AGU PUBLICATIONS

Earth's Future

RESEARCH ARTICLE
10.1002/2015EF000330

Response of SO_2 and particulate air pollution to local and regional emission controls: A case study in Maryland

Hao He[1], Konstantin Y. Vinnikov[1], Can Li[2,3], Nickolay A. Krotkov[3], Andrew R. Jongeward[1], Zhanqing Li[1,2], Jeffrey W. Stehr[1], Jennifer C. Hains[4], and Russell R. Dickerson[1,2]

[1]Department of Atmospheric and Oceanic Science, University of Maryland, College Park, Maryland, USA, [2]Earth System Science Interdisciplinary Center, University of Maryland, College Park, Maryland, USA, [3]NASA Goddard Space Flight Center, Greenbelt, Maryland, USA, [4]Maryland Department of the Environment, Baltimore, Maryland, USA

Key Points:
- Identify the long-term trend of sulfur pollution in the eastern U.S.
- Response of SO2 and sulfate pollution to local and regional control measures
- Remotely sensed satellite products capture the impacts of recent air pollution control

Supporting Information:
- Supporting Information S1
- Table S1

Corresponding author:
H. He, Department of Atmospheric and Oceanic Science, University of Maryland, College Park, MD 20742, USA. (haohe@umd.edu)

Citation:
He, H., K. Y. Vinnikov, C. Li, N. A. Krotkov, A. R. Jongeward, Z. Li, J. W. Stehr, J. C. Hains, and R. R. Dickerson (2016), Response of SO_2 and particulate air pollution to local and regional emission controls: A case study in Maryland, *Earth's Future*, *4*94–109, doi:10.1002/2015EF000330.

Received 19 OCT 2015
Accepted 6 MAR 2016
Accepted article online 11 MAR 2016
Published online 12 APR 2016

Abstract This paper addresses the questions of what effect local regulations can have on pollutants with different lifetimes and how surface observations and remotely sensed data can be used to determine the impacts. We investigated the decadal trends of tropospheric sulfur dioxide (SO_2) and aerosol pollution over Maryland and its surrounding states, using surface, aircraft, and satellite measurements. Aircraft measurements indicated fewer isolated SO_2 plumes observed in summers, a ~40% decrease of column SO_2, and a ~20% decrease of atmospheric optical depth (AOD) over Maryland after the implementation of local regulations on sulfur emissions from power plants (~90% reduction from 2010). Surface observations of SO_2 and particulate matter (PM) concentrations in Maryland show similar trends. OMI SO_2 and MODIS AOD observations were used to investigate the column contents of air pollutants over the eastern U.S.; these indicate decreasing trends in column SO_2 (~60% decrease) and AOD (~20% decrease). The decrease of upwind SO_2 emissions also reduced aerosol loadings over the downwind Atlantic Ocean near the coast by ~20%, while indiscernible changes of the SO_2 column were observed. A step change of SO_2 emissions in Maryland starting in 2009–2010 had an immediate and profound benefit in terms of local surface SO_2 concentrations but a modest impact on aerosol pollution, indicating that short-lived pollutants are effectively controlled locally, while long-lived pollutants require regional measures.

1. Introduction

Many areas of the United States, and increasingly the developing world, suffer air pollution that adversely affects the environment and human health. The U.S. Environmental Protection Agency (EPA) has established "Criteria Pollutants" with demonstrated effects on morbidity and mortality. These include sulfur dioxide (SO_2) and particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$). SO_2 has been linked to respiratory ailments [*Ware et al.*, 1981]. Its residence time, also called atmospheric lifetime, in the lower atmosphere ranges from ~2 days in the winter to less than 1 day in the summer [e.g., *Hains et al.*, 2008; *Lee et al.*, 2011]. $PM_{2.5}$, the main source of air pollution mortality in the United States [*EPA*, 2004, 2009; *Ware et al.*, 1981], is mainly removed in precipitation processes and has an atmospheric lifetime of about 10 d. $PM_{2.5}$ is also responsible for haze, which usually dictates the limit of visual range [*Hand and Malm*, 2007], impacts the hydrological cycle [*EPA*, 2009; *Fan et al.*, 2013; *Ramanathan et al.*, 2005], and affects the weather and climate [*IPCC*, 2013]. Conversion of gaseous SO_2 to condensed phase sulfate (SO_4^{2-}) has historically been a major source of $PM_{2.5}$ in many areas of the world, including the eastern United States [*Hand et al.*, 2012a, 2012c; *Tsigaridis et al.*, 2006].

In the United States as a whole, anthropogenic SO_2 emissions come mainly from power plants and other coal combustion facilities and have decreased by ~6%/yr in the last decade [*Hand et al.*, 2012b]. Sulfate aerosols from SO_2 usually peak in summer [*Hidy et al.*, 1978] and historically account for 50%–60% of the ground-level $PM_{2.5}$ observed in the eastern United States [*Hand et al.*, 2012c]. The highest concentrations of air pollution tend to occur in urban areas, but due to the relatively long lifetime of tropospheric sulfate aerosols, the regional transport of sulfur pollutants becomes important to downwind air quality [e.g., *Hains et al.*, 2008; *Taubman et al.*, 2006]. In the United States, air quality violations are issued at the municipal level; the responsibility for reducing emissions falls to a large extent on the states, but the chemistry and meteorology leading to unhealthy air can be regional in nature. Recently, the U.S. EPA ruled to uphold

the Cross-State Air Pollution Rule (CSAPR), designed to protect states downwind of major SO_2 and nitrogen oxides (NO_x) sources [*EPA*, 2014]. The temporal and spatial scales of air pollutant formation and loss are becoming increasingly important as the role of regional and inter-hemispheric transport becomes more apparent [*Adhikary et al.*, 2010; *Cooper et al.*, 2011; *Hsu et al.*, 2012; *Singh et al.*, 2009].

For the State of Maryland, a milestone for air pollution regulation, the Healthy Air Act (HAA), was passed by the Maryland General Assembly in 2006, with the goal of improving the local air quality. The first phase of the HAA was implemented in 2009–2010 and reduced SO_2 emissions from power plants by 80%–85% from 2002 levels [*MDE*, 2013]. The HAA was expected to reduce both SO_2 and aerosol pollution, especially the sulfate loading. Whether it leads to changes that are observable from satellite instruments designed to monitor SO_2 and aerosols in the atmosphere is an intriguing scientific question. The goal of this paper is to evaluate observed changes in the mentioned atmospheric parameters by comparing their values during two 3-year intervals, 2006–2008 prior to and 2010–2012 subsequent to the implementation of the HAA. However, previous studies show that the air quality of Maryland is substantially influenced by upwind anthropogenic sources, especially emissions from power plants in the Ohio River Valley region [*Brent et al.*, 2015; *Hains et al.*, 2008; *He et al.*, 2013b; *Taubman et al.*, 2006]. Thus, the HAA provides an exceptional opportunity to test the impact of a step change of emissions on levels of pollutants with different lifetimes.

In the late 1990s, the U.S. EPA established the Continuous Emission Monitoring System (CEMS) to continuously measure emissions, including SO_2 and CO_2 from point sources, mainly power plants. Surface SO_2 and $PM_{2.5}$ concentrations are monitored by the EPA Air Quality System (AQS) network. Starting in 2000, the Regional Atmospheric Measurement Modeling and Prediction Program (RAMMPP, http://www.atmos.umd.edu/~RAMMPP) has conducted regular aircraft measurements of SO_2, other trace gases, and aerosols in summer, providing a long-term record of tropospheric pollution in the eastern United States. Satellite sensors, such as the National Aeronautics and Space Administration (NASA) Ozone Monitoring Instrument (OMI) and Moderate Resolution Imaging Spectroradiometer (MODIS), also provided more than 10 years of global SO_2 and atmospheric aerosol records [*King et al.*, 2003; *Levelt et al.*, 2006]. We conducted a comprehensive study, combining long-term observations to identify the relative contribution of local emissions versus regional transport of air pollutants to air pollution in Maryland. Our research focused on air pollution in summer, when SO_2 is quickly oxidized to sulfur aerosols with a lifetime shorter than 1 day [*Hains*, 2007; *Hains et al.*, 2008; *Lee et al.*, 2011]. With sulfate aerosols peaking in the summer and assuming no long-term trends in other aerosol components such as organic aerosols with major contributions from natural sources, trends in $PM_{2.5}$ mass concentrations can reflect the changes of sulfate aerosol i.e., reduction of power plant SO_2 emissions.

2. Data and Methods

We employed datasets from monitors on point sources, surface and aircraft *in-situ* measurements, and satellite remote sensing products. Hourly CEMS data including SO_2 emissions, CO_2 emissions, and electricity generated from power plants in five states of the eastern U.S., Maryland (MD), Ohio (OH), Pennsylvania (PA), Virginia (VA), and West Virginia (WV) (hereafter named the research domain), were downloaded from the EPA Clean Air Markets Program database (http://ampd.epa.gov/ampd), and monthly mean SO_2 emissions were computed (see details in [*He et al.*, 2013b]). Data from 1998 to 2012 were used, and monthly averages were calculated to track the reduction of power plant emissions of SO_2. Hourly measurements of surface SO_2 (available after 2006: Essex, Beltsville, Piney Run, MD, and Washington, D.C.) and $PM_{2.5}$ (available after 2010: Rockville, Beltsville, Oldtown, Fairhill, Hagerstown, MD, and Washington, DC) concentrations were obtained from the EPA AQS website (https://ofmext.epa.gov/AQDMRS/aqdmrs.html). Daily filter-based observations of sulfate concentration in $PM_{2.5}$ at eight Maryland (Glen Burnie, Essex, Beltsville, Bowie, Frostburg, and three sites in the Baltimore metropolitan area) and one District of Columbia (McMillian) monitoring sites were available from http://www.epa.gov/airdata/ad_viz_plotval.html. Monthly mean aerosol optical depth (AOD) at a wavelength of 500 nm were obtained from observations of clear sky solar radiation at three AERONET stations (GSFC, SERC, and Maryland Science Center) in Maryland (http://aeronet.gsfc.nasa.gov) [*Holben et al.*, 1998].

The RAMMPP airborne measurements have been discussed extensively [e.g., *He et al.*, 2012; *He et al.*, 2013b; *Taubman et al.*, 2006], so only a brief summary is provided here. Ambient SO_2 was measured by a modified,

commercially available trace-level pulsed fluorescence analyzer (Model 43C, Thermo Environmental Instruments, Franklin, Massachusetts) [*Luke*, 1997]. Aerosol scattering was measured using an integrating Nephelometer (Trust Science Innovation, TSI Model 3563) at 450, 550, and 700 nm [*Anderson et al.*, 1996]. Research flights were usually carried out on summer days with poor air quality forecasted, and research spirals (defined as spirals over fixed locations to measure the vertical distribution of air pollutants) were conducted upwind and downwind of the Washington–Baltimore area (see details about the RAMMPP aircraft project in [*He et al.*, 2013b]). We integrated the altitude profile (from surface to the top of the research spiral, around 3 km above the ground) to obtain the column contents of tropospheric SO_2 in Dobson Units ($1\ DU = 2.69 \times 10^{16}$ molecules/cm^2) and aircraft aerosol scattering measurements to calculate AOD values, which have been compared to the NASA research aircraft measurements [*He et al.*, 2014].

Previous studies show that OMI can identify large point sources, such as power plants, and track changes of SO_2 emissions [*Fioletov et al.*, 2011; *Li et al.*, 2010]. Level 2 SO_2 products have been widely used but need preprocessing, such as filtering out cloudy scenes [e.g., *Fioletov et al.*, 2011; *Li et al.*, 2010]. In this study, we used the Level 3 daily data (OMSO2e) downloaded from the NASA Goddard Earth Sciences Data and Information Service Center (http://daac.gsfc.nasa.gov/), which are gridded from cloud-screened Level 2 SO_2 data. In addition, an air mass factor (AMF) correction has been applied to Level 3 products to more appropriately account for different observation conditions, such as viewing geometry and surface reflectivity. We used

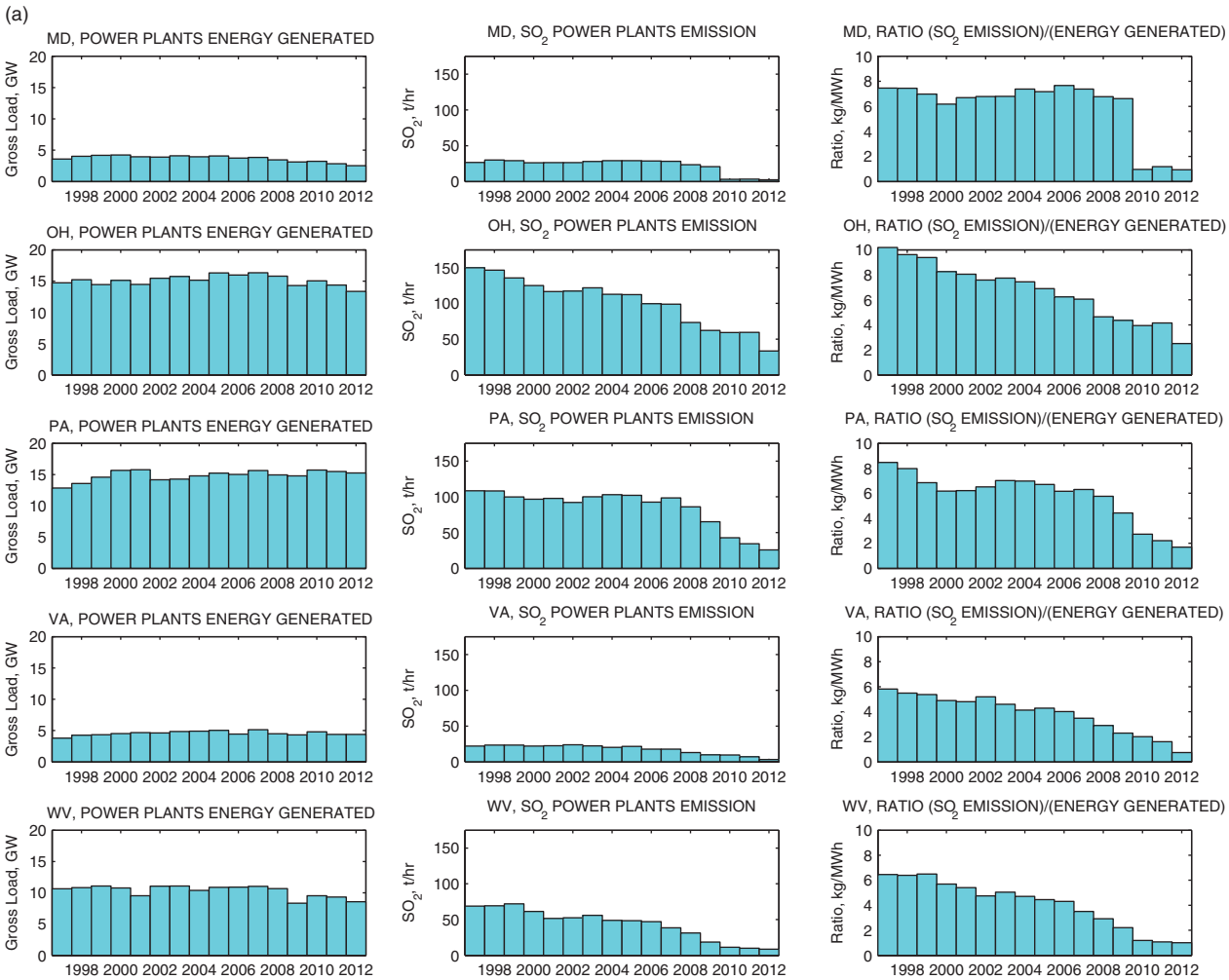

Figure 1. Historic power plant electrical power output and emissions of SO_2 in the research domain. (a) Power plant electricity generated per hour (left), SO_2 emissions (center), ratios of emissions to energy production (right). (b) Locations of major SO_2 point sources, with the color and size of each circle reflecting the magnitude of emissions (as indicated, Unit: 10^3 tons/yr); (c) Map of SO_2/CO_2 ratios (unitless). Figures 1b and 1c are based on the annual 2010 CEMS data. Locations of states Maryland, Ohio, Pennsylvania, Virginia, and West Virginia are shown in Figure 1b.

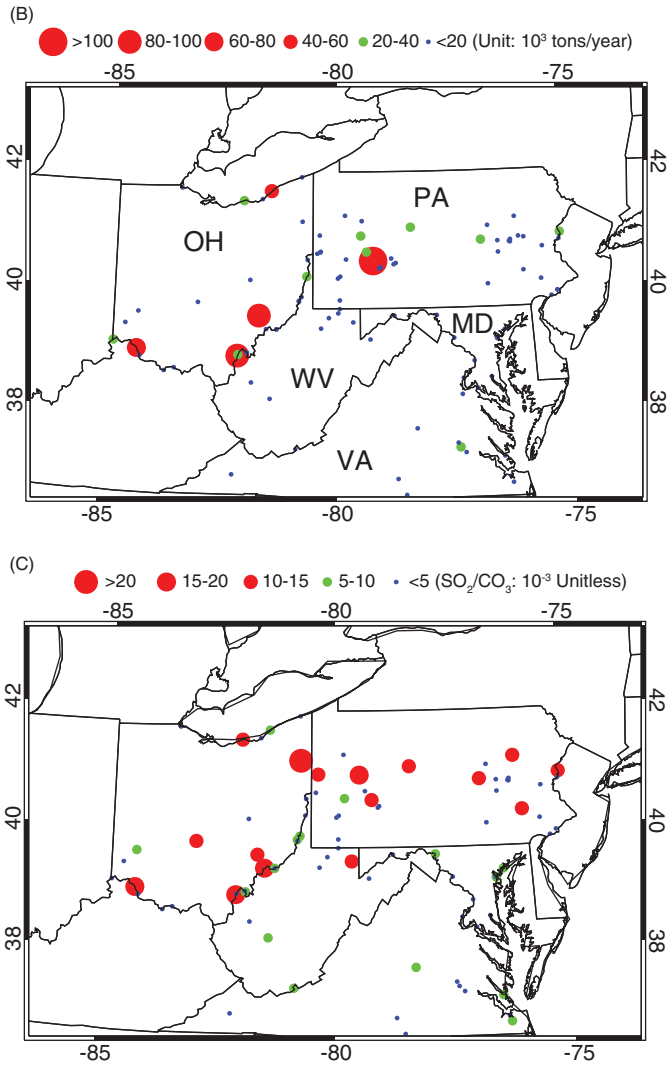

Figure 1. (continued)

two operational OMI planetary boundary layer (PBL) SO_2 products, one version based on the band residual difference (BRD) algorithm [*Krotkov et al.*, 2008] and the new version based on the principal component analysis (PCA) algorithm [*Li et al.*, 2013]. Monthly mean SO_2 columns were calculated for the trend analysis. The level 3 MODIS monthly products (onboard Aqua, MYD08_M3) were downloaded from the NASA Level 1 and Atmosphere Archive and Distribution System (http://ladsweb.nascom.nasa.gov/). MODIS AOD retrievals were processed with the Collection 5 algorithm [*Levy et al.*, 2007; *Remer et al.*, 2008] and have been validated over land and ocean [*Levy et al.*, 2010; *Remer et al.*, 2008].

3. Results and Discussion: Pollution Trends in Maryland and Neighboring States

3.1. Sulfur Emissions and Surface Observations

Figure 1 shows the long-term trend of SO_2 emissions in the five states of the research domain. In the last decade, SO_2 emissions in the research domain have decreased substantially. Starting in 2010, power plant emissions of SO_2 in Maryland fell to less than 20% of their historic values, from ~25 t/hr to ~3 t/hr. The drop resulted from the installation of flue gas desulfurization (FGD) under the HAA, while electricity production was essentially unchanged. This step change contrasts sharply with the gradual reduction in emissions observed in other states in the research domain i.e., PA, OH, VA, and WV. However, the power plant emissions

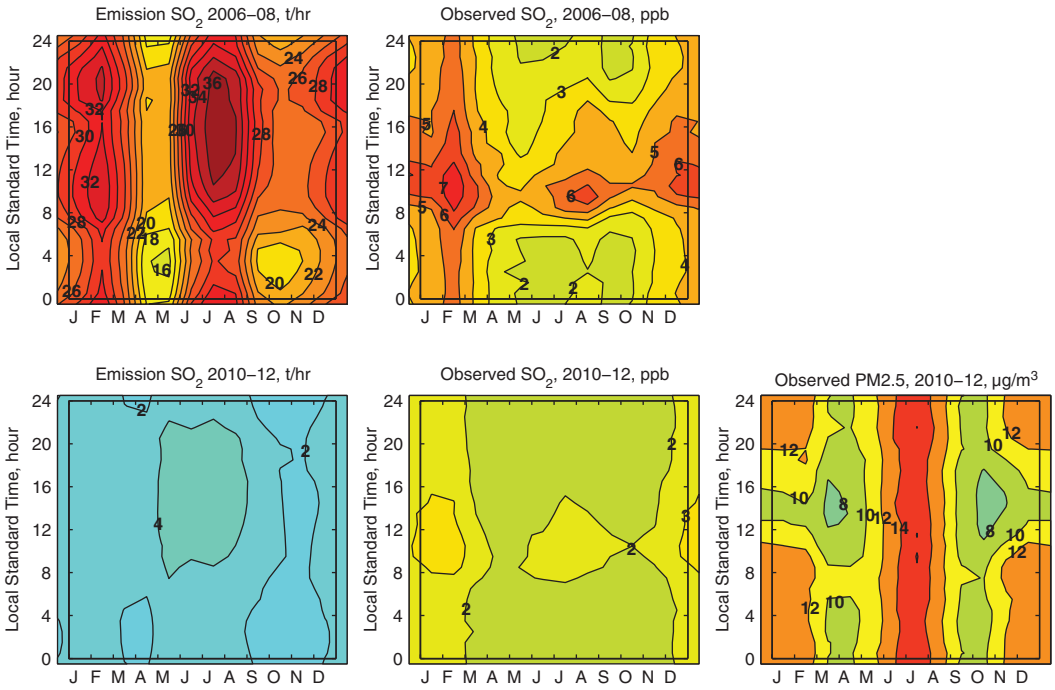

Figure 2. Contour plots of seasonal and diurnal cycles of SO_2 emissions (left), SO_2 concentrations (middle) and $PM_{2.5}$ levels (right) in Maryland before (top, 2006–2008 average) and after (bottom, 2010–2012 average) the implementation of the Healthy Air Act. The Y-axis shows local time and the X-axis shows month of the year.

of SO_2 in these upwind states, especially OH and PA, are an order of magnitude higher than emissions in Maryland, so regional transport is expected to influence sulfur pollution in Maryland.

We also present the locations of major point sources summarized in the 2010 CEMS dataset in the research domain, with annual total SO_2 emissions (Fig 1b) and the ratio of SO_2 to CO_2 emissions (Fig 1c). Compared to the point sources in upwind states, especially the Ohio River Valley, power plants in Maryland emit much less SO_2 and also have substantially lower SO_2 to CO_2 emission ratios. Previous studies demonstrated that under the prevailing westerly winds in summer, upwind emission sources could significantly influence the air quality in Maryland [*Hains et al.*, 2008; *He et al.*, 2013b; *Taubman et al.*, 2006].

Hourly power plant emissions show significant diurnal and seasonal variations, determined by variations in electricity demand and supply (Figure 2, left panels). The seasonal variation in SO_2 emissions is greater than the daily variation in SO_2 emissions. For the two selected 3-year intervals, the seasonal cycle of SO_2 emission rates in Maryland had two maxima (>30 t/hr) in winter and summer before the HAA, while after the HAA, it only has one maximum, with an emission rate of ~4 t/hr in summer. The diurnal cycle of power plant SO_2 emission rates show maxima in the afternoon hours during summer, when electricity demand is high due to air conditioner usage [*He et al.*, 2013a].

If air quality is only controlled by local emissions, then ambient pollutant concentrations should follow emissions, and this is indeed seen for SO_2 in Maryland. Figure 2 (middle panels) displays seasonal/diurnal patterns of SO_2 concentration for the two selected 3-year intervals. After the implementation of the HAA, concentrations of SO_2, with a relatively short lifetime of several days or less, fell dramatically for all times of the day and over all seasons; in contrast, concentrations of $PM_{2.5}$, with a relatively long lifetime of about 10 d, fell only modestly. The seasonal cycle of observed SO_2 follows the seasonal cycle of power plant emissions, with peak SO_2 surface concentrations observed in the coldest months and in the middle of the day when PBL processes transport the pollutants down to the surface, and a secondary maximum in summer is related to rising demand for electricity. The broad winter maximum reflects slower losses. The summer maximum has decreased by ~60%, from ~6 ppb to ~2 ppb, after the HAA.

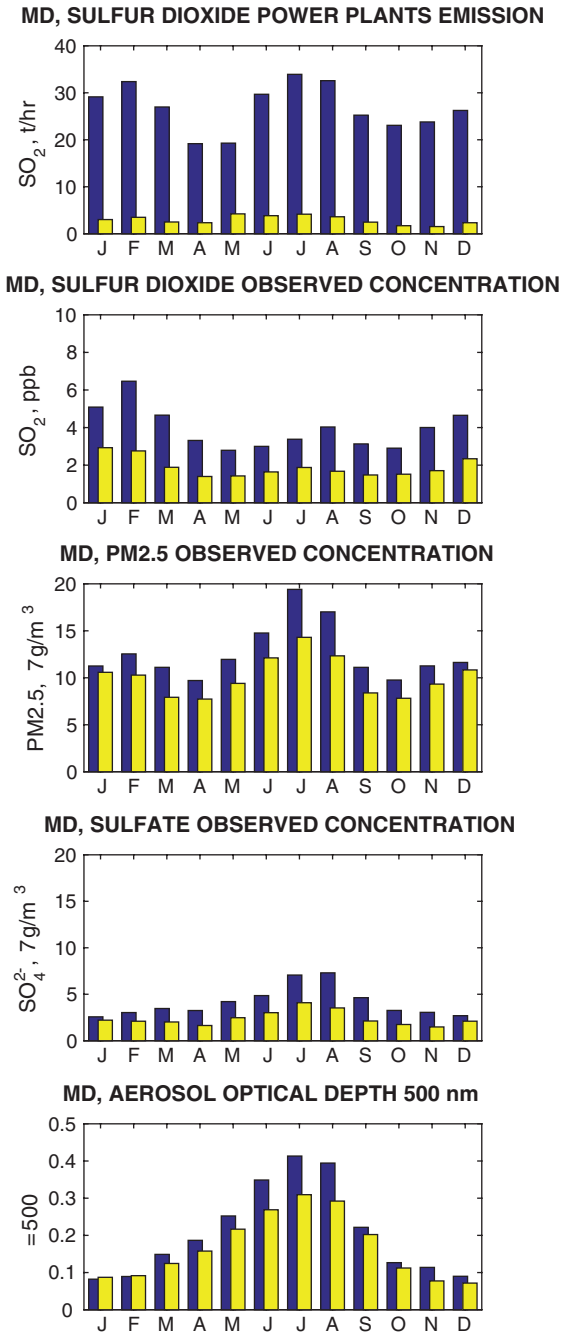

Figure 3. Seasonal variations of Maryland mean SO_2 power plant emissions, observed ambient SO_2, $PM_{2.5}$, and sulfate concentrations as well as AERONET AOD at 500 nm wavelength. Presented data were averaged for two 3-year time intervals, before (2006–2008, blue) and after (2010–2012, yellow) after implementation of the Maryland Healthy Air Act.

Hourly data of $PM_{2.5}$ concentrations in Maryland are available after 2009, when the U.S. EPA designated the automated Federal Equivalent Method for hourly $PM_{2.5}$ measurements. $PM_{2.5}$ variations in 2010–2012 (Figure 2, bottom-right panel) show that the diurnal cycle of $PM_{2.5}$ concentrations is significantly weaker than the seasonal cycle. Peak $PM_{2.5}$ concentrations are observed in the summer, when the rate of oxidation of gaseous precursors, SO_2, and volatile organic compounds (VOC) to particulate matter peaks.

Power plants emissions demonstrate diurnal and seasonal variations resulting from variations in electricity demand (Figure 2), and ambient SO_2 responds to these fluctuations. The decay time (t) of temporal fluctuations in hourly time series of emissions and pollutant concentrations was calculated as the least square estimate of the parameter t in the exponential approximation of lag-correlation function $r(\tau) = \exp(-|\tau|/t)$, where τ is lag. The least square condition is $\Sigma[ln(r(\tau))-|\tau|/t]^2 = min$. To obtain time series of fluctuations, time-dependent expected values with diurnal and seasonal variations were computed and subtracted from time series of observed data. The decay time based on observed temporal fluctuations of surface SO_2 is 2–8 hours depending on season and daytime, substantially shorter than the 0.5–3.0 d lifetime of tropospheric SO_2 [*Chin et al.*, 2000; *Lee et al.*, 2011]. Statistically evaluated decay time of fluctuations from time-dependent expected values can be interpreted as the combination of chemical lifetime and dispersal, similar to that discussed in *de Foy et al.* [2015], which is necessarily shorter than the atmospheric lifetime or residence time as usually defined. The $PM_{2.5}$ decay time based on temporal fluctuations of concentration in Maryland is ~18–20 hours, which is longer than the decay time of SO_2. These results support the hypothesis that SO_2 pollution in Maryland is more locally controlled than $PM_{2.5}$ pollution i.e., mitigation of Maryland power plant emissions has less impact on Maryland $PM_{2.5}$ concentrations than on SO_2 levels.

Before hourly observations of $PM_{2.5}$ were available, other techniques such as $PM_{2.5}$ measurements based on filter packs provided daily to weekly records of $PM_{2.5}$ in Maryland. Data from nine stations in Maryland were used to construct long-term monthly mean $PM_{2.5}$ concentrations for the periods 2006–2008 and 2010–2012, which show a significant decrease of $PM_{2.5}$ concentrations, especially sulfate aerosols, after the implementation of the HAA (Figure 3, middle and lower panel). $PM_{2.5}$ concentrations show the

Table 1. Maryland Averaged Monthly and Annual Quantities for 2006–2008

	Emission of SO_2, t/h	ΔE/E, %	Observed SO_2, ppb	$\Delta SO_2/SO_2$, %	Observed $PM_{2.5}$, μg/m^3	$\Delta PM_{2.5}/PM_{2.5}$, %	Observed SO_4, μg/m^3	$\Delta SO_4/SO_4$, %	Observed τ_{500}	$\Delta\tau_{500}/\tau_{500}$, %
January	29.1	−90	5.1	−42 ± 4	11.3	−6 ± 10	2.6	−14 ± 13	0.08	6 ± 15
February	32.4	−89	6.5	−57 ± 2	12.5	−18 ± 6	3.0	−31 ± 14	0.09	2 ± 10
March	27.0	−91	4.7	−60 ± 2	11.1	−29 ± 7	3.5	−41 ± 1	0.15	−16 ± 16
April	19.2	−88	3.3	−58 ± 1	9.7	−20 ± 9	3.3	−50 ± 6	0.19	−15 ± 12
May	19.3	−78	2.8	−49 ± 3	12.0	−21 ± 7	4.2	−41 ± 9	0.25	−14 ± 16
June	29.7	−87	3.0	−45 ± 1	14.8	−18 ± 7	4.9	−38 ± 10	0.35	−23 ± 13
July	33.9	−88	3.4	−44 ± 2	19.4	−26 ± 8	7.1	−42 ± 5	0.41	−25 ± 10
August	32.6	−89	4.0	−58 ± 2	17.0	−28 ± 10	7.3	−52 ± 10	0.39	−26 ± 22
September	25.2	−90	3.1	−53 ± 1	11.1	−25 ± 7	4.6	−54 ± 6	0.22	−9 ± 30
October	23.1	−93	2.9	−48 ± 3	9.8	−20 ± 7	3.3	−46 ± 10	0.13	−11 ± 15
November	23.8	−94	4.0	−57 ± 2	11.4	−18 ± 12	3.1	−51 ± 7	0.11	−31 ± 6
December	26.2	−91	4.7	−50 ± 5	11.6	−7 ± 11	2.7	−22 ± 15	0.09	−20 ± 5
ANN	**26.8**	**−89**	**4.0**	**−53** ± 1	**12.7**	**−20** ± 9	**4.1**	**−42** ± 11	**0.21**	**−19** ± 6

E, Power Plants Emissions of SO_2; SO_2, observed concentrations of SO_2 at air quality stations; $PM_{2.5}$, observed concentration of $PM_{2.5}$ particles; SO_4, sulfate concentrations in $PM_{2.5}$ particles; τ_{500}, AOD at 500 nm. Δ, Mean difference between 3-year averages 2010–2012 and 2006–2008. Relative changes in the variables are expressed in % ± standard error.

largest decrease during the warm months (from April to September) i.e., the seasonal cycle of $PM_{2.5}$ weakened after the implementation of the HAA. As only half of summertime $PM_{2.5}$ is sulfate and assuming other components of $PM_{2.5}$ are not substantially affected by the HAA, the relative decrease of $PM_{2.5}$ after the HAA is much smaller than the decrease of power plant emissions and ambient levels of SO_2 in Maryland (Figure 3, upper panel). During the cold months (October to March), relatively small reductions of $PM_{2.5}$ and sulfate concentrations are observed as the production of a secondary species is generally low. AERONET observations in Maryland show that AOD has a strong seasonal cycle with a maximum in summer; the values have decreased moderately since the HAA (Figure 3, lower panel). The magnitude of AOD decrease is consistent with the changes of surface $PM_{2.5}$ observations, especially in summer.

Results depicted in Figures 2 and 3 are quantified in Table 1, which shows monthly and annual average emissions and observed ambient SO_2, $PM_{2.5}$, AOD, and sulfate as well as trends for all available sites in Maryland. Time periods are again chosen to represent conditions before (2006–2008) and after (2010–2012) the implementation of the HAA. Uncertainty indicated is the standard error of the percent change. In response to the recorded ~90% reduction in emissions, ambient SO_2 fell by 53 ± 1%, suggesting that a large fraction of this trace gas is under local control. $PM_{2.5}$ and AOD showed decreases of ~20%, identical to each other within uncertainty but smaller than the decrease in ambient SO_2. The change in sulfate (−42%) is intermediate between the change in SO_2 and the change in $PM_{2.5}$ (−20%).

Essentially, all of the improvement in $PM_{2.5}$ can be attributed to the decline in sulfate. The decrease (1.7 μg m^{-3}, 42% out of 4.1 μg m^{-3}) in sulfate is smaller than that for total $PM_{2.5}$ (2.5 μg m^{-3}, 20% out of 12.7 μg m^{-3}), but most of the sulfate in the eastern United States is neutralized, and when corrected for the greater molecular weight of $(NH_4)_2SO_4$ (132 g/mole) compared to SO_4^{2-} (96 g/mole), the resulting change is −2.4 μg m^{-3} and can account for most (93%) of the observed drop in $PM_{2.5}$. The EPA's National Emissions Inventory reports almost no change in VOC emissions for this time period; thus, the organic aerosols would not be expected to improve, and this is consistent with the observed change in $PM_{2.5}$ arising from control of SO_2 emissions.

In summary, surface SO_2 concentrations respond more readily than $PM_{2.5}$ pollution, under control of their different lifetimes. The summertime tropospheric SO_2 lifetime is less than 1 day [*Hains et al*., 2008; *Lee et al*.,

2011]. SO_2 emissions in upwind states (especially Ohio and Pennsylvania) are several times higher than in Maryland (Figure 1a), but pollutant loss and dispersion [*He et al*., 2013b] reduces the amount of SO_2 imported from out of state to the Maryland monitoring sites. On the other hand, sulfate oxidized from SO_2 has a lifetime in days, and cross-state transport can play an important role in determining the Maryland $PM_{2.5}$ pollution.

3.2. Sulfur Pollution Observed by Aircraft

To investigate the long-term trends in tropospheric SO_2 and aerosol pollution, we selected five locations with consistent airborne measurements between 2000 and 2012: Luray, Winchester, and Cumberland are upwind of the Baltimore–Washington area; Harford and Easton are in the downwind areas (see location information in Figure 4 and details on RAMMPP flight planning in [*He et al.*, 2013b]). During summer in the eastern United States, SO_2 has a lifetime shorter than 1 day and is unevenly distributed, with greater concentration in plumes from power plants [*Hains et al.*, 2007; *Taubman et al.*, 2006]. So, airborne measurements of tropospheric SO_2 are significantly influenced by the stochastic probability of hitting or missing these plumes. Isolated SO_2 plumes were observed frequently in the free troposphere (FT) during RAMMPP research flights. After identifying these plumes, we found that the probability of intercepting an SO_2 plume during research flights is ~10% (see Discussion 1 in the supplementary material) before the HAA. Even though the chance is low, these isolated plumes are important for estimating the tropospheric SO_2 column from the space, such as OMI SO_2 products, which are more sensitive to SO_2 aloft [*Krotkov et al.*, 2006]. RAMMPP flights observed fewer plumes over the downwind locations after 2010, demonstrating the effectiveness of the HAA on sulfur pollution in Maryland, and also, the lower OMI SO_2 columns are anticipated.

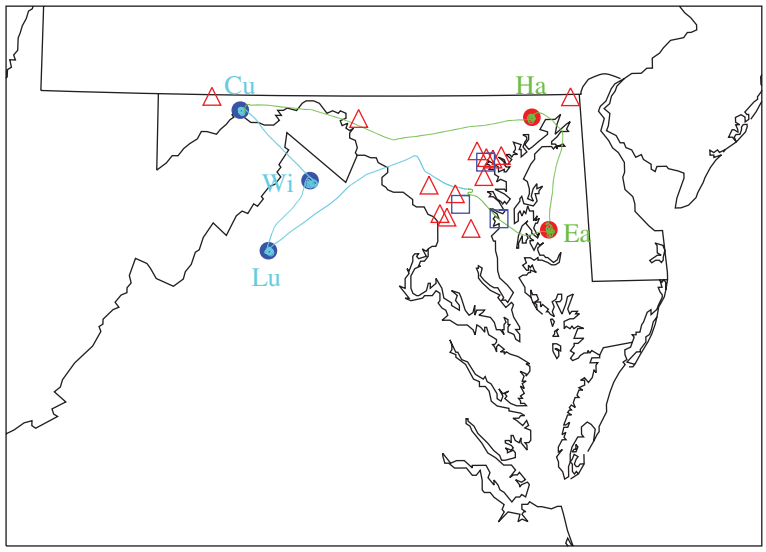

Figure 4. Locations of selected RAMMPP research spirals in the Mid-Atlantic States during a typical "westerly transport" flight pattern. Cyan and green lines show the morning and afternoon flight routes, respectively. Blue and red dots show the locations of morning and afternoon spirals, respectively. The dominant regional transport is from west to east for Maryland [*He et al.*, 2013b]. Five airports (the spiral locations) extensively covered by this flight pattern are: Luray, VA (38.67°N, 78.50°W, "Lu", 68 spirals), Winchester, VA (39.14°N, 78.14°W, "Wi", 60 spirals), Cumberland, MD (39.62°N, 78.76°W, "Cu", 74 spirals), Harford County, MD (39.57°N, 76.20°W, "Ha", 69 spirals), and Easton, MD (38.80°N, 76.07°W, "Ea", 79 spirals). This figure is adapted from [*He et al.*, 2013b] with additional research flights in 2012. Red triangles and blue squares show the locations of EPA AQS monitoring sites and AERONET sites used in this study.

Long-term aircraft measurements of column SO_2 and AOD (Figure S1, Supporting Information) show low values of SO_2 column content consistently found over Luray, a relatively remote and clean area. Winchester is downwind of the Ohio River Valley, and a larger variability of long-term SO_2 column was observed due to the isolated plumes transported from upwind sources. Cumberland has consistently high SO_2 columns due to a local power plant (see Discussion 1 in the supplementary material). In eastern Maryland, Harford and Easton have lower SO_2 columns and are less influenced by upwind sources. For tropospheric AOD, less variation among different locations is observed, suggesting that aerosol pollution is more regional.

To quantify the effect of the HAA, we calculated the average SO_2 column and AOD for two time periods, 2000–2009 and 2010–2012, before and after the HAA, respectively. Table 2 shows large reductions of air pollution in Maryland after 2010, ~40% in column SO_2 and ~20% in AOD, similar to results gleaned from

Table 2. Changes of Tropospheric SO_2 Column and AOD Observed by Aircraft Before and After the HAA

	Luray	Winchester	Cumberland	Harford	Easton
			SO_2		
2000–2009	0.36 (0.06[a])	0.62 (0.11)	0.69 (0.06)	0.41 (0.05)	0.54 (0.06)
2010–2012	0.18 (0.06)	0.38 (0.06)	0.52 (0.24)	0.24 (0.03)	0.29 (0.01)
% Change[b]	−49.3	−39.1	−24	−40.8	−47.1
			AOD		
2000–2009	0.27 (0.03)	0.31 (0.04)	0.27 (0.03)	0.30 (0.04)	0.30 (0.05)
2010–2012	0.26 (0.01)	0.25 (N/A)	0.32 (0.02)	0.20 (0.08)	0.24 (0.07)
% Change	−3.9	−19.6	+18.3	−33.3	−20.0

[a]Standard Deviation of the mean.
[b]2000–2009 data as baseline.

surface observations. One exception is weaker SO_2 reduction and increased AOD observed in Cumberland, which could be caused by the local source and a change of flight patterns (see Discussion 1 in the supplementary material).

3.3. SO_2 and Aerosol Pollution Observed From Space

To investigate the large-scale trends of sulfur pollution in the eastern United States, we conducted a linear regression analysis of OMI SO_2 and MODIS AOD observations in July from 2005 to 2012. Figure 5 shows the multi-year mean OMI PCA SO_2 column and the slope from a least squares linear regression of the data in the eastern United States. OMI PCA products successfully capture the high SO_2 loading over the Ohio River Valley, with a multi-year mean of around 0.2 DU, and show a 5%/yr decreasing trend (> −0.03 DU/yr) over the Ohio River Valley. However, when conducting a similar analysis using the previous operational OMI products based on the BRD algorithm (Figure S3), we noticed noisy SO_2 columns especially over the Great Lakes, and OMI BRD SO_2 products cannot capture the trends of decreasing emissions in the eastern United States. These results confirm the improvement of the new PCA algorithm [*Li et al.*, 2013]. In the following sections, we will use the OMI PCA SO_2 products for further analysis.

The linear regression analysis of long-term MODIS Aqua AOD data shows decreasing trends similar to OMI SO_2 observations over the eastern U.S. (Figure 6). The largest reduction is observed over the downwind coastal area including Maryland and the Atlantic Ocean, while a smaller decrease is found over the source regions of the Ohio River Valley. The decreasing trend of MODIS AOD over Maryland is around −0.02/yr or ~6%/yr. Therefore, satellite observations suggest that the upwind reduction of SO_2 column (~5%/yr) in the Ohio River Valley corresponds to the improvements of summertime AOD in the downwind areas (~6%/yr). Because sulfate aerosols dominate the summertime $PM_{2.5}$ concentrations in the eastern U.S., these results demonstrate the effectiveness of national regulations on power plant SO_2 emissions for improving regional aerosol pollution. However, our analysis also suggests that for a small state like Maryland, a state policy such as the HAA has limited effects on longer-lived regional air pollution problems.

To investigate the changes of summertime sulfur pollution due to the HAA, we examined changes of the OMI PCA SO_2 column and MODIS AOD averaged over the summers of 2006–2008 and 2010–2012. Figure 7 shows ~0.40 DU of SO_2 detected by OMI over the Baltimore–Washington area in Maryland before the HAA and a steep reduction (>0.35 DU) after 2010. MODIS AOD over the Baltimore–Washington area only decreased by ~0.05 (from ~0.15 in 2006–2008). There was an 85% decrease in the SO_2 column over Maryland following the implementation of the HAA, and the decreasing trend in satellite observations is consistent with the reduction of SO_2 emissions and ambient observations discussed in Section 3.1; however, MODIS AOD only decreased by ~30%. These results confirm that the SO_2 pollution in Maryland is mainly controlled by local sources, while the $PM_{2.5}$ pollution is regional.

3.4. Quantitative Analysis of Trends in Remotely Sensed Sulfur Pollution Over the Eastern United States

To quantitatively investigate the trends of sulfur pollution, we grouped the satellite observations into three domains: the EUS, MD, and coastal ocean, which cover five states in the research domain, central Maryland

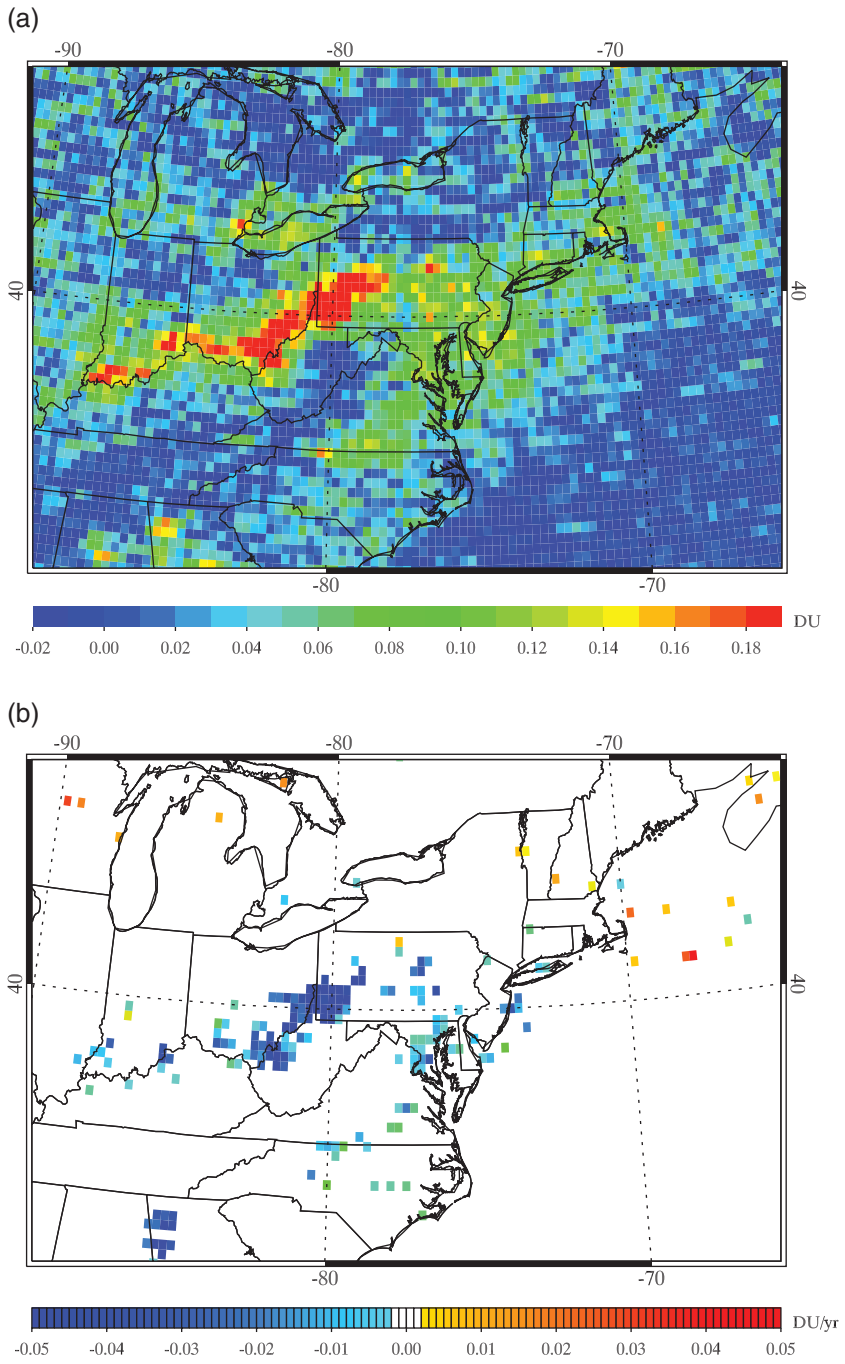

Figure 5. Linear regression analysis of SO_2 column via OMI PCA over the eastern United States from 2005 to 2012. Data of July are selected to represent summer pollution. (a) Multi-year mean from 2005 to 2012; (b) Slope of linear regression trend (showing only grid boxes with column $SO_2 > 0.1$ DU, $R^2 > 0.5$, and $p < 0.05$).

(mainly the Baltimore–Washington area), and the downwind Atlantic Ocean, respectively (Figure S4). We conducted time series linear regressions on the summertime (JJA) CEMS emissions, OMI PCA SO_2 column, and MODIS AOD (Figure 8 and Figure S5) in these domains and summarized the annual trends in Table 3. Over the last decade, power plant emissions of SO_2 decreased substantially, by 16.7%/yr and 26.5%/yr in EUS and MD, respectively. The local HAA reduced the Maryland emissions of SO_2 by ~90% after 2010. From space, substantial decreases in OMI SO_2 column contents of 13.5%/yr and 21.9%/yr were observed over EUS and MD, respectively. The OMI SO_2 column over MD fell by ~60% after 2010, even though the local reduction

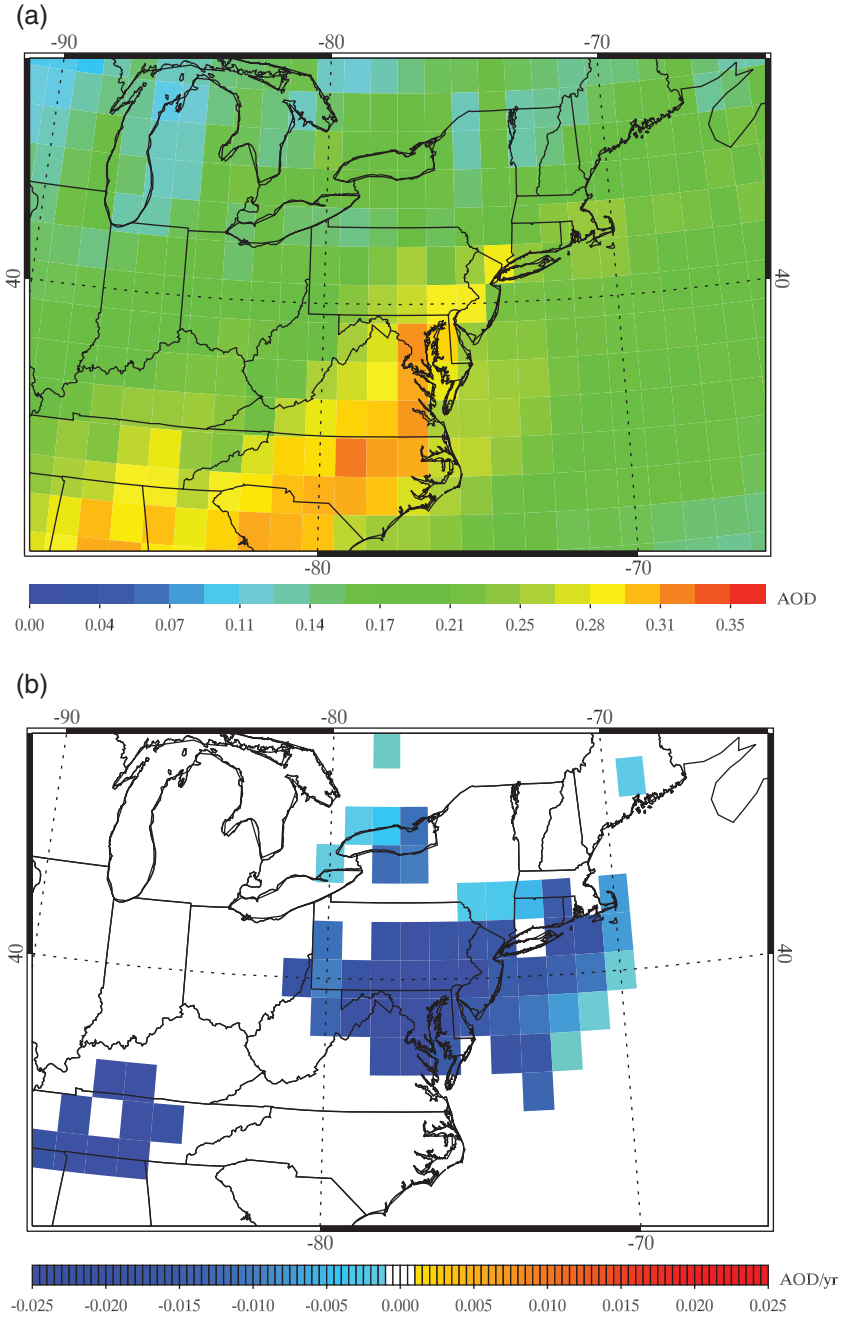

Figure 6. Linear regression analysis of MODIS Aqua AOD over the eastern United States from 2005 to 2012. Data from July were selected to represent summer pollution. (a) Multi-year mean from 2005 to 2012; (b) Slope of linear regression trend (only showing grid boxes with column $SO_2 > 0.05$, $R^2 > 0.5$, and $p < 0.05$).

of Maryland power plant emissions was ~90%. The reason for this discrepancy is that SO_2 pollution over MD is influenced by the regional transport of emissions from upwind regions such as the Ohio River Valley [*Hains et al.*, 2008; *He et al.*, 2013b], and the SO_2 columns decreased to a level that is hardly detectable by OMI. The trend in MODIS AOD shows slight decreases over EUS and MD, ~7%/yr and ~20%–25%, respectively, after 2010. SO_2 column reductions are almost double those of AOD reductions, and sulfate aerosols consist of around half of the total aerosol loadings over the eastern United States [*Hand et al.*, 2012c]. Over the Atlantic Ocean near the East Coast, no statistically significant trend in SO_2 column was observed ($p > 0.20$), likely because the lifetime of summertime tropospheric SO_2 is short, and SO_2 concentrations over the Atlantic Ocean are near the detection limit of OMI. However, we observed substantial decreases of AOD (~4%/yr

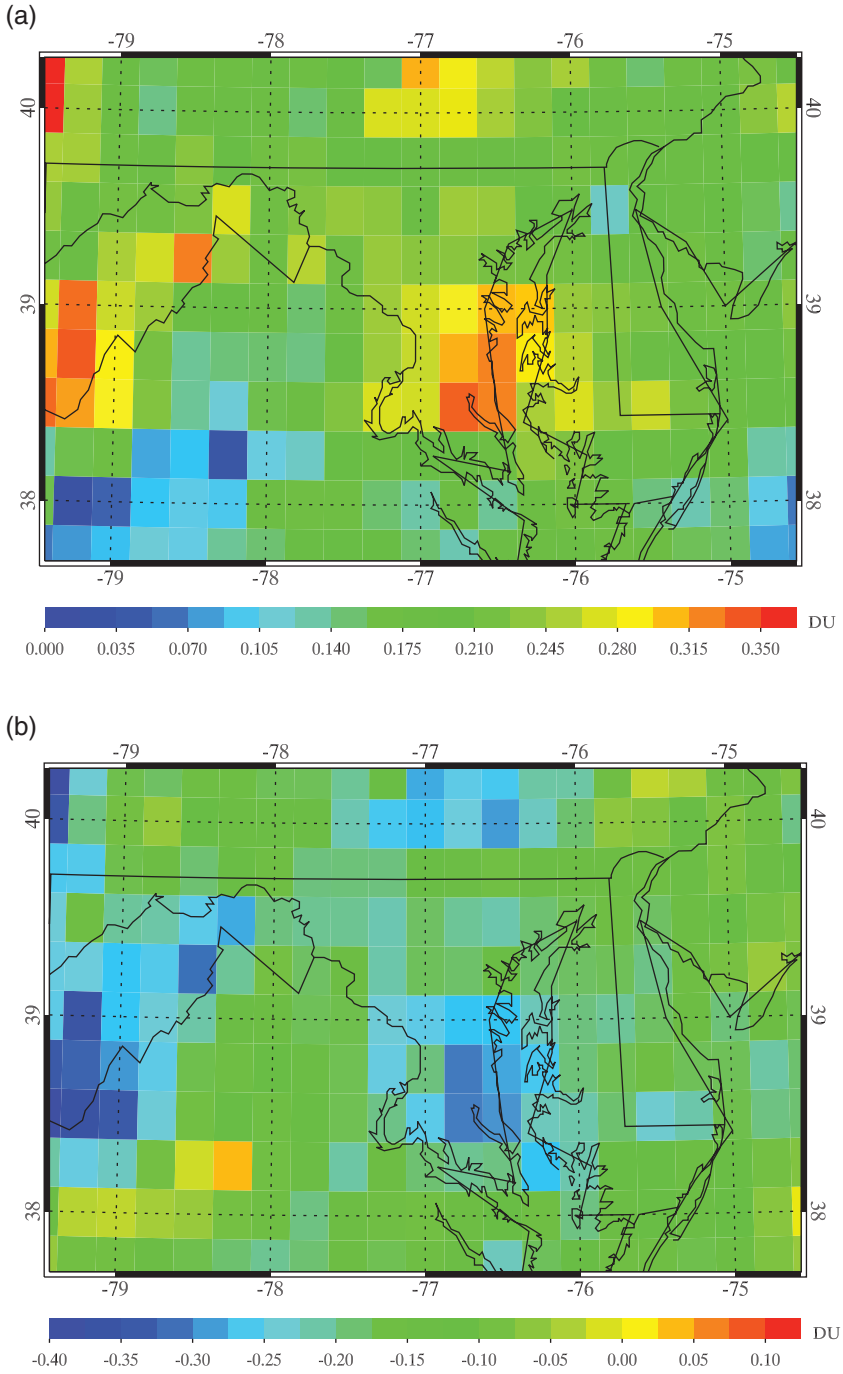

Figure 7. Satellite observations of summertime SO_2 column and MODIS AOD over the Baltimore–Washington area before and after implementation of the HAA. (a) OMI PCA SO_2 column in 2006–2008; (b) Changes of OMI SO_2 column between 2006–2008 and 2010–2012; (c) MODIS AOD in 2006–2008; (d) Changes of MODIS AOD between 2006–2008 and 2010–2012.

and ~20% after 2010) over the coastal ocean. Previous studies reported no statistically significant trend in global over-ocean AOD [*Remer et al.*, 2008; *Zhang and Reid*, 2010], but our study area over the Atlantic Ocean is small and close to the coast, so regional transport of sulfur pollutants from the continent can be identified.

4. Concluding Remarks

Long-term observations from surface monitors, aircraft, and satellites tell a consistent story of sulfur and $PM_{2.5}$ pollution over Maryland in the past decade. Due to recent local and national regulations of

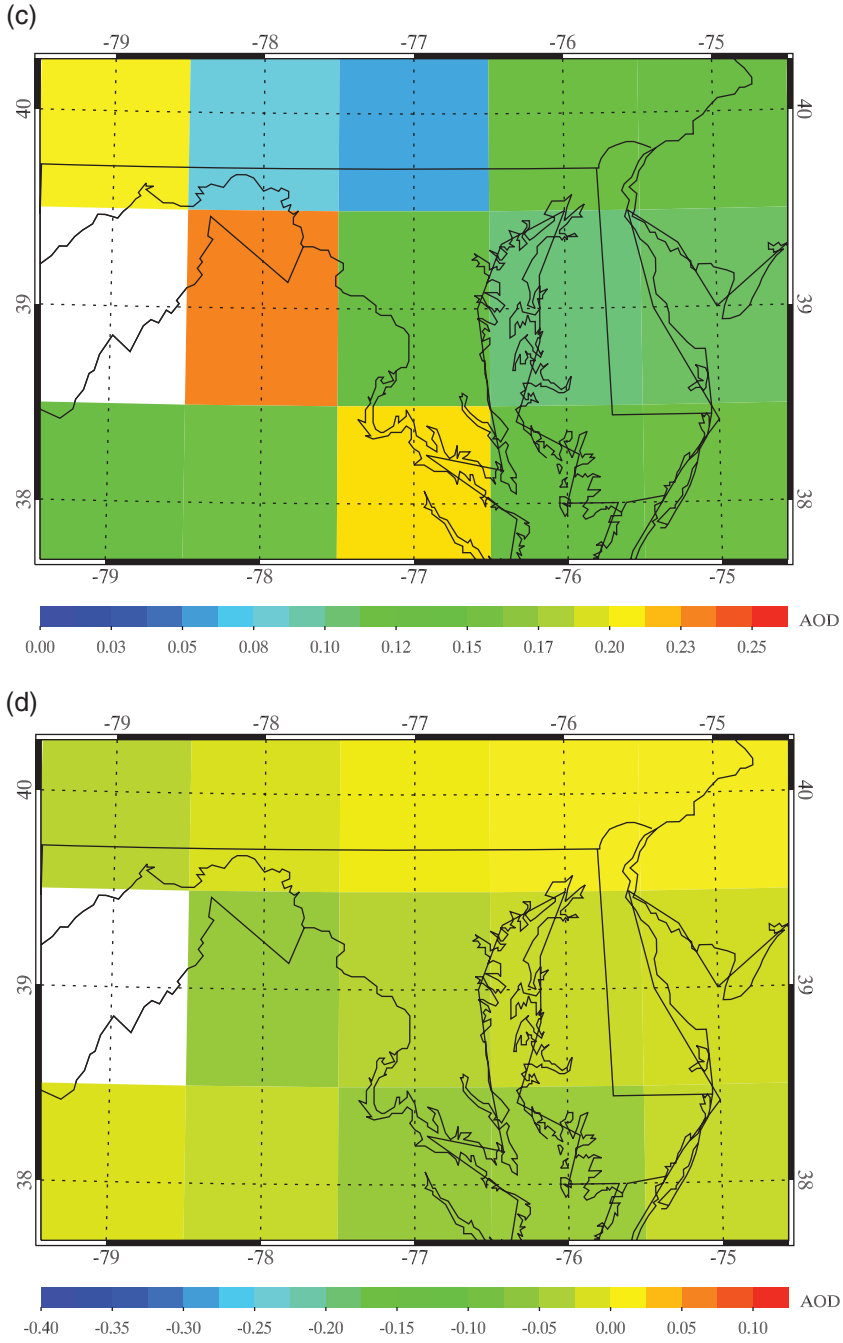

Figure 7. (continued)

power plant SO_2 emissions, summertime tropospheric SO_2 columns, $PM_{2.5}$ levels, and AOD decreased substantially over the eastern United States. The Healthy Air Act, implemented in Maryland in 2009–2010, provided an exceptional opportunity to test the impact of a step change in local emissions on ambient pollution levels. This Maryland regulation reduced in-state power plant emissions of SO_2 by ~90%; long-term aircraft measurements, surface observations, and satellite products showed a reduction in column SO_2 of ~50%, while $PM_{2.5}$ and AOD showed a reduction of ~25%. Even though the regulations on Maryland power plants successfully reduced local SO_2 pollution, PM pollution in Maryland is more regional, and regional/national regulations are needed for further improvement of air quality.

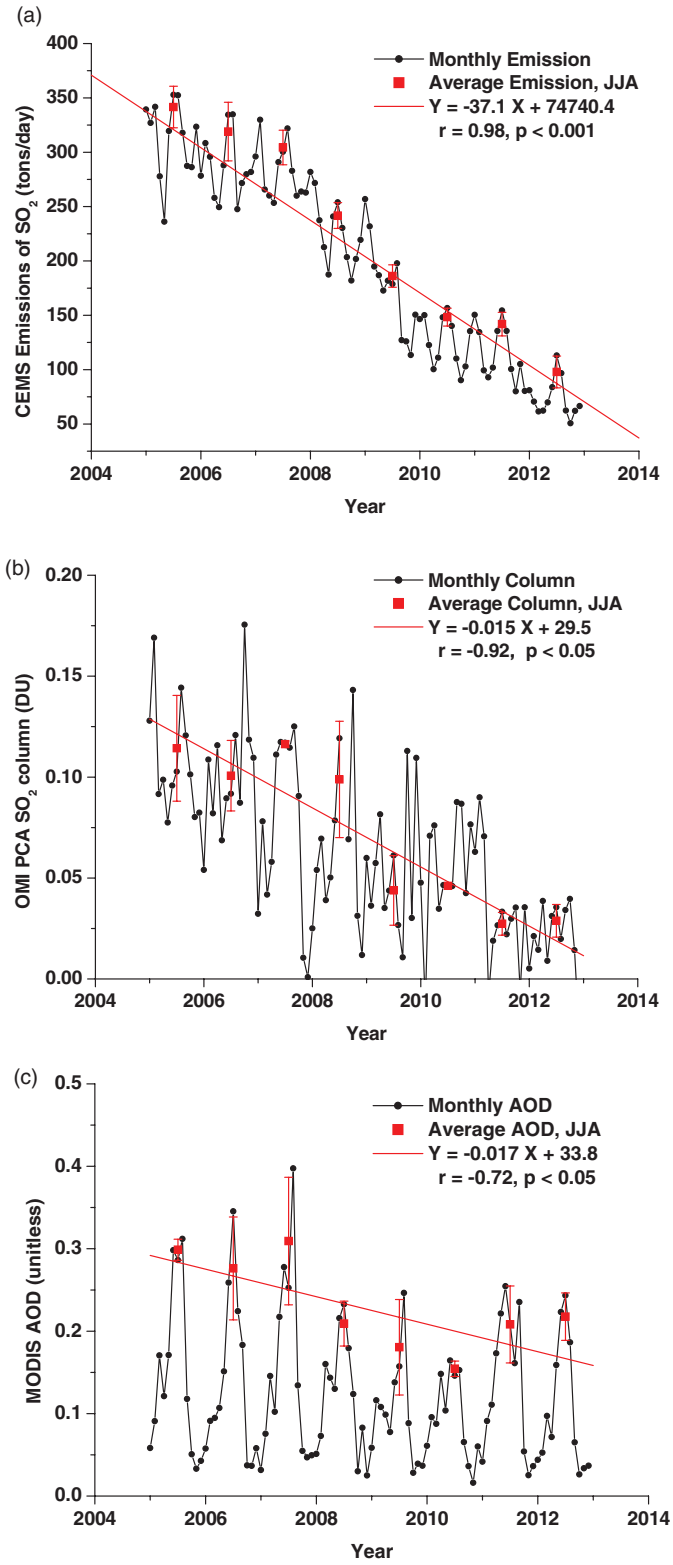

Figure 8. Long-term trends in CEMS emissions, OMI SO_2, and MODIS AOD over the EUS. Summer average values were calculated, and linear regression analysis was conducted. Errors are one standard deviation of the mean normalized by the valid data number.

Table 3. Changes of OMI SO_2 Column and MODIS AOD Over the Eastern United States

	CEMS Emissions		OMI SO_2 column			MODIS AOD		
	EUS	MD	EUS	MD	Ocean	EUS	MD	Ocean
Mean[a]	222.6	21.1	0.21	0.21	0.10	0.23	0.30	0.23
R^2 [b]	0.96	0.86	0.60	0.58	0.25	0.52	0.66	0.69
P[b]	<0.001	<0.001	<0.05	<0.05	>0.20	<0.05	<0.05	<0.05
Trend(%/yr)[c]	−16.7	−26.5	−13.5	−21.9	−7.4	−7.3	−7.8	−4.3
2005–2009[d]	278.6	31.5	0.23	0.28	0.11	0.25	0.33	0.25
2010–2012	129.4	3.9	0.17	0.10	0.09	0.19	0.24	0.20
% Change	−53.6	−87.7	−26.3	−63.5	−15.5	−24.1	−26.4	−19.2

[a]Multiple-year mean is calculated from summers in 2005–2012.
[b]R^2 and P values are based from the time series linear regression analysis.
[c]Trend is defined as slope of linear regression divided by multiple-year mean.
[d]2005–2009 value as baseline.

Acknowledgments
We thank the Maryland Department of the Environment (MDE) for support of the RAMMPP project. We thank Dr. D. Goldberg for helpful comments. K. Vinnikov acknowledges NASA support through a GEO-CAPE grant NNX12AD03A. This research was also supported by NASA AQAST and NASQ Aura Science Team grants NNX11AK34G and NNX12AI18G. We acknowledge the AERONET Team for the production of the data used in this research. CEMS data of SO_2 and CO_2 emissions from power plants are available at the EPA Clean Air Markets Program database (http://ampd.epa.gov/ampd). Observations of surface SO_2 and $PM_{2.5}$ have been obtained freely from the EPA Air quality System network website (https://ofmext.epa.gov/AQDMRS/aqdmrs.html). Aircraft measurements are available at the UMD RAMMPP website (http://www.atmos.umd.edu/~RAMMPP, per request). AERONET AOD observations are available through the NASA AERONET website (http://aeronet.gsfc.nasa.gov). Operation OMI SO2 products have been obtained from the NASA Goddard Earth Sciences Data and Information Service Center (http://daac.gsfc.nasa.gov/). The MODIS level 3 products are available from the NASA Level 1 and Atmosphere Archive and Distribution System (http://ladsweb.nascom.nasa.gov/).

The policy-relevant conclusions of these atmospheric chemistry experiments are that local regulations are indeed effective in reducing air pollution but only within the limits dictated by tropospheric advection and removal processes. Local concentrations of species such as SO_2, with a lifetime similar to the transport time over a single state (~1 day), respond dramatically. Longer-lived species, such as $PM_{2.5}$, respond only modestly as they depend more on regional emissions. Effective control of $PM_{2.5}$, for improved health, visibility, and acid deposition, must consider transport across national borders and state lines.

References

Adhikary, B., et al. (2010), A regional scale modeling analysis of aerosol and trace gas distributions over the eastern Pacific during the INTEX-B field campaign, *Atmos. Chem. Phys.*, *10*(5), 2091–2115, doi:10.5194/acp-10-2091-2010.

Anderson, T. L., et al. (1996), Performance characteristics of a high-sensitivity, three-wavelength, total scatter/backscatter nephelometer, *J. Atmos. Oceanic Technol.*, *13*(5), 967–986, doi:10.1175/1520-0426(1996)013<0967:PCOAHS>2.0.CO;2.

Brent, L. C., et al. (2015), Evaluation of the use of a commercially available cavity ringdown absorption spectrometer for measuring NO2 in flight, and observations over the Mid-Atlantic States, during DISCOVER-AQ, *J. Atmos. Chem.*, *72*, 503–521, doi:10.1007/s10874-013-9265-6.

Chin, M., R. B. Rood, S. J. Lin, J. F. Muller, and A. M. Thompson (2000), Atmospheric sulfur cycle simulated in the global model GOCART: Model description and global properties, *J. Geophys. Res. Atmos.*, *105*(D20), 24671–24687, doi:10.1029/2000JD900384.

Cooper, O. R., et al. (2011), Measurement of western U.S. baseline ozone from the surface to the tropopause and assessment of downwind impact regions, *J. Geophys. Res. Atmos.*, *116*(D21), D00V03, doi:10.1029/2011JD016095.

de Foy, B., Z. Lu, D. G. Streets, L. N. Lamsal, and B. N. Duncan (2015), Estimates of power plant NOx emissions and lifetimes from OMI NO2 satellite retrievals, *Atmos. Environ.*, *116*, 1–11, doi:10.1016/j.atmosenv.2015.05.056.

EPA (2004), Air quality criteria for particulate matter, *Final Rep. EPA 600/P-99/002aF-bF*, U.S. Environmental Protection Agency, Washington, D. C., Oct. 2004.

EPA (2009), Integrated science assessment for particulate matter, *Final Rep. EPA/600/R-08/139F*, Washington, D. C.

EPA (2014), EPA vs. EME: Homer city generation, 12–1183, US, 11–1302, 2014.b.

Fan, J., L. R. Leung, D. Rosenfeld, Q. Chen, Z. Li, J. Zhang, and H. Yan (2013), Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds, *Proc. Natl. Acad. Sci. U. S. A.*, *110*(48), E4581–E4590, doi:10.1073/pnas.1316830110.

Fioletov, V. E., C. A. McLinden, N. Krotkov, M. D. Moran, and K. Yang (2011), Estimation of SO2 emissions using OMI retrievals, *Geophys. Res. Lett.*, *38*, 5, doi:10.1029/2011GL049402.

Hains, J. C. (2007), A chemical climatology of lower tropospheric trace gases and aerosols over the mid-atlantic region, PhD Dissertation, Univ. of Maryland, College Park.

Hains, J. C., L. W. A. Chen, B. F. Taubman, B. G. Doddridge, and R. R. Dickerson (2007), A, side-by-side comparison of filter-based PM2.5 measurements at a suburban site: A closure study, *Atmos. Environ.*, *41*(29), 6167–6184, doi:10.1016/j.atmosenv.2007.04.008.

Hains, J. C., B. F. Taubman, A. M. Thompson, J. W. Stehr, L. T. Marufu, B. G. Doddridge, and R. R. Dickerson (2008), Origins of chemical pollution derived from Mid-Atlantic aircraft profiles using a clustering technique, *Atmos. Environ.*, *42*(8), 1727–1741, doi:10.1016/j.atmosenv.2007.11.052.

Hand, J. L., and W. C. Malm (2007), Review of aerosol mass scattering efficiencies from ground-based measurements since 1990, *J. Geophys. Res. Atmos.*, *112*(D18), 24, doi:10.1029/2007jd008484.

Hand, J. L., K. A. Gebhart, B. A. Schichtel, and W. C. Malm (2012a), Increasing trends in wintertime particulate sulfate and nitrate ion concentrations in the Great Plains of the United States (2000-2010), *Atmos. Environ.*, *55*, 107–110, doi:10.1016/j.atmosenv.2012.03.050.

Hand, J. L., B. A. Schichtel, W. C. Malm, and M. L. Pitchford (2012b), Particulate sulfate ion concentration and SO2 emission trends in the United States from the early 1990s through 2010, *Atmos. Chem. Phys.*, *12*(21), 10353–10365, doi:10.5194/acp-12-10353-2012.

Hand, J. L., B. A. Schichtel, M. Pitchford, W. C. Malm, and N. H. Frank (2012c), Seasonal composition of remote and urban fine particulate matter in the United States, *J. Geophys. Res. Atmos*, *117*, 22, doi:10.1029/2011jd017122.

He, H., et al. (2012), SO2 over central China: Measurements, numerical simulations and the tropospheric sulfur budget, *J. Geophys. Res. Atmos.*, *117*, 15, doi:10.1029/2011jd016473.

He, H., L. Hembeck, K. M. Hosley, T. P. Canty, R. J. Salawitch, and R. R. Dickerson (2013a), High ozone concentrations on hot days: The role of electric power demand and NOx emissions, *Geophys. Res. Lett.*, *40*(19), 5291–5294, doi:10.1002/grl.50967.
He, H., et al. (2013b), Trends in emissions and concentrations of air pollutants in the lower troposphere in the Baltimore/Washington airshed from 1997 to 2011, *Atmos. Chem. Phys.*, *13*(15), 7859–7874, doi:10.5194/acp-13-7859-2013.
He, H., et al. (2014), An elevated reservoir of air pollutants over the Mid-Atlantic States during the 2011 DISCOVER-AQ campaign: Airborne measurements and numerical simulations, *Atmos. Environ.*, *85*, 18–30, doi:10.1016/j.atmosenv.2013.11.039.
Hidy, G. M., P. K. Mueller, and E. Y. Tong (1978), Spatial and temporal distributions of airborne sulfate in parts of United-States, *Atmos. Environ.*, *12*(1-3), 735–752, doi:10.1016/0004-6981(78)90255-X.
Holben, B. N., et al. (1998), AERONET—A federated instrument network and data archive for aerosol characterization, *Remote Sens. Environ.*, *66*(1), 1–16, doi:10.1016/S0034-4257(98)00031-5.
Hsu, N. C., C. Li, N. A. Krotkov, Q. Liang, K. Yang, and S. C. Tsay (2012), Rapid transpacific transport in autumn observed by the A-train satellites, *J. Geophys. Res. Atmos.*, *117*, 13, doi:10.1029/2011jd016626.
IPCC (2013), Climate change 2013: The physical science basis, *Contribution of Working Group I to the Fifth Assessment Report (AR5) of the Intergovernmental Panel on Climate Change*, 1535 pp.
King, M. D., W. P. Menzel, Y. J. Kaufman, D. Tanre, B. C. Gao, S. Platnick, S. A. Ackerman, L. A. Remer, R. Pincus, and P. A. Hubanks (2003), Cloud and aerosol properties, precipitable water, and profiles of temperature and water vapor from MODIS, *IEEE Trans. Geosci. Remote Sens.*, *41*(2), 442–458, doi:10.1109/TGRS.2002.808226.
Krotkov, N. A., S. A. Carn, A. J. Krueger, P. K. Bhartia, and K. Yang (2006), Band residual difference algorithm for retrieval of SO2 from the aura Ozone Monitoring Instrument (OMI), *IEEE Trans. Geosci. Remote Sens.*, *44*(5), 1259–1266, doi:10.1109/TGRS.2005.861932.
Krotkov, N. A., et al. (2008), Validation of SO2 retrievals from the Ozone Monitoring Instrument over NE China, *J. Geophys. Res. Atmos.*, *113*(D16), D16S40, doi:10.1029/2007JD008818.
Lee, C., R. V. Martin, A. van Donkelaar, H. Lee, R. R. Dickerson, J. C. Hains, N. Krotkov, A. Richter, K. Vinnikov, and J. J. Schwab (2011), SO2 emissions and lifetimes: Estimates from inverse modeling using in situ and global, space-based (SCIAMACHY and OMI) observations, *J. Geophys. Res. Atmos.*, *116*, 13, doi:10.1029/2010jd014758.
Levelt, P. F., E. Hilsenrath, G. W. Leppelmeier, G. H. J. van den Oord, P. K. Bhartia, J. Tamminen, J. F. de Haan, and J. P. Veefkind (2006), Science objectives of the ozone monitoring instrument, *IEEE Trans. Geosci. Remote Sens.*, *44*(5), 1199–1208, doi:10.1109/TGRS.2006.872336.
Levy, R. C., L. A. Remer, and O. Dubovik (2007), Global aerosol optical properties and application to moderate resolution imaging spectroradiometer aerosol retrieval over land, *J. Geophys. Res. Atmos.*, *112*, D13210, doi:10.1029/2006JD007815.
Levy, R. C., L. A. Remer, R. G. Kleidman, S. Mattoo, C. Ichoku, R. Kahn, and T. F. Eck (2010), Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, *Atmos. Chem. Phys.*, *10*(21), 10399–10420, doi:10.5194/acp-10-10399-2010.
Li, C., Q. Zhang, N. A. Krotkov, D. G. Streets, K. B. He, S. C. Tsay, and J. F. Gleason (2010), Recent large reduction in sulfur dioxide emissions from Chinese power plants observed by the Ozone Monitoring Instrument, *Geophys. Res. Lett.*, *37*, 6, doi:10.1029/2010gl042594.
Li, C., J. Joiner, N. A. Krotkov, and P. K. Bhartia (2013), A fast and sensitive new satellite SO2 retrieval algorithm based on principal component analysis: Application to the ozone monitoring instrument, *Geophys. Res. Lett.*, *40*(23), 6314–6318, doi:10.1002/2013GL058134.
Luke, W. T. (1997), Evaluation of a commercial pulsed fluorescence detector for the measurement of low-level SO2 concentrations during the gas-phase sulfur intercomparison experiment, *J. Geophys. Res. Atmos.*, *102*(D13), 16255–16265, doi:10.1029/96JD03347.
MDE (2013), The Maryland Healthy Air Act. [Available at http://www.mde.md.gov/programs/Air/ProgramsHome/Pages/air/md_haa.aspx.]
Ramanathan, V., C. Chung, D. Kim, T. Bettge, L. Buja, J. T. Kiehl, W. M. Washington, Q. Fu, D. R. Sikka, and M. Wild (2005), Atmospheric brown clouds: Impacts on South Asian climate and hydrological cycle, *Proc. Natl. Acad. Sci. U. S. A.*, *102*(15), 5326–5333, doi:10.1073/pnas.0500656102.
Remer, L. A., et al. (2008), Global aerosol climatology from the MODIS satellite sensors, *J. Geophys. Res. Atmos*, *113*, D13210, doi:10.1029/2006JD007815.
Singh, H. B., W. H. Brune, J. H. Crawford, F. Flocke, and D. J. Jacob (2009), Chemistry and transport of pollution over the Gulf of Mexico and the Pacific: Spring 2006 INTEX-B campaign overview and first results, *Atmos. Chem. Phys.*, *9*(7), 2301–2318, doi:10.5194/acp-9-2301-2009.
Taubman, B. F., J. C. Hains, A. M. Thompson, L. T. Marufu, B. G. Doddridge, J. W. Stehr, C. A. Piety, and R. R. Dickerson (2006), Aircraft vertical profiles of trace gas and aerosol pollution over the mid-Atlantic United States: Statistics and meteorological cluster analysis, *J. Geophys. Res. Atmos.*, *111*, D10S07, doi:10.1029/2005JD006196.
Tsigaridis, K., M. Krol, F. J. Dentener, Y. Balkanski, J. Lathière, S. Metzger, D. A. Hauglustaine, and M. Kanakidou (2006), Change in global aerosol composition since preindustrial times, *Atmos. Chem. Phys.*, *6*(12), 5143–5162, doi:10.5194/acp-6-5143-2006.
Ware, J. H., L. A. Thibodeau, F. E. Speizer, S. Colome, and B. G. Ferris (1981), Assessment of the health-effects of atmospheric sulflur-oxides and particulate matter: Evidence from observational studies, *Environ. Health Perspect.*, *41*(October), 255–276, doi:10.1289/ehp.8141255.
Zhang, J., and J. S. Reid (2010), A decadal regional and global trend analysis of the aerosol optical depth using a data-assimilation grade over-water MODIS and Level 2 MISR aerosol products, *Atmos. Chem. Phys.*, *10*(22), 10949–10963, doi:10.5194/acp-10-10949-2010.