with decreases in baseline mortality rates: the ensemble mean PM2.5-related mortality burden decreases by 46% if the baseline mortality rate is kept constant at 2002 levels, and by 36% if PM2.5 concentration is kept constant (figure S6). Among all causes, IHD is the leading cause of PM2.5-related mortality in NYS, which contributes 87% of the total mortality (figure S7). The IHD related ensemble mean mortality decreases from 6230 (CI: 3680–8830) deaths in 2002–2030 (CI: 564–4080) deaths in 2012. NYC, the most populated and polluted region in NYS, contributes about half of the total PM2.5-related mortality, where the ensemble mean PM2.5-related mortality burden decreases by 62% from 4090 (CI: 2480–5690) deaths in 2002 to 1560 (CI: 525–2730) deaths in 2012 (figure S8).

4. Discussion

4.1. Which is the ‘best’ PM2.5 product?
Determining which PM2.5 product is the ‘best’ should take into account at least three criteria—resolution, availability and accuracy (table S2). The statistical satellite-based PM2.5 product (PM2.5_Emory) has the finest spatial and temporal resolution, which captures some of the fine-scale patterns of PM2.5 by incorporating land use and traffic-related information. Our evaluation with independent observations shows PM2.5_Emory best agrees with ground-based observations for the urban area (PM2.5_CAS) and the rural external SRMT site that is closer to an AQS monitor. Jerrett et al (2017) compare the PM2.5 mortality risk estimated using multiple exposure assessment methods, and they also find the best fit with statistical land use regression model. However, PM2.5_Emory is a localized product designed for a small region (e.g. NYS in this study). The expansion of this product to wider regions is limited by the availability of ground-based monitors and consistent ancillary data. PM2.5_FAQSD and PM2.5_CDC are available for the entire US with daily resolution but at coarser spatial resolution (~10 km); we find PM2.5_FAQSD performs better over urban areas, while PM2.5_CDC performs better over remote areas (table 2). The global Dalhousie product (PM2.5_Dal_GL), while limited in temporal resolution, has the widest coverage, which is valuable for assessing the PM2.5-related global burden of disease (Cohen et al 2017). The regional Dalhousie product (PM2.5_DalNA) is available monthly for North America, and it best correlates with the rural SRMT site farther from any AQS monitor (table 2). Lee et al (2012) compare the predictive capabilities of the Dalhousie product versus spatially interpolated PM2.5, and they similarly find the Dalhousie product is more accurate than spatially interpolated data for areas 100 km or further away from monitors. In summary, there is no single product that stands out in all three criteria. Depending on the study design, the choice of PM2.5 product for epidemiological studies should reflect a trade-off among these criteria.
4.2. How do PM$_{2.5}$ exposure estimates depend on ground-based measurements?

All of the PM$_{2.5}$ products in table 1 (except PM$_{2.5,\text{CMAQ}}$) either merge AQS observations or use AQS observations to train the model, and their temporal variability is thus almost identical to PM$_{2.5,\text{AQS}}$ at AQS sites ($R > 0.97$, table 2), indicating the important role of AQS in driving the temporal variability of these products. Areas surrounding AQS monitors typically have smaller exposure uncertainties than areas where monitors are sparse (figure 4(a)). The largest uncertainty is found over northern NYS, where only one AQS monitor is available. We find all products show better correlation and smaller RMSE$_{N}$ with PM$_{2.5,\text{SRMT}}$ at the St. Lawrence site than the AQS observations ($R > 0.97$, table 2), indicating the important role of AQS in driving the temporal variability of these products. Among all products, PM$_{2.5,\text{CMAQ}}$ has the least uncertainty in exposure estimates, consistent with Jerrett et al. (2017). PM$_{2.5,\text{FAQS}}$, which fuses CMAQ with AQS data, shows a stronger correlation with other products. It should be noted that we only evaluate one single model version (CMAQ v4.7) in this study. A newer version of CMAQ (v5.2) improves the organic carbon scheme (Appel et al. 2017, Murphy et al. 2017), which is expected to improve the simulation of the seasonal cycle of PM$_{2.5}$. Despite the uncertainties, CTMs have the unique advantage of providing information on aerosol speciation (Li et al. 2014, Li et al. 2017, van Donkelaar et al. 2016). The increasing availability of observations and space-based monitors is important even in the product with the least reliance on ground-based monitors. Much of NYS has sufficient monitors: more than 90% of the state area contains at least one monitor within 100 km. PM$_{2.5}$ products derived with similar approaches are likely to have larger discrepancies over regions where ground-based monitors are sparse.

4.3. What is the value of satellite remote sensing and model simulations?

Our evaluation with independent observations from SRMT suggests the inclusion of satellite remote sensing improves the representativeness of PM$_{2.5}$ in remote areas (table 2). Of the four satellite-based products, only the statistical approach (PM$_{2.5,\text{Emory}}$) captures some of the urban spatial variability measured by NYCCAS. For the geophysical approach (PM$_{2.5,\text{Dal,NA}}$ and PM$_{2.5,\text{Dal,GL}}$), satellite AOD provides observational constraints over the globe with fine spatial resolution, which outperforms unconstrained model simulations (i.e. PM$_{2.5,\text{CMAQ}}$), though the model simulated relationship between AOD-PM$_{2.5}$ often introduces large uncertainties (Jin et al. 2019). For the AQS-Remote Sensing merged approach (PM$_{2.5,\text{CDC}}$), incorporating satellite-AOD better resolves urban-rural gradients of PM$_{2.5}$ than the product spatially interpolated from AQS observations (i.e. PM$_{2.5,\text{IDW}}$). For the statistical approach, the contribution from satellite AOD is small, less important than land use and meteorological variables (Bi et al. 2019). Bi et al. (2019) suggest larger enhancement of PM$_{2.5}$ over roads after incorporating satellite AOD, but the difference is generally small ($<0.2\,\mu g\,m^{-3}$). Other studies that use statistical models to predict PM$_{2.5}$ find that models with satellite-based AOD better predict PM$_{2.5}$ than without (Beckerman et al. 2013, Ma et al. 2014).

Among all products, PM$_{2.5,\text{CMAQ}}$ has the least accuracy, whose monthly temporal variability is almost uncorrelated with the others, suggesting that the direct use of this CTM without observational constraints in epidemiological studies will introduce larger uncertainties in exposure estimate, consistent with Jerrett et al. (2017). PM$_{2.5,\text{FAQS}}$, which fuses CMAQ with AQS data, shows a stronger correlation with other products. It should be noted that we only evaluate one single model version (CMAQ v4.7) in this study. A newer version of CMAQ (v5.2) improves the organic carbon scheme (Appel et al. 2017, Murphy et al. 2017), which is expected to improve the simulation of the seasonal cycle of PM$_{2.5}$. Despite the uncertainties, CTMs have the unique advantage of providing information on aerosol speciation (Di et al. 2016, Li et al. 2017, van Donkelaar et al. 2019), source attribution (Lelieveld et al. 2015, Silva et al. 2016a, Hu et al. 2017), and historical and future trends beyond the period of observations (Silva et al. 2016b).

4.4. Does the choice of PM$_{2.5}$ products matter for health impact analysis?

Depending on the choice of PM$_{2.5}$ products, we show the estimated mortality burden varies by 43% (equation (S6)). On average, uncertainty in exposure-response function causes 130% uncertainty (equation (S10)) in the estimated mortality burden, which is more than a factor of 4 larger than the uncertainty due to the choice of PM$_{2.5}$ products ($\delta_{PM}=28\%$). Previous studies similarly suggest uncertainties in exposure-response functions have larger impacts than uncertainty in exposure estimates (Silva et al. 2013, Ford and Heald 2016). The increasing availability of observations (both in situ and space-based) is expected to better constrain the exposure estimate, thus to further reduce uncertainty in PM$_{2.5}$ estimates. All products show consistent decreasing trends in PM$_{2.5}$, and thus decrease in the PM$_{2.5}$-related mortality burden that varies by 26% across the different products. At low PM$_{2.5}$ levels, the relationship between PM$_{2.5}$ and relative risk is approximately linear (Burnett et al. 2014, Di et al. 2017), and thus the uncertainty in the exposure-response function should not strongly influence the long-term trend in the mortality burden. However, it should be noted that the integrated model of Burnett et al. (2014) relies on pooling exposure-response functions from studies using different exposure assessment methods, and uncertainty in exposure could cause errors in building the exposure-response functions (Kiourmourtzoglou et al. 2019).
et al 2014), Hart et al 2015). Besides, we only consider the uncertainties in the ambient concentration of PM$_{2.5}$, but the measured ambient concentration differs from the true personal exposure, and such difference is expected to introduce larger biases in the estimates of relative risks (Zeger et al 2000).

5. Conclusions

We examined seven long-term (2002–2012) publicly available PM$_{2.5}$ products over NYS, which cover the most common exposure assessment methods used in health studies. We use independent ground-based observations to evaluate these products over both urban and rural environments. Among the seven products, the localized statistical satellite-based PM$_{2.5}$ data have the finest spatial and temporal resolution, and best accuracy over areas with dense monitors, while the geophysical satellite-based product correlates best with ground-based PM$_{2.5}$ at the remote site. Inclusion of satellite remote sensing improves the representativeness of PM$_{2.5}$ estimates in a remote area. All products, however, have limited capability to resolve the spatial patterns of PM$_{2.5}$ at the intra-urban scale captured by NYCCAS. While the uncertainty in the state-level PWA PM$_{2.5}$ is small ($\delta_{PM} <5\%$ after excluding outlier products), we find larger uncertainties over upstate NY where ground-based monitors are sparse. We highlight the importance of ground-based observations to reduce the uncertainties in PM$_{2.5}$ exposure estimate, as well as the independent (i.e. not used to develop the product) observations for objective assessment.

Despite these uncertainties summarized above, all products show a significant decrease of PM$_{2.5}$ by 28%–37% from 2002 to 2012, which we attribute to the implementation of emission controls. We conclude that emission controls have improved public health across NYS: the multi-product ensemble mean PM$_{2.5}$-related mortality burden decreased by 5660 deaths (67%) from 8410 (CI: 4570–12 400) deaths in 2002 to 2750 (CI: 700–5790) deaths in 2012. We estimate a 28% uncertainty in the state total mortality burden due to the choice of exposure assessment method, much less than the uncertainty in the integrated exposure–response function (130%). Overall, we conclude that exposure estimates for PM$_{2.5}$ using combinations of ground-based measurements, remotely sensed and modeled data hold substantial promise, and are rapidly becoming the state of the art for exposure assessment in epidemiological and health impact studies.

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