Recent changes in particulate air pollution over China observed from space and the ground: Effectiveness of emission control

Jintai Lin¹, Chris Nielsen¹, Yu Zhao¹, Yu Lei¹, Yang Liu², Michael B. McElroy¹*

¹ School of Engineering and Applied Sciences and Harvard China Project, Harvard University, 19 Oxford St., Cambridge, MA, 02138, U.S.

² Department of Environmental and Occupational Health, Emory University, Rollins School of Public Health, 1518 Clifton Road NE, Atlanta, GA, 30322, U.S.

*Corresponding author. Email: mbm@seas.harvard.edu. Tel: 1-617-4954359

Key words: aerosol optical depth, primary aerosols, secondary aerosols, emission control effectiveness, particulate air quality

Brief: Relatively weak controls on secondary aerosols delay improvements of particulate air quality in industrialized regions of China.

The Chinese government has moved aggressively since 2005 to reduce emissions of a number of pollutants including primary particulate matter (PM) and sulfur dioxide (SO₂), efforts inadvertently aided since late 2008 by economic recession. Satellite observations
of aerosol optical depth (AOD) and column nitrogen dioxide (NO2) provide independent indicators of emission trends, clearly reflecting the sharp onset of the recession in the fall of 2008 and rebound of the economy in the latter half of 2009. Comparison of AOD with ground-based observations of PM over a longer period indicate that emission-control policies have been generally successful in reducing concentrations of aerosol pollutants in less industrialized, interior regions of China, but not in more industrialized regions. The lack of success in industrialized regions is attributed to the increasing importance of secondary aerosols formed from precursor species including nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOC), and ammonia (NH3). To bring about a long-term improvement in air quality, it is suggested that future regulations in China emphasize not only emissions of primary PM and SO2, but also NOx, NMVOC, and NH3.

1. Introduction

Aerosols have a significant impact not only on climate (1-3) but also on public health (4). The impact is particularly important for particles of small sizes, formed for example from precursor chemicals including sulfur dioxide (SO2), nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOC), and ammonia (NH3). These particles, which are generally hydroscopic and reflective, can have a significant impact on the energy budget of the climate system, responsible for negative radiative forcing offsetting positive forcing from greenhouse gases. Given their small sizes, they can penetrate readily into human lungs triggering a variety of respiratory and cardiovascular problems. Sulfate and nitrate aerosols make an important contribution also to the phenomenon of
acid deposition with negative impacts not only on agriculture but also on natural ecosystems (5).

The Chinese government has strong incentives to reduce emissions of aerosols and precursors. China is a major source region of anthropogenic aerosols as a consequence of fast industrialization and urbanization, the reliance of the Chinese economy on coal and relatively weak emission controls (5-8). The resulting high aerosol loadings (9-10) and strong acid deposition (5) have posed a major domestic environmental problem. To tackle these issues, China has taken aggressive steps since 2005 to reduce emissions of primary aerosols and SO$_2$ (see Supporting Information section 1).

Observations from space and in situ can be used to provide information on aerosols over source regions. Here we use such data to evaluate the effect of recent emission controls on aerosol loading in East China (see Figure 1a for region specifications) over the course of Oct 2004 – Dec 2009. Data employed here include daily aerosol optical depth (AOD) at 483.5 nm measured by the Ozone Monitoring Instrument (OMI) onboard the Aura satellite and surface observations for daily mean mass concentrations of particulate matter with diameters not more than 10 $\mu$m ([PM$_{10}$]) in 66 major cities. OMI AOD is particularly sensitive to PM $\leq$ 2.5 $\mu$m (PM$_{2.5}$) derived both from primary emissions and from secondary formation in the atmosphere. Particles of larger sizes derived typically from surface emissions, while making an important contribution to the mass of PM$_{10}$, contribute to a much less extent to OMI AOD. In addition, OMI passes China in the afternoon (around 2:00 pm) when the mixing is strongest and pollutants more evenly distributed in the boundary layer. It offers thus valuable information specifically on the abundance of aerosols in the near-surface region.
2. Descriptions and processing of atmospheric measurements

The OMI AOD data were taken from the daily level-3 dataset OMAEROe (available at http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omaeroe_v003.shtml) with a horizontal resolution of 0.25° lat x 0.25° long based on the level-2 retrieval using the multiwavelength algorithm, OMAERO (11). Validation of OMAERO has been discussed by Curier et al. (11) and Livingston et al. (12). The detailed methodology for generating the OMAEROe dataset can be found at the online readme file (13). More descriptions of the data and comparisons with ground-based AOD measurements are available in Supporting Information section 2 and 4.

Daily mean mass concentrations of ground-level PM$_{10}$ ([PM$_{10}$]) were derived using the Air Pollution Index (API) data from the Ministry of Environmental Protection (MEP) of China (http://datacenter.mep.gov.cn/TestRunQian/air_dairy_en.jsp). The API records for previous years (2000-2006) have been used to study aerosol loadings and their impacts on meteorology in China (14-18). Detailed descriptions of the API data and the conversion from API scores to [PM$_{10}$] are discussed in Supporting Information section 2.

Concentrations of aerosols vary significantly both in time and space as a consequence of changes in a variety of factors including emissions of aerosols and precursors, atmospheric lifetimes of aerosols, and meteorological conditions. To evaluate the interannual trends of OMI AOD and [PM$_{10}$], a 365-day moving average was applied to the daily data to eliminate the effect of temporal variations within a year (see Supporting Information section 3).

3. Results and discussion
Analysis of the AOD data indicates an approximately linear trend over the initial four years, Oct 2004 – Sep 2008. (The behavior as we show later is more complicated since late 2008 reflecting the influence of the economic recession.) A simple fit for the interannual trend, derived by linear regression on the moving averaged data over the first four years, results in a trend of rapid growth in the northeastern region contrasted with a reduction in the interior-south region of East China (Figure 1b). We propose in what follows to focus on those two regions separately.

A general downward trend in [PM$_{10}$] is observed over the northeastern region during the initial period offset by an upward trend in AOD (Figure 2a). Across the 37 cities within the region, mean [PM$_{10}$] decreased from $\sim 107$ $\mu$g/m$^3$ to $\sim 96$ $\mu$g/m$^3$, while AOD increased from a minimum value of $\sim 0.96$ to a level of $\sim 1.08$. Averaged over the region, AOD increased from $\sim 0.93$ to $\sim 1.02$. These opposite trends can be attributed to the varying effectiveness of emission controls addressing primary aerosols and secondary aerosol precursors in China. First, as presented in more detail in the Supporting Information section 1, most national control efforts to date have targeted emissions of primary PM and SO$_2$. Approximately 60% of anthropogenic emissions of PM in 2005 are derived from three main sources including cement production, coal combustion for power generation, and biofuel burning for residential use (Supporting Figure S1). The recent controls on these sources are estimated to have resulted in an overall reduction in emissions of primary PM from 2005 to 2010. The targeted control of sulfur in the Eleventh Five-Year Plan (11$^{th}$ FYP) has reportedly led to a decrease in emissions of SO$_2$ by $\sim 9\%$ from 2005 to 2008 (Supporting Figure S2). In contrast, controls for emissions of other aerosol precursors have been much less strict. As a result, emissions of NO$_x$ are
estimated to have grown dramatically in recent years; and emissions of NMVOC and NH₃ are estimated to have increased from 2005 to 2010 by 18% and by up to 10%, respectively (Supporting Information section 1 and Figure S2). Second, the industrialization and urbanization of China is relatively more advanced in this region (19), where booming transportation and industrial sectors have resulted in large and rapidly increasing anthropogenic emissions of NOₓ and NMVOC (7, 20-23). The consequent increase in nitrate and secondary organic aerosols (SOAs) makes an important and growing contribution to the AOD observed by OMI. Thus, while controls on emissions of primary aerosols and SO₂ resulted in reductions of [PM₁₀], their impact on AOD has been more than offset by the increases in nitrate, SOAs and ammonium resulting in greater observed values for AOD. Consistent with our analysis, Zhao et al. (5) suggested that the impact of sulfur control on acid deposition from 2005 to 2010 might be negated by the increase in emissions of NOₓ and NH₃ under current policies.

Changes in the source of nitrate aerosols can be inferred from measurements of monthly mean tropospheric vertical column densities (VCDs) of nitrogen dioxide (NO₂) retrieved from OMI (available at http://www.temis.nl/airpollution/no2col/no2regioomimonth_col3.php; see Supporting Information section 2 and 3 for data descriptions and processing). Averaged over the northeastern region, the VCD of NO₂ increased by ~27% during the four years prior to the economic downturn (Figure 2a), a factor of 2 – 3 larger than the increase in OMI AOD. This implies a significant increase in emissions of NOₓ and, consequently, the concentration of nitrate aerosols, responsible partially for the increase in OMI AOD.
The effect of emission reductions associated with the economic recession that set in during late 2008 and persisted until late 2009 is evident in the AOD and NO$_2$ VCD data for the northeastern region (Figure 2a). In particular, regional mean AOD decreased from a value exceeding 1.0 prior to the economic recession to a comparatively low level of 0.87 in late 2009. Concurrently, the VCD of NO$_2$ decreased by about 15%. More recent data, for Oct – Dec 2009, indicate a recovery in both AOD and NO$_2$ VCD, in agreement with the resumption of growth in the Chinese economy during this period.

The short-term emission restrictions targeting the 2008 Beijing Summer Olympics had an important impact for the Beijing-Tianjin area (part of the northeastern region as defined here), where both AOD and NO$_2$ VCD decreased after 2007 following an increase in the prior years (Figure 2b). The reduction is most evident in the data for the cities of Beijing and Tianjin: AOD dropped by ~ 9% from the 2007 mean to the 2008 mean and NO$_2$ VCD declined by ~ 12% (15-20% for AOD and 20-30% for NO$_2$ VCD if the summertime values alone are considered).

Over the interior-south region, the overall level of industrialization and urbanization is much lower than that over the northeastern region, although growth in recent several years has also been rapid (19). Anthropogenic emissions of NO$_x$ and NMVOC from these sources are relatively minor: ratios of NO$_x$ emissions in 2005 to emissions of PM$_{10}$, PM$_{2.5}$ and SO$_2$ are 0.82, 1.14 and 0.4, respectively, about 28-42% lower than the corresponding ratios for the northeastern region (Supporting Information Table S1). The likely increases in production of nitrate and SOAs resulting from the recent industrial and urban development had a relatively small impact on both AOD and [PM$_{10}$] as compared to the impact of the decrease in emissions of primary aerosols and SO$_2$. Thus, in contrast to the
northeastern region, a general downward trend is observed for the interior-south region both for OMI AOD and [PM$_{10}$] (Figure 2c).

As discussed in the Supporting Information section 5, the conclusions drawn here are relatively insensitive to the influence of changes in meteorological conditions including precipitation, temperature, wind speed, mixing in the boundary layer, and associated changes in emissions and transport of natural dust aerosols.

Our analysis highlights the increasing importance both for public health and for the environment of emissions of NO$_x$ and NMVOC as industrialization and urbanization advances in China. China has begun drafting pollution control targets for the 12$^{\text{th}}$ FYP (for 2011-2015). This study indicates that the 12$^{\text{th}}$ FYP should include stricter and more effective emission control measures for NO$_x$, NMVOC and NH$_3$, in addition to continuing limitations on emissions of primary aerosols and SO$_2$.

Reducing secondary aerosols has a significant positive effect for public health. The potential impact on climate is more complicated since a reduction in secondary aerosols may contribute to a net increase in radiative forcing. Effective regulations to control emissions of black carbon and greenhouse gases are essential if we are to minimize future disturbance of the global climate.

Acknowledgments

This research is supported by the National Science Foundation, grant ATM-0635548, and Energy Foundation. We acknowledge the free use of OMI AOD data from NASA and ground-based AOD data from AERONET and EAST-AIRE (Project NNX08AH71G). We thank Xi Lu and Junling Huang for constructive discussion.
Supporting Information Available

Analysis of controls and magnitudes of anthropogenic emissions of aerosols and precursors in China; detailed data descriptions and processing for OMI AOD, [PM$_{10}$], OMI NO$_2$ VCDs, and surface measurements of AOD and meteorological parameters; evaluation of OMI AOD using ground-based AOD measurements; and analysis of effects of changes in meteorology on aerosols. This material is available free of charge via the Internet at http://pubs.acs.org.

References:


(13) README for OMAEROe (OMI Daily L3e for OMAERO).

http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/documents/v003/OMAEROe_OSIPS_README_V003.doc


(23) Wei, W. Research and Forecast on Chinese Anthropogenic Emissions of Volatile Organic compounds. Tsinghua University, Beijing, China, 2009.

**Figure 1.** Regional specifications based on linear trend analysis. (a) Definition of East China (103°E-124°E, 21°N-42°N; the entire domain) and its sub-regions. The ‘northeast’ and ‘interior-south’ are denoted as regions bounded by brown and yellow lines, respectively; and the ‘southeast’ consists of the remaining areas in the southeast. The Beijing-Tianjin area, part of the northeastern region, is bounded by green lines. Also shown are provinces and province-level municipalities (Beijing (‘BJ’), Tianjin (‘TJ’), Shanghai (‘SH’), and Chongqing (‘CQ’)) in East China. Jurisdictions in red are mentioned in the paper. (b) Interannual trend of OMI AOD over Oct 2004 – Sep 2008 (prior to the economic downturn) derived with a simple fit using linear regression (see Supporting Information section 3).

**Figure 2.** Changes in daily data for OMI AOD and [PM$_{10}$] and in monthly data for OMI NO$_2$ VCD during Oct 2004 – Dec 2009. The black line denotes the regional mean AOD from OMI; the red line denotes the mean of OMI AOD sampled in the cities with [PM$_{10}$]
measurements; the green line denotes the mean of \([\text{PM}_{10}]\) measurements. The black ‘+’ line denotes the regional mean \(\text{NO}_2\) VCD from OMI; the red ‘+’ line denotes the mean \(\text{NO}_2\) VCD sampled in the cities with \([\text{PM}_{10}]\) measurements. There are 37, 2 and 21 cities with \([\text{PM}_{10}]\) measurements in the three regions, respectively. Data shown here are based on a 365-day moving average for OMI AOD and \([\text{PM}_{10}]\) and a 12-month moving average for OMI \(\text{NO}_2\) VCD, i.e., each data point represents the mean over the prior year (Supporting Information section 3). The dashed vertical line depicts the approximate start of the Chinese economic downturn.
Figure 1.
Figure 2.