Assessing Trends in Urban Ground-level Concentrations of Nitrogen Dioxide

By

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Abstract

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A review of the National Ambient Air Quality Standards (NAAQS) is required of the U.S. Environmental Protection Agency every five years. Included in the review typically is an assessment of health effects associated with air quality adjusted to meet the existing and potential alternative standards. In 2010, a 1-hour standard for nitrogen dioxide (NO_2) was established to protect the population from short-term exposure found to cause adverse health effects in sensitive populations. Ambient monitoring data collected over the last 30+ years indicate NO_2 concentrations have progressively lowered throughout all urban areas in the United States such that no areas are currently above the existing daily maximum 1-hour (DM1H) standard of 100 parts per billion. For the current and likely subsequent reviews of the NO_2 NAAQS, it is necessary to adjust monitored concentrations upward to meet the existing standard (or some alternative if greater than existing conditions) in a manner that estimates realistic concentrations across the entire distribution. A prior analysis of ambient air quality data strongly supported that concentrations could reasonably be adjusted proportionally across the entire distribution, though there was evidence that the relationship might be nonlinear in the lower and upper percentiles of the distribution. This study revisits the trends in the reduction of ambient NO_2 over time and compares two methods for adjusting NO_2 concentrations upward. The first method assumes a proportional relationship exists across the distribution of DM1H concentrations while a second method assumes the same proportional relationship up to and including the 98^{th} percentile DM1H and a nonlinear relationship above the 98^{th} percentile. While this study is not intended to definitively define the appropriateness of either adjustment (proportional vs. proportional with nonlinear above 98^{th} percentile DM1H) in representing the hypothetical high concentration air

quality scenario, there is evidence to suggest assuming proportionality across the entire distribution could result in a significant underestimation of concentrations at the upper percentiles of the distribution.

Biography

I am a native of North Carolina and a two-time graduate of North Carolina State University. I completed a B.A. degree in Sociology with an Anthropology concentration in 1987 and returned to earn a B.S. degree in Meteorology, completed in 1997. I have worked for a private consulting firm (through numerous corporate mergers and name changes) for the past 18 years as an air quality meteorologist and scientific programmer supporting Federal and State environmental agencies as well as private industry. While my career as a consultant has provided many opportunities to work on diverse projects over the years, my primary professional duties include air dispersion modeling and the ongoing maintenance of regulatory air dispersion environmental models, related data preprocessors, and the development of new modeling tools. With so much of my work focused on air quality and related issues, my participation in the Master of Environmental Assessment program at North Carolina State University has broadened my view and experience of environmental issues, the regulatory process to manage these issues, and their global impact on our world.

Acknowledgements

First, I would like to thank my wife and children for their continued patience and support of my educational pursuits over the years, as well as my extended family and friends for their constant encouragement, all of which has kept me motivated. I would also like to thank Ms. Linda Taylor for her advisement and helping me navigate may way through the program the past three years and Dr. Stephen Graham for his expert guidance and patience with me through the completion of this project. I am sincerely grateful for the education and experience I have gained through this process.

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INTRODUCTION

For nearly 40 years (1971 – 2010) the United States Environmental Protection Agency's (EPA's) primary (health-based) National Ambient Air Quality Standards (NAAQS) for nitrogen dioxide (NO_2) addressed only long-term exposure based on a limit on the arithmetic annual average, set at 53 parts per billion (ppb). The Clean Air Act (CAA) requires a review of the NAAQS at 5-year intervals for six criteria pollutants, one of which is oxides of nitrogen (NO_X), specified in terms of NO_2. The Integrated Science Assessment (ISA) performed during the last completed review of the NO_2 NAAQS concluded that there is an association with short-term exposure to NO_2 and adverse health effects, in particular, increased airway responsiveness (AR) (EPA, 2008a). The ISA findings were based on results from controlled human exposure studies and epidemiologic studies that evaluated hospital emergency department visits for respiratory symptoms, particularly most often experienced by children and asthmatics. In addition, there have been numerous field studies, some of which were conducted in areas where the entire annual distribution of ambient 24-hour average concentrations of NO_2 was below the annual standard of 53 ppb (EPA, 2008a). This indicates that the then-existing annual standard alone did not protect against the observed effects associated with short-term exposure to NO_2.

In 2010, as a result of the findings of the NAAQS review, the EPA retained the existing annual standard and added a new 1-hour standard to protect against acute effects from short-term exposure (FR, 2010). The new 1-hour standard was set at a level of 100 ppb, a probabilistic standard based on a 3-year average of the 98th percentile values of a complete years' distribution of daily maximum 1-hour (DM1H) NO_2 concentrations. In addition, the EPA updated the NO_2 monitoring requirements to increase the number of monitors throughout the U.S., focusing on both near-road and area-wide NO_2 concentrations to better represent ambient air quality in

locations having high concentrations concomitant with potentially susceptible and vulnerable populations. As many as 125 monitors would be added to the existing NO_2 monitoring network as a result of this change. The new monitors were originally required to be operational by January 1, 2013 (FR, 2010); however, that requirement has since been revised. The updated schedule is based on a phased approach which now requires certain monitors to be operational by January 1, 2015; January 1, 2016; and January 1, 2017 (US FR, 2013).

Central to all NAAQS reviews is determining the adequacy of the existing standard; that is, whether or not the standard provides requisite health protection to the public. To support conclusions regarding this determination during the last completed review, EPA performed an assessment of the health risks associated with ambient concentrations occurring at the standard level, an annual average of 53 ppb (EPA, 2008c). As all areas across the United States were well below the then-existing annual standard, ambient concentrations were adjusted upward to simulate concentrations that would just meet an annual average concentration of 53 ppb. One key issue from that review was the proportional approach used to adjust concentrations, an approach that assumed proportionality across the entire distribution of ambient concentrations. Changes observed for a given concentration metric (e.g., median, minimum, maximum) were assumed constant across the entire distribution of hourly values, such that quantitative factors used to adjust air quality to just meet the then-existing annual standard were appropriate to use in adjusting any hourly ambient concentration (EPA, 2008c). Support for the approach included a linear regression analysis of ambient monitoring data in several urban areas that demonstrated a mostly linear relationship in 1-hour daily maximum values when comparing an historical high concentration year with a more recent low concentration year (EPA, 2008b). There were, however, instances of non-linearity, including the presence of statistically significant linear

regression intercepts and curvature across the upper and lower percentiles of the concentration distribution.

This current study further assessed trends in ambient concentrations of NO_2 in urban areas throughout the U.S. using a similar approach to that used for the previous NAAQs review. In particular, this study evaluated the potential impact of nonlinearities at the upper percentiles of the DM1H distributions of ambient ground-level concentrations of NO_2 comparing two different methods to adjust concentrations upward to just meet the existing DM1H standard of 100 ppb. The focus on these upper percentile concentrations is important in that controlled human exposure studies indicated increases in airway responsiveness following exposures to NO_2 concentrations of 100 to 300 ppb (or higher) for 30 minutes to 2 hours (EPA, 2008c). These selected concentrations serve as a comparative indicator of concentrations of potential concern, often referred to previously as health effect benchmarks (EPA, 2008c). It is the goal that insights gained from this study can inform subsequent NAAQS reviews when performing risk and exposure analyses to determine the adequacy of existing or alternative standards and serve as a baseline study for comparison when data become available from new monitoring sites. With new monitors sited where short-term exposure is assumed to be higher, trends in ambient concentrations may differ from what has been observed with the prior ambient monitoring network.

METHODS

This study extends the prior analysis conducted during the last completed review of the NO_2 NAAQS (EPA, 2008b). In that assessment, EPA evaluated the historical trends in the distribution of hourly ambient concentrations of NO_2 in six cities across the United States including: Los Angeles, Atlanta, New York, Philadelphia, Denver, and Chicago. In each city,

hourly ambient NO_2 data for equivalent monitors within a single Core Based Statistical Area (CBSA) were evaluated by comparing an historical high concentration year, prior to 1986, to a recent low concentration year, for which 2007 was used, for all six cities. To examine the trend in the distribution of 1-hour concentrations of NO_2 over the 20+ year period analyzed, for each monitoring site, high and low concentration years were plotted and linear regressions were performed using the 0-100 percentiles of the DM1H concentration distribution. The regression results generally supported a linear relationship in the decrease over time in ground-level ambient concentrations of NO_2 across the mid-range of percentiles in the distributions. On occasions, particularly evident at the lower and upper percentiles, however, the plots exhibited a more pronounced curvature, indicative of a nonlinear relationship.

For the current study, two analyses were performed to further our understanding of trends in NO_2 concentrations and evaluate the potential impact of nonlinearities when adjusting ambient concentrations to meet the existing or alternative standards. The first analysis repeated the linear regression completed by EPA (2008b) to reassess the trends in NO_2 across the distribution over time, using data more recent than 2007 for the low concentration year, where available. The second analysis compared two different methods to adjust ambient 1-hour concentrations upward to just meet the 1-hour NO_2 NAAQS of 100 ppb. The first method of adjustment assumed proportionality across the entire distribution of concentrations while the second method assumed proportionality up to and including the DM1H 98^{th} percentile, and nonlinearity above the DM1H 98^{th} percentile. The number of days per year were then counted in which the DM1H concentration equals or exceeds a concentration of 100 ppb, prior to and after the application of each adjustment method. The current study was completed in four steps or phases: 1) data acquisition and completeness review; 2) linear regression analysis of high versus low

concentration years; 3) concentration adjustments upward to just meet the existing DM1H standard of 100 ppb; and 4) counts of the number of days where the DM1H concentration exceeds 100 ppb. The remainder of this section provides a detailed discussion of the methods used to complete each of the phases of the study.

Monitoring Sites

To leverage off of the prior work performed by EPA (2008b), this study focused on the same six urban areas and analyzed hourly ambient concentrations of NO_2 collected at the same monitoring sites. Table 1 lists the monitoring sites and locations that were included in the former analysis and in this study. These sites share a long history of monitoring ambient NO_2 levels which makes it possible to analyze trends over multiple decades.

Data Acquisition and Completeness Review

All ambient NO_2 data used in this study were obtained as yearly files of 1-hour averages, downloaded in plain text format via the internet from the EPA's Air Quality System (AQS) (EPA, 2014a), accessed on November 1, 2014. Each data file was evaluated for completeness. A Fortran program was developed to read each file and determine the completeness of the data using the following criteria: 1) at least 75% of the hourly values in a day must be valid for an individual day to be complete; 2) 75% of the days in a quarter year must be complete for the quarter to be complete; and 3) all four quarters in the year must be complete. Each data file represented a calendar year (January - December), and quarter years were defined as: Quarter 1 = January-March; Quarter 2 = April-June; Quarter 3 = July-September; and Quarter 4 = October-December. In general, only complete years of data were used for this study; though, there are a couple of exceptions discussed later in this report.

When possible, the regression analysis was repeated for each monitor site using the same high concentration year used in the EPA (2008b) analysis. The one exception is Denver, for which 1977 was used originally. At the time the data were obtained for this study, data were only available back to 1980 via the AQS website. The regression for Denver was repeated using the first year that complete data were available for both monitors analyzed within the CBSA.

The selection of the low concentration year for the sites within a CBSA was based on the most recent year in which ambient data were available and complete for all monitors analyzed within a CBSA. When data more recent than 2007 were not available for all monitors analyzed within the CBSA or more recent data were incomplete for any one monitor within the CBSA, 2007 was used as the low concentration year for all monitors within the CBSA.

For all but the Denver sites, the data used in the prior analysis for the high concentration year were downloaded. Data from 1980 through 1985 were obtained for Denver, from which a high concentration year was selected based on data completeness. Table 2 lists, by monitoring site, the data downloaded as the high concentration year for use in the regression analysis and the results of the completeness determination for each dataset. Note that multiple years were downloaded for Denver. Data from 2000 through 2013 were also downloaded for all sites and evaluated for completeness. Table 3 contains the results of the completeness determination for years 2000 through 2013. In both Table 2 and Table 3, "Y" indicates the data are complete for the site and year specified, and "N" indicates the data are not complete. An empty cell in Table 2 means the data were not required and no attempt was made to acquire the data. A dash or hyphen ("-") in Table 3 means the monitor was not active for the year and the data were not available.

Table 1. Monitoring Sites Analyzed by Study City and Core Based Statistical Area.

			Physical Location			
AQS ID	**Study City**	**CBSA**	**State**	**County**	**Latitude** *Degrees*	**Longitude** *Degrees*
131210048	Atlanta	Atlanta-Sandy Springs-Marietta, GA	GA	Fulton	33.779330	-84.395760
170314002	Chicago	Chicago-Naperville-Joliet, IL-IN-WI	IL	Cook	41.855243	-87.752470
080013001	Denver	Denver-Aurora-Boulder, CO	CO	Adams	39.838119	-104.949840
080310002	Denver	Denver-Aurora-Boulder, CO	CO	Denver	39.751184	-104.987625
060370002	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.136500	-117.923910
060370113	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.051110	-118.456360
060371002	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.176050	-118.317120
060371103	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.066590	-118.226880
060371201	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.199250	-118.532760
060371301	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	33.928990	-118.210710
060371701	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.067030	-117.751400
060372005	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	34.132600	-118.127200
060374002	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Los Angeles	33.823760	-118.189210
060595001	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Orange	33.925130	-117.952640
060655001	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Riverside	33.852750	-116.541010
060658001	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	Riverside	33.999580	-117.416010
060710001	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	San Bernardino	34.895007	-117.024484
060712002	Los Angeles	Los Angeles-Long Beach-Santa Ana, CA	CA	San Bernardino	34.100020	-117.492010
340131003	New York	New York-Northern New Jersey-Long Island, NY-NJ-PA	NJ	Essex	40.757501	-74.200500
340170006	New York	New York-Northern New Jersey-Long Island, NY-NJ-PA	NJ	Hudson	40.670250	-74.126081
340273001	New York	New York-Northern New Jersey-Long Island, NY-NJ-PA	NJ	Morris	40.787628	-74.676301
340390004	New York	New York-Northern New Jersey-Long Island, NY-NJ-PA	NJ	Union	40.641440	-74.208365
340070003	Philadelphia	Philadelphia-Camden-Vineland, PA-NJ-DE-MD	NJ	Camden	39.923042	-75.097617
420170012	Philadelphia	Philadelphia-Camden-Vineland, PA-NJ-DE-MD	PA	Bucks	40.107222	-74.882222
420450002	Philadelphia	Philadelphia-Camden-Vineland, PA-NJ-DE-MD	PA	Delaware	39.835556	-75.372500
420910013	Philadelphia	Philadelphia-Camden-Vineland, PA-NJ-DE-MD	PA	Montgomery	40.112222	-75.309167
421010004	Philadelphia	Philadelphia-Camden-Vineland, PA-NJ-DE-MD	PA	Philadelphia	40.008889	-75.097780
421010047	Philadelphia	Philadelphia-Camden-Vineland, PA-NJ-DE-MD	PA	Philadelphia	39.944651	-75.165206

Table 2. Completeness Determination for High Concentration Years of 1-hour NO_2.

AQS ID	Area	1980	1981	1982	1983	1984	1985
131210048	Atlanta						Y
170314002	Chicago				Y		
080013001	Denver	N	Y	N	Y	N	Y
080310002	Denver	Y	Y	Y	Y	Y	N
060370002	Los Angeles						Y
060370113	Los Angeles						Y
060371002	Los Angeles						Y
060371103	Los Angeles						Y
060371201	Los Angeles						Y
060371301	Los Angeles						Y
060371701	Los Angeles						Y
060372005	Los Angeles						Y
060374002	Los Angeles						Y
060595001	Los Angeles						Y
060655001	Los Angeles						Y
060658001	Los Angeles						Y
060710001	Los Angeles						N
060712002	Los Angeles						Y
340131003	New York					Y	
340170006	New York					Y	
340273001	New York					Y	
340390004	New York					Y	
340070003	Philadelphia					Y	
420170012	Philadelphia					Y	
420450002	Philadelphia					N	
420910013	Philadelphia					Y	
421010004	Philadelphia					Y	
421010047	Philadelphia					Y	
"Y" = Complete; "N" = Incomplete							

Table 3. Completeness Determination of 1-hour NO_2 Data (2000-2013).

AQS ID	Area	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013
131210048	Atlanta	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	-	-	-	-
170314002	Chicago	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	Y
080013001	Denver	Y	N	N	N	Y	Y	Y	Y	Y	N	N	Y	Y	Y
080310002	Denver	N	Y	Y	N	Y	Y	N	Y	N	N	Y	N	Y	Y
060370002	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	Y	N	Y	Y
060370113	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	N
060371002	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	N	N	N	N
060371103	Los Angeles	Y	Y	Y	Y	N	Y	Y	Y	N	N	Y	Y	N	N
060371201	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	N
060371301	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	N	-	-	-	-	-
060371701	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
060372005	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	Y	N	N
060374002	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	Y	N	N	N
060595001	Los Angeles	N	Y	N	Y	Y	Y	Y	Y	Y	Y	N	N	N	N
060655001	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
060658001	Los Angeles	Y	Y	N	Y	Y	Y	Y	Y	Y	Y	N	N	N	N
060710001	Los Angeles	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	N
060712002	Los Angeles	Y	Y	N	Y	N	Y	Y	Y	Y	Y	Y	Y	Y	Y
340131003	New York	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
340170006	New York	Y	Y	Y	Y	Y	Y	Y	N	N	Y	Y	Y	N	N
340273001	New York	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
340390004	New York	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
340070003	Philadelphia	Y	Y	Y	N	Y	N	Y	Y	N	-	-	-	-	-
420170012	Philadelphia	Y	Y	Y	Y	Y	Y	Y	Y	N	Y	Y	Y	Y	Y
420450002	Philadelphia	Y	Y	Y	Y	Y	N	N	Y	Y	Y	Y	Y	N	y
420910013	Philadelphia	Y	Y	Y	Y	Y	Y	Y	Y	Y	-	-	-	-	-
421010004	Philadelphia	Y	Y	Y	N	Y	N	Y	Y	Y	Y	Y	Y	Y	Y
421010047	Philadelphia	N	Y	Y	N	Y	Y	N	Y	Y	Y	Y	Y	Y	Y
"Y" = Complete; "N" = Incomplete; "-" = Inactive															

Linear Regression Analysis

A linear regression was performed for each monitoring site pairing the high concentration year (prior to 1986) with the low concentration year (2007 or later). As noted previously, for all but the Denver area, the same high concentration year used in the prior study (EPA, 2008b) was used for this study. Based on the results of the data completeness evaluation shown in Table 2, 1981 was selected as the high concentration year for Denver since it is the earliest year during which the NO_2 data for both monitors is complete. The most recent year in which ambient data are available and complete for all monitors analyzed within a CBSA was used as the low concentration year (2007 or later). Table 4 identifies the high and low concentration years, by city, used for the regression analysis. Note in Table 2 that both Los Angeles and Philadelphia each have one monitor where the high concentration year was determined to be incomplete. The data for each of these monitors were found to be just under 75% complete for a single quarter while all other quarters were found to be complete. For this reason, and for consistency with EPA (2008b) analysis, it was decided to retain these data as the high concentration year for each of these cities. It is possible that since the original analysis, performed in 2008, the data have been updated which could be the reason for the difference in the completeness determination between then and now.

Table 4. 1-hour Ambient NO_2 High and Low Concentration Years.

Area City	High Concentration Year	Low Concentration Year
Atlanta	1985	2008
Chicago	1983	2013
Denver	1981	2013
Los Angels	1985	2007
New York	1984	2011
Philadelphia	1984	2007

The linear regression was performed using the R programming language for statistical computing and graphics (R, 2015). For each monitor site, the high and low concentration years were read into R, and the maximum daily 1-hour concentration was identified for each day of the year, for each dataset. Next, the 0^{th} through 100^{th} percentile values of the maximum daily 1-hour concentrations for every 1% increment were computed for the high concentration and low concentration datasets. The concentration for each paired percentile across the distributions were plotted, one against the other. R was then used to fit a line to the plotted points for which the equation of the line and the coefficient of determination (r-squared or r^2) were calculated. r^2, a value between 0.00-1.00, is a measure of the fit of the data to the line. An r^2 value of 0.95 and above is generally interpreted as a good fit of the data to the line.

Adjustment of Concentrations to Just Meet the Existing 1-hour Standard

The 1-hour ambient concentrations of NO_2 were adjusted upward using two different methods applied to the 2011 through 2013 hourly NO_2 concentrations for those monitoring sites determined to be complete (see Table 3). 2011 through 2013 were chosen because they were the most recent three consecutive years for which a 1-hour design value had been computed at the time the analysis was performed. Recall that the existing 1-hour NO_2 primary NAAQS is a probabilistic standard based on a 3-year average of the annual 98^{th} percentile of the DM1H values. The first method (Method 1) assumes a proportional decrease in concentrations from the high concentration year to the low concentration year across the entire distribution of monitored values at each site within each CBSA or city. The second method (Method 2) assumes a proportional decrease in concentrations up to the 98^{th} percentile and a nonlinear decrease beyond the 98^{th} percentile.

Method 1 applied a single factor to each DM1H concentration for all monitors analyzed within a city. The factor for each city was computed based on the highest 1-hour NO_2 design value among the group of sites within the city that were included in this study. The design value is the computed concentration based on the form of the standard. Design values for the 2011-2013 period were previously calculated for each monitor site and published by the EPA (EPA, 2014b). The Method 1 adjustment factor for each city was calculated as the quotient of the area design value divided by 100 ppb, which is the target concentration.

To address the nonlinearity in the decrease in concentrations at the upper end of the distribution, Method 2 started with the Method 1 resultant dataset and further adjusted the DM1H concentrations that are above the 98th percentile using a set of unique site-specific factors. The 98th percentile of the DM1H concentrations for a complete year of data is approximately the 8th highest maximum daily value based on 365 days in a year. The remainder of the distribution above the 98th percentile is made up of the seven DM1H values, here referred to as the H1H, H2H,…, and H7H (read as the high first high, high second high,…, and high seventh high). For Method 2, a separate adjustment factor was derived for each of the seven maximum 1-hour concentrations above the 98th percentile, and the set of seven factors were derived specific to each monitoring site. The seven factors were each computed from the unadjusted 1-hour concentrations of yearly data from 2004 through 2014. Each of the seven factors represent the average of the ratio of the yearly high value of the maximum daily 1-hour concentrations (e.g., H3H) divided by the 98th percentile of maximum daily 1-hour concentrations for the year, computed each year for the set of complete years of data from 2004 through 2013. In other words, the factor used to further adjust the H3H maximum daily 1-hour value at a given site after the Method 1 factor was applied, is the mean value computed from the

set of ratios of the H3H divided by the 98th percentile, calculated for each year from 2004 through 2013. The years 2004 through 2013 were selected as the set of years on which to base the Method 2 factors in order to reduce any bias that might exist prior to 2004 due to the implementation of the NO_X State Implementation Plan (SIP) Call which required 22 states and the District of Columbia to address the regional transport of ozone (RTO) through their SIPs. This rule was expected to lead to greater reductions of ambient NO_2 at around 2004 and beyond (EPA, 2015).

Exceedances of Potential Health Effect Benchmark Levels

A count of the number of days in which the 1-hour average ambient concentration of NO_2 exceeds 100 ppb prior to and after applying Methods 1 and 2 was determined at each monitoring site. The counts were determined separately for each year from 2011 through 2013 by counting the number days the maximum daily 1-hour concentration exceeds 100 ppb.

RESULTS

Analysis of High Concentration Years to Low Concentration Years

Figure 1 through Figure 7 illustrate the patterns in the decrease in 1-hour ambient concentrations of NO_2 over the last several decades in the six cities analyzed. These figures present the 1-hour percentile concentrations for the high year plotted against the low year for each monitoring site. Percentile concentrations were calculated across the entire distribution of maximum daily 1-hour concentrations at single percent increments from 0-100%. Each plot includes the best fit line, the equation of the line to show the slope and y-intercept, and the r^2 value which is a measure of the fit of the line to the plotted points.

The figures confirm that at every site analyzed there is a decrease in ambient 1-hour NO_2 concentrations across the distribution over time. The earlier year, termed as the high concentration year, does in fact represent higher concentrations across the distribution relative to the more recent year, termed as the low concentration year. For 16 of the 28 sites analyzed, r^2 is greater than or equal to 0.95. Recall, an r^2 value greater than or equal to 0.95 is generally considered a good fit which suggests a strongly linear relationship across the distribution.

For all sites, the slope is a statistically significant positive value less than 1.0, which indicates the magnitude of the linear reduction across the distribution of percentile concentrations from the high year to the low year. In addition, the y-intercept is positive for all but four monitoring sites. The y-intercept is a constant offset that is either added (positive) or subtracted (negative) to the linearly reduced percentile concentrations. Even though the y-intercept represents a constant value, the effect of the offset is greater at the lower end of the distribution, where percentile concentrations are lower, relative to the upper end where the concentrations are higher. That said, even if there is a strong linear relationship, the presence of

a significant intercept indicates the relationship is not entirely proportional. Note also that for those sites with a positive y-intercept, this translates to smaller reductions in the concentrations at the lower end of the distribution and larger reductions at the upper end where the offset represents a smaller percentage of the total concentration.

A common pattern seen in Figure 1 through Figure 7 is pronounced curvature through the upper and lower ends of the distribution below the fitted line and flattening through the middle portion of the distribution above the line. Points below the line indicate a greater reduction in concentrations while points above the line indicate reductions to a lesser extent. Here again the figures illustrate a greater reduction at the upper end of the distribution than through the middle of the distribution.

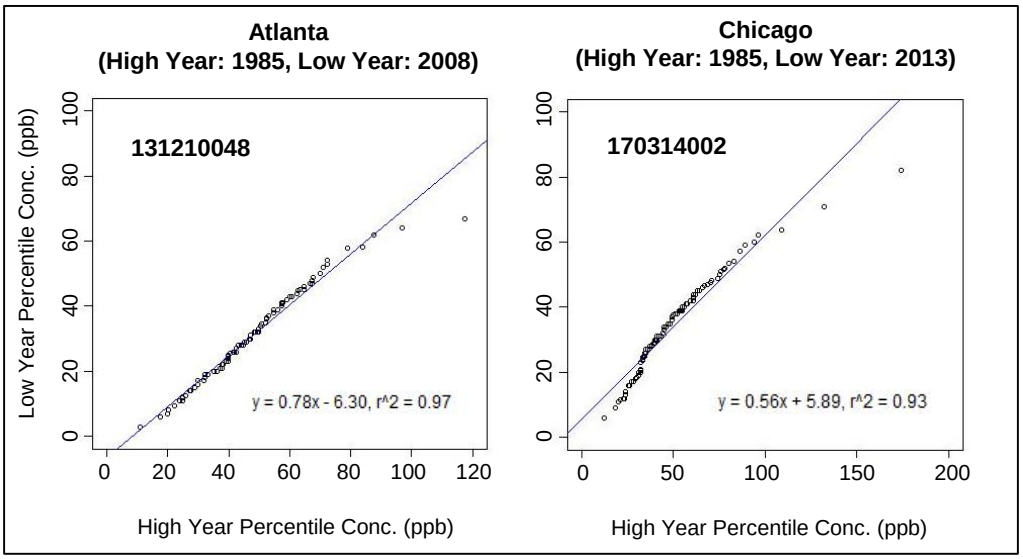

Figure 1. Comparison of High to Low Year Concentration Distribution, Atlanta and Chicago.

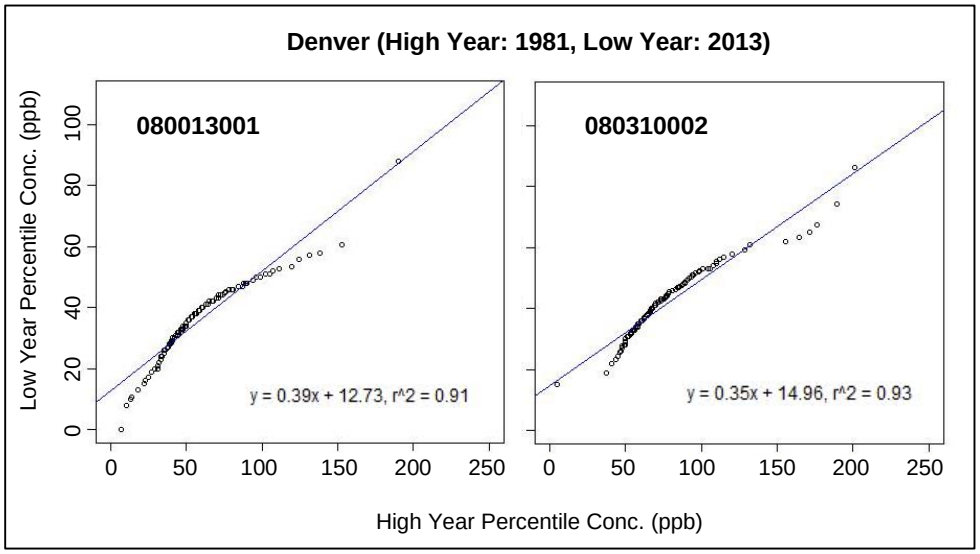

Figure 3. Comparison of High to Low Concentration Distributions, Denver.

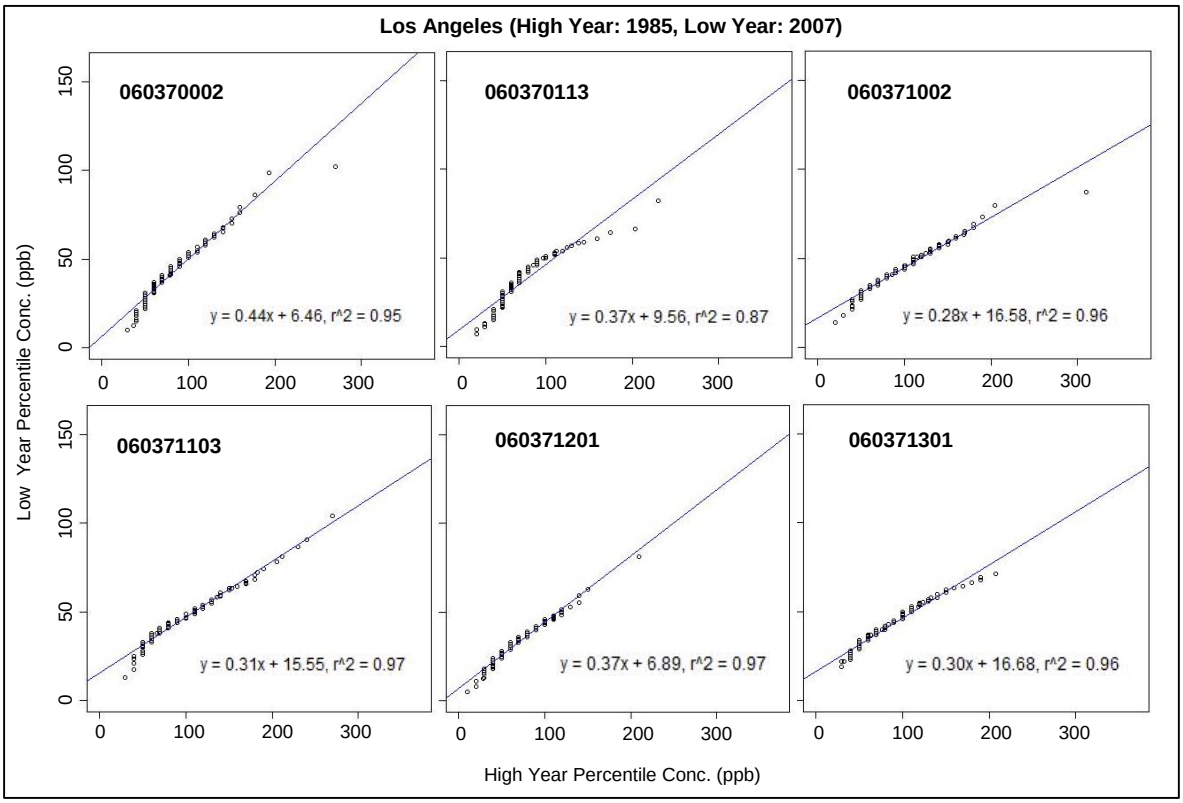

Figure 2. Comparison of High to Low Concentration Distributions, Los Angeles.

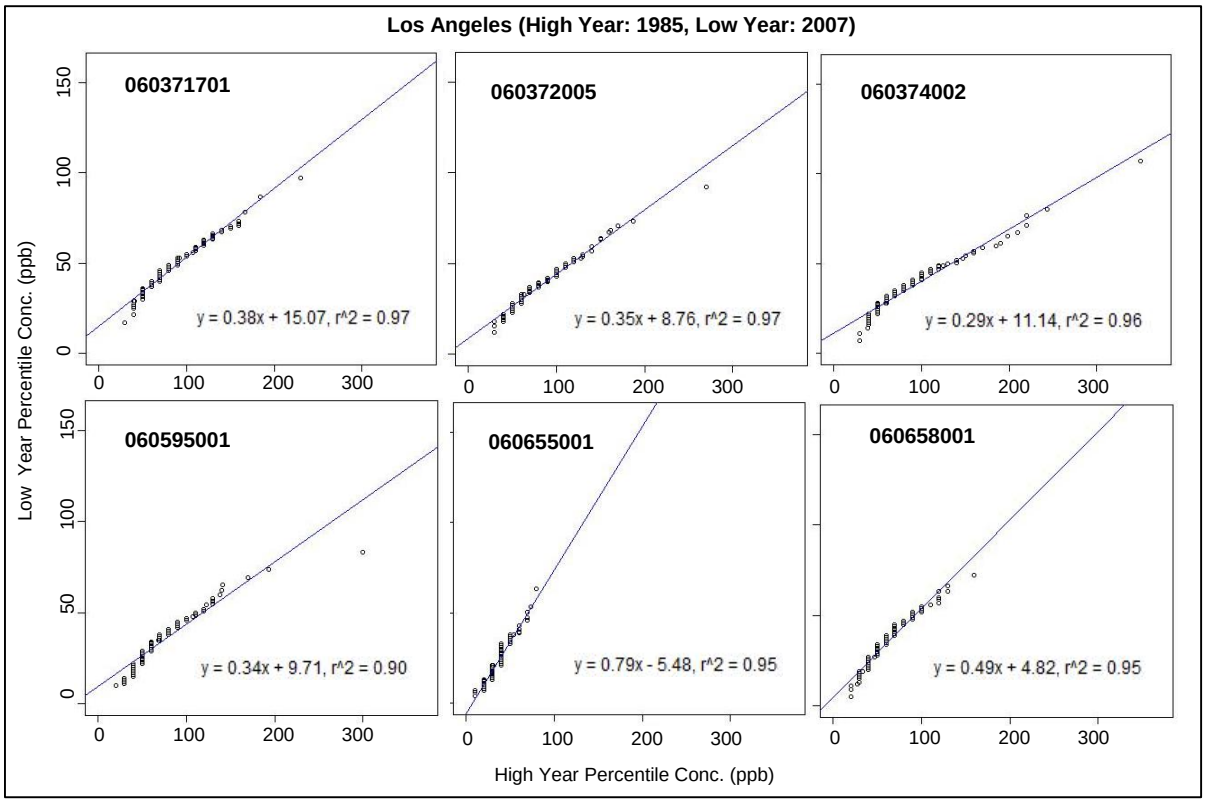

Figure 5. Comparison of High to Low Concentration Distributions, Los Angeles (cont.).

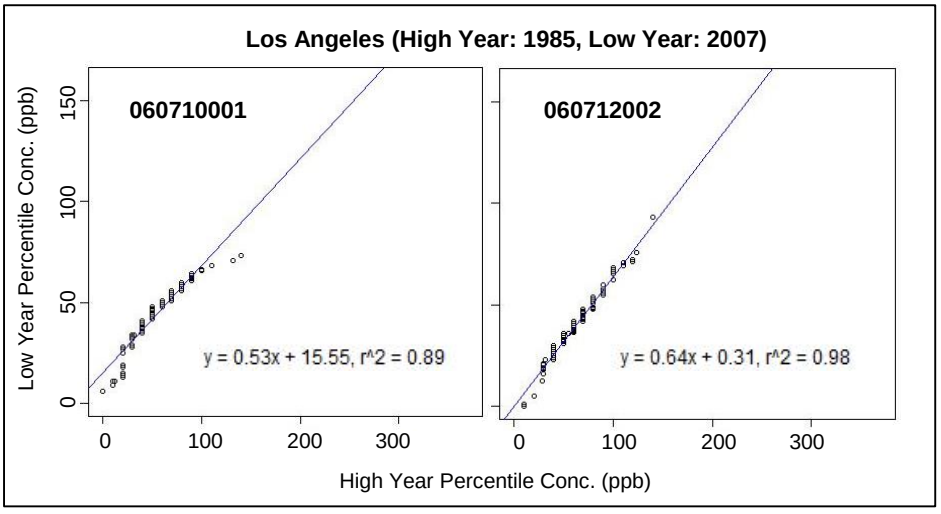

Figure 4. Comparison of High to Low Concentration Distributions, Los Angeles (cont.).

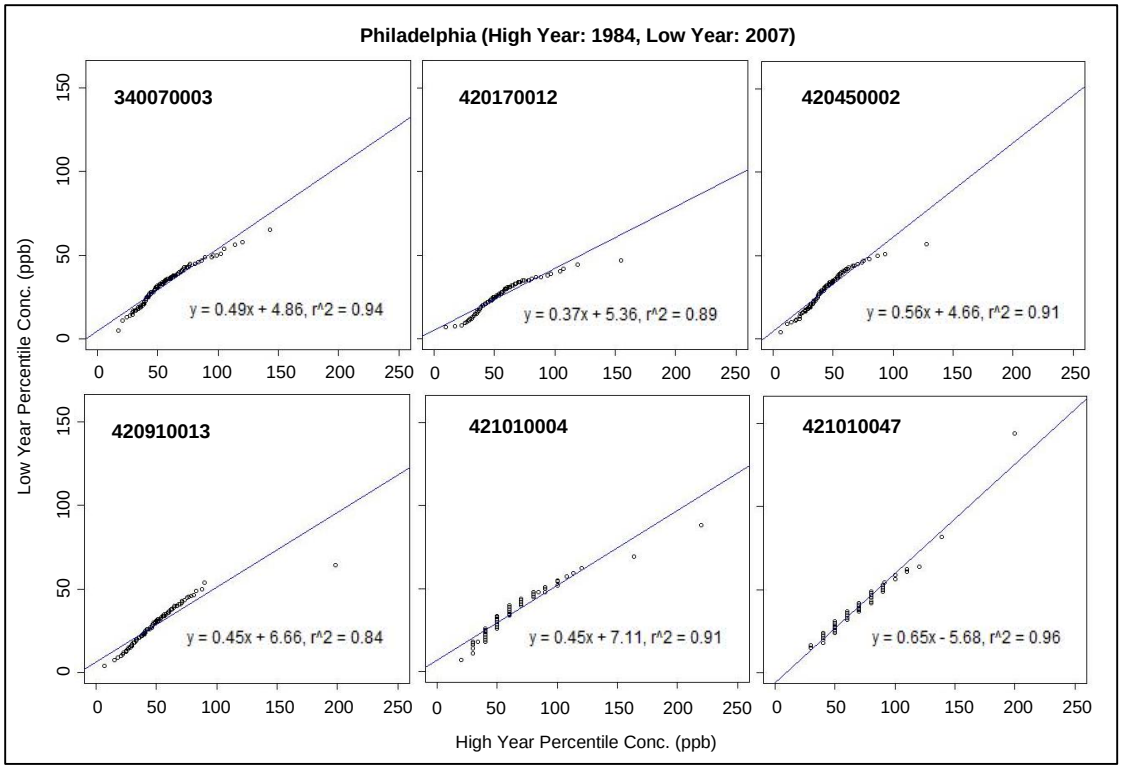

Figure 7. Comparison of High to Low Concentration Distributions, Philadelphia.

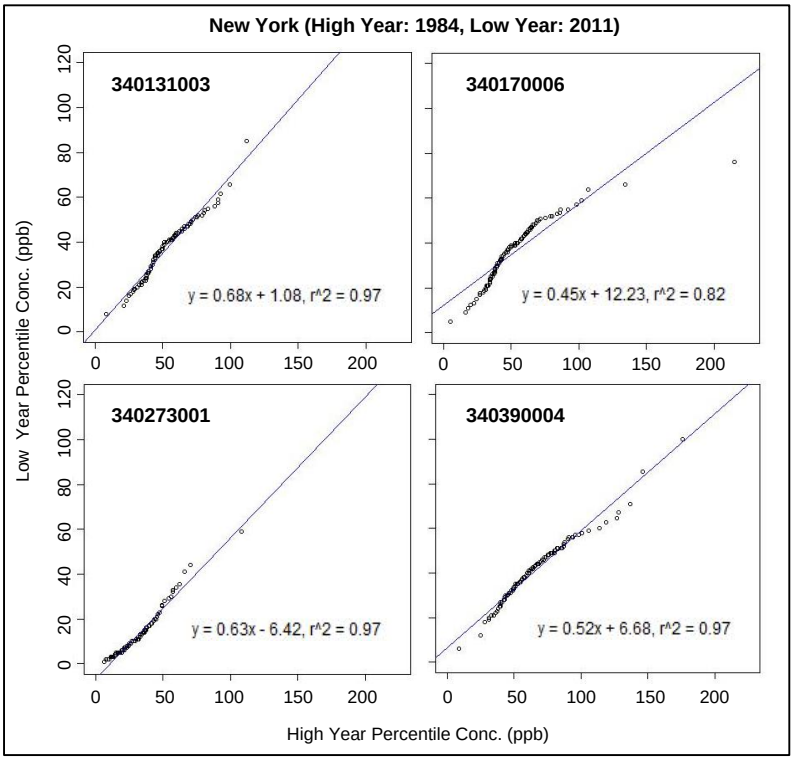

Figure 6. Comparison of High to Low Concentration Distributions, New York.

Adjustment of Concentrations to Just Meet the Existing 1-hour Standard

Recall that Method 1 assumes a proportional relationship in the decrease of NO_2 over time, and the DM1H are adjusted upward using a single area-wide adjustment factor across the entire distribution and for all monitors in a CBSA. Method 2 assumes a proportional adjustment up to and including the DM1H 98th percentile along with applying a non-proportional adjustment above the 98th percentile. The seven DM1H concentrations, which represent the distribution above the 98th percentile, were further adjusted after applying Method 1, based on a set of individual site-specific factors. The adjustment factors derived for both methods are listed in Table 5. Also included in Table 5 are the area-wide design values used to compute the Method 1 adjustment factors. The yellow highlights identify the site on which the area-wide Method 1 factor is based, the corresponding design value, and the resultant factor. Note that Atlanta and Chicago are omitted from Table 5. The Atlanta monitor has been inactive since 2010, and the 2012 data from the Chicago monitor was not complete. For both of these cities, only a single monitor was included in this study, and there were no other equivalent monitors for either city. Without complete data for each of the three years (2011-2013) a design value could not be computed. Thus, the concentrations could not be adjusted for Atlanta or Chicago. In all, concentrations were adjusted using Method 1 and Method 2 for 43 datasets, from 2011-2013, across four urban areas.

Table 5. Method 1 and Method 2 Concentration Adjustment Factors

AQS ID	Area City	Design Value	Method 1 Adj Factors	Method 2 Upper Distribution Adjustment Factors						
				H1H	H2H	H3H	H4H	H5H	H6H	H7H
080013001	Denver	62	1.61	1.32	1.22	1.17	1.13	1.09	1.05	1.03
080310002	Denver		1.61	1.39	1.17	1.15	1.12	1.07	1.04	1.03
060370002	Los Angeles		1.56	1.26	1.17	1.15	1.1	1.08	1.05	1.02
060370113	Los Angeles		1.56	1.27	1.16	1.13	1.1	1.09	1.04	1.02
060371103	Los Angeles		1.56	1.41	1.24	1.17	1.12	1.06	1.04	1.02
060371201	Los Angeles		1.56	1.32	1.19	1.12	1.06	1.04	1.03	1.01
060371701	Los Angeles	64	1.56	1.25	1.16	1.11	1.09	1.05	1.04	1.02
060372005	Los Angeles		1.56	1.43	1.27	1.16	1.12	1.1	1.07	1.05
060655001	Los Angeles		1.56	1.27	1.17	1.08	1.06	1.04	1.02	1.01
060710001	Los Angeles		1.56	1.21	1.14	1.09	1.08	1.05	1.04	1.02
060712002	Los Angeles		1.56	1.34	1.19	1.12	1.07	1.05	1.03	1.02
340131003	New York		1.49	1.39	1.26	1.19	1.12	1.09	1.04	1.02
340170006	New York		1.49	1.52	1.25	1.2	1.14	1.08	1.04	1.02
340273001	New York		1.49	1.44	1.24	1.16	1.13	1.08	1.05	1.02
340390004	New York	67	1.49	1.44	1.29	1.18	1.11	1.07	1.04	1.02
420170012	Philadelphia		1.64	1.25	1.15	1.08	1.06	1.04	1.04	1.01
420450002	Philadelphia		1.64	1.55	1.34	1.16	1.1	1.05	1.04	1.02
421010004	Philadelphia	61	1.64	1.39	1.22	1.15	1.09	1.06	1.04	1.02
421010047	Philadelphia		1.64	1.55	1.36	1.2	1.15	1.1	1.05	1.02

Figure 8 through Figure 23 illustrate the DM1H NO_2 percentile concentrations prior to and after the Method 1 and Method 2 adjustments were applied to all available and complete datasets from 2011 through 2013. As the focus of this study is primarily on the differences in the middle and upper end of the distribution, shown are the percentile concentrations for the 80th percentile and above. The figures show the proportional linear increase based on Method 1 and the additional nonlinear increase in the upper distribution above the 98th percentile using the site-specific derived Method 2 factors. At any given site, the Method 1 and Method 2 resultant percentile values are identical at and below the 98th percentile down the entire distribution. As the figures illustrate, the Method 2 factors result in a noticeable increase in the percentile concentrations over and above the Method 1 factors.

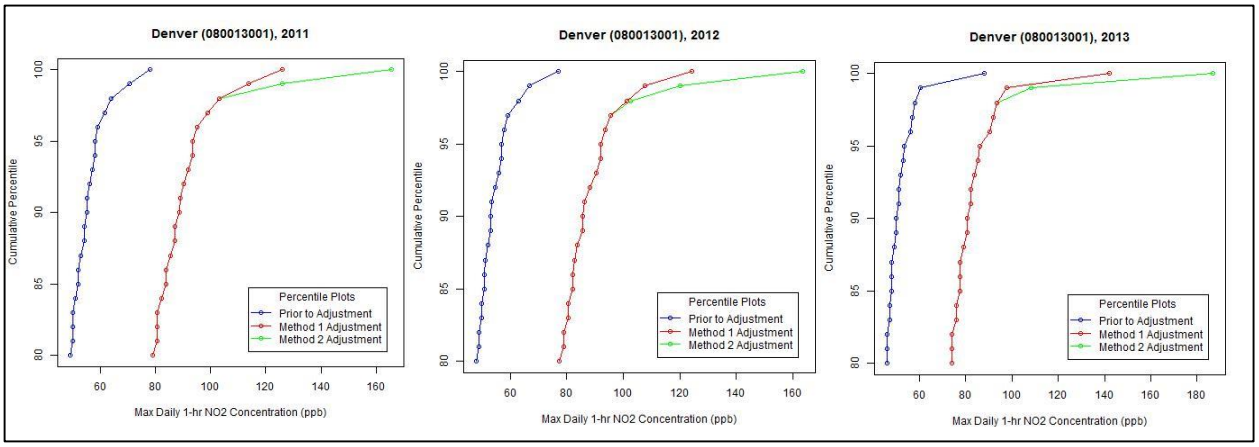

Figure 8. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Denver (080013001; 2011-2013).

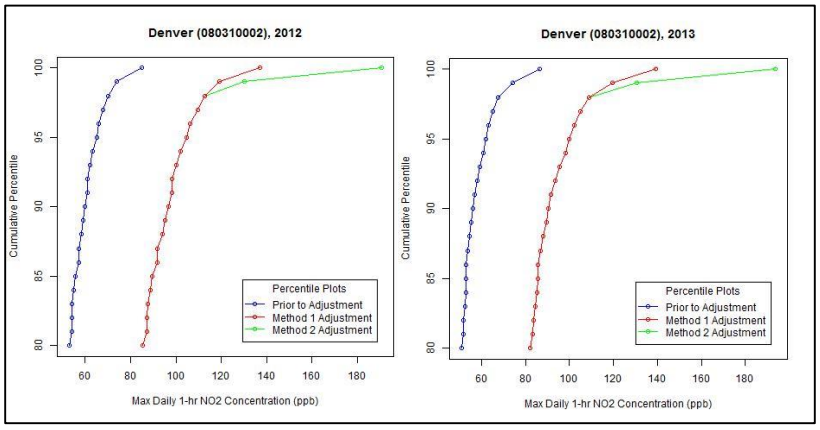

Figure 9. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Denver (080010002; 2012, 2013).

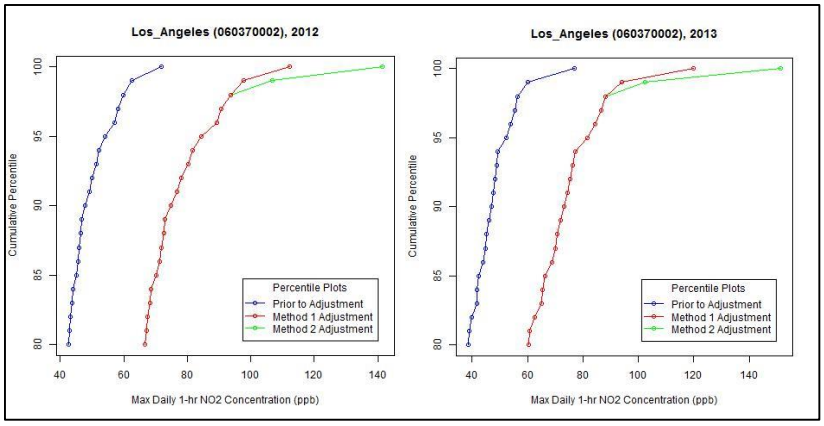

Figure 10. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060370002; 2012, 2013).

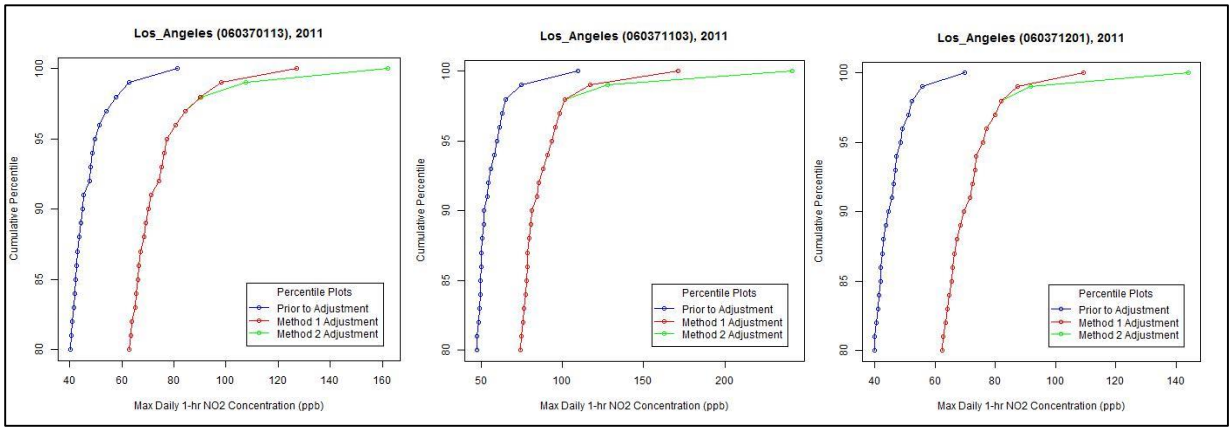

Figure 11. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060370113, 060371103, and 060371201; 2011).

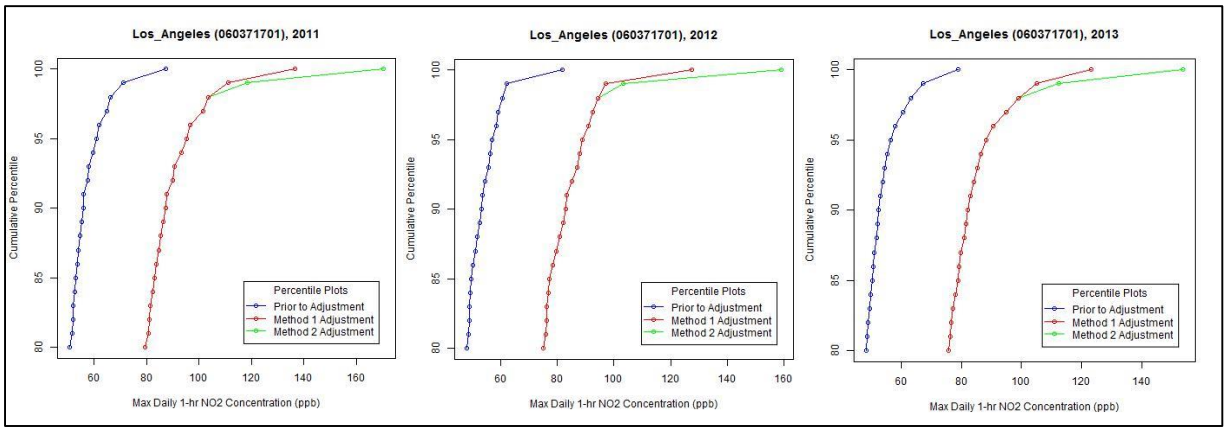

Figure 12. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060371701; 2011-2013).

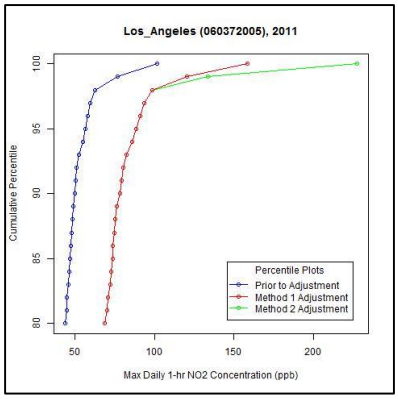

Figure 13. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060372005; 2011).

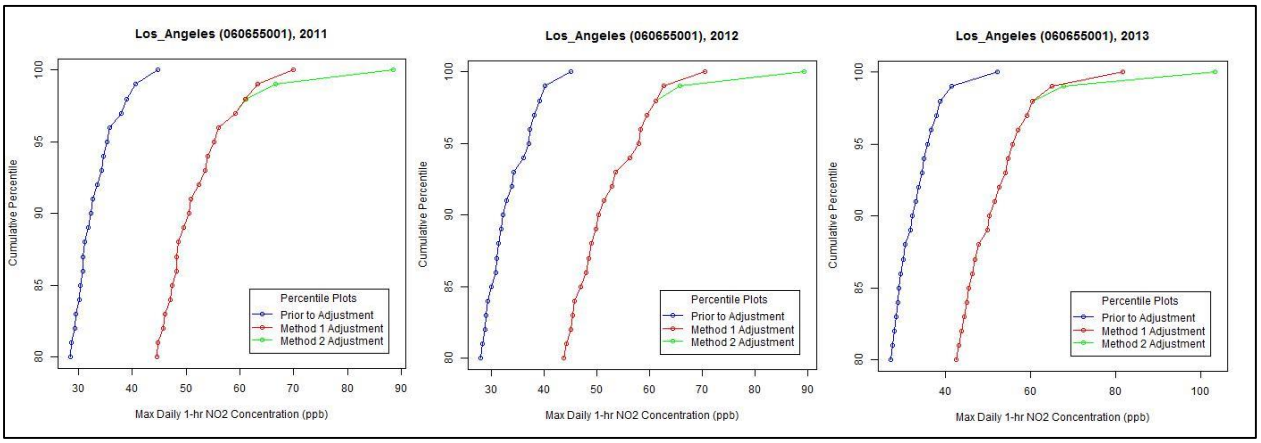

Figure 14. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060655001; 2011-2013).

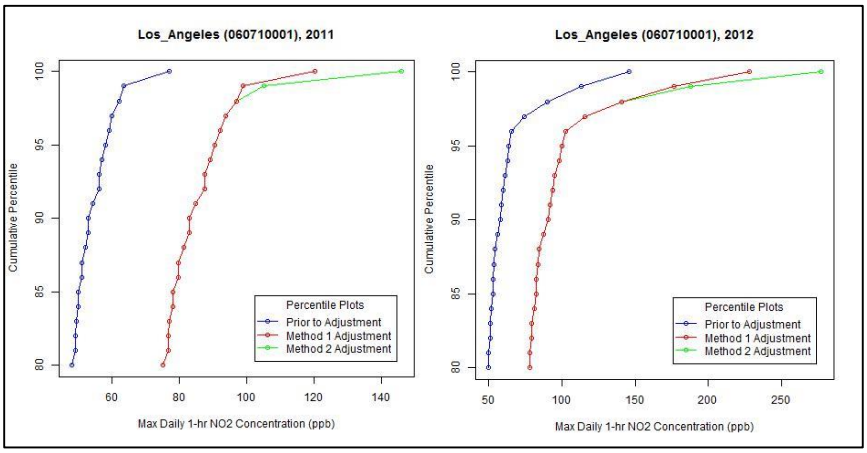

Figure 15. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060710001; 2011-2012).

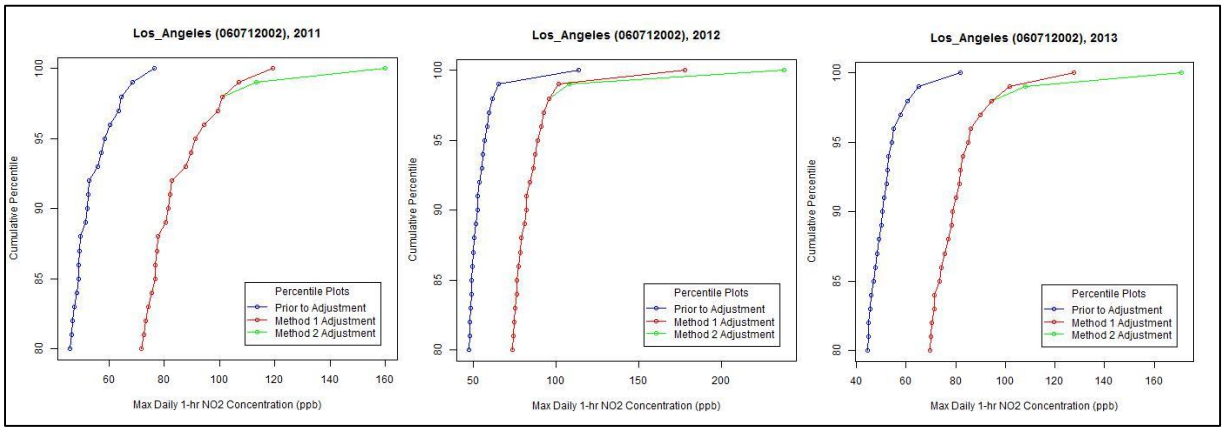

Figure 16. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Los Angeles (060712002; 2011-2013).

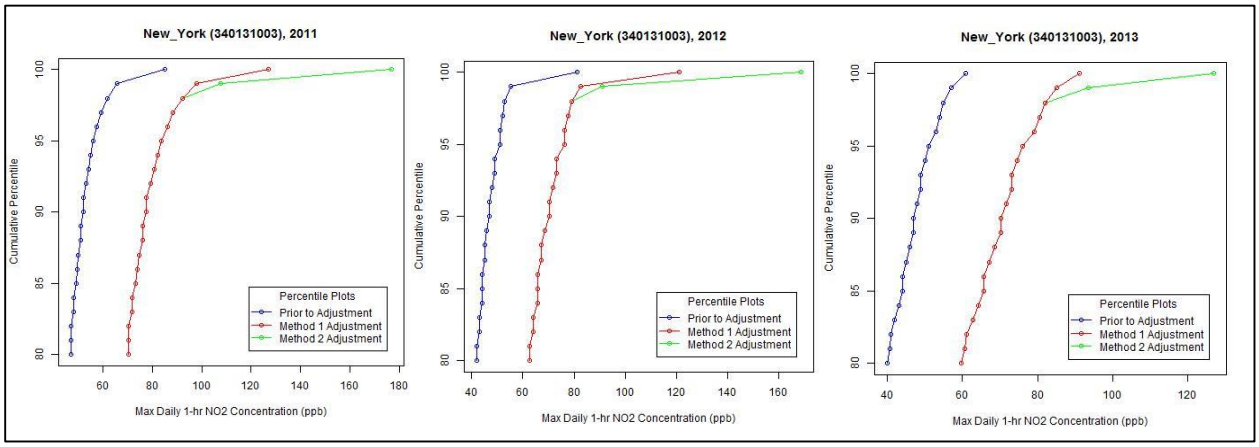

Figure 17. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, New York (340131003; 2011-2103).

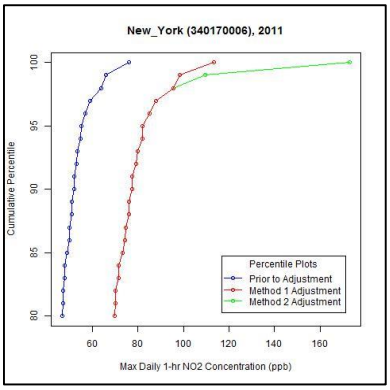

Figure 18. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, New York (340170006; 2011).

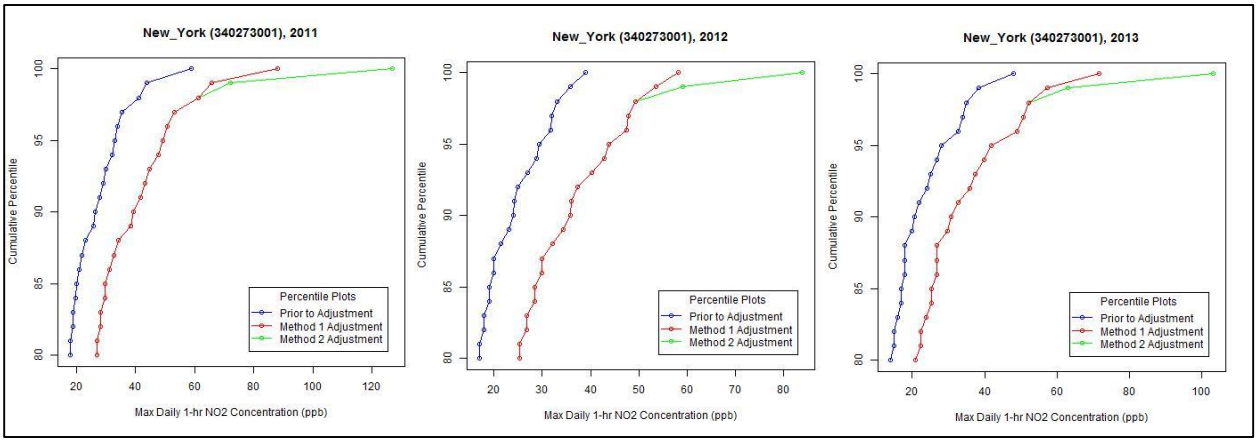

Figure 19. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, New York (340273001; 2011-2013).

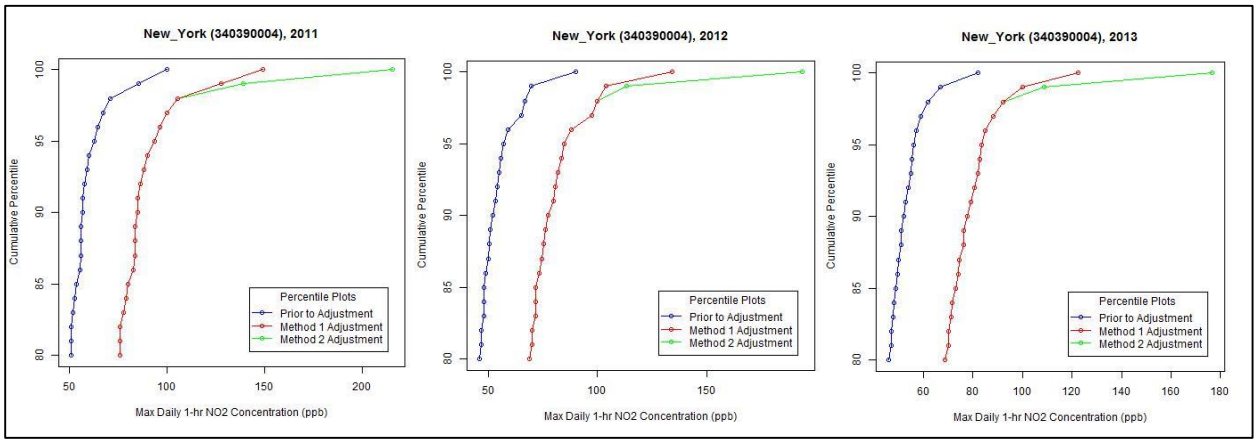

Figure 20. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, New York (340390004; 2011-2013).

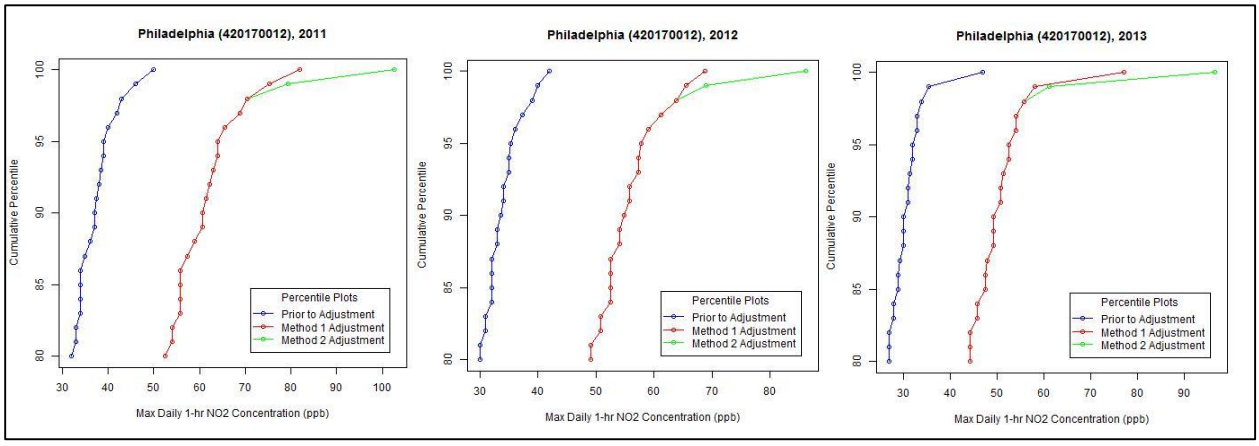

Figure 21. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Philadelphia (420170012; 2011-2013).

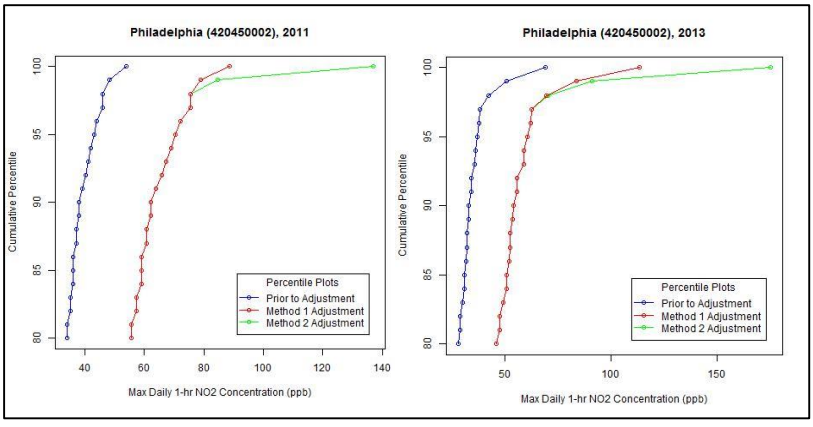

Figure 22. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Philadelphia (420450002; 2011, 2013).

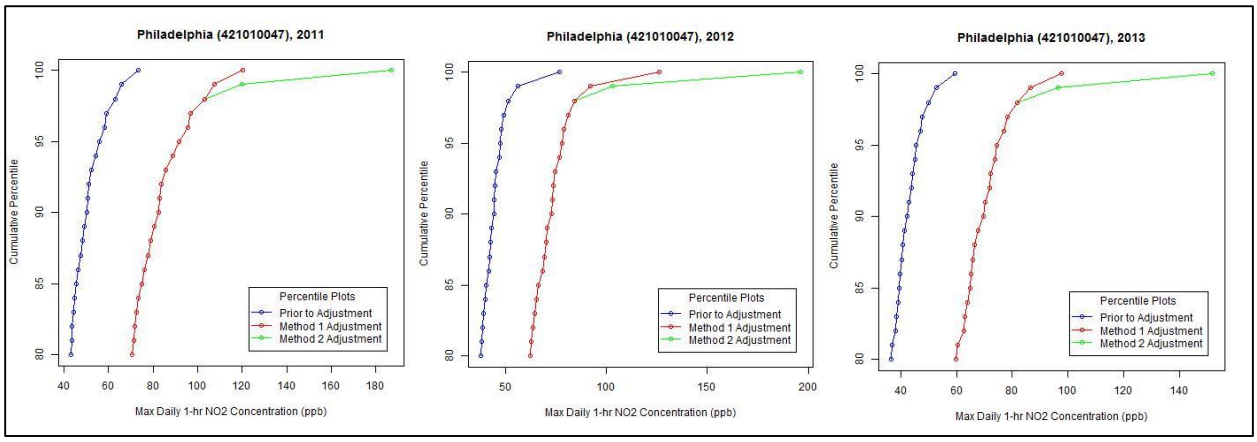

Figure 23. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Philadelphia (421010047; 2011-2013).

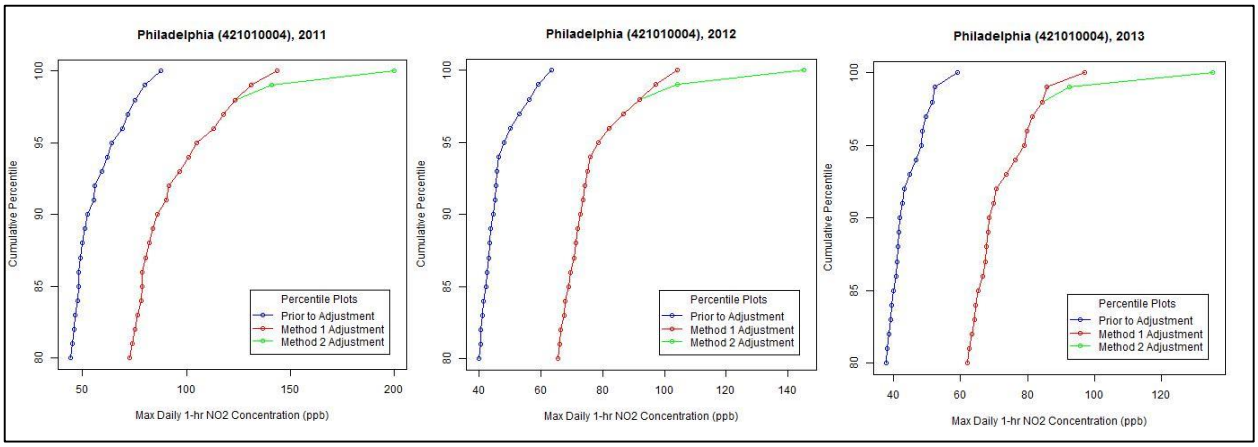

Figure 24. Percentile Concentrations Prior to and after Method 1 and Method 2 Adjustments, Philadelphia (421010004; 2011-2013).

Exceedances of Potential Health Effect Benchmark Levels

After applying Method 1 and Method 2 to adjust 1-hour ambient NO_2 concentrations, the number days in which the DM1H concentration exceeds 100 ppb were counted for each of the annual datasets in which concentrations were adjusted. In all, concentrations were adjusted for 43 yearly datasets across 19 monitoring sites in four cities. The counts were performed prior to and after applying each of the two concentration adjustment methods. The results of the counts are shown in Table 6 for each dataset adjusted in 2011-2013 in which the data are complete.

The most noticeable result from Table 6 is the number of datasets in which there are zero days that exceed 100 ppb prior to and after adjusting the concentrations to just meet the existing 1-hour standard, regardless whether Method 1 is applied solely or Method 2 is subsequently applied. Prior to adjusting the concentrations, there are only four datasets with one or more days exceeding 100 ppb. All four are in the Los Angeles area. Thus, recent ambient concentration measurements would likely not be associated with adverse health outcomes when using the lowest 1-hour health effect benchmark level of 100 ppb.

The Method 1 concentration adjustment approach resulted in 27 of the 43 yearly datasets having days with exceedances while 38 datasets show exceedances as a result of applying Method 2. The largest number days in a year that exceed a 1-hour health effect benchmark of 100 ppb from Methods 1 and 2 is 23 for a site in Philadelphia which originally had zero days of exceedances. Prior to adjusting the data to just meet the existing 1-hour standard, the largest number of days in a year that exceed 100 ppb is seven for a site in Los Angeles, which increased to 20 after both Method 1 and Method 2 were applied. The application of Method 2 results in an increase in the number days that exceed 100 ppb for 28 of the 43 datasets over Method 1.

Table 6. Number of Days per year with an Exceedance of a 1-hour Health Effect Benchmark of 100 ppb (2011-2013).

			# Days Exceed 100 ppb		
AQS ID	Area City	Year	No Adjustment	Method 1	Method 2
080013001	Denver	2011	0	0	3
080013001	Denver	2012	0	0	2
080013001	Denver	2013	0	1	2
080310002	Denver	2012	0	0	2
080310002	Denver	2013	0	1	3
340131003	New_York	2011	0	4	5
340131003	New_York	2012	0	2	3
340131003	New_York	2013	0	0	3
340170006	New_York	2011	0	3	6
340273001	New_York	2011	0	0	1
340273001	New_York	2012	0	0	0
340273001	New_York	2013	0	0	1
340390004	New_York	2011	0	12	12
340390004	New_York	2012	0	9	9
340390004	New_York	2013	0	4	5
420170012	Philadelphia	2011	0	0	1
420170012	Philadelphia	2012	0	0	0
420170012	Philadelphia	2013	0	0	0
420450002	Philadelphia	2011	0	0	2
420450002	Philadelphia	2013	0	1	3
421010004	Philadelphia	2011	0	23	23
421010004	Philadelphia	2012	0	2	5
421010004	Philadelphia	2013	0	0	3
421010047	Philadelphia	2011	0	8	8
421010047	Philadelphia	2012	0	2	4
421010047	Philadelphia	2013	0	0	3
060370002	Los_Angeles	2012	0	3	6
060370002	Los_Angeles	2013	0	3	5
060370113	Los_Angeles	2011	0	3	5
060371103	Los_Angeles	2011	1	8	8
060371201	Los_Angeles	2011	0	2	3
060371701	Los_Angeles	2011	0	12	12
060371701	Los_Angeles	2012	0	2	5
060371701	Los_Angeles	2013	0	7	7
060372005	Los_Angeles	2011	1	7	7
060655001	Los_Angeles	2011	0	0	0
060655001	Los_Angeles	2012	0	0	0
060655001	Los_Angeles	2013	0	0	1
060710001	Los_Angeles	2011	0	4	6
060710001	Los_Angeles	2012	7	20	20
060712002	Los_Angeles	2011	0	9	9
060712002	Los_Angeles	2012	1	5	6
060712002	Los_Angeles	2013	0	4	6

DISCUSSION

The regression analysis shows strong support for a linear relationship in the reduction of ambient levels of NO_2 over multiple decades across the entire distribution at the urban monitoring sites. This is consistent with findings in the previous 2008 study (EPA, 2008b). However, the support is not overwhelming, and there are sufficient differences in the concentration patterns at many of the sites to suggest the relationship is closer to linear through the middle portion of the distribution and nonlinear at upper and lower ends of the distribution, primarily above the 98th percentile of the maximum daily 1-hour concentrations. As noted in the Results section above, many of the sites show a pronounced curvature at the upper and lower ends of the distribution that falls below the best fit line and a flattening above the line through the middle of the distribution. General regression performance statistics (r^2 values) are the lowest where the points fall below the line and continue to track farther below the line through the tails of the distribution. For those cities in which multiple sites were available and analyzed (Denver, Los Angles, New York, and Philadelphia), there is a mix of results with regard to the distribution patterns and r^2 values such that it is difficult to conclude there is an area-wide linear or nonlinear relationship in the reductions of NO_2 at the ends of the distribution of concentrations. The differences in the patterns appear to be site-dependent rather than city dependent.

There is, however, further support for a nonlinear reduction in the upper end of the distribution found even in many of the sites with higher r^2 values well above 0.95. Most datasets, regardless of the r^2 value, exhibit curvature in which one or more data points dip below the fitted line at the upper end of the distribution. For some sites, the data points remain below the line through the tail of the distribution, others trend back above the line at the very tail, and

some do not trend below the fitted line until the last few data points. Those areas where the data fall below the fitted line represent the largest reductions in NO_2 concentrations over time. In general, these patterns show that reductions are greater and disproportionate at the upper end of the distribution, above the 98th percentile, than in the middle of the distribution.

Referring back to Table 6, when comparing the counts of the number of days that exceed 100 ppb prior to and after applying each of the two concentration adjustment methods, Method 1 produced one or more days of exceedances for a majority of the datasets in which there were no exceedances in the original data. The number of days per year with exceedances increased substantially as a result of using the proportional adjustment (Method 1) for each of the four study areas in which there were days with exceedances in the original data. As expected, the number of days per year increased further across most datasets after applying Method 2, but interestingly, increases using Method 2 only occurred in datasets with a count of 6 days or less after applying Method 1. The counts of the number of days per year with exceedances ranged from 0 to 23 after applying Method 1. None of the 10 datasets having a count of 7 or more days with exceedances after applying Method 1 experienced an increase from Method 2, while 28 of the datasets having a count of 6 or less days after applying Method 1 did show an increase after applying Method 2. Thus, more than one-fourth of the datasets that have days with exceedances after applying Method 1 were not affected by the use of Method 2.

Given that Method 2 has to occur subsequent to applying Method 1 (i.e., Method 1 must be applied prior to Method 2), Method 2 may not provide any benefit for those datasets that experience the largest increases in the number of days with exceedances from Method 1. However, for those datasets that experience fewer increases from Method 1, relying solely on Method 1 could result in a gross underestimation in concentrations at the upper end of the

distribution. Further, if the array of health concentrations of interest in the future is further stratified (e.g., concentrations above 100 ppb, in 10 ppb increments), Method 1 would also generate fewer concentration exceedances in comparison with Method 2. It is also possible that, with additional concentration adjustments to account for other high concentration environments (such as on or near roads), a greater number of exceedances and perhaps at higher benchmark levels (e.g., 200 ppb) would be estimated using Method 2 compared with Method 1. Given the evidence from this study that supports nonlinearity at the upper end of the distribution of the reduction in ambient NO_2 concentrations over time, relying solely on Method 1 to adjust concentrations upward could significantly affect estimated air quality, exposures, and possibly risk results when reviewing the health protection associated with the existing or alternative standards.

CONCLUSION

Trends in the changes in the levels of urban ambient NO_2 were analyzed. While results from this analysis and a previous study support a proportional relationship in the reduction of NO_2 over the last several decades, this study also provides strong evidence that the relationship is nonlinear through the lower and upper percentiles, primarily above the 98^{th} percentile of DM1H concentrations, on which the 1-hour NO_2 NAAQS is based. Two methods were compared to adjust ambient concentrations of NO_2 upward to just meet a DM1H standard of 100 ppb. Method 1, a simple method, assumes a linear relationship across the entire distribution while Method 2 also assumes a linear relationship, though only up to the 98^{th} percentile, and includes a nonlinear relationship for concentrations above the 98^{th} percentile. The number days in a year were counted for which a DM1H concentration exceeded 100 ppb prior to and after the

application of each adjustment method as a means to evaluate the potential for health effects associated with short-term exposure.

This study does not definitively indicate that the relationship in the reduction of NO_2 over time is either proportional across the entire percentile distribution or that there is definitely a transition to a more nonlinear pattern in the upper percentiles. However, a comparison of results from the two methods used to adjust the concentrations upward suggest that neither method should be ignored in approximating alternative air quality scenarios requiring such an adjustment, and each method might be a viable approach depending on the circumstances. In cases where Method 1 produces large counts of the number of days per year that exceed potential health effect benchmark levels, the subsequent application of Method 2 may not provide any significant benefit for the effort required to apply Method 2. Conversely, for those datasets in which Method 1 results in small increases in the number of days that exceed potential health effect benchmark levels, omitting the additional application of Method 2 to the upper percentiles may result in a significant underestimation of concentrations which could adversely affect an exposure assessment that relies heavily on the adjusted concentration data.

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