

Improvements in Urban Air Quality: Case Studies from New York State, USA

Shannon M. Buckley · Myron J. Mitchell

Received: 27 August 2009 / Accepted: 17 March 2010 / Published online: 16 April 2010
© Springer Science+Business Media B.V. 2010

Abstract Air quality levels vary over regions due to meteorological factors, proximity to sources, and local conditions (i.e., topography). The Northeast USA is subjected to pollution inputs from both local sources and those from the upwind Midwest USA that are transported by prevailing meteorological patterns. With the passage of the Clean Air Act in 1970 and the establishment of the National Ambient Air Quality Standards (NAAQS), national levels of air pollutants have declined significantly. Our study compared air quality time trends between five of the largest cities within New York State (Albany, Buffalo, New York City, Rochester, and Syracuse) and statewide means to national trends. Data were obtained from the NYS Department of Environmental Conservation (DEC) Bureau of Air Quality Surveillance for six criteria pollutants: carbon monoxide (CO), sulfur dioxide (SO₂), nitrogen dioxide (NO₂), ozone (O₃), particulate matter (PM_{2.5}), and lead (Pb). Regional Kendall tests found significant downward trends for each pollutant statewide from 1980 to 2007, while trends by city varied by decade and pollutant. The evaluation of historical trends of pollution in industrialized nations is useful in showing recent air quality improvements and also in the understanding what can be the result in

air pollutant controls in those developing nations currently experiencing high levels of pollution.

Keywords Urban · Air quality · CO · SO₂ · NO₂ · O₃ · PM_{2.5} · Pb

1 Introduction

In the USA, a major advancement to improve air quality began in 1970 when Congress passed the Clean Air Act (CAA) and created the Environmental Protection Agency (EPA) to enforce and support environmental regulations. Under the CAA, sources of air pollution have been required to employ technologies and methods to reduce emissions to meet National Ambient Air Quality Standards (NAAQS). The NAAQS identify and address six criteria pollutants: carbon monoxide (CO), sulfur dioxide (SO₂), nitrogen dioxide (NO₂), ozone (O₃), particulate matter <2.5 μm in diameter (PM_{2.5}), and lead (Pb) by setting primary and secondary standards. Primary standards protect against negative human health effects while secondary standards protect against adverse effects on public welfare. A history of NAAQS can be seen in Table 1. Despite these regulations and general success in reducing both emissions and concentrations of pollutants in the atmosphere, there are still non-attainment areas throughout the USA, mainly in highly urbanized locations (EPA 2008a).

S. M. Buckley (✉) · M. J. Mitchell
Department of Environmental and Forest Biology, SUNY,
College of Environmental Science and Forestry,
1 Forestry Drive,
Syracuse, NY, USA
e-mail: shbuckle@syr.edu

Table 1 Evolution of Air Quality Standards. Standards and sampling times provided for the criteria pollutants under the USEPA NAAQS

| Pollutant | 1971 | 1978/1979 | 1987 | 1997 | 2006/2008 | Current | WHO Air Quality Guidelines (2006) |
|-------------------|---|----------------------------------|---|--|--|--|--|
| CO | 9 ppm 8 h 35 ppm 1 h | Same | Same | Same | Same | 9 ppm 8 h 35 ppm 1 h | – |
| SO ₂ | 0.03 ppm annual 0.14 ppm 24 h | Same | Same | Same | Same | 0.03 ppm annual 0.14 ppm 24 h | 0.008 ppm 24 h 0.19 10 min |
| NO ₂ | 0.053 ppm annual | Same | Same | Same | Same | 0.053 ppm annual | 0.02 ppm annual 0.11 ppm 1 h |
| O ₃ | 0.08 ppm max 1-h level | 0.12 ppm max 1-h value | Same | 0.08 ppm 8 h 3 yr mean of the 4th max value | 0.075 ppm 8 h 3 yr mean of the 4th max value | 0.075 ppm 8 h 3 yr mean of the 4th max value | 0.05 ppm 8 h |
| TSP | 75 µg m ⁻³ annual 260 µg m ⁻³ 24 h | Same | – | – | – | – | – |
| PM ₁₀ | – | – | 50 µg m ⁻³ annual 150 µg m ⁻³ 24 h | 150 µg m ⁻³ 99th percentile annual | 150 µg m ⁻³ 24 h | 150 µg m ⁻³ 24 h | 20 µg m ⁻³ annual 50 µg m ⁻³ 24 h |
| PM _{2.5} | – | – | – | 15 µg m ⁻³ annual 65 µg m ⁻³ 24 h | 15 µg m ⁻³ annual 35 µg m ⁻³ 24 h | 15 µg m ⁻³ annual 35 µg m ⁻³ 24 h | 10 µg m ⁻³ annual 25 µg m ⁻³ 24 h |
| Pb | – | 1.5 µg m ⁻³ quarterly | Same | Same | 0.15 µg m ⁻³ rolling 3-month mean | 0.15 µg m ⁻³ rolling 3-month mean | – |

WHO standards provided for reference (SO₂, NO₂, O₃ converted to ppm based on the WHO conversion factors provided in Harrop (2002). Sources: Bachmann 2007; WHO 2006)

Elevated levels of air pollutants have been found to cause harm to both the environment and human health. A recent report by the EPA (2008a) documents a 50% decline in aggregate pollutant emissions from 1980 to 2006 despite a 121% increase in gross domestic product, 101% increase in vehicle distances traveled, and 32% increase in population. Despite these successes, future levels of pollutants may be influenced by climate change (i.e., Mickley et al. 2004; Jacob and Winner 2009) and pollution transported from developing nations around the globe (Fenger 1999). In order to document these impacts, it is useful to have an understanding on the recent trends in atmospheric concentration of these pollutants.

Air pollution varies over areas due to meteorological factors, proximity to sources, and local conditions (i.e., topography). The Northeast USA has been often referred to as the ‘tailpipe’ of the country because air pollution from the Midwest is transported to this region by prevailing meteorological patterns (Bari et al. 2003). Additionally, this region is subject to local pollution that is capable of contributing comparable amounts of pollutants as imported to the region (Rahn and Lownethal 1985). Urban centers and the surrounding areas are often associated with elevated pollution levels resulting from a high density of sources including traffic and industry (Fenger 1999).

Air pollution in urban areas has long been a concern due to the high density of sources and human populations exposed to those sources. Industrialized nations have been successfully improving air quality over the last few decades (Fenger 1999). However, at the same time, rapidly developing nations are now facing the same dangerously high levels of air pollution, following many of the same patterns developed nations experienced during their respective periods of industrialization. A relationship has been found between income and levels of air pollution with lower incomes generally associated with poorer air quality (Fenger 1999; Sivertsen 2006). By examining the historical trends of developed nations, developing nations can learn important lessons to limit impacts of air pollution on their rapidly expanding urban populations.

This study examines historical air quality records of the ‘criteria’ pollutants in five of the largest cities within New York State (NYS): Albany, Buffalo, New

York City, Rochester, and Syracuse. We evaluated trends in criteria pollutants concentrations from 1980 to 2007 and made comparisons among NYS cities. These patterns were also evaluated with respect to the US national trends and global issues related to air pollutants.

2 Methods

Air monitoring data were obtained from the New York State Department of Environmental Conservation (DEC), Bureau of Air Quality Surveillance for five locations across New York State: Albany, Buffalo, New York City, Rochester and Syracuse. Data from the Borough of Manhattan were chosen to represent the city due to its central location, with the realization that this selection has associated problems with data limitations and methodological issues. Records from the World Trade Center roof have been removed from this study, due to the anomalous nature of readings from that elevated height.

Chemical species included in this study are the criteria pollutants (CO, SO₂, NO₂, O₃, PM_{2.5}, and Pb). The data used are based on the NAAQS averaging time (see Table 1) and are not necessarily representative of the mean annual concentrations. For example, CO 8 hour (8-h) values are based on the maximum value recorded during an 8-h period for the given year, while CO 1 hour (1-h) are based on the highest value recorded during 1 h. Sulfur dioxide and PM_{2.5} data are from annual means and the maximum value for any given 24-h period in a year. Ozone data are based on the fourth maximum value of each year. Annual means are used for NO₂. Lead data for each year consist of four quarterly means. Data identified by the DEC as not meeting summary criteria (i.e., not sufficient entries to provide a representative mean) were not included in the analysis. Specific information regarding instrumentation and data collection protocols can be found on the DEC’s website (<http://www.dec.ny.gov/chemical/8541.html>; February 2010). Table 2 provides a summary of the pollutants, cities, and data range included in our study.

Due to the non-normality and heterogeneous variances of the data, Mann–Kendall tests for trend were performed on each chemical pollutant concen-

Table 2 Changes in New York State air quality in five cities

| Chemical species | City | Year range | Regional M–K test results (tau, <i>n</i> , annual median rate of change) |
|--|---------------|------------|---|
| CO 8 h (ppm) | Albany | 1997–2007 | –0.75, <i>n</i> =244, –0.25 |
| | Buffalo | 1980–2007 | |
| | New York City | 1980–2007 | |
| | Rochester | 1980–2007 | |
| | Syracuse | 1980–2007 | |
| CO 1 h (ppm) | Albany | 1997–2007 | –0.77, <i>n</i> =244, –0.53 |
| | Buffalo | 1980–2007 | |
| | New York City | 1980–2007 | |
| | Rochester | 1980–2007 | |
| | Syracuse | 1980–2007 | |
| SO ₂ annual (ppm) | Albany | 1980–2007 | –0.79, <i>n</i> =159, –0.0004 |
| | Buffalo | 1980–2007 | |
| | New York City | 1980–2007 | |
| | Rochester | 1980–2007 | |
| | Syracuse | 1980–2007 | |
| SO ₂ 24 h (ppm) | Albany | 1980–2007 | –0.73, <i>n</i> =222, –0.002 |
| | Buffalo | 1980–2007 | |
| | New York City | 1980–2007 | |
| | Rochester | 1980–2007 | |
| | Syracuse | 1980–2007 | |
| O ₃ 8 h 4th max value (ppm) | Albany | 1987–2007 | –0.28, <i>n</i> =72, –0.0005 |
| | New York City | 1980–2007 | |
| | Rochester | 1980–2007 | |
| | Syracuse | 1980–2007 | |
| | | | |
| NO ₂ annual (ppm) | Albany | 1990–2007 | –0.56, <i>n</i> =63, –0.0003 |
| | Buffalo | 1980–2007 | |
| | New York City | 1982–2007 | |
| | | | |
| PM _{2.5} annual (μg m ⁻³) | Albany | 2000–2007 | –0.44, <i>n</i> =67, –0.33 |
| | Buffalo | 2001–2007 | |
| | New York City | 2000–2007 | |
| | Rochester | 2000–2007 | |
| | Syracuse | 2000–2007 | |
| PM _{2.5} 24 h (μg m ⁻³) | Albany | 1999–2007 | –0.39, <i>n</i> =108, –0.94 |
| | Buffalo | 1999–2007 | |
| | New York City | 1999–2007 | |
| | Rochester | 1999–2007 | |
| | Syracuse | 1999–2007 | |
| Lead quarterly (μg m ⁻³) | Albany | 1983–1998 | –0.62, <i>n</i> =320, –0.01 |
| | Buffalo | 1982–1998 | |
| | New York City | 1983–1998 | |
| | Rochester | 1982–1998 | |
| | Syracuse | 1983–1998 | |

Results from Regional Kendall test are included, and all are significant ($\alpha=0.05$). Data from New York State DEC Bureau of Air Quality Surveillance

tration at each site, for the full time period, and for each decade of the record (1980–1989, 1990–1999, and 2000–2007) (Helsel et al. 2006). The analysis of the results from the three time periods provides additional information on the historical changes in temporal patterns. Regional Mann–Kendall test for trend was used on all sites for each pollutant and averaging time to detect the potential statewide trends (Helsel and Frans 2006). Data from these tests, and reported in associated tables, include Kendall’s tau correlation coefficient and slope (or median rate of change for Regional M–K tests). Significance was based on $\alpha=0.05$.

3 Results

In New York State, all six criteria pollutants exhibited statistically significant downward trends (see Table 2 for Regional Mann–Kendall (M–K) test results). Significant decreases were found at all sites for 1-h and 8-h CO for the entire data range (1980–2007; Table 3 and Fig. 1). For CO 1 h, NYC saw the greatest decrease (-0.81 , $p<0.0001$), followed by Syracuse (-0.62 , $p<0.0001$), both above the regional rate of change, -0.53 ($p<0.0001$). The same trend

appears in the 8-h data, with NYC (-0.51 , $p<0.0001$) and Syracuse (-0.32 , $p<0.0001$) above the regional median rate of change, -0.25 ($p<0.0001$). As seen in Table 3, there were no significant trends during the 1980s at any site for CO 1 h. Rochester was the only city that saw significant trends in CO 8 h in the 1980s (-0.23 , $p=0.0091$). However, significant downward trends appear in the 1990s through 2000s for both 1-h and 8-h records. The steepest decline in any decade was found in Syracuse, 1990–1999, with a slope of -1.45 ($p=0.0024$) for CO 1 h and -0.9 ($p=0.0024$) for CO 8 h.

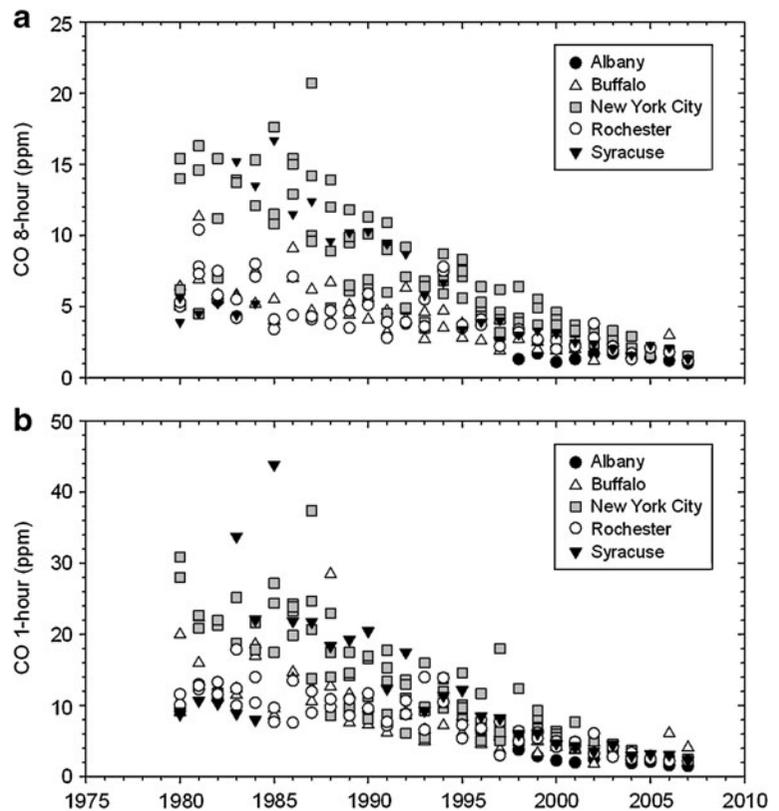
Albany, despite having an overall trend for the data range in both CO 1 h and 8 h, had no significant trends during the 1990s or 2000s individually and had the smallest downward trend of the five cities. Buffalo only experienced significant declines during the 1990s for both 1 h and 8 h (-0.43 , $p=0.0006$ and -0.22 , $p=0.0032$, respectively).

All cities showed similar downward trends in annual SO₂ values from 1980 to 2007, especially in the 24-h data (Fig. 2; Table 4). A greater number of significant downward trends per decade were seen in the annual data compared to the 24-h data, with Albany, Rochester and Syracuse each having one decade each of significant decline in SO₂ 24-h data.

Table 3 CO Mann–Kendall test results for five New York State cities from 1980 to 2007

| | | 1980–2007 | 1980–1989 | 1990–1999 | 2000–2007 | |
|--|--------------|-----------|------------------------------------|------------------------------------|------------------------------------|------------------------------------|
| Eight hour and 1-h averaging times are included. Data in table include (in order): Kendall's tau correlation coefficient, number of records used in test, and slope for entire time period, as well as individual decades (1980s, 1990s, and 2000s). Italicized data are statistically significant annual trends ($\alpha=0.05$), ‘–’ represents non-significant results | CO 1 h (ppm) | Albany | <i>-0.67, n=11</i> <i>-0.23</i> | <i>No data</i> | – | – |
| | | Buffalo | <i>-0.71, n=51</i> <i>-0.47</i> | – | <i>-0.56, n=20</i> <i>-0.43</i> | – |
| | | NYC | <i>-0.66, n=94</i> <i>-0.81</i> | – | <i>-0.31, n=44</i> <i>-0.42</i> | <i>-0.65, n=17</i> <i>-0.47</i> |
| | | Rochester | <i>-0.68, n=57</i> <i>-0.40</i> | – | <i>-0.46, n=20</i> <i>-0.47</i> | <i>-0.67, n=14</i> <i>-0.4</i> |
| | | Syracuse | <i>-0.59, n=31</i> <i>-0.62</i> | – | <i>-0.78, n=10</i> <i>-1.45</i> | <i>-0.71, n=8</i> <i>-0.3</i> |
| | CO 8 h (ppm) | Albany | <i>-0.36, n=11</i> <i>-0.05</i> | <i>No data</i> | – | – |
| | | Buffalo | <i>-0.67, n=51</i> <i>-0.20</i> | – | <i>-0.48, n=20</i> <i>-0.22</i> | – |
| | | NYC | <i>-0.68, n=94</i> <i>-0.51</i> | – | <i>-0.40, n=44</i> <i>-0.34</i> | <i>-0.61, n=17</i> <i>-0.30</i> |
| | | Rochester | <i>-0.65, n=57</i> <i>-0.17</i> | <i>-0.39, n=23</i> <i>-0.23</i> | <i>-0.44, n=20</i> <i>-0.17</i> | <i>-0.50, n=14</i> <i>-0.15</i> |
| | | Syracuse | <i>-0.59, n=31</i> <i>-0.32</i> | – | <i>-0.78, n=10</i> <i>-0.90</i> | <i>-0.71, n=8</i> <i>-0.20</i> |

Fig. 1 CO levels in five New York State cities from 1980 to 2007. Maximum values for each year are used for the 8-h (a) and 1-h (b) averaging times



Albany showed the greatest downward trend of annual data (-0.0006 , $p < 0.0001$), while Albany, Buffalo, and NYC all had similar downward trends for the 24-h data (0.002 , $p < 0.0001$).

Ozone levels were available from all cities except Buffalo (Table 2 and Fig. 3). All cities except Syracuse experienced significant downward trends when using the entire data range (1980–2007; Table 5). New York City had the largest downward trend (-0.002 , $p = 0.011$), four times the regional median rate of change (-0.0005 , $p = 0.0003$). No location showed significant trends in any decade.

Results for NO_2 were only available for Albany, Buffalo, and New York City (Table 2). Buffalo and New York City showed the same significant downward trend from 1980 to 2007 (-0.0003 , $p < 0.0001$ for both; Table 6 and Fig. 4). Buffalo's only significant decadal trend was in the 2000s (-0.0008 , $p = 0.0094$), while New York City experienced its only significant downward trend in the 1990s (-0.0007 , $p = 0.036$).

Although the EPA began to research $\text{PM}_{2.5}$ as early as 1978, and standards recommended as early as

1980–82, action was deferred until promulgation in 1997 (Bachmann 2007). As the national monitoring network developed from 1998 to 2001, $\text{PM}_{2.5}$ data in New York State were only available from 2000 to 2007 (Table 7 and Fig. 5). Significant trends in annual data were only shown in NYC (-0.53 , $p < 0.0001$) and Rochester (-0.23 , $p = 0.037$), while 24-h data saw significant declines in NYC (-0.57 , $p = 0.0003$) and Syracuse (-1.4 , $p = 0.047$).

Atmospheric Pb concentrations between 1980 and 1998 have declined significantly, with a majority of reductions occurring in the 1980s (Table 8 and Fig. 6). The greatest downward trend was in Syracuse (-0.041 , $p < 0.0001$) and the smallest trend in Albany (-0.004 , $p < 0.0001$).

4 Discussion

Air pollution can negatively impact both human health and the environment (e.g., ALA 2008; EPA 2008a; Grantz et al. 2003; Samet et al. 2000). The World Health Organization (WHO 2006) reports that

Fig. 2 SO₂ levels in five New York State cities from 1980 to 2007. Annual (a) and 24-h (b) averaging times shown

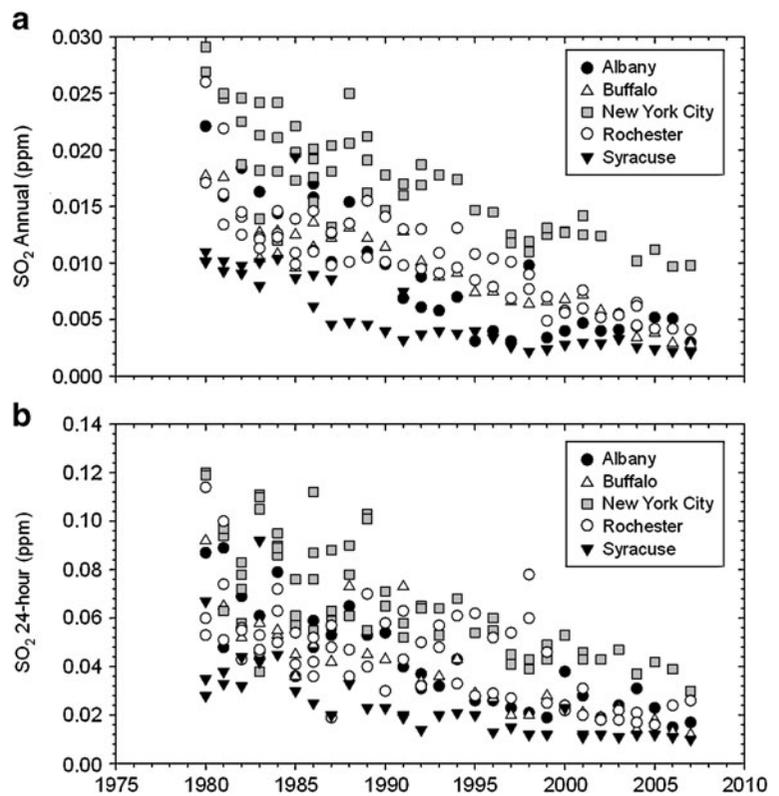
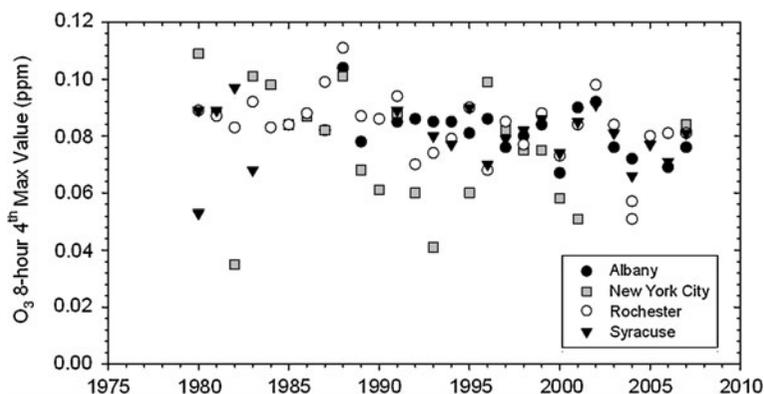


Table 4 SO₂ Mann–Kendall test results for five New York State cities from 1980 to 2007

| | | 1980–2007 | 1980–1989 | 1990–1999 | 2000–2007 |
|------------------------------|-----------|--|--|--|--|
| SO ₂ annual (ppm) | Albany | <i>-0.66, n = 30</i> <i>-0.0006</i> | – | – | – |
| | Buffalo | <i>-0.77, n = 32</i> <i>-0.0004</i> | – | <i>-0.80, n = 10</i> <i>-0.0007</i> | <i>-0.86, n = 8</i> <i>-0.0007</i> |
| | NYC | <i>-0.64, n = 55</i> <i>-0.0005</i> | <i>-0.33, n = 30</i> <i>-0.0006</i> | <i>-0.53, n = 16</i> <i>-0.0006</i> | <i>-0.72, n = 9</i> <i>-0.0004</i> |
| | Rochester | <i>-0.70, n = 61</i> <i>-0.0004</i> | <i>-0.37, n = 27</i> <i>-0.0005</i> | <i>-0.55, n = 20</i> <i>-0.0005</i> | <i>-0.43, n = 14</i> <i>-0.0003</i> |
| | Syracuse | <i>-0.78, n = 37</i> <i>-0.0003</i> | <i>-0.54, n = 18</i> <i>-0.0005</i> | <i>-0.53, n = 11</i> <i>-0.0002</i> | <i>-0.64, n = 8</i> <i>-0.0001</i> |
| SO ₂ 24 h (ppm) | Albany | <i>-0.71, n = 31</i> <i>-0.002</i> | – | <i>-0.78, n = 11</i> <i>-0.003</i> | – |
| | Buffalo | <i>-0.74, n = 32</i> <i>-0.002</i> | – | <i>-0.64, n = 10</i> <i>-0.003</i> | <i>-0.79, n = 8</i> <i>-0.002</i> |
| | NYC | <i>-0.58, n = 62</i> <i>-0.002</i> | – | <i>-0.54, n = 18</i> <i>-0.002</i> | <i>-0.71, n = 10</i> <i>-0.002</i> |
| | Rochester | <i>-0.48, n = 62</i> <i>-0.001</i> | <i>-0.36, n = 28</i> <i>-0.002</i> | – | – |
| | Syracuse | <i>-0.73, n = 35</i> <i>-0.001</i> | – | <i>-0.55, n = 11</i> <i>-0.001</i> | – |

Annual and 24-h averaging times are included. Data in table include (in order): Kendall's tau correlation coefficient, number of records used in test, and slope for entire time period and individual decades (1980s, 1990s, and 2000s). Italicized data are statistically significant annual trends ($\alpha=0.05$), '–' represents non-significant results

Fig. 3 O₃ levels in four New York State cities from 1980 to 2007, data represent the fourth-highest daily maximum 8-h average in a given year



urban air pollution (both indoor and outdoor) contributes to more than two million premature deaths every year worldwide, with over half occurring in developing nations. In the USA, the EPA's most recent report on National Air Quality (2008b) reports that despite improvements in concentrations of air pollutants, over 158 million people are still exposed to levels of pollution (mainly from O₃ and PM_{2.5}) that exceed NAAQS levels. Although New York City has showed overall improvements in air quality since 1980, there are substantial variations in the changes among pollutants and locations; for example, New York City continues to experience levels of PM_{2.5} that violate current NAAQS. Within our study, there were large variations between individual locations and timeframes (as seen in Tables 3, 4, 5, 6, 7 and 8).

Table 5 Ozone Mann–Kendall test results for New York State cities from 1980 to 2007 based on a 3-year average of the fourth-highest daily maximum 8-h average

| | | 1980–2007 |
|----------------------|-----------|--------------------------------------|
| O ₃ (ppm) | Albany | <i>-0.34, n=21</i> <i>-0.0006</i> |
| | NYC | <i>-0.42, n=20</i> <i>-0.002</i> |
| | Rochester | <i>-0.30, n=10</i> <i>-0.0004</i> |
| | Syracuse | – |

Data in table include (in order): Kendall's tau correlation coefficient, number of records used in test, and slope for entire time period. Italicized data are statistically significant annual trends ($\alpha=0.05$), '–' represents non-significant results. There were no significant trends in any single decade at any location

In urban areas, 95% of all CO emissions may come from incomplete internal combustion within vehicles (ALA 2002; EPA 2008a). Yet, since 1990, CO concentrations declined 62% in the USA despite a 24% increase in vehicle distance traveled, suggesting that increased efficiency of vehicles (lower pollutant emissions) is at least partially responsible for the improved air quality (EPA 2008a). Levels within all five cities used in our study showed similar downward trends in CO concentrations. However, these CO concentrations were elevated above natural background levels and can still pose a threat to the environment and human health. For example, CO undergoes chemical reactions in the atmosphere that produce ozone or react with hydroxyl radicals to form CO₂ thus contributing to gases associated with global climate change and other deleterious environmental impacts (Rasmussen et al. 1975; Khalil and Rasmussen 1990).

Most atmospheric SO₂ (87% of total emissions, EPA 2008a) is associated with the combustion of fossil fuels, two thirds of which are associated with electricity generation (Wake et al. 1998). Ambient SO₂ concentrations can impact human health by aggravating asthma and other respiratory problems (EPA 2008b). Due to its conversion to SO₃ and its high solubility in water, a majority of the SO₂ is converted to sulfuric acid and hence is a major contributor to "acidic deposition". With residence times of SO₂ in the atmosphere ranging from 20 min to 7 days (Rasmussen et al. 1975), acidic deposition produced from SO₂ can have a large geographical impact. Ecosystem impacts can include decreasing pH in sensitive water bodies, subsequent effects of fish and aquatic biota population, damage to vegetation, and depletion on the availability

Table 6 Annual NO₂ Mann–Kendall test results for three New York State cities from 1980 to 2007

| | | 1980–2007 | 1980–1989 | 1990–1999 | 2000–2007 |
|-----------------------|---------|--------------------------------------|----------------|--------------------------------------|-------------------------------------|
| NO ₂ (ppm) | Albany | – | <i>No data</i> | – | <i>No data</i> |
| | Buffalo | <i>–0.73, n=26</i> <i>–0.0003</i> | – | – | <i>–0.79, n=8</i> <i>–0.0008</i> |
| | NYC | <i>–0.41, n=30</i> <i>–0.0003</i> | – | <i>–0.41, n=15</i> <i>–0.0007</i> | – |

Data in table include (in order): Kendall's tau correlation coefficient, number of records used in test, and slope for entire time period and individual decades (1980s, 1990s, and 2000s). Italicized data are statistically significant annual trends ($\alpha=0.05$), '–' represents non-significant results

of nutrient cations (i.e., calcium and magnesium), as well as increased aluminum mobility and sulfur and nitrogen content, all of which have major environmental impacts (Driscoll et al. 2001, 2003; Wake et al. 1998). Sulfur dioxide can also be transformed into sulfate aerosols through a number of processes (Saxena and Seigneu 1987), introducing associated problems of haze and respiratory problems (as discussed below), as well as climate implications.

Due to the CAA and subsequent amendments, SO₂ emissions and resultant air concentrations have declined throughout New York State (as shown in this study), within the Northeast USA and within the contiguous states (ALA 2002; Butler et al. 2001; EPA 2008a; Wake et al. 1998). Butler et al. (2001) found a strong relationship between SO₂ emissions and atmospheric concentration in the Northeast USA and New York State. The US EPA (2008a) has estimated that nationally, SO₂ concentrations have decreased over 50% since 1990 in the USA. Although levels of SO₂ have dramatically decreased, there is evidence of continued acidification in surface waters due to previous accumulation of pollutants in soils and the

presence of other acidifying pollutant species (i.e. nitrogen), resulting in delayed acidification as solutes responsible for acidification and/or eutrophication are mobilized to soils and surface waters (Cape et al. 2003).

Currently, within the USA, a majority of NAAQS violations are associated with elevated O₃ levels, irritating human lungs and harming plants by damaging leaves and reducing growth rate and yields for crops (EPA 2008b). Ozone is a unique pollutant to regulate as it is formed from other pollutants as they are transported away from the source, with suburban areas exhibiting higher concentrations than that within the urban center (Katsouyanni 2003). The importance of these interactions suggests the necessity of addressing NO_x and volatile organic compound (VOCs) levels. Wang et al. (2009) found that high levels of O₃ in NYS can be associated with elevated levels transported from Canada, and that western New York is among the most affected/vulnerable in the USA to these imports.

In our study, O₃ showed very small downward trends and no significant trends during any one decade. The difficulty in reducing O₃ levels can be the result of

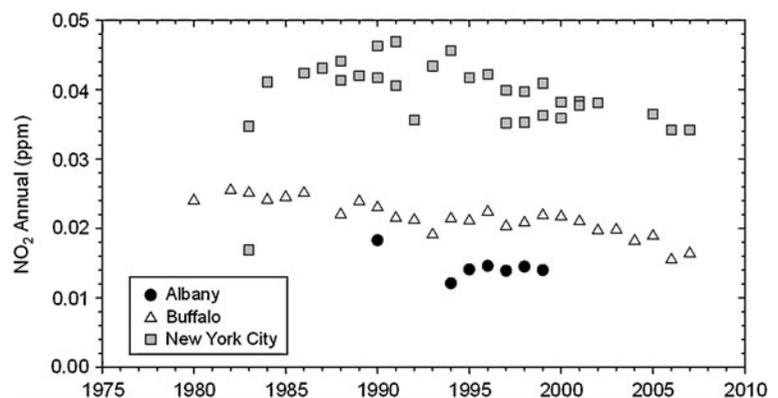
Fig. 4 Annual NO₂ levels in three New York State cities from 1980 to 2007

Table 7 PM_{2.5} Mann–Kendall test results for five New York State cities from 1999 to 2007 for the annual and 24-h averaging time

| | | 2000–2007 |
|--|-----------|----------------------|
| PM _{2.5} annual ($\mu\text{g m}^{-3}$) | Albany | – |
| | Buffalo | – |
| | NYC | <i>–0.55, n = 33</i> |
| | | <i>–0.53</i> |
| | Rochester | <i>–0.53, n = 10</i> |
| PM _{2.5} 24 h ($\mu\text{g m}^{-3}$) | Syracuse | – |
| | Albany | – |
| | Buffalo | – |
| | NYC | <i>–0.36, n = 48</i> |
| | | <i>–0.57</i> |
| | Rochester | – |
| | Syracuse | <i>–0.37, n = 16</i> |
| | | <i>–1.4</i> |

Data in table include (in order): Kendall's tau correlation coefficient, number of records used in test, and slope for entire time period and individual decades (1980s, 1990s, and 2000s). Italicized data are statistically significant annual trends ($\alpha = 0.05$), '–' represents non-significant results

Fig. 5 PM_{2.5} levels in five New York State cities from 1999 to 2007 for the annual (a) and 24-h averaging time (b)

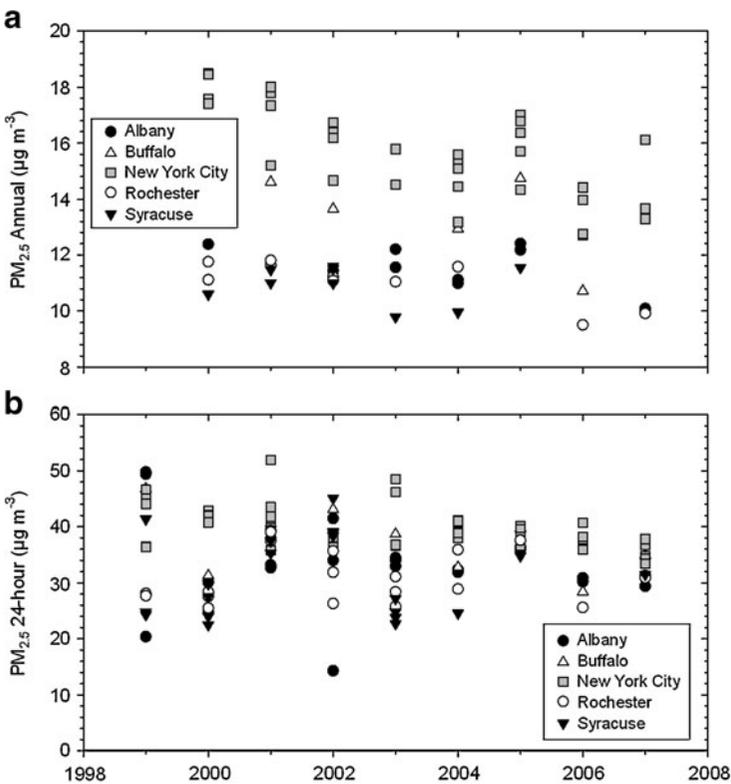


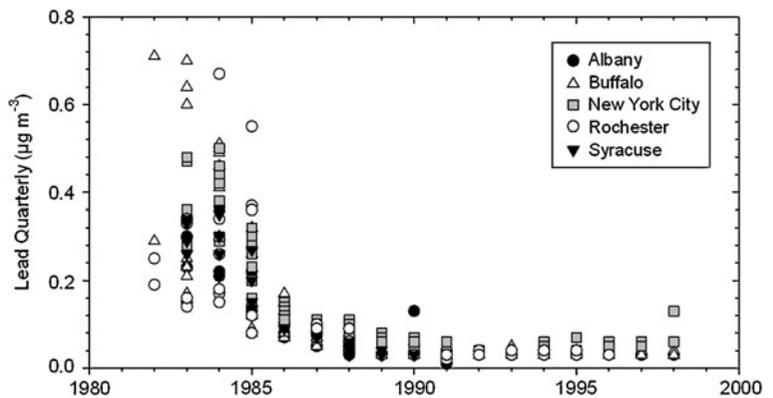
Table 8 Lead Mann–Kendall test results for five New York State cities from 1980 to 1998 based on a quarterly average

| | | 1980–1999 | 1980–1989 |
|----------------------------------|---------------|-----------------------|----------------------|
| Lead ($\mu\text{g m}^{-3}$) | Albany | <i>–0.55, n = 55</i> | <i>–0.77, n = 22</i> |
| | | <i>–0.004</i> | <i>–0.033</i> |
| | Buffalo | <i>–0.60, n = 111</i> | <i>–0.73, n = 57</i> |
| | | <i>–0.01</i> | <i>–0.038</i> |
| | NYC | <i>–0.73, n = 73</i> | <i>–0.76, n = 46</i> |
| | <i>–0.017</i> | <i>–0.05</i> | |
| | Rochester | <i>–0.53, n = 56</i> | <i>–0.47, n = 30</i> |
| | | <i>–0.011</i> | <i>–0.022</i> |
| | Syracuse | <i>–0.77, n = 25</i> | <i>–0.75, n = 22</i> |
| | | <i>–0.041</i> | <i>–0.047</i> |

Data in table include (in order): Kendall's tau correlation coefficient, number of records used in test, and slope for entire time period and individual decades (1980s and 1990s). Italicized data are statistically significant trends ($\alpha = 0.05$). There were no significant trends in the 1990s

many interacting circumstances, ranging from climate change, to local emissions of NO_x and VOCs, to transport of pollution from surrounding areas (Wang et al. 2009), to stratospheric influence and increase levels

Fig. 6 Lead levels in five New York State cities from 1980 to 1998 based on a quarterly average. All slopes are significant ($\alpha=0.05$)



of methane (Vingarzan 2004). Some studies suggest that climate change will likely increase pollution events associated with elevated O₃ concentrations (Jacob and Winner 2009; Hogrefe et al. 2004). However, other studies (i.e., Tagaris et al. 2007; Leung and Gustafson 2005) have found mixed results regarding climate change's impact on pollution levels.

Changes in NO₂ concentrations were based on limited data, but appear to vary by location. Within the USA, between 1990 and 2006, NO₂ concentrations have been reduced 30% (EPA 2008a). Fenger (2009) reports that NO_x and O₃ are the main pollutant problems in developing nations due to the increased use and availability of vehicles in urban areas. Yet still, little is known about the long-term health impacts of NO₂ outside the increased vulnerability of asthmatics and irritation caused within the lungs (Brunekreef and Holgate 2002; EPA 2008a; Katsouyanni 2003). Elevated NO₂ concentrations can contribute to acidic deposition and O₃ formation (EPA 2008a; Galloway et al. 2003; Katsouyanni 2003; WHO 2006).

The interest in particulate pollution has evolved over time (as seen in Table 1) with the development of more sophisticated instrumentation and long-term research (WHO 2006), beginning with initial interest in total suspended particles (TSP), to PM₁₀, to the current focus on fine (PM_{2.5}, PM₁) and ultrafine particulates (<0.1 µm). Generally, particles can influence the amount of solar radiation reaching vegetation or through changing the soil chemistry by impacting nutrient cycling, but the impact of particles on environments and ecosystems depends largely on its constituents (i.e., heavy metals, sulfur, and nitrogen compounds; Grantz et al. 2003). Fine particulates are of concern to policymakers and

epidemiologists due to linkages with mortality and morbidity (Dockery et al. 1993; Englert 2004; Pope 2000; Pope et al. 2002; Samet et al. 2000; WHO 2006), as well as contributing to poor visibility (Sivertsen 2006). Ezzati et al. (2002) estimated that about one million premature deaths per year are associated with particulate pollution (in Katsouyanni 2003). Pope et al. (2009) noted that a reduction in fine particulates significantly improved life expectancy in the USA by as much as 15%.

In the USA, mobile sources are one of the leading contributors to PM_{2.5} in the atmosphere (Gertler et al. 2003) and are the largest source of PM_{2.5} emissions in densely populated urban areas. Annual PM_{2.5} concentrations have been reduced 14% between 2000 and 2006 across the USA, and similar trends are found for daily concentrations (EPA 2008a). The downward trends within some, but not all our study sites, reflect the current national struggle to decrease atmospheric levels of particles. According to the most recent American Lung Association State of the Air Report (2008), New York City has been removed from the top 25 list of most polluted cities due to reduction of annual PM_{2.5} values. Yet, as particulate pollution can be influenced by meteorological conditions in addition to proximity to sources, DeGaetano and Doherty (2004) suggested the difficulty ascribing improving concentrations with regulatory actions. Moreover, the impacts of climate change on PM levels have been difficult to predict (Jacob and Winner 2009) emphasizing the need for continued monitoring and research on this pollutant.

The slower responses in concentrations of O₃, NO₂ and PM_{2.5} compared to the other pollutants seen in our study have been noted elsewhere (e.g., Brunekreef and Holgate 2002; Fenger 2009) and suggest that the

evaluations of these pollutants especially with respect to changes in the contributions of mobile sources on global air quality are needed. If cities within the USA are having difficulty maintaining 'safe' levels of these concentrations, developing nations will be faced with an even greater challenge. These three pollutants, along with Pb and CO, are also closely associated with vehicle emissions and should be monitored closely as vehicle usage continues to increase (EPA 2008a).

The American Lung Association (2002) reports that over the last 20 years, Pb levels across the nation have declined 93%. Sources of Pb are likely to be local due to the relatively short atmospheric lifetime (days) that makes it unlikely for atmospheric Pb to be transported long distances (Elsenreich et al. 1986). The decline in Pb seen across New York State is likely due to decreased use of leaded gasoline within the USA (Daines et al. 1970; Elsenreich et al. 1986), as Table 8 reflects that the majority of the downward trend in Pb occurred in the 1980s, as there was the gradual phase out of leaded gasoline.

5 Conclusions

The result of our study shows the varying success of pollutant reductions throughout New York State. Regionally, all of the pollutants examined in this study showed a significant downward trend in levels, yet results varied by location. Reductions in CO did not appear to be underway until the 1990s, as was the case with 24-h max concentrations of SO₂. Ozone and NO₂ temporal patterns showed the least consistency among sites, with some regions experiencing downward trends while others do not. The results for PM_{2.5} show that further improvements are needed.

By examining these trends in the New York State and similar records from around the world, developing nations can learn from the experiences of industrialized nations and limit the deleterious environmental impacts of levels of air pollutants during their industrialization. As an increasing proportion of the world's population is living in cities, more people are being exposed to potentially dangerous levels of air pollutants. The standards proposed by the WHO are targeted at developing nations and their obtainment should be made a priority with respect to the improvement of global environmental health. Air pollution in developing

nations needs to remain an important issue as role of the global transport of pollutants becomes more evident (Fenger 2009). Developed nations such as the USA are learning which methods and policies are most effective at reducing pollutant concentrations, and these results need to be shared and used to provide important information on the potential for atmospheric pollutant reductions in rapidly industrializing nations.

Acknowledgements The authors gratefully acknowledge the support for this work by Syracuse Center of Excellence CARTI project award, which is supported by a grant from U.S. Environmental Protection Agency [Award No: X-83232501-0]. This work has not been subjected to the Agency's required peer and policy review and therefore does not necessarily reflect the views of the agency and no official endorsement shall be inferred. We would also like to thank the Russ Twaddle at the New York State Department of Environmental Conservation Bureau of Air Quality Surveillance for providing the data and assistance. We also want to thank Phil Hopke at Clarkson University for his helpful comments on these analyses and interpretations. And a final thanks to the two anonymous reviewers for their useful comments, questions and suggestions.

References

- American Lung Association (2002). Trends in air quality. <http://www.lungusa.org>. Accessed 10 October 2008.
- American Lung Association (2008). State of the air: 2008. <http://www.lungusa.org>. Accessed 10 October 2008.
- Bachmann, J. (2007). Chronology of the U.S. National Ambient Air Quality Standards Supplemental Tables to A&WMA 2007 Critical Review. *Journal of Air and Waste Management*, available online: http://secure.awma.org/journal/pdfs/2007/6/10.3155-1047-3289.57.6.652_supplmaterial.pdf. Accessed 11 October 2008.
- Bari, A., Dutkiewicz, V. A., Judd, C. D., Wilson, L. R., Luttinger, D., & Husain, L. (2003). Regional sources of particulate sulfate, SO₂, PM_{2.5}, HCl, and HNO₃, in New York, NY. *Atmospheric Environment*, 37, 2837–2844.
- Brunekreef, B., & Holgate, S. T. (2002). Air pollution and health. *Lancet*, 360, 1233–1242.
- Butler, T. J., Likens, G. E., & Stunder, B. J. B. (2001). Regional-scale impacts of Phase I of the Clean Air Act Amendments in the USA: The relation between emissions and concentrations, both wet and dry. *Atmospheric Environment*, 35, 1015–1028.
- Cape, J. N., Fowler, D., & Davidson, A. (2003). Ecological effects of sulfur dioxide, fluorides, and minor air pollutants: Recent trends and research needs. *Environment International*, 29, 201–211.
- Daines, R. H., Motto, H., & Chilko, D. M. (1970). Atmospheric lead: Its relationship to traffic volume and proximity to highways. *Environmental Science & Technology*, 4(4), 318–322.

- DeGaetano, A. T., & Doherty, O. M. (2004). Temporal, spatial and meteorological variations in hourly PM_{2.5} concentration extremes in New York City. *Atmospheric Environment*, *38*, 1547–1558.
- Dockery, D. W., Pope, C. A., Xu, X., Spengler, J. D., Ware, J. H., Fay, M. E., et al. (1993). An association between air pollution and mortality in six U.S. cities. *New England Journal of Medicine*, *329*, 1753–1759.
- Driscoll, C. T., Lawrence, G. B., Bulger, A. J., Butler, T. J., Cronan, C. S., Eagar, C., et al. (2001). Acidic deposition in the Northeastern United States: Sources and inputs, ecosystem effects, and management strategies. *BioScience*, *51*(3), 180–198.
- Driscoll, C. T., Driscoll, K. M., Mitchell, M. J., & Raynal, D. J. (2003). Effects of acidic deposition on forest and aquatic ecosystems in New York State. *Environmental Pollution*, *123*, 327–336.
- Elsenreich, S. J., Metzger, N. A., & Urban, N. R. (1986). Response of atmospheric lead to decreased use of lead in gasoline. *Environmental Science & Technology*, *20*, 171–174.
- Englert, N. (2004). Fine particles and human health—a review of epidemiological studies. *Toxicology Letters*, *149*, 235–242.
- Ezzati, M., Lopez, A. D., Rodgers, A., Vander Hoorn, S., Murray, C. J. L., & CRACG (2002). Selected major risk factors and global and regional burden of disease. *Lancet*, *360*, 1347–1360.
- Fenger, J. (1999). Urban air quality. *Atmospheric Environment*, *33*, 4877–4900.
- Fenger, J. (2009). Air pollution in the last 50 years—from local to global. *Atmospheric Environment*, *43*, 13–22.
- Galloway, J. N., Aber, J. D., Erisman, J. W., Seitzinger, S. P., Howarth, R. H., Cowling, E. B., et al. (2003). The nitrogen cascade. *BioScience*, *53*, 341–356.
- Gertler, A. W., Gillies, J. A., & Pierson, W. R. (2003). An assessment of the mobile source contribution to PM₁₀ and PM_{2.5} in the United States. *Water, Air, and Soil Pollution*, *123*, 203–214.
- Grantz, D. A., Garner, J. H. B., & Johnson, D. W. (2003). Ecological effects of particulate matter. *Environment International*, *29*, 213–239.
- Harrop, O. (2002). *Air quality assessment and management: A practical guide* (p. 353). London: Taylor & Francis.
- Helsel, D. R., & Frans, L. M. (2006). Regional Kendall test for trend. *Environmental Science and Technology*, *40*(13), 4066–4073.
- Helsel, D. R., Mueller, D. K., & Slack, J. R. (2006). Computer program for the Kendall family of trend tests: U.S. Geological Survey Scientific Investigations Report 2005–5275, p. 4.
- Hogrefe, C., Lynn, B., Civerolo, K., Ku, J.-Y., Rosenthal, J., Rosenzweig, C., et al. (2004). Simulating changes in regional air pollution over the eastern United States due to changes in global and regional climate and emissions. *Journal of Geophysical Research*, *109*, D22301. doi:10.1029/2004JD004690.
- Jacob, D. J., & Winner, D. A. (2009). Effect of climate change on air quality. *Atmospheric Environment*, *43*, 51–63.
- Katsouyanni, K. (2003). Ambient air pollution and health. *British Medical Bulletin*, *68*, 143–156.
- Khalil, M. A. K., & Rasmussen, R. A. (1990). The global cycle of carbon monoxide: Trends and mass balance. *Chemosphere*, *20*(1–2), 227–242.
- Leung, L. R., & Gustafson, W. I. (2005). Potential regional climate change and implications to U.S. air quality. *Geophysical Research Letters*, *32*, L16711. doi:10.1029/2005GL022911.
- Mickley, L. J., Jacob, D. J., & Field, B. D. (2004). Effects of future climate change on regional air pollution episodes in the United States. *Geophysical Research Letters*, *31*, L24103. doi:10.1029/2004GL021216.
- Pope, C. A. (2000). Epidemiology of fine particulate air pollution and human health: Biologic mechanism and who's at risk? *Environmental Health Perspectives*, *108*(4), 713–723.
- Pope, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., et al. (2002). Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *Journal of the American Medical Association*, *287*(8), 1132–1141.
- Pope, C. A., Ezzati, M., & Dockery, D. W. (2009). Fine-particulate air pollution and life expectancy in the United States. *New England Journal of Medicine*, *360*(4), 376–386.
- Rahn, K. A., & Lownethal, D. H. (1985). Pollution aerosol in the Northeast: Northeastern–Midwestern contributions. *Science*, *228*(4697), 275–284.
- Rasmussen, K. H., Taheri, M., & Kabel, R. L. (1975). Global emissions and natural processes for removal of gaseous pollutants. *Water, Air, and Soil Pollution*, *4*, 33–64.
- Samet, J. M., Dominici, F., Currier, I., Coursac, I., & Zeger, S. L. (2000). Fine particulate air pollution and mortality in 20 U.S. cities. *New England Journal of Medicine*, *343*(24), 1742–1749.
- Saxena, P., & Seigneur, C. (1987). On the oxidation of SO₂ to sulfate in atmospheric aerosols. *Atmospheric Environment* (1967), *21*(4), 807–812.
- Sivertsen, B. (2006). Chapter 2: Global ambient air pollution concentrations and trends, in *Air Quality Guidelines: Global Update 2005*. World Health Organization. <http://www.euro.who.int/Document/E90038.pdf>. Accessed 12 November 2008.
- Tagaris, E., Manomaiphiboon, K., Liao, K.-J., Leung, L. R., Woo, J.-H., He, S., et al. (2007). Impact of global climate change and emissions on regional ozone and fine particulate matter concentrations over the United States. *Journal of Geophysical Research*, *112*, D14312. doi:10.1029/2006JD008262.
- U.S. Environmental Protection Agency (2008a). Latest Findings on National Air Quality: Status and Trends through 2006. Office of Air Quality Planning and Standards, Air Quality Assessment Division. Publication No. EPA-454/R-07-007. http://www.epa.gov/air/airtrends/2007/report/trends_report_full.pdf. Accessed 10 October 2008.
- U.S. Environmental Protection Agency (2008b). Latest Findings on National Air Quality: Status and Trends through 2007. Office of Air Quality Planning and Standards, Air Quality Assessment Division. Publication No. EPA-454/R-08-006. <http://www.epa.gov/airtrends/2008/report/TrendsReportfull.pdf>. Accessed 20 March 2009.

- Vingarzan, R. (2004). A review of surface ozone background levels and trends. *Atmospheric Environment*, 38, 3431–3442.
- Wake, C., Carpenter, K., Cox, J., Souney, J., Sanborn, P., & Rodgers, M. (1998). Chapter 4: Air quality in New England. In *New England's Changing Climate, Weather and Air Quality*. Climate Change Research Center. <http://neci.sr.unh.edu/neccwaq.html>. Accessed 20 November 2008.
- Wang, H., Jacob, D. J., LeSager, P., Streets, D. G., Park, R. J., Gillibrand, A. B., et al. (2009). Surface ozone background in the United States: Canadian and Mexican pollution influences. *Atmospheric Environment*, 43, 1310–1319.
- World Health Organization (2006). WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: Global update 2005, summary of risk assessment. WHO/SDE/PHE/OEH/06.02.