Atmospheric Environment 117 (2015) 259–270

Contents lists available at ScienceDirect

Atmospheric Environment

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

Variability and potential sources of summer $PM_{2.5}$ in the Northeastern United States

Rolando O. Saunders*, Darryn W. Waugh

Department of Earth and Planetary Sciences, The Johns Hopkins University, Baltimore, MD, USA

HIGHLIGHTS

- Summer concentrations of aerosol (PM) have decreased in the Northeastern U.S.
- High-PM events in the Northeastern U.S. are generally caused by wildfires or stagnation of anthropogenic emissions.
- Organic carbon is the main component in events caused by wildfires.
- Ammonium sulfate is the main component in events of anthropogenic origin.
- The occurrence of PM events in the region has decreased since 1999.

ARTICLE INFO

Article history:
Received 2 February 2015
Received in revised form
29 June 2015
Accepted 8 July 2015
Available online 11 July 2015

Keywords:
Air pollution
Transport
Fine aerosol
Wildfires
Variability
MERRA

ABSTRACT

The variability of ground-level concentrations of fine suspended particulate matter ($PM_{2.5}$) in the Northeastern United States is examined using observed $PM_{2.5}$ from multiple data networks together with output from the Modern-Era Retrospective Analysis for Research and Applications Aerosol Reanalysis (MERRAero). The long-term variations as well as the occurrence of short-term high-concentration episodes in the region are investigated for the period 1999–2013. This analysis shows that over this period there has been a significant decrease in summer-mean $PM_{2.5}$. A decrease in the occurrence and magnitude of high-$PM_{2.5}$ events in the Northeastern U.S. region is also observed. The potential sources of $PM_{2.5}$ are analyzed using MERRAero aerosol optical depth for two of the main components of the pollutant: organic carbon and ammonium sulfate. The analysis indicates that high-$PM_{2.5}$ events in the Northeastern U.S. are, generally, the result of long range transport of smoke from large boreal wildfires, Midwestern industrial emissions, or a combination of both. There are roughly equal numbers of events due to natural or anthropogenic sources for the 2002–2012 period for this region. The events that have an anthropogenic source are characterized by a strong high pressure system in the Southern U.S. that cause aerosols to be advected from the Midwest into the Northeastern U.S. The meteorology related to wildfire events is more variable, consistent with the varied locations of the fires that cause aerosol events in the Northeastern U.S.

1. Introduction

There is a well-established association between short-term exposure to suspended particulate matter with aerodynamic diameters less than 2.5 μm ($PM_{2.5}$) and cardiopulmonary morbidity and mortality (Brook, 2008; de Hartog et al., 2009; Dominici et al., 2006; Jalava et al., 2006; Pope III and Dockery, 2006). There is also growing evidence that some constituents of $PM_{2.5}$ are more harmful than others (Bell et al., 2009, 2014; Peng et al., 2009; Strak et al., 2012, 2013). It is therefore important to understand and model the spatial and temporal variations of not only the total particulate matter concentration, but also of individual constituents.

Here we examine the variability of $PM_{2.5}$ in the Northeastern United States (NEUS). There is large temporal variability in the $PM_{2.5}$ in the NEUS, on daily through to interannual time scales, which is related to changes in both sources and meteorology. Ammonium sulfate ($AmmSO_4$) and organic carbon (OC) are the major components of $PM_{2.5}$ in the NEUS (Chin et al., 2007; Hand et al., 2011), with $AmmSO_4$ coming primarily from anthropogenic sources (in particular, electricity power plants in the Ohio Valley) (Hand et al., 2011) and the organic matter coming from

* Corresponding author.
E-mail address: rolivas1@jhu.edu (R.O. Saunders).

http://dx.doi.org/10.1016/j.atmosenv.2015.07.007
1352-2310/

a combination of anthropogenic (residential and industrial biofuel) and natural (e.g. wildfires) sources (e.g. Park et al., 2007). The concentration and occurrence of episodes with high concentrations of $PM_{2.5}$ depend on changes in these sources as well meteorology. For example, previous studies have linked daily variability in $PM_{2.5}$ to synoptic meteorology, with low values when frontal systems ventilate the boundary layer and higher values during stagnation events with weak winds, no precipitation and high temperatures (e.g. Tai et al., 2010, 2012; Dharshana et al., 2010). Another cause of variability, and high $PM_{2.5}$, are wildfires. Several studies have reported events where long-range transport from wildfires, primarily from Canada, have lead to rapid increases in $PM_{2.5}$ in the NEUS (DeBell et al., 2004; Colarco et al., 2004; Duck et al., 2007; Bein et al., 2008; Miller et al., 2011). We are, however, not aware of any study that has examined the occurrence and potential sources of high-$PM_{2.5}$ events in the Northeastern United States over a long time period. We perform such a study here.

In this work we explore the variability of summer $PM_{2.5}$ in the NEUS region for a 15-year period, focusing on the occurrence, composition, and source of high-concentration events. We examine the measured regional and state-wide ground-level concentrations of $PM_{2.5}$ from daily to decadal scales and identify the frequency and intensity of high-concentration events. We then use the observed $PM_{2.5}$ from multiple data networks together with output from the Modern-Era Retrospective Analysis for Research and Applications Aerosol Reanalysis (MERRAero) (Buchard et al., 2014) to examine the composition and source of the high events. Our analysis indicates that the relative concentrations of $AmmSO_4$ and OC varies among the high-concentration events, and that this depends on whether the events are the result of anthropogenic emissions, long range transport of wildfires, or a combination of these two processes.

The air quality data, reanalysis output and details of the calculations are described in the following section. In Section 3, the variability of $PM_{2.5}$ in the Northeastern U.S. is explored using station measurements from the U.S. Environmental Protection Agency (EPA). In Section 4, the sources and the spatial variability of high-$PM_{2.5}$ events are analyzed using MERRAero reanalysis. The reanalysis output and station measurements are also compared in this section. Concluding remarks are in Section 5.

2. Data and methods

2.1. $PM_{2.5}$ data

To analyze the variability of ground level $PM_{2.5}$ we use daily mean $PM_{2.5}$ measured by the Federal Reference Method that are archived on the EPA Air Quality System website (www.epa.gov/airquality/airdata/). We use 24-h average $PM_{2.5}$ concentrations at local conditions (Code 88101), and consider all the available stations in the Northeastern United States (NEUS) extending from West Virginia to Maine (see Fig. 1). For most of our analysis we focus on the daily-mean $PM_{2.5}$ concentration averaged over the stations shown in Fig. 1, which we refer to as the NEUS-mean $PM_{2.5}$. We also consider averages over stations within individual states. The NEUS-mean and state-mean $PM_{2.5}$ are calculated for each day of the period 1999–2013. The number of measurements available varies between days. There are over 200 EPA $PM_{2.5}$ – monitoring stations active for the NEUS during the 1999–2013 period, however, only about a third of them sample $PM_{2.5}$ on a daily basis, with the majority of the stations sampling every 3 days.

Data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) were used to analyze individual components of $PM_{2.5}$, specifically $AmmSO_4$ and OC. The stations from this network are much more sparse than those from the EPA (see Fig. 1) and the sampling frequency is also lower than the EPA stations with $PM_{2.5}$ data available only every 3 days.

Data from the Chemical Speciation Network (CSN) were also

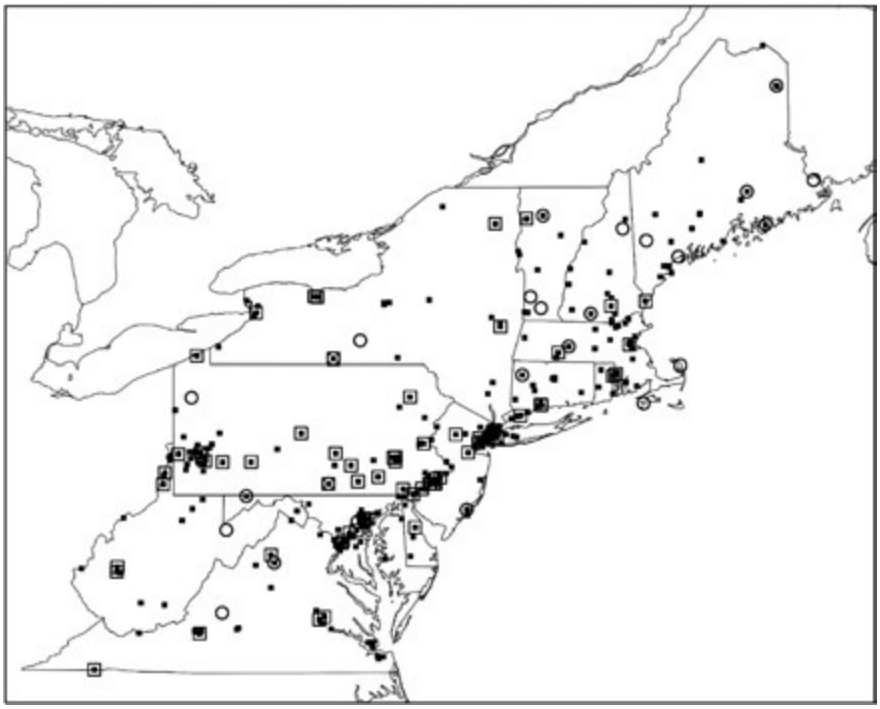

Fig. 1. Data sampling locations. EPA's $PM_{2.5}$ Network monitoring stations for the period 1999–2013 (small squares), Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring stations for the period 2002–2012 (circles), and EPA's Chemical Speciation Network (CSN) (large squares). MERRAero $PM_{2.5}$ values were sampled at grid points nearest to the $PM_{2.5}$ Network stations, CSN, or to the IMPROVE stations according to the analysis performed. In this study all of the analyses were performed for the Northeastern United States (NEUS) region encompassing the region from Virginia (including West Virginia) to Maine.

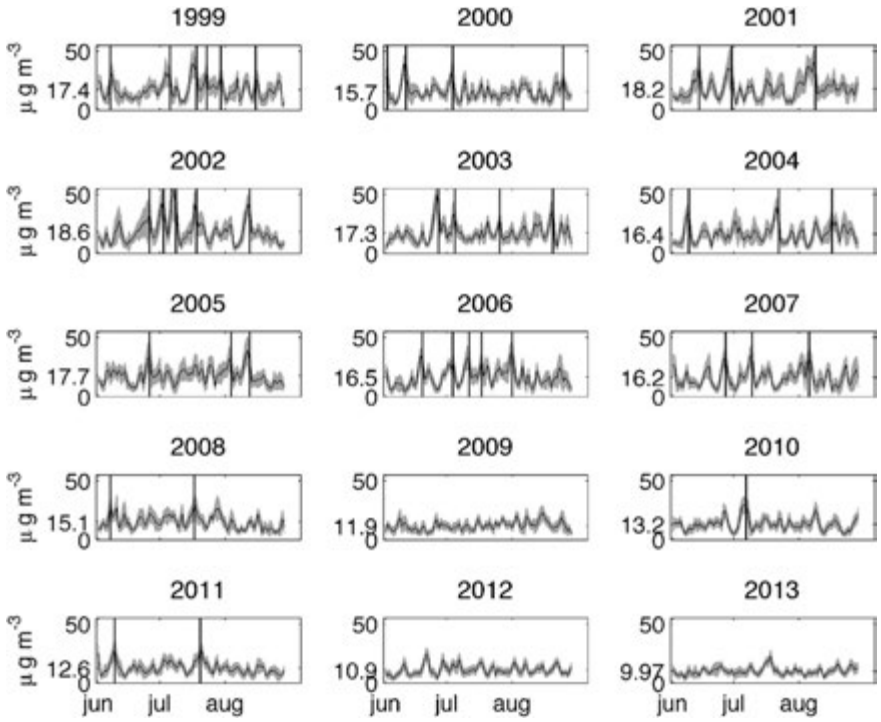

Fig. 2. Daily regional mean EPA Network $PM_{2.5}$ for the NEUS. 1 − standard deviation error bars. The vertical lines indicate events that are higher than the 95% of the concentration distribution for the summers of 1999–2013. The summer mean value is indicated in the vertical axis for each year.

used in the individual component analysis for sulfate (SO_4) (Code 88403) and OC (Code 88305) and comparisons of $PM_{2.5}$ speciation mass (Code 88502). These data were obtained from the Air Quality System. The locations of the CSN stations are shown in Fig. 1. Current speciation data files contain the raw speciation data from the national $PM_{2.5}$ network. The organic carbon data are reported as measured and have not been blank-corrected.

2.2. MERRAero Aerosol Reanalysis

Together with stations measurements we use aerosol output from the Modern-Era Retrospective Analysis for Research and Applications (MERRA) Aerosol Reanalysis (MERRAero) (Buchard et al., 2015). This reanalysis provides an estimate of the three-dimensional distribution of aerosol, including information on aerosol speciation, at high spatial (around 50 km horizontal resolution) and temporal (hourly) resolution, and enables examination of the evolution of aerosol distributions not possible from available ground data.

The MERRAero reanalysis product is obtained by running the Goddard Earth Observing System Version 5 (GEOS-5) (Rienecker et al., 2008) with the Goddard Chemistry Aerosol Radiation and Transport (GOCART) module in replay mode, i.e., driven by MERRA meteorology, and by assimilating aerosol optical depth (AOD) data from the Moderate Resolution Imaging Spectroradiometer (MODIS) satellite instruments (Buchard et al., 2014, 2015). MERRAero covers the period 2002–2014 at a nominal horizontal resolution of about 50 km (0.5° latitude by 0.625° longitude) with 72 vertical layers

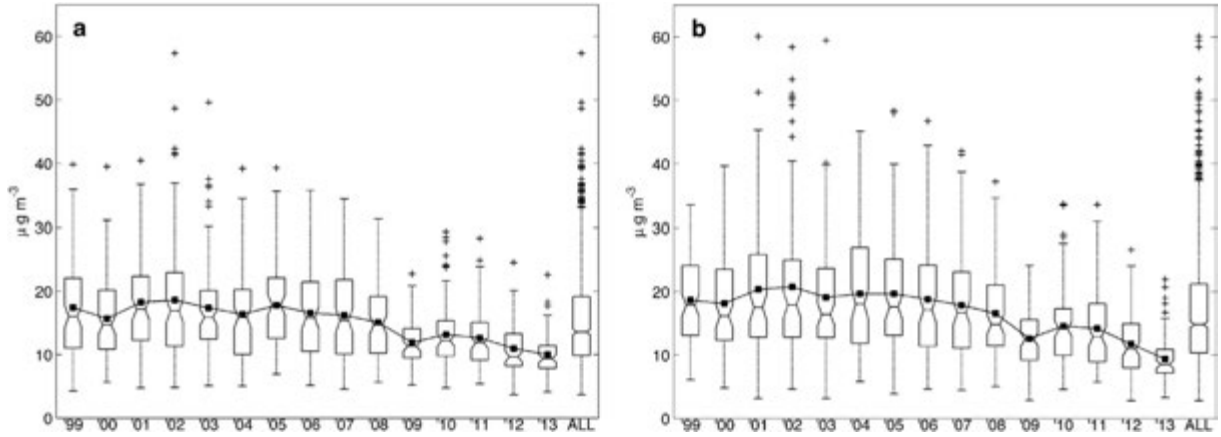

Fig. 3. Distributions of daily regional $PM_{2.5}$ mean from the EPA Network for (a) the Northeastern U.S. region and (b) Maryland, for the summers of the 1999–2013 period. On each box, the central mark is the median and the edges of the box are the 25th and 75th percentiles. The dot markers indicate the summer mean for each year. The whisker length corresponds to 99.3% of data coverage if the data were normally distributed. The ALL box plot shows the distribution of the data for the entire summer $PM_{2.5}$ population for the 1999–2013 period.

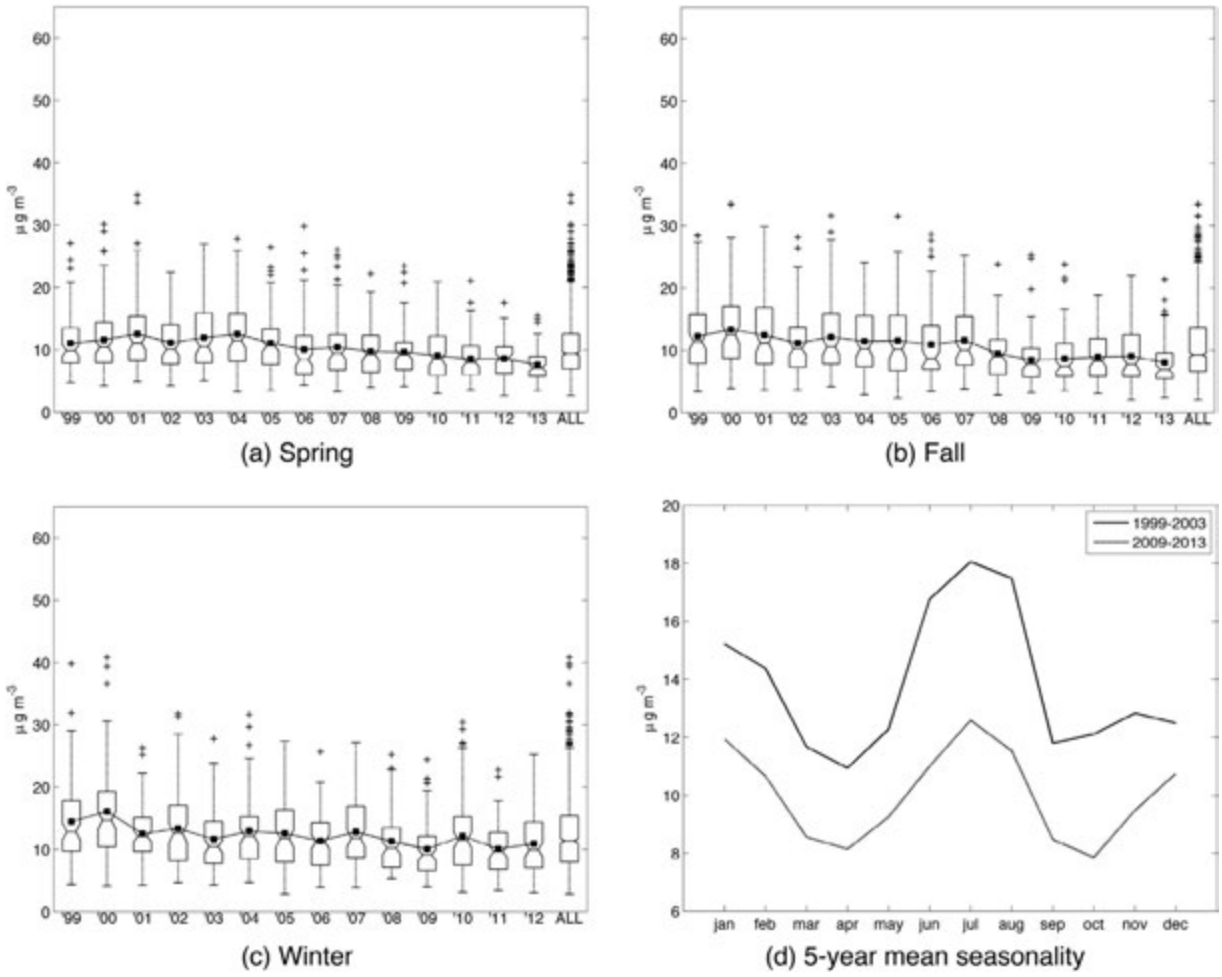

Fig. 4. Distributions of daily regional $PM_{2.5}$ mean from the EPA Network for the Northeastern U.S. region for the (a) springs (b) falls and (c) winters of the 1999–2013 period. On each box, the central mark is the median and the edges of the box are the 25th and 75th percentiles. The dot markers indicate the summer mean for each year. The whisker length corresponds to 99.3% of data coverage if the data were normally distributed. The ALL box plot shows the distribution of the data for the entire $PM_{2.5}$ population for the season of the 1999–2013 period. (d) 5-year monthly regional $PM_{2.5}$ Network mean for the Northeastern U.S. for a high-concentration period (1999–2003) is compared to a low concentration period (2009–2013).

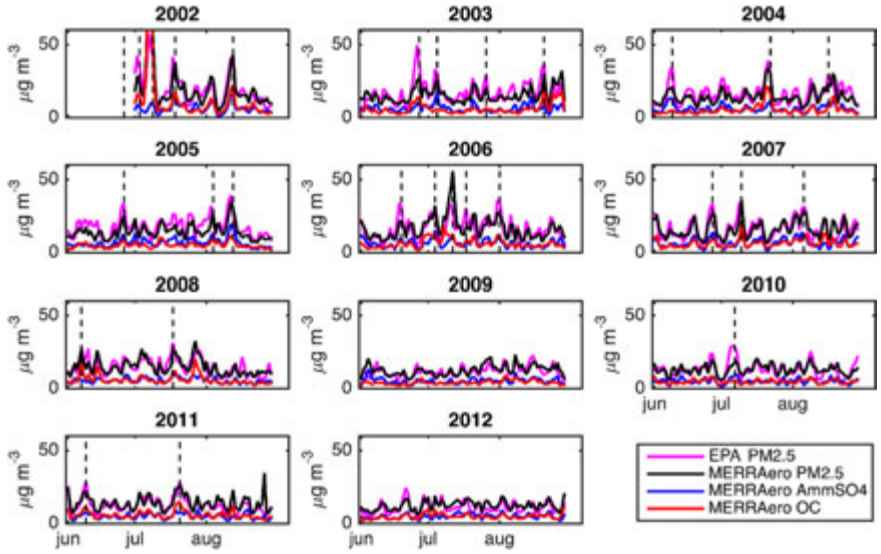

Fig. 5. Daily regional mean $PM_{2.5}$ from the EPA Network and MERRAero. The vertical lines indicate events that are higher than the 95% of the $PM_{2.5}$ concentration distribution for the summers of 1999–2013. Two of the components of the model output, organic carbon (OC) and ammonium sulfate ($AmmSO_4$) are also shown. The MERRAero regional daily averages were generated by sampling the model output at grid points nearest to the EPA's $PM_{2.5}$ Network stations as shown in Fig. 1.

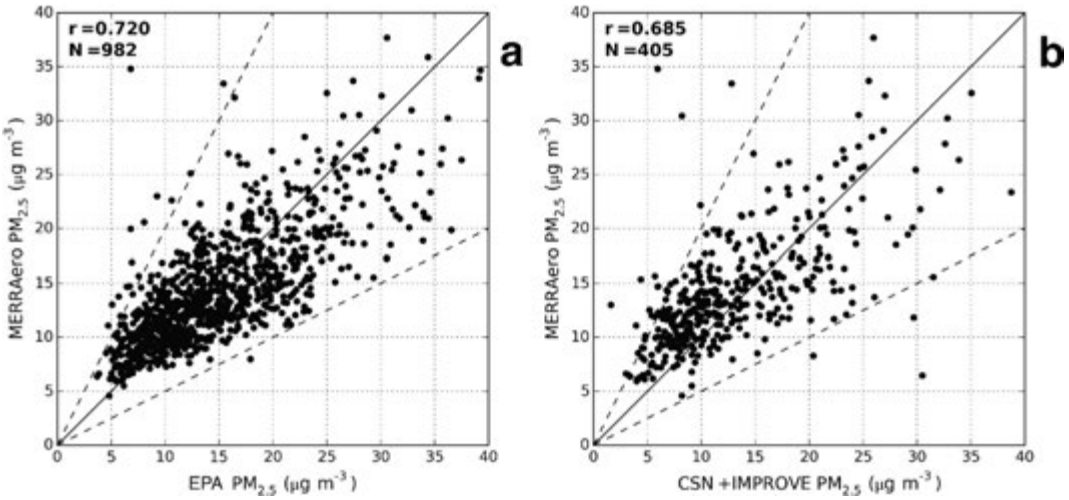

Fig. 6. Scatterplots of daily regional (NEUS) means of $PM_{2.5}$ showing the agreement between (a) MERRAero $PM_{2.5}$ vs. EPA's Network $PM_{2.5}$ and (b) MERRAero $PM_{2.5}$ vs. CSN and IMPROVE $PM_{2.5}$. The daily regional means are for the summers of the 2002–2012 period (N = 982 days). Given that IMPROVE and CSN stations don't sample daily, the data points are reduced to N = 405. For (a) the MERRAero output was sampled at grid points nearest to the EPA Network stations whereas for (b) the MERRAero grid points were sampled nearest to the CSN and IMPROVE stations.

between the surface and about 80 km. In the present work, the meteorological conditions during high $PM_{2.5}$ events were analyzed using MERRA meteorological fields.

The GOCART module simulates five aerosol species: organic carbon (OC), black carbon (BC), sulfate (SO_4), dust, and sea-salt aerosols treated as external mixtures that do not interact with each other (Chin et al., 2002; Colarco et al., 2010). The GOCART module does not include ammonium nitrate. Removal for all species occurs via convective and large-scale wet deposition and dry deposition, and additionally sedimentation for the coarser dust and sea-salt particles. MERRAero $PM_{2.5}$ is calculated by adding the ground-level concentrations of the individual species: OC, BC, dust, sea salt and $AmmSO_4$. $AmmSO_4$ mass is obtained by multiplying SO_4 mass by a factor of 1.375. This assumes that sulfate exists mainly in ammonium sulfate, with only a small fraction in sulfuric acid or ammonium bisulfate.

Emissions of OC, BC, SO4, and relative precursor gases are prescribed in MERRAero, see (Buchard et al., 2014) for details. The anthropogenic carbon dioxide emissions are from the Emission Database for Global Atmospheric Research (EDGAR) Version 4.1 inventory, and biomass burning emissions are from the NASA Quick Fire Emission Dataset (QFED) Version 2.1. The production of sea-salt aerosols follows the Gong (2003) formulation of size dependent number flux with a modified windspeed term equal to $u_*^{2.41}$ (u_* is the friction velocity) and a new sea surface temperature correction term to modulate the strength of sea-salt emissions.

Daily averages of modeled $PM_{2.5}$ were calculated from sampling the hourly MERRAero fields using the grid points nearest to the EPA's $PM_{2.5}$ Network stations (see Fig. 1). In some instances, the MERRAero grid points were sampled nearest to the CSN and IMPROVE stations to make equitable comparisons. Data from CSN and IMPROVE were combined to calculate daily regional averages of $PM_{2.5}$ (Fig. 6b), OC, and $AmmSO_4$ (Fig. 7) for the NEUS.

3. $PM_{2.5}$ variability

3.1. Summer $PM_{2.5}$

We first examine the temporal variability of $PM_{2.5}$ in the Northeastern United States (NEUS) for the summers between 1999 and 2013. As described in Section 2, the regional mean $PM_{2.5}$

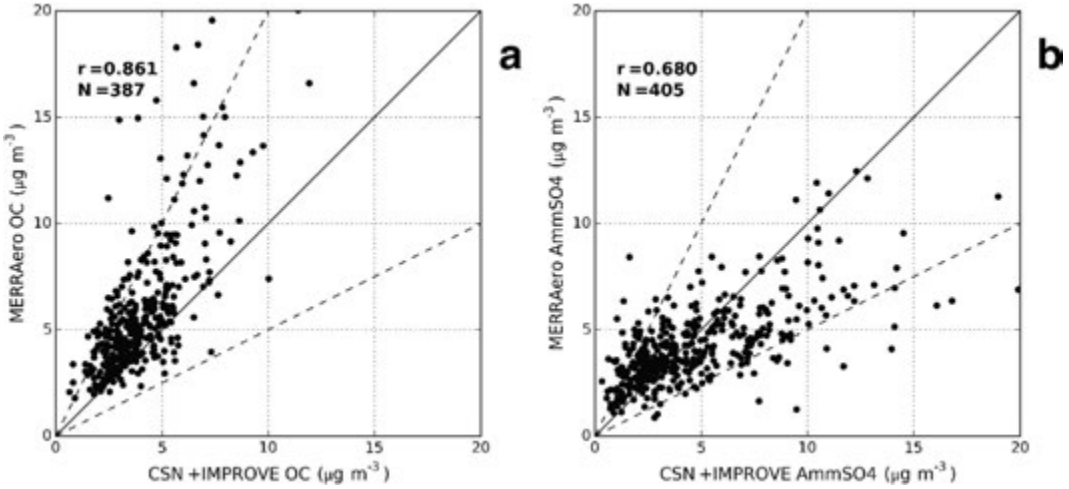

Fig. 7. Scatterplots of daily regional (NEUS) means of two $PM_{2.5}$ components showing the agreement between MERRAero and CSN and IMPROVE (a) OC and (b) $AmmSO_4$. The MERRAero grid points were sampled nearest to the CSN and IMPROVE stations.

concentration for the NEUS was calculated from 24-h average concentrations at local conditions sampled at the EPA's $PM_{2.5}$ Network stations in the NEUS. Fig. 2 shows the time series of the mean and standard deviation of $PM_{2.5}$ concentrations in the NEUS for each summer (June–August) between 1999 and 2013. There is considerable day-to-day variability of the NEUS-mean $PM_{2.5}$ within a summer, as well as some large differences between years.

Focusing first on the daily variability, the most striking feature is the occurrence of short (1–3 day) events where there are high regional-mean $PM_{2.5}$ (e.g., exceeding 30 $\mu g\ m^{-3}$) and periods of similar or longer length where the mean $PM_{2.5}$ is low (less than 10 $\mu g\ m^{-3}$). The frequency of events with high $PM_{2.5}$ for the regional mean concentration varies between years, with less frequent high events in later years. For example, the vertical lines in Fig. 2 correspond to events when the daily NEUS-mean $PM_{2.5}$ concentrations are larger than the 95% for the summer $PM_{2.5}$ concentrations of the period 1999–2013 (around 28 $\mu g\ m^{-3}$). There are 3–5 of these events for each summer between 1999 and 2007, but for later years there are between 0 and 2 events each summer. Another interesting feature is the variability in the magnitude of the high-$PM_{2.5}$ events. Whereas the regional concentrations of $PM_{2.5}$ exceed 35 $\mu g\ m^{-3}$ for many events before 2008, the regional mean concentration has not been over 29 $\mu g\ m^{-3}$ after 2008. See below for further discussion.

There is also considerable interannual variability in the summer NEUS-mean values (marked on the lower y-axis of plots in Fig. 2), including a decline from 1999 to 2013. This is more clearly seen quantified in "box–whisker" plots for each summer, shown in Fig. 3a. The box plot marked as ALL, shows the distribution of the entire summer $PM_{2.5}$ population for the 1999–2013 period. On each box, the central mark is the median and the edges of the box are the 25th and 75th percentiles, while the whiskers show the range of 99.3% of the data if it were normally distributed. The dot markers and plot indicate the mean values for the individual summers. Fig. 3a shows a decline in the summer-mean NEUS $PM_{2.5}$, as well as a decrease in the variability (interquartile range) over the summer and the magnitude of extreme events. Fitting a linear trend to the summer mean $PM_{2.5}$ yields a decrease of 0.54 $\mu g\ m^{-3}$ per year, which corresponds to a decrease of 7.5 $\mu g\ m^{-3}$ over the 14 year period. The extreme concentrations, as indicated by the whisker length and the outliers, have also declined over this period. There has also been a considerable reduction in the interquartile range since 2008 indicating a reduction in the variability of regional summer $PM_{2.5}$ concentrations.

The above analysis has focused on the NEUS regional-average $PM_{2.5}$, but very similar variability occurs for averages over individual states. For example, Fig. 3b shows the distributions of daily-mean PM averaged over stations within Maryland. The values for Maryland are slightly higher than the NEUS average, but the decreases in mean values, range, and extreme events from 1999 to 2013 are similar. This also holds for other states in the Northeastern US region. This consistency among states implies that most high-$PM_{2.5}$ events have a large spatial coverage.

A likely contributor to the reduction in summer-mean $PM_{2.5}$ is reduced SO_2 emissions. SO_2 emissions are precursors of sulfates which are one of the main components of aerosols in the NEUS, and there has been an observed decrease in SO_2 emissions over the last decade (U.S. Environmental Protection Agency, 2012; Hand et al., 2012; de Gouw et al., 2014). This includes a large decrease in emissions from 2008 to 2009, due to the economic recession, that likely contributes to the drop in NEUS $PM_{2.5}$ in 2009. Changes in SO_2 emissions are also likely a factor in the decrease in occurrence and magnitude of high-$PM_{2.5}$ events. While there is qualitative agreement, it is notable that the percentage decrease in regional summer $PM_{2.5}$ concentrations (46%) is less than the percentage decrease in SO_2 emissions (63%) (de Gouw et al., 2014). This is likely because there are other aerosol emission sources (see below), that have not decreased during this period.

3.2. Other seasons

There is also similar interannual variability for NEUS $PM_{2.5}$ for different seasons. Fig. 4a–c show analogous box–whisker plots to Fig. 3 except for spring, fall, and winter respectively. In all seasons there is a decrease in mean values, range, and extreme events from 1999 to 2013. However, the magnitude of the long-term decrease in summer (over 7 $\mu g\ m^{-3}$) is much larger than other seasons (around 5 $\mu g\ m^{-3}$). The anomalous high values have also decreased: in the summer the outlying regional $PM_{2.5}$ values would reach 60 $\mu g\ m^{-3}$, while in the winter they would only reach 40 $\mu g\ m^{-3}$.

The seasonality and the reduction in regional mean $PM_{2.5}$ can be seen more clearly when comparing the monthly means for the first five years of the data record with means for the last 5 years, see Fig. 4d. For both periods the spring and fall regional mean concentrations are smaller than in summer and winter months, but there has been a change in the summer-winter contrast. The values for summer months are much higher than for winter months for the 1999–2003 period but, because of the larger summer decrease, in the 2009–2013 period the summer levels are similar to those of the winter months.

The cause of the large reduction in summer $PM_{2.5}$ compared to other seasons is unknown. Examination of power plant SO_2 emissions between 2000 and 2013 shows similar decreasing trends for all seasons. Thus changes in these emissions do not appear to be the cause of the change in seasonality of $PM_{2.5}$.

4. High-$PM_{2.5}$ events

We now examine the characteristics and sources of the high-$PM_{2.5}$ events identified above. To explore the causes of these events,

Table 1
High $PM_{2.5}$ events in the Northeastern US with corresponding OC to $AmmSO_4$ ratio and potential sources. The labels fire, anthropogenic or mix indicate high-$PM_{2.5}$ events that were caused by wildfires, anthropogenic pollution or a mix of both respectively. For some events, a source has not been identified using the described methodology, these are labeled as undetermined.

Event	Date	OC/$AmmSO_4$ ratio	Source
01	2002-07-09	4.32	Wildfire
02	2002-07-19	1.09	Mix
03	2002-08-14	1.43	Undetermined
04	2003-06-27	1.53	Wildfire
05	2003-07-05	0.33	Anthropogenic
06	2003-07-27	1.10	Wildfire
07	2003-08-22	2.18	Wildfire
08	2004-06-09	0.32	Anthropogenic
09	2004-07-23	1.62	Wildfire
10	2004-08-18	0.73	Mix
11	2005-06-26	0.72	Mix
12	2005-08-05	0.74	Anthropogenic
13	2005-08-14	0.58	Anthropogenic
14	2006-06-19	0.80	Mix
15	2006-07-04	0.86	Mix
16	2006-07-12	1.06	Dust
17	2006-07-18	1.26	Mix
18	2006-08-02	1.19	Mix
19	2007-06-27	0.69	Anthropogenic
20	2007-07-10	1.50	Undetermined
21	2007-08-07	0.96	Mix
22	2008-06-07	3.13	Wildfire
23	2008-07-18	0.87	Mix
24	2010-07-07	0.61	Anthropogenic
25	2011-06-09	1.62	Wildfire
26	2011-07-21	1.95	Wildfire

we use $PM_{2.5}$ output from the MERRAero reanalysis. MERRAero provides estimates of the three-dimensional distribution of major tropospheric aerosol components, including $AmmSO_4$ and OC, at a constant spatial and temporal resolution. This enables an examination of the evolution of the three-dimensional aerosol distribution in the days leading up to the high-$PM_{2.5}$ events, that is not possible from data alone.

4.1. MERRAero evaluation

Before analyzing the high-$PM_{2.5}$ events we briefly examine how well MERRAero reproduces the temporal variations in the $PM_{2.5}$ measurements. Buchard et al. (2015) compared the MERRAero $PM_{2.5}$ and ground-based measurements over the USA for a similar period (2003–2012) and showed that there is a negative bias of MERRAero $PM_{2.5}$ as compared to EPA's Network $PM_{2.5}$, particularly in the Midwest (a positive bias was found in the Northeastern US). They also found a stronger positive correlation (in 2010) between MERRAero and station $PM_{2.5}$ in the Eastern US (lower correlations were found in the West Coast). In this work we focus on summer $PM_{2.5}$ over the NEUS.

Fig. 5 shows the temporal variability in the NEUS mean $PM_{2.5}$ from the EPA's $PM_{2.5}$ Network (dashed) and MERRAero (solid) for summers between 2002 and 2012, as well as for other $PM_{2.5}$ components. (The MERRAero mean is calculated using the model grid point closest to the $PM_{2.5}$ Network stations shown in Fig. 1). There is a high bias (20%) when the regional $PM_{2.5}$ mean is calculated using MERRAero sampled nearest to the $PM_{2.5}$ Network stations as opposed to using all the grid points within the NEUS domain. This can be attributed to the sensors being located in

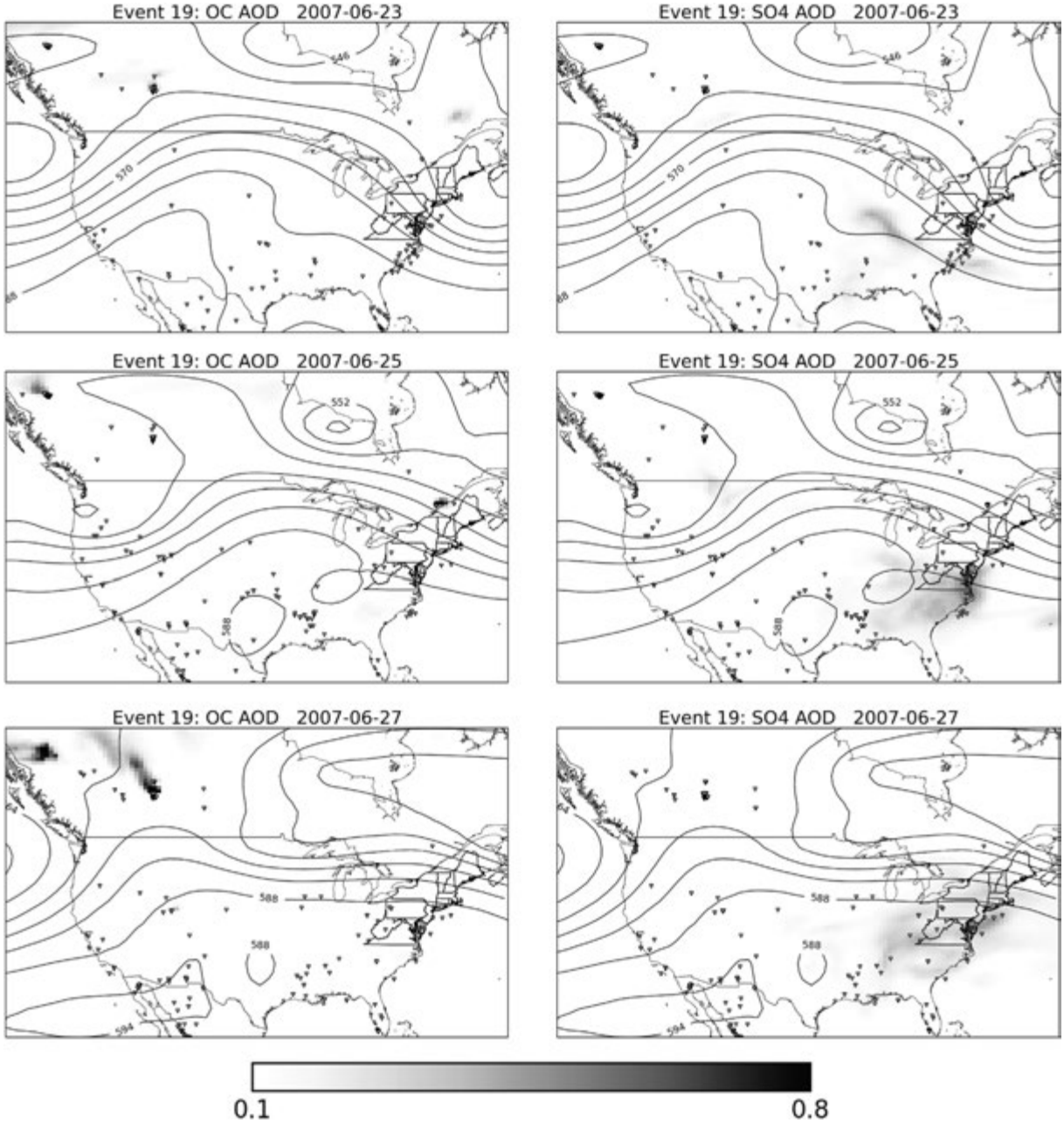

Fig. 8. Source analysis for the 2007-06-27 event of anthropogenic pollution ($OC/AmmSO_4 = 0.69$) indicated in Table 1 as Event #19. The shaded surface is MERRAero aerosol optical depth (AOD) for OC (left column) and SO_4 (right column). The contours are geopotential height in decameters at the 500 hPa level. Triangles show MODIS fire hotspots.

highly populated areas and also to the events not covering the complete extent of the region. MERRAero closely reproduces the observed temporal variability of the observed regional mean $PM_{2.5}$. In particular, the modeled $PM_{2.5}$ has similar daily, interannual and longer term variability. Furthermore, MERRAero reproduces the occurrence of short periods with high $PM_{2.5}$, as well as the long term decrease in summer mean values from 2002 to 2012.

There is good correlation ($r = 0.72$) between EPA's $PM_{2.5}$ and MERRAero $PM_{2.5}$ concentrations. However, there is a tendency for MERRAero $PM_{2.5}$ to be less than the $PM_{2.5}$ Network measurements (i.e., MERRAero has a low bias) at high $PM_{2.5}$ values and a high bias at low $PM_{2.5}$ values, see Fig. 6a. A bias is also found between MERRAero $PM_{2.5}$ and $PM_{2.5}$ from IMPROVE and CSN, see Fig. 6b.

We now examine the capability of MERRAero to reproduce the concentration of individual $PM_{2.5}$ components by comparing the reanalysis output to data from the IMPROVE and CSN networks. As described in Section 2, MERRAero output includes five aerosol constituents: SO_4, OC, BC, dust, and sea-salt. In this analysis we focus our attention on $AmmSO_4$ and OC which are the predominant components of $PM_{2.5}$ in the NEUS (e.g., IMPROVE 2011). As has been described, data from the CSN and IMPROVE stations were combined to estimate daily regional means of $PM_{2.5}$ (from speciation networks), OC and $AmmSO_4$. As seen in Fig. 7, there is good agreement between the reanalysis and the station data for both components. MERRAero tends to overestimate $AmmSO_4$ concentrations at low values but underestimates $AmmSO_4$ at high values. Also, there is a high bias for the estimates of OC from MERRAero when compared to station data. This is consistent with the findings of previous authors (Buchard et al., 2015).

The above analysis shows that although there are some biases between MERRAero and surface aerosol measurements, MERRAero reproduces the observed temporal variations and the relative

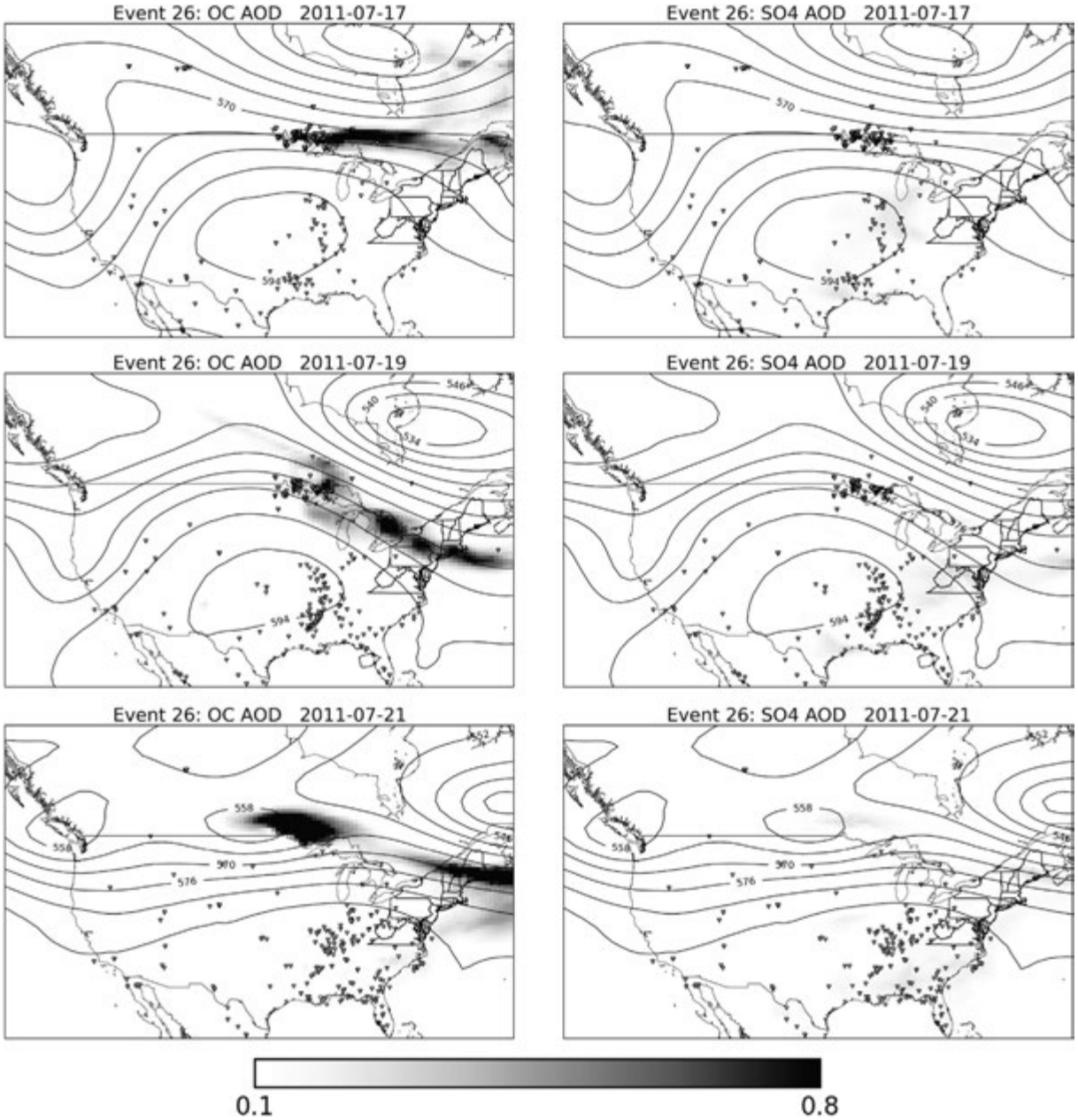

Fig. 9. Source analysis for the 2011-07-21 wildfire event ($OC/AmmSO_4 = 1.95$) indicated in Table 1 as Event #26. As in Fig. 8, the shaded surface is MERRAero AOD for OC (left column) and SO_4 (right column). The contours are geopotential height in decameters at the 500 hPa level. Triangles show MODIS fire hotspots.

magnitude of OC to $AmmSO_4$. This agreement indicates that MERRAero can be used to examine the source of high-$PM_{2.5}$ events.

4.2. $PM_{2.5}$ source attribution

In Fig. 5 we can see that the relative contribution of $AmmSO_4$ and OC varies between high-$PM_{2.5}$ events. For some events, there is a large increase in OC but little change in $AmmSO_4$ (e.g., 9 July 2002, 23 July 2004), whereas the opposite occurs for other events (e.g., 5 July 2003, 9 June 2004). This contrast in composition suggests differences in aerosol sources. We expect aerosols from anthropogenic sources to have higher $AmmSO_4$ concentrations, while we expect high concentrations of OC from biomass burning sources (Kang et al., 2014). With this in mind, we use the OC/$AmmSO_4$ ratio as a simple metric to identify potential aerosol sources. The above characterization of events by their OC/$AmmSO_4$ ratio is only a rough estimate of the source and cause of the events (especially as MERRAero does not assimilate information on the aerosol speciation).

The OC/$AmmSO_4$ ratio in MERRAero $PM_{2.5}$ for the events identified in Fig. 5 are listed in Table 1. The ratio values vary from 0.39 to 4.31 among the events, with roughly equal number of events with OC/$AmmSO_4$ less than or larger than one. This variation in the OC/$AmmSO_4$ ratio suggests a range of causes for the high-$PM_{2.5}$ episodes and roughly equal contributions from anthropogenic (industrial) and natural (wildfire) sources.

To explore the potential sources of the high-$PM_{2.5}$ events we take advantage of the full time-varying three-dimensional aerosol fields from MERRAero. For each event, we use these fields together with MERRA meteorological fields and trajectory calculations to examine the evolution of aerosol before the events. In the presentation below we focus on the MERRAero AOD for OC and for SO_4 and the MERRA 500 hPa geopotential height fields. The AOD and geopotential height illustrate the flow and transport of $PM_{2.5}$ into the NEUS region.

First consider the June 27, 2007 event. This event has a ratio OC/$AmmSO_4$ = 0.69 and potentially has an anthropogenic source. As shown in Fig. 8, a region of high $AmmSO_4$ concentration starts to generate on June 22 and 23 in the Midwest. On June 24, a high pressure system causes the pollution to accumulate. Between June 25 and 27, $AmmSO_4$ that has been transported into the NEUS further accumulates due to a strengthening high pressure system. Even though there are high concentrations of OC in Canada, likely due to wildfires (June 25 and 26), only a small amount of it is advected into the NEUS (Fig. 8). Thus, analysis of the evolution of MERRAero aerosol distributions supports the hypothesis that this high-$PM_{2.5}$ event was due to local anthropogenic sources. Further support for this hypothesis is found in back trajectory calculations which shows the majority of air parcels coming from the Midwest (not shown).

Next consider the high-concentration event of July 21, 2011. This event has a ratio OC/$AmmSO_4$ = 1.95, which suggests a wildfire source for this event. As shown in Fig. 9, there is a distinct plume of OC that can be attributed to Canadian wildfires (the triangles show MODIS fire hotspots). A large amount of material is advected from Canada to the NEUS, suggesting that wildfires are the main source of the high $PM_{2.5}$ in this event. While $AmmSO_4$ is also traced to the fire, some of it is generated in the Midwest and locally and its concentration increased by a high pressure system.

Similar analyses to that above were performed for all of the

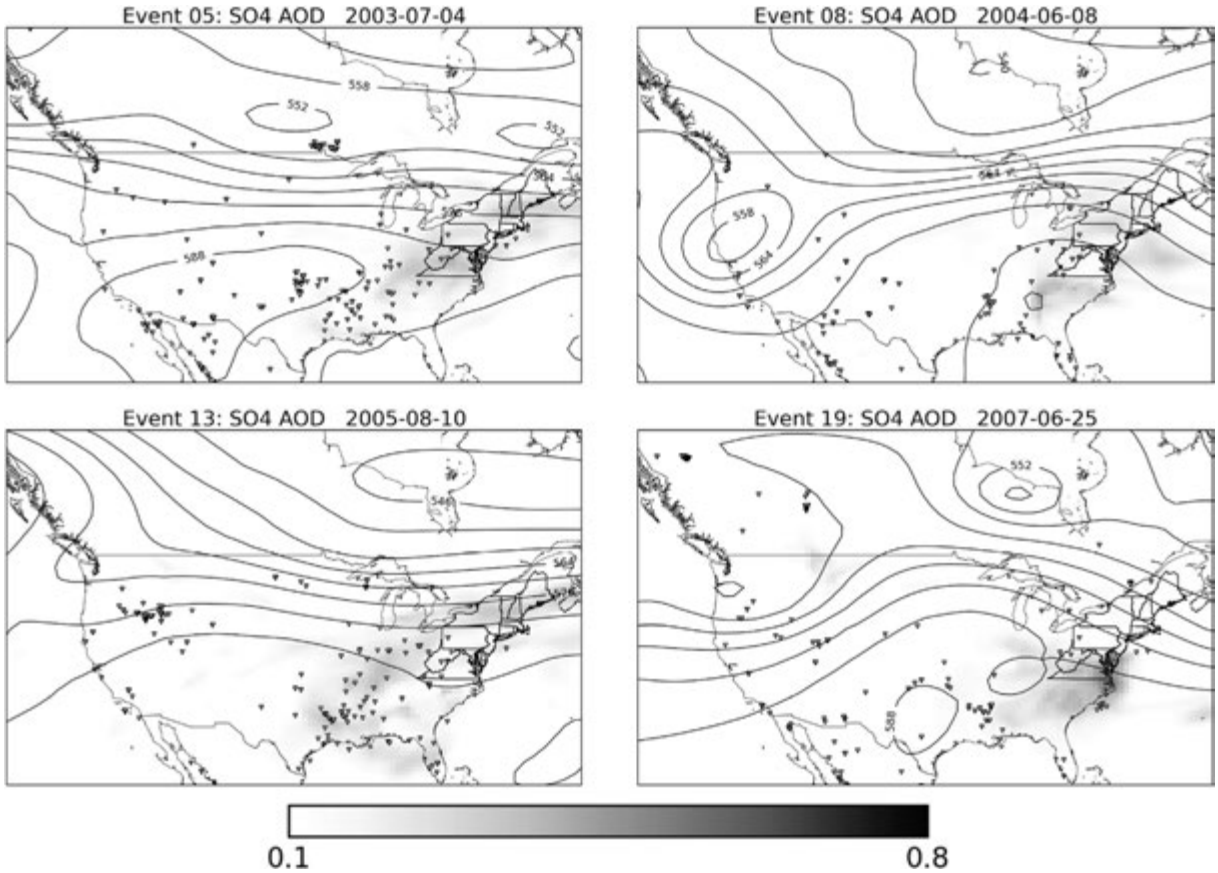

Fig. 10. Examples of meteorology associated with high-$PM_{2.5}$ events of anthropogenic sources. The shaded surface is MERRAero AOD for SO_4. The contours are geopotential height in decameters at the 500 hPa level. Triangles show MODIS fire hotspots.

events listed in Table 1. In general, events with an OC/$AmmSO_4$ ratio less than 0.7, have anthropogenic sources, whereas events with OC/$AmmSO_4$ ratios larger than 1.5 are caused by biomass burning. For events with an OC/$AmmSO_4$ ratio nearer to 1 the causes appear to be a mixture of these two types of sources.

For events with OC/$AmmSO_4$ ratios less than 0.7, meteorology from MERRA shows a consistent pattern. As shown in Fig. 10 the events are characterized by a developing high pressure system in the Southern U.S. that strengthens throughout the duration of the event. Aerosols from the Midwest are advected into the NEUS, increasing the concentration in the region.

On the other hand, events with an OC/$AmmSO_4$ ratio larger than 1.5 are usually caused by boreal fire emissions accompanied by a strong low-pressure system in Eastern Canada and an off-phased high pressure system in the Midwest. The meteorology involved in these events and the locations of the sources are much more varied than those for anthropogenic events. This is illustrated in Fig. 11, and further discussion for individual events can be found in Colarco et al. (2004), Mathur (2008) and Miller et al. (2011). Pollution transported from wildfires can have a strong impact in the air quality of regions already burdened by high background aerosol concentrations, such as populated areas in the NEUS, particularly when coupled with high pressure systems that cause pollution accumulation (Bein et al., 2008). A crucial factor that determines the transport of aerosols from a wildfire to a remote location is the altitude of the emission injection. Pollution transport is enhanced when the emission are injected at high elevations above the boundary layer. However, these high altitude injections are not expected in smoldering or peat fires (Turquety et al., 2007). Colarco et al. (2004) have found that an important mechanism for transporting pollutants from elevated layers to the surface is by entrainment into the planetary boundary layer of a gradually subsiding plume, such as in the July 9, 2002 event (Event #1).

In the case of mixed events, the meteorology consists of westerly flow over the Midwest with air already laden with OC from biomass burning, as can be seen in Fig. 12 which shows two examples of meteorology associated with high-$PM_{2.5}$ events of mixed anthropogenic and biomass burning sources. For both examples shown (events #10 and #21), the 500 hPa level height over the NEUS is not particularly high as compared to the anthropogenic events, but some accumulation can still be observed, specially in the southern portion of the region.

5. Conclusions

Analysis of EPA $PM_{2.5}$ station measurement shows large temporal variability in the $PM_{2.5}$ averaged over the Northeastern United States (NEUS), on daily, interannual and longer time scales. On the daily time scales the most striking feature is the occurrence of 1–3 day events with high NEUS-mean $PM_{2.5}$. The frequency and intensity of these high-concentration events varies between years, and there is a decrease in both the frequency and intensity over the period considered. There is also a significant decrease in summer mean $PM_{2.5}$ between 1999 and 2013. There are much smaller decreases in other seasons, resulting in an overall decrease in the seasonality of $PM_{2.5}$.

The composition of the $PM_{2.5}$ varies among high-concentration events, with $PM_{2.5}$ in some events primarily composed of $AmmSO_4$,

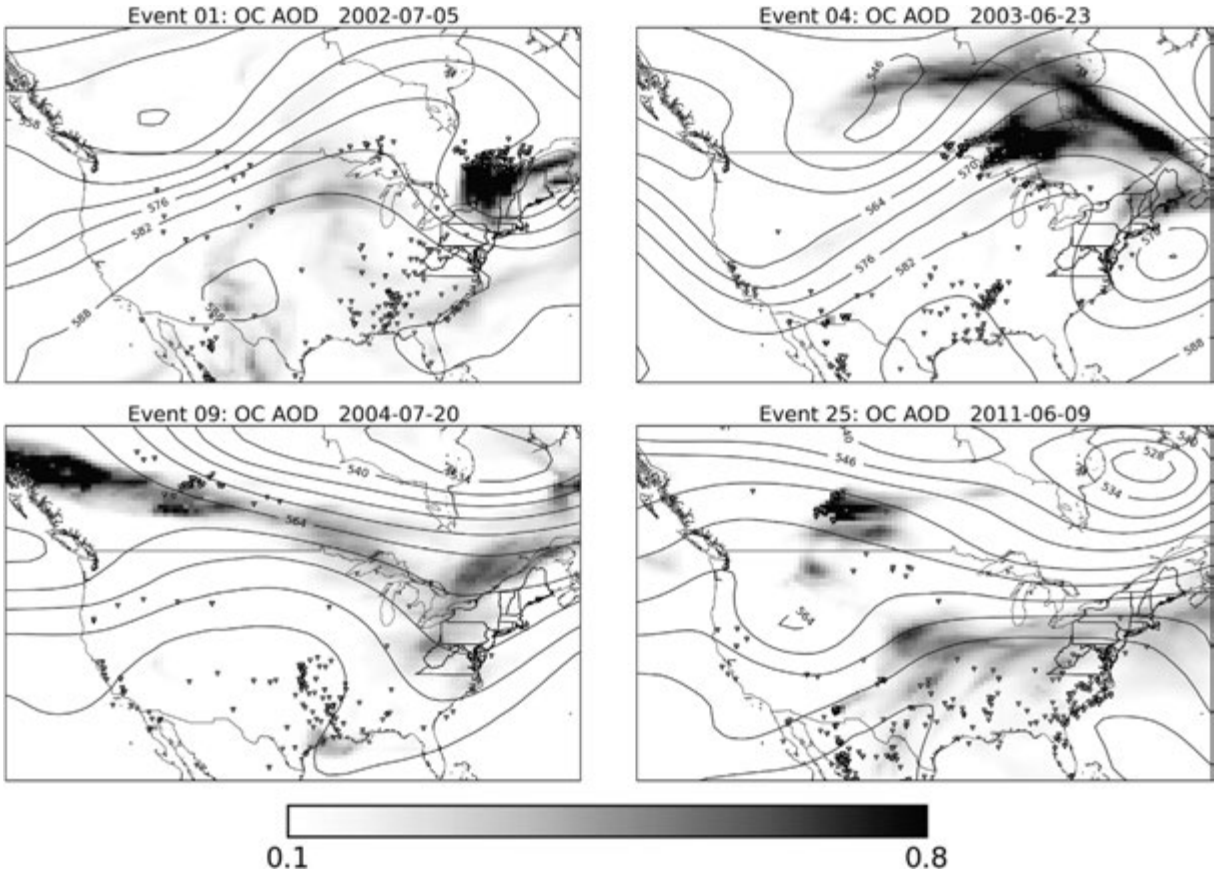

Fig. 11. Examples of meteorology associated with high-$PM_{2.5}$ events of biomass burning sources. The shaded surface is MERRAero AOD for OC. The contours are geopotential height in decameters at the 500 hPa level. Triangles show MODIS fire hotspots.

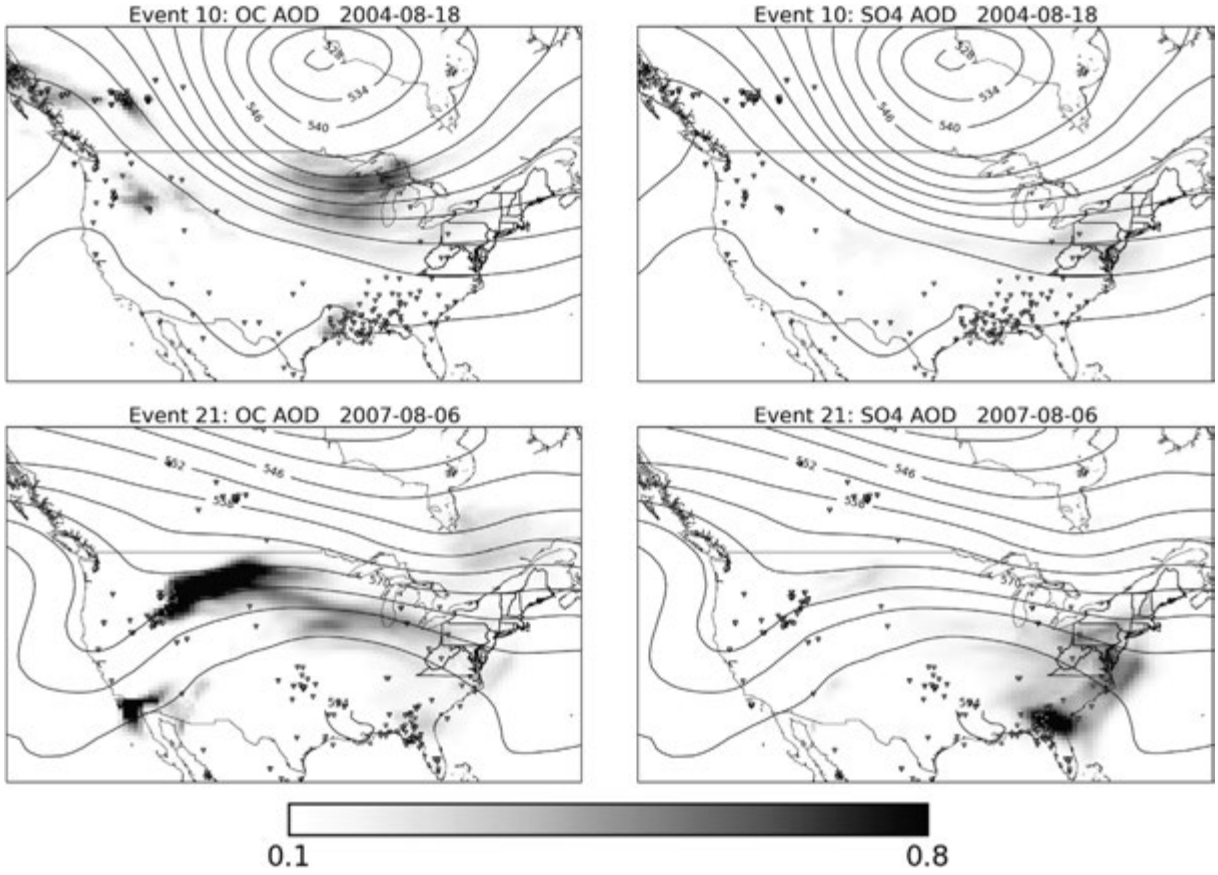

Fig. 12. Examples of meteorology associated with high-$PM_{2.5}$ events of mixed anthropogenic and biomass burning sources. As in Fig. 8, the shaded surface is MERRAero AOD for OC (left column) and SO_4 (right column). The contours are geopotential height in decameters at the 500 hPa level. Triangles show MODIS fire hotspots.

while in other events OC is the main component. These differences in $PM_{2.5}$ composition are linked to differences in the cause of the high $PM_{2.5}$. Events where $AmmSO_4$ is the main aerosol component, are caused by a combination of anthropogenic emissions in the Midwest and slow-moving high pressure systems. These high pressure systems develop in the Southeastern U.S., advect emissions from the Midwest and cause accumulation of $PM_{2.5}$ in the Northeastern U.S. In contrast, OC-dominated events are generally due to long-range transport from wildfires in Canada, Alaska or western US. For the 2002–2012 period about half of the high-$PM_{2.5}$ events had either an anthropogenic or a wildfire origin. The other half of the events was caused by a mixture of these sources.

Future work is planned to extend the analysis presented here in several ways. First, we plan a more detailed analysis of the meteorology that leads to the different types of high-concentration events. We will also examine other potential sources (e.g. vehicle emissions) and extend the analysis to consider other regions within the USA, where there is a different composition of $PM_{2.5}$ (e.g. Hand et al., 2011). Finally, analysis of the health impacts of high-concentration events (e.g. Le et al., 2014) could provide information on whether some constituents of $PM_{2.5}$ are more harmful than others.

Acknowledgments

We would like to acknowledge the support from the Glenadore and Howard L. Pim Postdoctoral Fellowship. We also would like to thank Peter R. Colarco, Mian Chin, Virginie Buchard and Arlindo M. Da Silva at the NASA Goddard Space Flight Center for providing the MERRAero output and helpful discussions.

References

Bein, K.J., Zhao, Y., Johnston, M.V., Wexler, A.S., 2008. Interactions between boreal wildfire and urban emissions. J. Geophys. Res. 113 (D7), D07304.

Bell, M.L., Ebisu, K., Peng, R.D., Samet, J.M., Dominici, F., 2009. Hospital admissions and chemical composition of fine particle air pollution. Am. J. Respir. Crit. Care Med. 179 (12), 1115–1120. http://dx.doi.org/10.1164/rccm.200808-1240OC.

Bell, M., Ebisu, K., Leaderer, B., Gent, J., Lee, H., Koutrakis, P., Wang, Y., Dominici, F., Peng, R., 2014. Associations of $PM_{2.5}$ constituents and sources with hospital admissions: analysis of four counties in Connecticut and Massachusetts (USA) for persons ≥65 years of age. Environ. Health Perspect. 122 (2), 138–144.

Brook, R.D., 2008. Cardiovascular effects of air pollution. Clin. Sci. 115, 175–187.

Buchard, V., da Silva, A.M., Colarco, P.R., Darmenov, A., Randles, C.A., Govindaraju, R., Torres, O., Campbell, J., Spurr, R., 2014. Using the OMI aerosol index and absorption aerosol optical depth to evaluate the NASA MERRA aerosol reanalysis. Atmos. Chem. Phys. Discuss. 14, 32177–32231.

Buchard, V., da Silva, A., Randles, C.A., Colarco, P., Ferrare, R., Hair, J., Hostetler, C., Tackett, J., Winker, D., 2015. Evaluation of the surface PM2.5 in Version 1 of the NASA MERRA Aerosol Reanalysis over the United States. Under review in Atmos. Environ.

Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B., Duncan, B., Martin, R., Logan, J., Higurashi, A., Nakajima, T., 2002. Tropospheric aerosol optical thickness from the gocart model and comparisons with satellite and sun photometer measurements. J. Atmos. Sci. 59 (3), 461–483.

Chin, M., Diehl, T., Ginoux, P., Malm, W., 2007. Intercontinental transport of pollution and dust aerosols: implications for regional air quality. Atmos. Chem. Phys. 7 (21), 5501–5517.

Colarco, P., Schoeberl, M., Doddridge, B., Marufu, L., Torres, O., Welton, E., 2004. Transport of smoke from Canadian forest fires to the surface near Washington, DC: injection height, entrainment, and optical properties. J. Geophys. Res. Atmos. 109, D06203.

Colarco, P., da Silva, A., Chin, M., Diehl, T., 2010. Online simulations of global aerosol distributions in the nasa geos-4 model and comparisons to satellite and ground-based aerosol optical depth. J. Geophys. Res. Atmos. 115, D14207.

Kang, Choong-Min, Gold, Diane, Koutrakis, Petros, October 2014. Downwind O_3 and $PM_{2.5}$ speciation during the wildfires in 2002 and 2010. Atmos. Environ. 95, 511–519. http://dx.doi.org/10.1016/j.atmosenv.2014.07.008. ISSN 1352-2310.

de Gouw, J.A., Parrish, D.D., Frost, G.J., Trainer, M., 2014. Reduced emissions of CO_2,

NO_x, and SO_2 from U.S. power plants owing to switch from coal to natural gas with combined cycle technology. Earth's Future 2 (2), 75–82.
de Hartog, J.J., Lanki, T., Timonen, K.L., Hoek, G., Janssen, N.A., Ibald-Mulli, A., Peters, A., Heinrich, J., Tarkiainen, T.H., Grieken, R.V., Wijnen, J.H.V., Brunekreef, B., Pekkanen, J., 2009. Associations between $PM_{2.5}$ and heart rate variability are modified by particle composition and beta-blocker use in patients with coronary heart disease. Environ. Health Perspect. 177 (1), 105–111.
DeBell, L., Talbot, R., Dibb, J., Munger, J., Fischer, E., Frolking, S., 2004. A major regional air pollution event in the Northeastern United States caused by extensive forest fires in Quebec, Canada. J. Geophys. Res. Atmos. 109 (D19).
Dharshana, K.G.T., Kravtsov, S., Kahl, J.D.W., 2010. Relationship between synoptic weather disturbances and particulate matter air pollution over the United States. J. Geophys. Res. 115 (D24), D24219. http://dx.doi.org/10.1029/2010JD014852.
Dominici, F., Peng, R., Bell, M., Pham, L., McDermott, A., Zeger, S., Samet, J., 2006. Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases. JAMA: J. Am. Med. Assoc. 295 (10), 1127–1134.
Duck, T.J., Firanski, B.J., Millet, D.B., Goldstein, A.H., Allan, J., Holzinger, R., Worsnop, D.R., White, A.B., Stohl, A., Dickinson, C.S., Donkelaar, A.V., 2007. Transport of forest fire emissions from Alaska and the Yukon territory to Nova Scotia during summer 2004. J. Geophys. Res. 112 (D10), D10S44.
Gong, S., 2003. A parameterization of sea-salt aerosol source function for sub- and super-micron particles. Glob. Biogeochem. Cycles 17 (4), 1094.
Hand, J.L., Copeland, A., Day, D.E., Dillner, A.M., Indresand, H., Malm, W.C., McDade, C.E., Moore, C.T., Pitchford, M.L., Schichtel, B.A., Watson, J.G., 2011. Interagency Monitoring of Protected Visual Environments (IMPROVE). Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States: Report V (Technical Report). Cooperative Institute for Research in the Atmosphere (CIRA), Colorado State University.
Hand, J.L., Schichtel, B.A., Malm, W.C., Pitchford, M.L., 2012. Particulate sulfate ion concentration and SO_2 emission trends in the United States from the early 1990s through 2010. Atmos. Chem. Phys. 12 (21), 10353–10365.
Jalava, P., Salonen, R., Halinen, A., Penttinen, P., Pennanen, A., Sillanpaa, M., Sandell, E., Hillamo, R., Hirvonen, M., 2006. In vitro inflammatory and cytotoxic effects of size-segregated particulate samples collected during long-range transport of wildfire smoke to Helsinki. Toxicol. Appl. Pharmacol. 215, 341–353.
Le, G., Breysse, P., McDermott, A., Eftim, S., Geyh, A., Berman, J., Curriero, F., 2014. Canadian forest fires and the effects of long-range transboundary air pollution on hospitalizations among the elderly. ISPRS Int. J. Geo Inf. 3 (2), 713–731.
Mathur, R., 2008. Estimating the impact of the 2004 Alaskan forest fires on episodic particulate matter pollution over the Eastern United States through assimilation of satellite-derived aerosol optical depths in a regional air quality model. J. Geophys Res. 113 (D17), D17302.
Miller, D.J., Sun, K., Zondlo, M.A., Kanter, D., Dubovik, O., Welton, E.J., Winker, D.M., Ginoux, P., 2011. Assessing boreal forest fire smoke aerosol impacts on U.S. air quality: a case study using multiple data sets. J. Geophys Res. 116, D22209.
Park, R., Jacob, D., Logan, J., 2007. Fire and biofuel contributions to annual mean aerosol mass concentrations in the United States. Atmos. Environ. 41, 7389–7400.
Peng, R.D., Bell, M.L., Geyh, A.S., McDermott, A., Zeger, S.L., Samet, J.M., Dominici, F., 2009. Emergency admissions for cardiovascular and respiratory diseases and the chemical composition of fine particle air pollution. Environ. Health Perspect. 117 (6), 957–963. http://dx.doi.org/10.1289/ehp.0800185.
Pope III, C.A., Dockery, D.W., 2006. Health effects of fine particulate air pollution: lines that connect. J. Air Waste Manage Assoc. 56, 709–742.
Rienecker, M.M., Suarez, M.J., Todling, R., Bacmeister, J., Takacs, L., Liu, H.C., Gu, W., Sienkiewicz, M., Koster, R.D., Gelaro, R., Stanjer, I., Nielsen, J.E., 2008. The GEOS-5 Data Assimilation System-documentation of Versions 5.0.1, 5.1.0, and 5.2.0. Technical Report Series on Global Modeling and Data Assimilation, 104606(27).
Strak, M., Janssen, N.A.H., Gosens, I., Cassee, F.R., Lebret, E., Godri, K.J., Mudway, I.S., Kelly, F.J., Harrison, R.M., Brunekreef, B., Steenhof, M., Hoek, G., 2012. Respiratory health effects of airborne particulate matter: the role of particle size, composition, and oxidative potential – the Raptes project. Environ. Health Perspect. 120 (8), 1183–1189.
Strak, M., Janssen, N.A.H., Gosens, I., Cassee, F.R., Lebret, E., Godri, K.J., Mudway, I.S., Kelly, F.J., Harrison, R.M., Brunekreef, B., Steenhof, M., Hoek, G., 2013. Airborne particulate matter and acute lung inflammation: Strak et al. respond. Environ. Health Perspect. 121, a11–a12.
Tai, A.P.K., Mickley, L.J., Jacob, D.J., 2010. Correlations between fine particulate matter ($PM_{2.5}$) and meteorological variables in the United States: Implications for the sensitivity of $PM_{2.5}$ to climate change. Atmos. Environ. 44 (32), 3976–3984.
Tai, A.P.K., Mickley, L.J., Jacob, D.J., Leibensperger, E.M., Zhang, L., Fisher, J.A., Pye, H.O.T., 2012. Meteorological modes of variability for fine particulate matter ($PM_{2.5}$) air quality in the United States: implications for $PM_{2.5}$ sensitivity to climate change. Atmos. Chem. Phys. 12 (6), 3131–3145.
Turquety, S., Logan, J.A., Jacob, D.J., Hudman, R.C., Leung, F.Y., Heald, C.L., Yantosca, R.M., Wu, S., Emmons, L.K., Edwards, D.P., Sachse, G.W., 2007. Inventory of boreal fire emissions for North America in 2004: importance of peat burning and pyroconvective injection. J. Geophys Res. 112 (D12), D12S03.
U.S. Environmental Protection Agency, 2012. Our Nation's Air: Status and Trends through 2010. Technical Report EPA-454/R-12-001. U.S. Environmental Protection Agency.