Environmental Research 133 (2014) 103–110

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Assessment of primary and secondary ambient particle trends using satellite aerosol optical depth and ground speciation data in the New England region, United States

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ARTICLE INFO

Article history:
Received 12 July 2013
Received in revised form
15 February 2014
Accepted 9 April 2014
Available online 4 June 2014

Keywords:
AOD
Emission
MODIS
$PM_{2.5}$
Trend analysis

ABSTRACT

The effectiveness of air pollution emission control policies can be evaluated by examining ambient pollutant concentration trends that are observed at a large number of ground monitoring sites over time. In this paper, we used ground monitoring measurements in conjunction with satellite aerosol optical depth (AOD) data to investigate fine particulate matter ($PM_{2.5}$; particulate matter with aerodynamic diameter ≤ 2.5 μm) trends and their spatial patterns over a large U.S. region, New England, during 2000–2008. We examined the trends in rural and urban areas to get a better insight about the trends of regional and local source emissions. Decreases in $PM_{2.5}$ concentrations (μg/m^3) were more pronounced in urban areas than in rural ones. In addition, the highest and lowest $PM_{2.5}$ decreases (μg/m^3) were observed for winter and summer, respectively. Together, these findings suggest that primary particle concentrations decreased more relative to secondary ones. This is also supported by the analysis of the speciation data which showed that downward trends of primary pollutants including black carbon were stronger than those of secondary pollutants including sulfate. Furthermore, this study found that ambient primary pollutants decreased at the same rate as their respective source emissions. This was not the case for secondary pollutants which decreased at a slower rate than that of their precursor emissions. This indicates that concentrations of secondary pollutants depend not only on the primary emissions but also on the availability of atmospheric oxidants which might not change during the study period. This novel approach of investigating spatially varying concentration trends, in combination with ground $PM_{2.5}$ species trends, can be of substantial regulatory importance.

1. Introduction

Fine particulate matter ($PM_{2.5}$; particulate matter with aerodynamic diameter ≤ 2.5 μm) is a mixture of local and regional pollutants including sulfate, nitrate, ammonium, organic and elemental carbon, metal oxides, and crustal compounds (U.S. EPA, 2004). The particles originating from anthropogenic and natural sources are released directly from the sources (i.e., primary pollution) or are formed in the atmosphere largely through photochemical reaction from precursor gases (i.e., secondary pollution). Numerous studies have demonstrated that ambient $PM_{2.5}$ concentrations are associated with adverse human health and environmental effects (Bell et al., 2007, 2010; Dockery et al., 1993; Gent et al., 2003, 2009; Ramanathan et al., 2001). The World Health Organization (WHO) annual and 24-h $PM_{2.5}$ guidelines are 10 and 25 μg/m^3, respectively (WHO, 2005). In the U.S. Environmental Protection Agency (EPA) (U.S. EPA, 2012) $PM_{2.5}$ standards are 12 μg/m^3 (annual) and 35 μg/m^3 (24-h) (U.S. EPA, 2013a). To comply with the standard, federal and state environmental protection agencies have planned and implemented emission mitigation strategies (e.g., National Clean Diesel Campaign; Cross-State Air Pollution Rule, formerly Clean Air Interstate Rule) to reduce particle pollution levels and thus protect human health and the environment (U.S. EPA, 2011a, 2011b).

The effectiveness of these policies can be evaluated by examining ambient pollutant concentration trends that are observed at a large number of ground compliance monitoring sites over time. However, the sampling frequency of these sites is usually every

Abbreviations: AOD, Aerosol Optical Depth; EIA, Energy Information Administration; EPA, Environmental Protection Agency; MODIS, Moderate Resolution Imaging Spectroradiometer; NASA, National Aeronautics and Space Administration; $PM_{2.5}$, Particulate Matter with aerodynamic diameter ≤ 2.5 μm.

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http://dx.doi.org/10.1016/j.envres.2014.04.006
0013-9351/

three or six days in the U.S., and $PM_{2.5}$ monitoring networks are typically sparsely distributed and there are many geographical areas without monitoring sites. This may cause less reliable environmental assessments, leading to the necessity of spatially and temporally resolved methods for comprehensively evaluating the regulatory efforts. Satellite remote sensing is increasingly used to provide $PM_{2.5}$ information to complement ground $PM_{2.5}$ monitoring networks. The satellite data include aerosol optical depth (AOD), which provides information about the amount of aerosol in the atmosphere, and are available for different geographical areas without spatial limitations. Recently, we introduced models that used AOD data to predict daily surface-level $PM_{2.5}$ concentrations with reasonably high accuracy (Lee et al., 2011a, 2012), as discussed below.

The objective of our study was to estimate the $PM_{2.5}$ concentration trends (with the assumption of an identical percent change in concentrations per year) using daily satellite-based $PM_{2.5}$ predictions in the New England region, U.S. during the period 2000–2008. We first investigated the location-specific $PM_{2.5}$ concentration trends that varied spatially using satellite AOD (10×10 km resolution) and statistical models. The spatially and temporally resolved satellite-based $PM_{2.5}$ concentrations led us to assess location-specific (i.e., grid-specific) $PM_{2.5}$ trends throughout the study region. In combination with the seasonal variation of $PM_{2.5}$ concentration changes, this enabled us to evaluate the relative contributions of primary and secondary particles to the $PM_{2.5}$ mass trends and thus assess the effectiveness of $PM_{2.5}$ emission reduction policies. We further examined the trends of ground $PM_{2.5}$ species concentrations observed in the Boston area and compared them with source emission trends, while analyzing the relative impacts of primary and secondary pollutants on the observed $PM_{2.5}$ trends.

2. Methods

2.1. Satellite-based fine particulate matter concentrations

The $PM_{2.5}$ mass trend estimates were based on the $PM_{2.5}$ concentration predictions using satellite AOD data, previously developed by Lee et al. (2011a, 2012) and updated in this study. To develop the prediction models, we obtained filter-based $PM_{2.5}$ mass concentrations from 69 EPA monitoring sites and Moderate Resolution Imaging Spectroradiometer (MODIS) AOD values from the National Aeronautics and Space Administration (NASA) in Massachusetts, Connecticut, Rhode Island, Southern Maine, New Hampshire, and Vermont for the period 2000–2008. AOD values were provided by the MODIS due to its relatively fine spatial (10 km) and temporal (1–2 days) resolution compared to other available satellite sensors. The MODIS AOD data were calibrated using ground $PM_{2.5}$ measurements on a daily basis using a mixed effects model to predict $PM_{2.5}$ in the study region (Lee et al., 2011a). This AOD daily calibration model generated day-specific $PM_{2.5}$-AOD relationships, as a combination of a fixed effect representing an average relationship for all days and a random effect explaining the daily variability of the relationship for each day. The model significantly improved the $PM_{2.5}$ predictive power in the study domain, which rendered AOD a robust predictor of $PM_{2.5}$. For days when AOD data were not available due to cloud, snow/ice cover, and retrieval errors (i.e., non-retrieval days), the spatial clustering method was shown to be useful to estimate the missing $PM_{2.5}$ concentrations (Lee et al., 2012). This modeling approach enabled all daily location-specific $PM_{2.5}$ concentrations to be estimated with reasonably high predictability. This study could contribute to acute and chronic health effect studies while providing spatially and temporally resolved $PM_{2.5}$ exposure estimates and thus reducing exposure errors. Taking advantage of the clustering method used in Lee et al. (2012), we performed the cluster analysis using the observed spatial variability of the $PM_{2.5}$ measurements over the study region in the current study. This cluster analysis identified groups (i.e., clusters) of days exhibiting similar spatial patterns of $PM_{2.5}$ concentrations, and a prediction model was developed for each cluster. In each cluster, we assumed that the relationship between average of $PM_{2.5}$ concentrations predicted from AOD data and average of regional $PM_{2.5}$ concentrations was constant and then calculated the ratio between the concentrations in each grid cell. This produced both cluster- and grid-specific ratios, and thus enabled us to predict all missing $PM_{2.5}$ concentrations. The regionally averaged $PM_{2.5}$ concentrations (i.e., average of all available $PM_{2.5}$ measurements on a given day) were available for all days. Using these two modeling approaches, daily ground $PM_{2.5}$ concentrations in each of the 579 grid cells (10×10 km^2) were estimated both for days with and without AOD data for a total of 3287 days.

2.2. Ground fine particulate matter species measurements

Ambient $PM_{2.5}$ samples were collected at the Harvard-EPA Clean Air Research Center monitoring site (42.34 °N, 71.10 °W) in Boston, MA from 2000 to 2008. This monitoring site has played a role in providing regionally representative data and thus $PM_{2.5}$ exposure estimates in epidemiological studies performed in the New England region, U.S. for more than a decade. At the monitoring site, daily 24-h integrated $PM_{2.5}$ samples were collected using the Harvard Impactor (Koutrakis et al., 1993). These samples were analyzed gravimetrically to determine daily $PM_{2.5}$ mass. Sulfate (SO_4^{2-}) concentrations were measured using ion chromatography before February 2, 2004. Since then, semi-continuous hourly measurements of SO_4^{2-} using a sulfate particulate analyzer (Thermo Electron Corporation, model 5020, Franklin, MA) were employed to calculate 24-h averages. These two different methods for SO_4^{2-} concentrations (i.e., ion chromatography and sulfate particulate analyzer) produced fairly equivalent results, as shown by Kang et al. (2010). SO_4^{2-} concentrations were not available for a number of days in 2004, 2005, and 2006. Thus, we estimated missing data from sulfur concentrations by X-ray fluorescence analysis using a simple linear regression (correlation $r=0.98$). Hourly ambient black carbon (a surrogate of elemental carbon) and particle number concentrations, measured with an Aethalometer (Magee Scientific Corporation, model AE-16, Berkeley, CA) and a Condensation Particle Counter (TSI Incorporated, model 3022 A, Shoreview, MN), respectively, were used to determine 24-h average black carbon and particle number concentrations. The same samplers and analytical methods for black carbon and particle number concentrations were constantly used for the entire study period. The black carbon concentrations measured by an Aethalometer may have bias caused by filter loading artifact (Arnott et al., 2005; Virkkula et al., 2007; Weingartner et al., 2003). However, in this study, we used the uncorrected black carbon concentrations because these values have been widely applied for health effect studies as exposure estimates in the study region. The total number of concentration is dominated by ambient ultrafine particles (<0.1 μm) due to the higher number of distribution in the nucleation- (size range less than 0.01 μm) and the Aitken-modes (size range between 0.01 and 0.1 μm). More details about Harvard-EPA Clean Air Research Center measurements are described in Kang et al. (2010). It is noted that the sampling frequency at the monitoring site was every day, which provided more frequent species measurements and thus potentially more reliable trend estimates, compared to Chemical Speciation Network (CSN) monitoring sites generally with the sampling schedule of every three days.

We also obtained ambient $PM_{2.5}$ mass, SO_4^{2-}, nitrate (NO_3^-), organic carbon, and elemental carbon concentrations measured at a Chemical Speciation Network site (42.33°N, 71.08°W) in the Boston area for the same period (2000–2008). We used this Chemical Speciation Network monitoring site primarily to complement unavailable or very limited species concentration information (NO_3^-, organic carbon, and elemental carbon) at the Harvard-EPA Clean Air Research Center monitoring site. This monitoring site is located approximately 1.8 km away from the Harvard-EPA Clean Air Research Center monitoring site. The concentrations were determined by analyzing 24-h integrated $PM_{2.5}$ samples every three days. It is noted that this site used an identical carbon sampler for the period 2000–2008 (U.S. EPA, 2013b).

2.3. Data analysis

An autoregressive model was used to examine the trends of satellite- ($PM_{2.5}$ mass) and ground-based ($PM_{2.5}$, SO_4^{2-}, black carbon, and particle number) concentrations due to potential autocorrelations of time-series data. The autoregressive model produces more reliable error estimates for independent variables compared to a linear regression model without autoregressive errors. For the model, we first employed a stepwise autoregression method to select the final order of the autoregressive error model. This method sequentially tested autoregressive error models from high-order ones, resulting in all significant error lags left in the model ($p<0.05$). Considering the residence time of $PM_{2.5}$ (days to weeks), $PM_{2.5}$ mass and species concentrations on lag 0 were much less likely to be correlated with those on lag day larger than 7 (one week). Due to potentially longer residence time, we decided to have an extra margin and thus ran the model with a starting lag of 25. The stepwise autoregressive model showed estimates of autocorrelations from lag 0 (correlation=1) to lag 25 with the corresponding p-values. This result demonstrated that autocorrelations generally became much smaller after the lag day of 3, indicating that the lag day of 3 was reasonable to estimate all the trend parameters in this analysis.

Because the concentration values were log-normally distributed we log-transformed them for the model calculations. Also, this transformation was based on the assumption that the percent change per year in the concentrations was constant throughout the study period. Despite the seasonal variations in ambient $PM_{2.5}$ mass and species concentrations, the assumption of constant annual percent changes is reasonable because the seasonal nature was evenly included in each year. To account for the annual trends, we considered the year as a continuous variable and used the year of 2000 as the baseline year. Daily satellite-predicted $PM_{2.5}$ mass concentrations in each of 10×10 km^2 grid cells allowed us to investigate grid-specific $PM_{2.5}$ mass trends. At the Harvard-EPA Clean Air Research Center monitoring site, there were more missing sampling days of the speciation compared to $PM_{2.5}$ mass. Thus, to compare trends for $PM_{2.5}$ mass and species concentrations, we selected only those days (a total of 2590 days) with valid