jwp00 | ACSJCA | JCA10.0.1465/W Unicode | research.3f (R3.6.i5 HF03:4230 | 2.0 alpha 39) 2014/07/15 09:23:00 | PROD-JCAVA | rq_3761196 | 7/24/2014 11:44:17 | 9 | JCA-DEFAULT

Environmental Science & Technology

Policy Analysis

pubs.acs.org/est

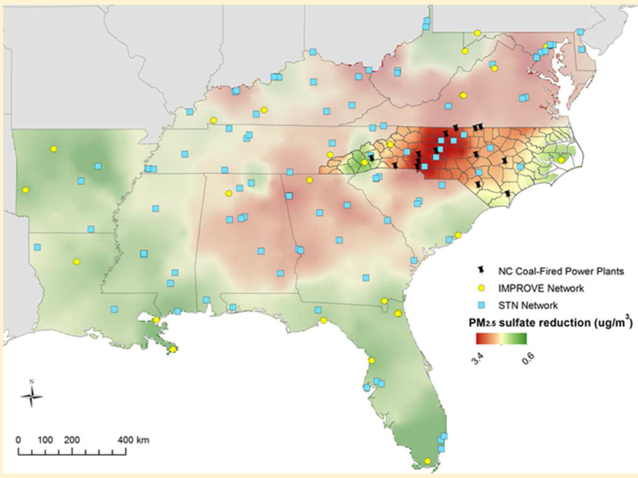
Health and Air Quality Benefits of Policies to Reduce Coal-Fired Power Plant Emissions: A Case Study in North Carolina

Ya-Ru Li* and Jacqueline MacDonald Gibson

Department of Environmental Sciences and Engineering, Gillings School of Global Public Health, University of North Carolina−Chapel Hill, Rosenau Hall, Campus Box 7431, Chapel Hill, North Carolina 27599-7431, United States

Ⓢ *Supporting Information*

ABSTRACT: We analyzed sulfur dioxide (SO_2) emissions and fine particulate sulfate ($PM_{2.5}$ sulfate) concentrations in the southeastern United States during 2002−2012, in order to evaluate the health impacts in North Carolina (NC) of the NC Clean Smokestacks Act of 2002. This state law required progressive reductions (beyond those mandated by federal rules) in pollutant emissions from NC's coal-fired power plants. Although coal-fired power plants remain NC's leading SO_2 source, a trend analysis shows significant declines in SO_2 emissions (−20.3%/year) and $PM_{2.5}$ sulfate concentrations (−8.7%/year⁻) since passage of the act. Emissions reductions were significantly greater in NC than in neighboring states, and emissions and $PM_{2.5}$ sulfate concentration reductions were highest in NC's piedmont region, where 9 of the state's 14 major coal-fired power plants are located. Our risk model estimates that these air quality improvements decreased the risk of premature death attributable to $PM_{2.5}$ sulfate in NC by about 63%, resulting in an estimated 1,700 (95% CI: 1500−1800) deaths prevented in 2012. These findings lend support to recent studies predicting that implementing the proposed federal Cross-State Air Pollution Rule (currently being evaluated by the U.S. Supreme Court) could substantially decrease U.S. premature deaths attributable to coal-fired power plant emissions.



■ INTRODUCTION

Recent regulation of particulate matter (PM) in ambient air has focused on controlling pollution sources that emit precursor pollutants. In the early 1990s, the U.S. Environmental Protection Agency (EPA) recognized that PM was particularly difficult for state and local governments to control because large amounts of PM can be produced from interstate sources of sulfur dioxide (SO_2) and nitrogen oxides (NO_x).[1] In response, the EPA developed more stringent controls on coal-fired power plant emissions in order to assist states in attaining the National Ambient Air Quality Standard (NAAQS) for PM.

The evolution of federal actions in regulation of power plants occurred in two phases. The first phase was the Acid Rain Program (ARP), which began in 1995 and affected power plants located in 21 eastern states.[2,3] The ARP implemented the first innovative cap-and-trade approach to control acid deposition. This approach sets an overall cap on SO_2 emissions but provides emission sources with flexibility in how they comply. The ARP required a 42% reduction in SO_2 emissions from power plants by 2010, relative to 1990 emissions.[3] In 2005, the second phase of controls, known as the Clean Air Interstate Rule (CAIR), began in response to the new NAAQS for $PM_{2.5}$ (PM with aerodynamic diameter ≤2.5 μm), set in 1997.[3,4] Specifically, the CAIR, developed under the "good neighbor" provision of the Clean Air Act, was designed to reduce the level of cross-border transport of $PM_{2.5}$ precursors. Similar to the ARP, the EPA also created trading programs to reduce power plant emissions of SO_2 and NO_x. CAIR affected power plants located in 27 eastern states; it set regional caps on SO_2 emissions to take effect in 2010, with lower caps to be promulgated in 2015.[4]

Since 1997, urban areas in the eastern states have experienced difficulty in attaining the new $PM_{2.5}$ standards due to transport of $PM_{2.5}$ precursors from sources in upwind states.[3] To address this challenge, EPA has proposed tighter federal limits on coal-fired power plant emissions, most recently under the Cross-State Air Pollution Rule, which would replace the CAIR. Anticipating tighter federal regulations in the future, and due to concerns about haze in the Appalachian Mountains, North Carolina (NC) moved ahead and enacted its own state regulation in 2002 to require pollutant emission reductions at coal-fired power plants.[5−7] In brief, this legislation, known as the Clean Smokestacks Act, required the state's 14 major coal-

Received: March 19, 2014
Revised: June 16, 2014
Accepted: July 21, 2014

 © XXXX American Chemical Society

fired power plants to progressively reduce NO_x emissions by 60% by 2009 and SO_2 emissions by 72% by 2013, relative to 2002 emissions. None of the states neighboring NC established similarly stringent legislation, although Maryland's Healthy Air Act required the state's coal-fired power plants to achieve 85% and 75% cuts in SO_2 and NO_x emissions, respectively, in 2013, relative to 2002 emissions.[8]

An increasing number of studies have investigated the responses of total $PM_{2.5}$ concentrations to U.S. power plant SO_2 emission reductions.[2,9,10] Few studies, however, have used observed $PM_{2.5}$ sulfate concentrations (a major component of $PM_{2.5}$, formed mainly from power plant emissions) or concentrations associated with specific pollution sources (e.g., coal-fired power plants) in their assessments of regulatory impacts on air quality[11−14] or public health.[15,16] Previous analyses using time series pollutant concentration data and/or air quality models have found that ambient $PM_{2.5}$ levels decreased over time following federally mandated SO_2 emissions reductions and suggested that the benefits of federal emissions control policies outweighed their costs. However, these previous studies have not considered the additional benefits from state policies more stringent than federal requirements. Furthermore, the previous studies assumed the health impacts of $PM_{2.5}$ are the same no matter what the source, despite mounting evidence that $PM_{2.5}$ toxicity differs by source due to differential PM composition.[17] Hence there is a need for analyses of air quality and health benefits that account for state policies and source-specific $PM_{2.5}$ toxicity.

This study evaluates the health and air quality benefits for NC of decreases in SO_2 emissions brought about by of the NC Clean Smokestacks Act. We compare observed $PM_{2.5}$ sulfate concentrations to SO_2 emissions over time and examine changes in the public health burden due to coal-fired power plant emissions using an approach that combines trend analysis,[18] modern spatiotemporal geostatistics,[19,20] and a health impact assessment accounting for the toxicity of $PM_{2.5}$ sulfate.[21] This analysis is the first to apply such an integrated assessment method to a given $PM_{2.5}$ component (i.e., $PM_{2.5}$ sulfate). We hypothesize that NC's ambient $PM_{2.5}$ levels and associated health burdens have decreased due to emission reductions achieved under the Clean Smokestacks Act.

■ MATERIALS AND METHODS

Air Pollution Data Sources and Preparation. SO_2 emissions data were acquired for 11 years, 2002 through 2012, from the EPA's National Emissions Inventory (NEI)[22] and EPA's Clean Air Markets Program Data (AMPD).[23] The NEI database collects air pollution emission data by source sectors and is updated every three years. The AMPD database provides continuous emissions monitoring (CEM) data at the facility level. To account for regional differences in emission trends, we partitioned NC into three distinct geographic regions: the coastal plain in the east, the piedmont in the center, and the mountains in the west (Supporting Information (SI), Figure S1). The CEM SO_2 emissions reported for each NC coal-fired power plant regulated by the Clean Smokestacks Act were aggregated to annual power plant SO_2 emissions from 2002 to 2012 for the whole state and each of these subregions. To evaluate impacts of interstate transport, the CEM data obtained covered not only NC but also the other 13 southeastern states and the District of Columbia (SI, Figure S1). For these other states, SO_2 emissions reported for each facility were aggregated to annual total SO_2 emissions at both the state and regional levels for the study period.

We acquired $PM_{2.5}$ sulfate monitoring data for 2002−2012 for the southeastern region from two sources: the EPA's Air Quality System (AQS)[24] and Federal Land Manager Database (FED).[25] These online databases contain data collected from two different air quality monitoring networks: the EPA Chemical Speciation Trends Network (STN or CSN) and the Interagency Monitoring of Protected Visual Environments (IMPROVE) network. Both networks collect and analyze 24 h samples every 3 days. There were a total of 133 $PM_{2.5}$ speciation monitoring sites across the southeastern US (SI, Figure S1). Over the time period analyzed, a total of 9545 and 72 112 daily measurements for NC and the whole southeastern region, respectively, were included in the analyses. Daily measurements were pooled to form annual average concentrations for trend comparison with annual SO_2 emissions at the subregional and state levels and for estimation of spatiotemporal variation in $PM_{2.5}$ sulfate concentrations.

Autoregressive Error Model for Air Pollution Trend Analysis. In order to test whether there is a statistically significant temporal trend in SO_2 emissions and $PM_{2.5}$ sulfate concentrations, trend analysis was used to model the 11 years of emission and concentration data. Autoregressive error model was employed to correct for autocorrelation of errors in time series of emissions and concentrations. A linear regression model with autoregressive errors can be written as[18]

$$y_t = x_t\beta + \varepsilon_t$$

with

$$\varepsilon_t = \phi_1\varepsilon_{t-1} - 1 + \phi_2\varepsilon_{t-2} + \cdots + \omega_t \text{and} \omega_t \sim iidN(0, \sigma^2) \quad (1)$$

where y_t is the annual emission or concentration, and x_t is the time period (i.e., years), β is the regression coefficient, ε_t is the autocorrelated regression error, φ_i is the autoregressive error model parameters, ω_t is the random error that is assumed to be normally and independently distributed with mean 0 and variance σ^2. To increase stability and interpretability of the analysis, both the emission and concentration data were log-transformed.[26] The regression errors were assumed to follow a first-order autoregressive process; that is, each error is correlated with the error immediately before it. To facilitate comparison of trends, the regression coefficient (β) and their 95% confidence intervals (CI) were presented as the percent change in emission or concentration for one year (i.e., average annual percent change) using the formula $(\exp(\beta \times 1) - 1) \times 100$.[26] The annual percent changes were intercompared and analyzed by Chow F-test.[27] This allowed us to test whether the trends differ significantly between NC and each of the other southeastern states and whether the trends differ in NC between the piedmont, mountain, and coast regions. Trends in emission and concentration were reported in tables for each of the subregions in NC and each of the southeastern states. Temporal patterns of annual emissions and concentrations were also plotted. The trend analyses were performed using SAS statistical software (version 9.2; SAS Institute Inc., Cary, NC).

Bayesian Maximum Entropy Method for Air Pollution Modeling. The Bayesian Maximum Entropy (BME) approach, an advanced function of space/time geostatistics, was employed to estimate spatiotemporal variation in $PM_{2.5}$ sulfate concentrations over the southeastern US. Complete descriptions of the

BME method have been published elsewhere.[19,28] In brief, the $PM_{2.5}$ sulfate concentration is modeled as a spatiotemporal random field (S/TRF). The BME method first applies maximum entropy theory to produce a prior probability density function (PDF) describing the S/TRF based on the general knowledge about the S/TRF. Then, BME updates this prior PDF, by employing a Bayesian conditionalization rule on the site-specific knowledge about the S/TRF, to yield a posterior PDF. The posterior PDF describes the spatiotemporal distribution of the $PM_{2.5}$ sulfate concentration, which serves as the input of air quality surfaces to be used in the health impact assessment.

In this study, the general knowledge for the S/TRF comprised the space/time mean trend and the covariance structure of the S/TRF; that is, we assumed that the ambient $PM_{2.5}$ sulfate concentration S/TRF can be modeled as the sum of a mean trend function and a residual S/TRF.[28] A mean trend is a spatiotemporal function that describes consistent patterns in the distribution of $PM_{2.5}$ sulfate concentrations, and this function was characterized by an additive space/time mean trend model. The mean trend was then subtracted from the original $PM_{2.5}$ sulfate concentration S/TRF to yield the residual $PM_{2.5}$ sulfate concentration S/TRF. The residual field is a spatiotemporal covariance function that describes the spatiotemporal variability of $PM_{2.5}$ sulfate concentrations that could not be explained by the mean trend function. We estimated values of the covariance function for different classes of spatial and temporal differences between any two space/time points, and then fitted a space/time covariance model to these estimated values.

The site-specific knowledge included hard data (accurate measures) and soft data (measures with uncertainty).[28] Since we were concerned with long-term health effects of $PM_{2.5}$ sulfate exposure, the annual average concentration was selected as the indicator of chronic exposure to $PM_{2.5}$ sulfate. Hard and soft data for yearly average concentration were constructed to account for uncertainty associated with the calculation of a yearly concentration from an incomplete set of daily measurements.[29,30] In this study, the yearly average concentration at any date t was defined as the average of daily measurements over the 365 days preceding date t. If the set of intended daily measurements for the 365 days prior to t was at least 75% complete (the number of intended measurements was 121 as the sampling frequency was every 3 days), the yearly average value calculated for date t was considered hard. Otherwise, the calculated value was considered soft. Soft data were assumed and characterized by the PDF of a normal distribution truncated below zero, as yearly concentrations cannot be negative. A full numerical description for constructing the hard and soft data is provided in the SI.

Since our general knowledge about the S/TRF consisted of its mean trend and covariance structure, the BME equation can be written as[28]

$$f_K(x_k) = A^{-1} \int dx f_S(x) f_G(x) \tag{2}$$

where x_k is the BME estimated residual $PM_{2.5}$ sulfate concentration at estimation points, x is the residual $PM_{2.5}$ sulfate concentrations at mapping points (i.e., the union of the hard/soft data points and the estimation point), A is a normalization constant, f_S is the truncated normal PDF characterizing the uncertainty of soft data, f_G is the prior PDF obtained from the general knowledge, and f_K is the posterior PDF describing residual $PM_{2.5}$ sulfate concentration at the estimation point. Ultimately, the expected value and corresponding estimation error variance of $PM_{2.5}$ sulfate concentration estimates were obtained by adding back the mean trend to the BME posterior PDF for residual $PM_{2.5}$ sulfate concentration. The BME interpolation was produced using the BMElib package[31] implemented by MATLAB software (R2011a; MathWorks, Natick, MA). Changes in concentrations across space and time were mapped for the southeastern U.S. using ArcGIS software (version 10.0; ESRI, Redlands, CA).

Estimation of Health Impacts. Health impact functions enable the quantification of health outcomes from changes in population exposure to a pollutant of interest. A log−linear function can be written as[32]

$$\Delta y = (\mathrm{AF})y_0 = (1 - e^{-\beta\Delta x})I_0 P \tag{3}$$

where AF is the attributable fraction (the fraction of observed adverse health outcomes that could be prevented if the pollutant exposure were reduced by Δx), y_0 is the baseline incidence of the health outcome, β is the coefficient of association between pollutant concentration and health outcome [i.e., the concentration−response (C−R) function], Δx is the estimated air pollution change, I_0 is the baseline incidence rate of the health outcome, P is the size of the exposed population, and Δy is the estimated change in the health outcomes due to the change in pollutant exposure.

There is growing evidence that PM toxicity varies by particle composition, but accounting for these differences in human health impact assessments remains quite challenging. Hence we conducted our impact analysis in two ways—one with $PM_{2.5}$ sulfate-specific C−R functions and another using the conventional approach with one C−R function for total $PM_{2.5}$ mass—to evaluate whether using chemical-specific risk coefficients changes our health impact estimates. Epidemiological literature for $PM_{2.5}$ sulfate- and total $PM_{2.5}$-attributed C−R functions for premature mortality was examined to summarize the association between fine particulate concentration and health (SI, Table S1). In this study C−R functions from prospective cohort studies were selected to estimate the long-term mortality risks of $PM_{2.5}$ sulfate[33−35] and total $PM_{2.5}$.[36−38] To obtain summary estimates of the health impacts, we pooled estimates of C−R functions from different studies into a single estimate using an inverse variance weighting approach, which takes into account the uncertainty of each estimate (SI, Table S1).

County-level population and mortality data for 2002 and 2012 were acquired from the Centers for Disease Control and Prevention's WONDER database.[39] The baseline incidence rates of premature mortality were age-adjusted based on the year 2000 US standard population, and the adjusted rates in 2010 (the latest rate) were used as a surrogate for baseline rates in 2012. We estimated exposures of $PM_{2.5}$ sulfate at the county level for 2002 and 2012 using the BME method and assumed that all individuals within a county experienced the same changes in exposure levels. Because we were concerned about the health impacts due to $PM_{2.5}$ sulfate from man-made sources, the estimated air pollution change in each county were the difference between the estimated $PM_{2.5}$ sulfate level and the estimated natural background level of $PM_{2.5}$ sulfate. We assumed a background level for nonanthropogenic $PM_{2.5}$ sulfates of 0.2 $\mu g/m^3$, which is the EPA estimate of background $PM_{2.5}$ sulfates for the eastern US.[40,41]

dx.doi.org/10.1021/es501358a | Environ. Sci. Technol. XXXX, XXX, XXX−XXX

Due to the substantial population growth in NC over the study period, we examined change in fractions, in addition to numbers, of deaths attributable to $PM_{2.5}$ sulfate (i.e., AF) between 2002 and 2012. The health impacts of $PM_{2.5}$ sulfate exposure were estimated at the county level by aggregating AF and number of deaths within county boundaries. To assess uncertainty in health impact estimates, we assumed that C-R functions and $PM_{2.5}$ sulfate exposure concentrations were normally and lognormally distributed, respectively. Monte Carlo simulation with an uncertainty sample size of 1000 was used to generate a 95% CI for each mean incidence estimate. The Monte Carlo simulations of health impacts were conducted using Analytica software (version 4.3; Lumina Decision Systems Inc., Los Gatos, CA), and mean estimates were mapped using ArcGIS software (version 10.0; ESRI, Redlands, CA).

■ RESULTS

Trends in SO_2 Emissions. Over the past decade, coal-fired power plants remained the dominant SO_2 source in NC and more generally in the southeastern U.S., although their contribution to total SO_2 emissions declined gradually (SI, Figure S2). In NC, the percentage of SO_2 emissions from coal-fired power plants decreased from 84% in 2002 to 64% in 2011. In contrast, in the southeastern US, coal-fired power plants' contribution was relatively stable over the same period, with percentages ranging between 66% and 76% of SO_2 emissions.

Since 2002, the major power plants regulated by the NC Clean Smokestacks Act have reduced their SO_2 emissions significantly (Table 1 and Figure 1). The Act set caps on power plant SO_2 emissions for 2009 and 2013; therefore, there was a steep decline from 2007 to 2009 and a further decrease after 2010. On average, annual SO_2 emissions from these power plants decreased by over 20% per year (−20.3% year^{-1}). Between 2002 and 2012, the annual power plant SO_2 emissions decreased from 459.7 thousand tons to 53.5 thousand tons—a reduction of nearly 90% (−88.4%). Most of the state's coal-fired power plants are in the piedmont region (SI, Figure S1), and the emissions reduction rate in this region was significantly faster (Chow $p < 0.05$) than in the coast and mountain regions. Specifically, emissions from these piedmont-located power plants decreased by about 14−35% each year except for in one plant, where the emissions decreased by 8% per year (data not shown). Total SO_2 emissions were also reduced in the Southeast over the same time period (Figure 2; SI, Table S2) but at a lower average rate (−13.6% year^{-1}) than in NC. The Chow test results further indicate that emissions decreased significantly faster (Chow $p < 0.05$) in NC than in its neighboring states (Georgia, South Carolina, Tennessee, and Virginia)—none of which had enacted legislation comparable to the NC Clean Smokestacks Act. Among other surrounding states in the Southeast, it appears that Maryland had a higher (but not significantly different) reduction rate (−22.6% year^{-1}) than NC, an indication that Maryland Healthy Air Act also achieved substantial emission reductions from power plants. Conversely, temporal trends in emissions did not vary significantly in some states, such as Arkansas and Louisiana, suggesting that flexibility offered by the federal trading programs might allow emissions to increase or to remain unchanged in some areas while decreasing in others.

Table 1. Annual (Mean and 95% CI) and Overall Percent Changes by Region for SO_2 Emissions and $PM_{2.5}$ Sulfate Concentrations (2002−2012)

pollutant trend	region	annual percent change (% year^{-1})	Chow p-value[a]	overall percent change[b] (%)
SO_2 emission	North Carolina	−20.3 (−27.0, −13.1)		−88.4
	Coast	−7.0 (−11.8, −1.9)	<0.05	−63.3
	Mountain	NS[c]	<0.05	−89.1
	Piedmont	−22.9 (−30.6, −14.3)		−91.1
$PM_{2.5}$ sulfate concentration	North Carolina	−8.7 (−12.3, −5.1)		−60.1
	Coast	−8.2 (−11.3, −5.1)	<0.05	−58.7
	Mountain	−8.8 (−12.4, −5.1)	<0.05	−59.8
	Piedmont	−9.5 (−12.8, −6.1)		−63.8

[a]Chow test was used to analyze whether the annual percent changes differ significantly in NC between the piedmont and mountain/coast regions. [b]Overall percent change was defined as the overall change of mean value (emission or concentration) from 2002 to 2012 using the formula $(\mathrm{Value}_{2002} - \mathrm{Value}_{2002})/\mathrm{Value}_{2002} \times 100$. [c]NS: Not significant at the 5% level ($p \geq 0.05$).

Trends in $PM_{2.5}$ Sulfate Concentrations. In accordance with SO_2 emission trends, the temporal trends in $PM_{2.5}$ sulfate concentrations demonstrated considerable reductions over the past decade (Table 1 and Figure 1). The average annual decrease in $PM_{2.5}$ sulfate in NC was around 9% per year (−8.7% year^{-1}), and the trend was statistically significant. As Figure 1 shows, this downward trend matched well with the period when the major emission cuts from the state's power plants occurred. The statewide annual average level decreased from 4.2 $\mu g/m^3$ in 2002 to 1.7 $\mu g/m^3$ in 2012, corresponding to an overall decrease of 60%. Again, the annual levels decreased significantly faster (Chow $p < 0.05$) in the piedmont than in other regions. Annual $PM_{2.5}$ sulfate concentrations also decreased in other southeastern states at rates of 5−10% per year (SI, Table S2).

Bayesian Maximum Entropy Estimation of $PM_{2.5}$ Sulfate. Figure 3 shows estimated annual mean $PM_{2.5}$ sulfate concentrations in 2002 and 2012 for the southeastern U.S. These maps illustrate the considerable declines in $PM_{2.5}$ sulfate concentrations from 2002 to 2012 in response to large-scale SO_2 emission reductions across the southeastern US. Temporal variations were substantial, but spatial patterns were generally consistent across years. High $PM_{2.5}$ sulfate concentrations tend to occur in areas where SO_2 emission densities are high. For example, concentrations were higher in the piedmont region of NC as the majority of coal-fired power plants are located in this region. Possibly due to the regulatory efforts of SO_2 emission reductions, the highest estimated $PM_{2.5}$ sulfate reductions between 2002 and 2012 also occurred in the central piedmont (SI, Figure S3), which is consistent with results from our trend analysis.

Statewide Premature Mortality Health Impacts. Consistent with the temporal trend in $PM_{2.5}$ sulfate concentrations, the annual percentage of premature deaths attributable to $PM_{2.5}$ sulfate exposure declined significantly from 2002 to 2012 (Table 2). Further, the health impact estimates are substantial regardless of the choice of C-R function on which they are based. According to the $PM_{2.5}$ sulfate risk function, the attributable fraction of all-cause deaths

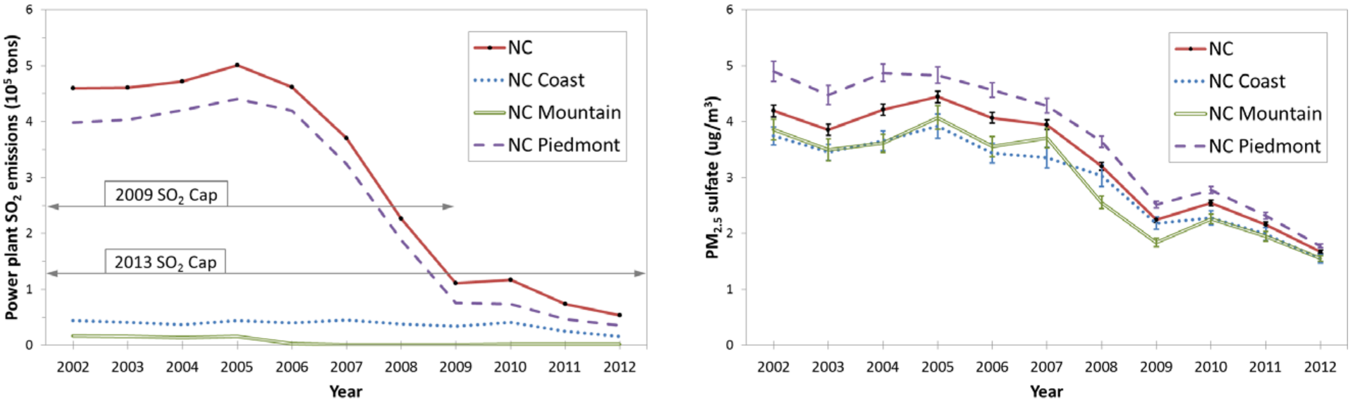


Figure 1. Annual power plant SO_2 emissions (left) and $PM_{2.5}$ sulfate concentrations (right) for NC (solid line) and each of its subregions (coast: dotted line; mountain: hollow line; piedmont: dashed line). The caps on power plant SO_2 emissions set by the Clean Smokestacks Act are indicated by solid line-solid arrows. The whiskers correspond to the standard error of the mean $PM_{2.5}$ sulfate concentration.

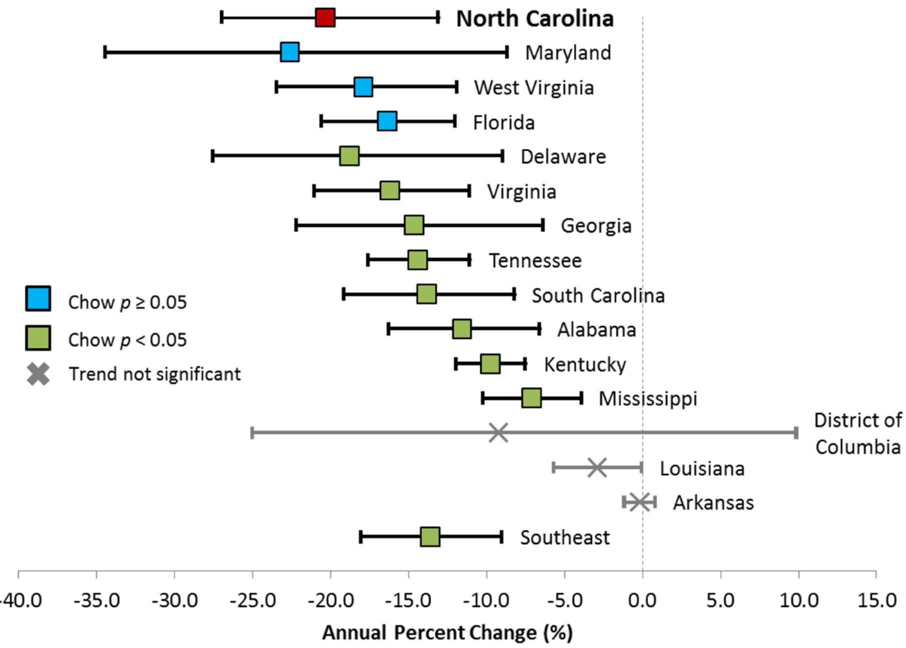


Figure 2. Annual percent changes in SO_2 emissions by state (2002–2012). The whiskers correspond to the upper and lower bounds of the 95% confidence interval. Chow test was used to analyze whether the annual percent changes differ significantly between NC and each of the other states.

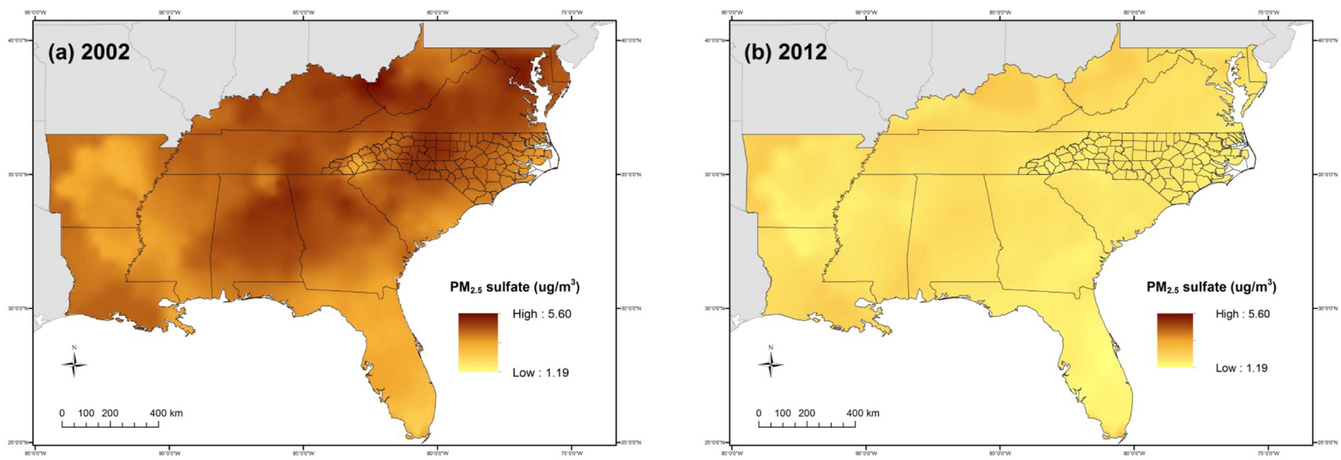


Figure 3. Spatial distribution of estimated $PM_{2.5}$ sulfate concentrations for the southeastern U.S. in (a) 2002 and (b) 2012.

dx.doi.org/10.1021/es501358a | *Environ. Sci. Technol.* XXXX, XXX, XXX–XXX

Table 2. Decrease in Fraction (AF) and Number of Premature Deaths Attributable to $PM_{2.5}$ Sulfate in NC

cause of death/C−R function type	AF (95% CI) 2002[a]	AF (95% CI) 2012[b]	overall decrease in AF[c]	attributable deaths prevented by clean air rules in 2012 (95% CI)
All-Cause				
$PM_{2.5}$ sulfate	3.2% (1.8, 4.5)	2.5% (1.6, 3.4)	−63%	1700 (1500, 1800)
total $PM_{2.5}$	1.2% (0.62, 1.8)	1.0% (0.55, 1.4)	−60%	1300 (1300, 1400)
Cardiopulmonary Disease[d]				
$Pm_{2.5}$ Sulfate	4.9% (2.9, 6.9)	1.9% (1.0, 2.7)	−61%	970 (910, 1,000)
total $PM_{2.5}$	4.8% (3.3, 6.2)	1.8% (1.1, 2.5)	−63%	940 (900, 980)
lung cancer[e]				
$PM_{2.5}$ sulfate	5.9% (1.9, 9.9)	2.3% (0.63, 3.9)	−61%	210 (190, 240)
total $PM_{2.5}$	5.5% (3.0, 8.0)	2.1% (1.0, 3.2)	−62%	200 (190, 210)

[a]Total number of cause-specific deaths (age ≥ 25) in 2002 for all-cause: 74 876; cardiopulmonary disease: 33 799; lung cancer: 5043. [b]Total number of cause-specific deaths (age ≥ 25) in 2012 for all-cause: 78 381; cardiopulmonary disease: 29 702; lung cancer: 5429. [c]Overall decrease was defined as the overall change of mean value (i.e., AF) from 2002 to 2012 using the formula $(\text{Value}_{2002} - \text{Value}_{2002})/\text{Value}_{2002} \times 100$. [d]International Classification of Diseases, Tenth Revision (ICD-10) codes I00−I78, J10-J18, J40-J47, and J67. [e]ICD-10 code C34.

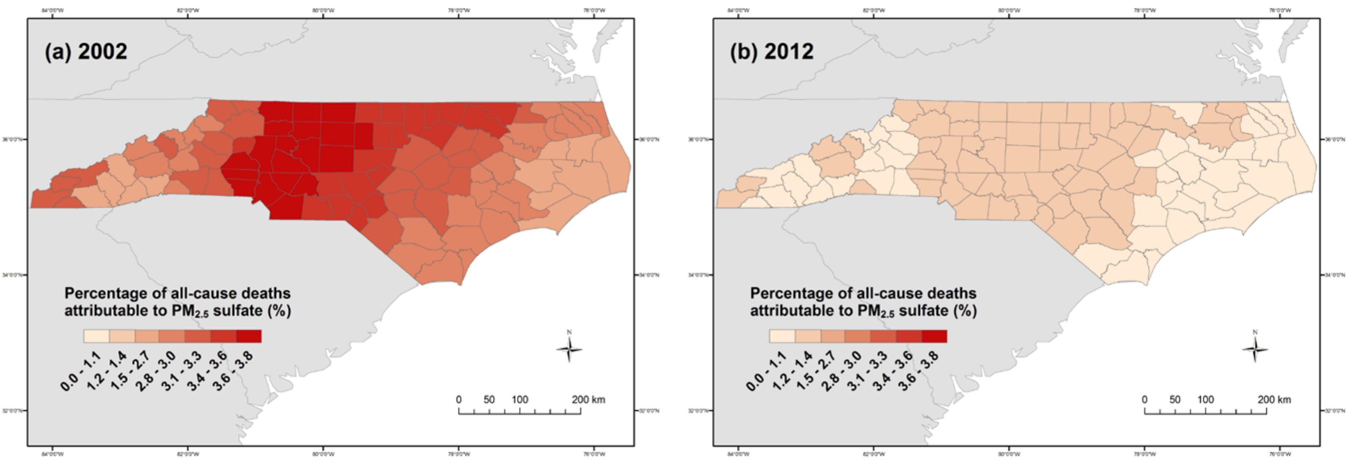


Figure 4. Percentage of annual all-cause deaths attributable to $PM_{2.5}$ sulfate in NC in (a) 2002 and (b) 2012.

decreased by 63%, from 3.2% (95% CI: 1.8%, 4.5%) in 2002 to 1.2% (95% CI: 0.62%, 1.8%) in 2012. This decline in health risks equates to about 1700 (95% CI: 1500, 1800) premature deaths avoided in 2012, compared to deaths expected if SO_2 emissions had remained unchanged; that is, if the premature mortality risk associated with $PM_{2.5}$ sulfate had remained the same in 2012 as in 2002, then an additional 1,700 deaths would have been expected. If the total $PM_{2.5}$ risk function was applied, the percentage of deaths decreased by 60%, and the risk model predicts that about 1,300 (95% CI: 1300, 1400) premature deaths were avoided in 2012. Similar trends were also observed for other cause-specific deaths, with about 60% reduction for both cardiopulmonary- and lung cancer-related causes between 2002 and 2012, irrespective of the C−R function used.

In addition to temporal reductions, there is also substantial geographic variation in mortality risk (Figure 4). In 2002, the estimated percentage of deaths attributed to $PM_{2.5}$ sulfate was above 2.4% for all counties (according to the $PM_{2.5}$ sulfate risk function). In 2012, no counties were above this level, and all counties were below 1.4%. This general trend holds true for cardiopulmonary and lung cancer mortality risk estimates (SI, Figures S4 and S5). In comparison to the mountain and coast regions, most counties in the piedmont region had higher percentages of all-cause deaths attributable to $PM_{2.5}$ sulfate exposure. Risk estimates based on the conventional total $PM_{2.5}$ risk functions displayed similar geographic patterns in NC (figures not shown).

Limitations. One limitation of this analysis is that the BME interpolation of $PM_{2.5}$ sulfate concentrations may be biased in areas that lack sufficient monitors. However, these areas are typically less populated, so the resulting bias in estimated health effects is expected to be small. Another limitation is uncertainty regarding the dose−response relation between $PM_{2.5}$ sulfate particles and health outcomes, as recent toxicological and epidemiologic research has yielded somewhat contradictory results with regard to the human health effects of $PM_{2.5}$ sulfate particles.[42,43] Nonetheless, we have endeavored to account for this uncertainty by using health impact functions from epidemiologic studies that have been subjected to extensive prior review. As a result of these limitations, the health benefits estimated are subject to additional aleatory and epistemic uncertainty.

■ DISCUSSION

Retrospective evaluation of the effectiveness of emission reduction programs can communicate the benefits of these programs to policymakers and the general public. The present study provides strong evidence that the combination of state and federal policies to reduce SO_2 emissions from coal-fired power plants has resulted in significant improvements in air quality and health in NC. $PM_{2.5}$ sulfate concentrations in ambient air decreased at an average annual rate of 8.7% during 2002–2012. As a result, in 2012, approximately 60% fewer premature deaths (about 1,700 all-cause deaths prevented) occurred than expected if $PM_{2.5}$ sulfate concentrations had remained the same as in 2002.

This study further suggests that implementation of the NC Clean Smokestacks Act reduced coal-fired power plant emissions more than would have occurred due to the federal policies alone. SO_2 emissions from coal-fired power plants decreased at an annual average rate of 20.3% during 2002–2012—a significantly greater rate than the 13.6% rate of decrease across all southeastern states and also significantly greater than the decreases observed in the four states neighboring NC. The peak rate of decrease in both SO_2 emissions and $PM_{2.5}$ sulfate concentrations, which occurred between 2007 and 2009, corresponds to the time period during which the Clean Smokestacks Act required the state's largest electricity providers (Duke Energy and Progress Energy) to substantially decrease SO_2 emissions: Duke Energy to 150 000 tons per year and Progress Energy to 100 000 tons per year from previous emissions of 223 098 and 147 269 tons, respectively.[5] The annual decrease in $PM_{2.5}$ sulfate concentrations was higher in the NC Piedmont region, where 9 of the state's 14 major coal-fired power plants are located, than in other regions, lending further support to the hypothesis that the Clean Smokestacks Act benefited air quality and health beyond the benefits of federal legislation alone.

The declining trends in regional $PM_{2.5}$ sulfate concentration reported in this study (−7.9% per year in the Southeast) are consistent with multiple recent studies illustrating the benefits of federal air quality policies. For example, Hand et al. found that $PM_{2.5}$ sulfate concentrations in the Southeast decreased at an annual rate of between 4.4% and 6.6% during 2001–2010.[13] Similarly, Blanchard et al. observed downward trends ranging from 3.7% to 6.2% per year during 1999–2010.[11] This work extends these previous studies by using modern geostatistical techniques to interpolate $PM_{2.5}$ sulfate concentrations across space and time, in order to support health impact assessment. The previous studies estimated trends and used simple interpolation algorithms (e.g., kriging) to estimate trends in unmonitored locations but did not employ the full power of space-time interpolation offered by the BME technique.

In this study, the relationship between SO_2 emission trends and ambient $PM_{2.5}$ sulfate concentrations followed a similar temporal pattern, with periods of decline in SO_2 emissions corresponding to periods of rapid decline in ambient $PM_{2.5}$ sulfate concentrations (Figure 1). This relationship also is consistent with the previous work by Hand et al.[13] and Blanchard et al.[11] Hand et al. found that power plant SO_2 emissions in the Southeast decreased at a similar rate as $PM_{2.5}$ sulfate concentrations from 2001 to 2010 (−6.4% per year), suggesting a linear relationship between emissions and concentrations. Blanchard et al. observed an annual emission reduction rate of 7.9% in the Southeast during 1999–2010, approximately linear with the downward trends in $PM_{2.5}$ sulfate concentrations.

This study found the rate of decrease in $PM_{2.5}$ sulfate concentrations was greater on average in NC than in the Southeast (8.7% per year as compared to 7.9% per year), but this difference was not statistically significant, despite the significantly greater reduction in SO_2 emissions in NC than in the Southeast. This result is also consistent with previous studies showing the important influence of long-range transport of SO_2 on local ambient $PM_{2.5}$ sulfate concentrations. For example, EPA reported that most $PM_{2.5}$ sulfates in the eastern United States are converted from regional SO_2 emissions, and power plants are the largest contributor to these regional emissions.[44] Specifically, Wagstrom and Pandis estimated that the average transport distance for SO_2 in the East ranges from 115 to 220 km.[45] It is possible that the reductions in SO_2 emissions in NC contributed substantially to the decreases in $PM_{2.5}$ sulfate concentrations in surrounding states and that, as a result, the benefits substantially exceed those in NC alone. Despite the lack of a significant difference in the rate of decline in $PM_{2.5}$ sulfate concentration in NC as compared to in the Southeast region, our spatiotemporal analysis nonetheless showed substantial geographic variation in $PM_{2.5}$ sulfate concentrations in the Southeast, with the highest concentrations occurring in areas of significant SO_2 emissions, including the NC Piedmont region. Thus, although the percentage rate of decline in $PM_{2.5}$ sulfate concentration is similar throughout much of the Southeast, our results indicate that local SO_2 emissions strongly influence the distribution of $PM_{2.5}$ sulfates and that, importantly, direct reductions from local sources appear to be effective in reducing $PM_{2.5}$ sulfate levels both locally and in surrounding areas.

Our health impact estimates also are consistent with a recent national health impact assessment by Fann et al.[16] The authors used an air quality model (CAMx) to estimate how U.S. air quality and health impacts attributable to 23 categories of emission sectors would change under new pollution emissions regulations. One of the proposed regulations Fann et al. considered is the Cross-State Air Pollution Rule, which would impose stricter limits on power plants in the eastern United States similar to those implemented under the NC Clean Smokestacks Act. The cross-state rule currently is under review by the U.S. Supreme Court; the EPA and the rule's opponents presented oral arguments in court in December 2013. Fann et al. estimated that if the new rule were implemented, then the total number of premature deaths in the U.S. attributable to power plant emissions would decrease from about 38 000 in 2005 to about 17 000 in 2016—a decline of 55%. This change is comparable to the decrease in premature mortality in NC that we estimated already has occurred at least in part as a result of the NC Clean Smokestacks Act (Table 2). The major difference between our approach and that of Fann et al. is that Fann et al. used an air quality model to *predict* air quality and health benefits if the Cross-State Air Pollution Rule were to be implemented, whereas we show the *observed* effects after NC's implementation of regulations comparable to the pending federal rule. Our results thus empirically validate the predictions of Fann et al. and lend further support for the health benefits of decreasing air pollutant emissions from power plants.

In summary, our findings suggest that the NC Clean Smokestacks Act, in conjunction with federal legislation, has substantially reduced coal-fired power plant emissions and, as a result, has improved air quality and public health in NC. SO_2 reductions in NC were significantly faster than the reductions across all southeastern states as well as the reductions in the four states neighboring NC, further suggesting that implementation of the Clean Smokestacks Act reduced coal-fired power plant emissions beyond what would have occurred due to federal legislation alone. The Clean Smokestacks Act positions NC to respond to more stringent NAAQS for $PM_{2.5}$ and could serve as a model for similar actions taken by other states. Furthermore, these results provide additional evidence of the benefits of the tightened standard proposed under the Cross-State Air Pollution Rule.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Details on data locations, hard and soft data construction, concentration−response functions, and health impact maps. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Phone: (919) 448-4776; e-mail: yaruli@live.unc.edu.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank Jeanette Reyes and Chieh-Han Lee for assisting in MATLAB code of BME interpolation.

■ REFERENCES

(1) Bachmann, J. Will the circle be unbroken: A history of the US national ambient air quality standards. *J. Air Waste Manage. Assoc.* **2007**, *57* (6), 652−697.

(2) Chestnut, L. G.; Mills, D. M. A fresh look at the benefits and costs of the US Acid Rain Program. *J. Environ. Manage.* **2005**, 77 (3), 252−266.

(3) Hubbell, B. J.; Crume, R. V.; Evarts, D. M.; Cohen, J. M. Policy Monitor: Regulation and Progress under the 1990 Clean Air Act Amendments. *Rev. Environ. Econ. Policy* **2010**, *4* (1), 122−138.

(4) Kruse, E. Case Comments: North Carolina v. Environmental Protection Agency. *Harvard Environ. Law Rev.* **2009**, *33* (1), 283−296.

(5) N.C. DENR (Department of Environment and Natural Resources). Clean Air Legislation: Clean Smokestacks Act. http://www.ncair.org/news/leg/ (accessed December 1, 2013).

(6) Hoppock, D.; Adair, S. K.; Murray, B.; Tarr, J. *Benifits of Early State Action in Environmental Regulation of Electric Utilities: North Carolina's Clean Smokestacks Act*; Nicholas Institute for Environmental Policy Solutions, Duke University: Durham, NC, 2012.

(7) Ross, W. G. The North Carolina Clean Smokestacks Act. *N. C. Med. J.* **2011**, *72* (2), 128−131.

(8) Maryland Department of the Environment. The Maryland Healthy Air Act. http://www.mde.state.md.us/programs/Air/Pages/MD_HAA.aspx (accessed December 14, 2013).

(9) Fann, N.; Risley, D. The public health context for $PM_{2.5}$ and ozone air quality trends. *Air Qual. Atmos. Health* **2013**, *6* (1), 1−11.

(10) U.S. EPA (Environmental Protection Agency). Regulatory Impact Analysis for the Federal Implementation Plans to Reduce Interstate Transport of Fine Particulate Matter and Ozone in 27 States; Correction of SIP Approvals for 22 States. http://www.epa.gov/crossstaterule/pdfs/FinalRIA.pdf (accessed September 27, 2013).

(11) Blanchard, C. L.; Hidy, G. M.; Tanenbaum, S.; Edgerton, E. S.; Hartsell, B. E. The Southeastern Aerosol Research and Characterization (SEARCH) study: Temporal trends in gas and PM concentrations and composition, 1999−2010. *J. Air Waste Manage. Assoc.* **2013**, *63* (3), 247−259.

(12) Blanchard, C. L.; Tanenbaum, S.; Hidy, G. M. Source attribution of air pollutant concentrations and trends in the Southeastern Aerosol Research and Characterization (SEARCH) Network. *Environ. Sci. Technol.* **2013**, *47* (23), 13536−13545.

(13) Hand, J. L.; Schichtel, B. A.; Malm, W. C.; Pitchford, M. L. Particulate sulfate ion concentration and SO_2 emission trends in the United States from the early 1990s through 2010. *Atmos. Chem. Phys.* **2012**, *12* (21), 10353−10365.

(14) Malm, W. C.; Schichtel, B. A.; Ames, R. B.; Gebhart, K. A. A 10-year spatial and temporal trend of sulfate across the United States. *J. Geophys. Res., Atmos.* **2002**, *107*, D22.

(15) Caiazzo, F.; Ashok, A.; Waitz, I. A.; Yim, S. H. L.; Barrett, S. R. H. Air pollution and early deaths in the United States. Part I: Quantifying the impact of major sectors in 2005. *Atmos. Environ.* **2013**, *79*, 198−208.

(16) Fann, N.; Fulcher, C. M.; Baker, K. The recent and future health burden of air pollution apportioned across U.S. sectors. *Environ. Sci. Technol.* **2013**, *47* (8), 3580−3589.

(17) U.S. EPA (Environmental Protection Agency). Integrated Science Assessment for Particulate Matter (Final Report). http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=216546 (accessed December 20, 2014).

(18) Helfenstein, U. The use of transfer function models, intervention analysis and related time series methods in epidemiology. *Int. J. Epidemiol.* **1991**, *20* (3), 808−815.

(19) Christakos, G. *Modern Spatiotemporal Geostatistics*; Oxford University Press: Oxford; New York, 2000.

(20) Christakos, G. A Bayesian maximum-entropy view to the spatial estimation problem. *Math. Geol.* **1990**, *22* (7), 763−777.

(21) WHO (World Health Organization). Health Impact Assessment (HIA). http://www.who.int/hia/en/ (accessed June 19, 2013).

(22) U.S. EPA (Environmental Protection Agency). CHIEF (Clearinghouse for Inventories & Emissions Factors). Emission Inventories. http://www.epa.gov/ttn/chief/eiinformation.html (accessed November 5, 2012).

(23) U.S. EPA (Environmental Protection Agency). Air Markets Program Data (AMPD). http://ampd.epa.gov/ampd/ (accessed November 21, 2013).

(24) U.S. EPA (Environmental Protection Agency). AQS (Air Quality System). Yearly Raw Data Files Retrieved From AQS. http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm (accessed March 9, 2012).

(25) Federal Land Manager Environmental Database (FED) Database Query Wizard. http://views.cira.colostate.edu/fed/QueryWizard/Default.aspx (accessed October 28, 2012).

(26) U.S. HHS (Department of Health & Human Services). HRSA (Health Resources and Services Administration). Trend Analysis and Interpretation. http://mchb.hrsa.gov/publications/ (accessed.

(27) Chow, G. C. Tests of equality between sets of coefficients in two linear regressions. *Econometrica* **1960**, *28* (3), 591−605.

(28) Christakos, G. *Temporal GIS: Advanced Functions for Field-Based Applications*; Springer: Berlin; New York, 2001.

(29) Reyes, J. M.; Serre, M. L. An LUR/BME framework to estimate $PM_{2.5}$ explained by on road mobile and stationary sources. *Environ. Sci. Technol.* **2014**, *48* (3), 1736−1744.

(30) Akita, Y.; Chen, J. C.; Serre, M. L. The moving-window Bayesian maximum entropy framework: Estimation of $PM_{2.5}$ yearly average concentration across the contiguous United States. *J. Exposure Sci. Environ. Epidemiol.* **2012**, *22* (5), 496−501.

(31) Serre, M. L.; Bogaert, P.; Christakos, G. *BMElib: The Bayesian Maximum Entropy Software for Space/Time Geostatistics, and Temporal GIS Data Integration*, 2.0b; 2001.

(32) WHO (World Health Organization). Quantifying environmental health impacts: Practical Guidance for Assessment of Disease Burden at National and Local Levels. http://www.who.int/quantifying_ehimpacts/national/en/ (accessed June 19, 2012).

(33) Dockery, D. W.; Pope, C. A.; Xu, X.; Spengler, J. D.; Ware, J. H.; Fay, M. E.; Ferris, B. G.; Speizer, F. E. An association between air pollution and mortality in six U.S. cities. *N. Eng. J. Med.* **1993**, *329* (24), 1753−1759.

(34) Pope, C. A.; Thun, M. J.; Namboodiri, M. M.; Dockery, D. W.; Evans, J. S.; Speizer, F. E.; Heath, C. W. Particulate air pollution as a predictor of mortality in a prospective study of US adults. *Am. J. Respir. Crit. Care Med.* **1995**, *151* (3), 669−674.

(35) Krewski, D. B.; Burnett, R. T.; Goldberg, M. S.; Hoover, K.; Siemiatycki, J.; Jerrett, M.; Abrahamowicz, M.; White, W. H. *Reanalysis of the Harvard Six Cities Study and the American Cancer Society Study of*

Particulate Air Pollution and Mortality; HEI (Health Effects Institute): Cambridge, MA, 2000.

(36) Pope, C. A.; Burnett, R. T.; Thun, M. J.; Calle, E. E.; Krewski, D.; Ito, K.; Thurston, G. D. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *J. Am. Med. Assoc.* **2002**, *287* (9), 1132−1141.

(37) Krewski, D. J., M.; Burnett, R. T.; Ma, R.; Hughes, E.; Shi, Y.; Turner, M. C.; Pope, III, CA; ; Thurston, G.; Calle, E. E.; Thun, M. J. *Extended Follow-Up and Spatial Analysis of the American Cancer Society Study Linking Particulate Air Pollution and Mortality*; HEI (Health Effects Institute): Boston, MA, 2009.

(38) Lepeule, J.; Laden, F.; Dockery, D.; Schwartz, J. Chronic exposure to fine particles and mortality: An extended follow-up of the Harvard Six Cities Study from 1974 to 2009. *Environ. Health Perspect* **2012**, *120* (7), 965−970.

(39) CDC (Centers for Disease Control and Prevention). Wide-ranging Online Data for Epidemiologic Research (WONDER). http://wonder.cdc.gov/ (accessed March 10, 2014).

(40) Trijonis, J. C.; Malm, W. C.; Pitchford, M.; White, W. H.; Charlson, R.; Husar, R. *Acidic Deposition: State of Science and Technology: Report 24 Visibility: Existing and Historical Conditions-Causes and Effects*; NAPAP (National Acid Precipitation Assessment Program): Washington, DC, 1990.

(41) U.S. EPA (Environmental Protection Agency). Guidance for Estimating Natural Visibility Conditions Under the Regional Haze Rule. http://www.epa.gov/ttn/caaa/t1/meta/m30624.html (accessed December 20, 2013).

(42) Grahame, T.; Schlesinger, R. Is ambient $PM_{2.5}$ sulfate harmful? *Environ. Health Perspect* **2012**, *120* (12), A454.

(43) Schwartz, J.; Lepeule, J. Is ambient $PM_{2.5}$ sulfate harmful? Schwartz and Lepeule Respond. *Environ. Health Perspect* **2012**, *120* (12), A454−A455.

(44) U.S. EPA (Environmental Protection Agency). The Particle Pollution Report: Current Understanding of Air Quality and Emissions through 2003. http://www.epa.gov/airtrends/aqtrnd04/pm.html (accessed December 26, 2013).

(45) Wagstrom, K. M.; Pandis, S. N. Source-receptor relationships for fine particulate matter concentrations in the Eastern United States. *Atmos. Environ.* **2011**, *45* (2), 347−356.