# Analysis of Primary Fine Particle National Ambient Air Quality Standard Metrics

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# **ABSTRACT**

In accordance with the Clean Air Act, the U.S. Environmental Protection Agency (EPA) is currently reviewing its National Ambient Air Quality Standards for particulate matter, which are required to provide an adequate margin of safety to populations, including susceptible subgroups. Based on the latest scientific, health, and technical information about particle pollution, EPA staff recommends establishing more protective health-based fine particle standards. Since the last standards review, epidemiologic studies have continued to find associations between short-term and long-term exposure to particulate matter and cardiopulmonary morbidity and mortality at current pollution levels. This study analyzed the spatial and temporal variability of fine particulate  $(PM_{2.5})$  monitoring data for the Northeast and the continental United States to assess the protectiveness of various levels, forms, and combinations of 24-hr and annual health-based standards currently recommended by EPA staff and the Clean Air Scientific Advisory Committee. Recommended standards have the potential for modest or substantial increases in protection in the Northeast, ranging from an additional 13– 83% of the population of the region who are living in areas not likely to meet new standards and thereby benefiting from compliance with more protective air pollution controls. Within recommended standard ranges, an optimal 24-hr (98th percentile)/annual standard suite occurs at  $30/12 \mu g/m^3$ , providing short- and long-term health protection for a substantial percentage of both Northeast (84%) and U.S. (78%) populations. In addition, the Northeast region will not benefit as widely as the nation as a whole if less stringent standards are selected. Should the 24-hr (98th percentile) standard be set at 35  $\mu$ g/m<sup>3</sup>, Northeast and U.S. populations will receive

#### **IMPLICATIONS**

The Clean Air Act calls on EPA to establish ambient air quality standards that protect public health with an adequate margin of safety. With respect to the forthcoming decision of EPA on whether to revise current  $PM<sub>2.5</sub>$  standards, this paper provides a set of methodological tools for regulatory agencies and decision-makers to determine which level, form, and combination of currently recommended health-based  $PM<sub>2.5</sub>$  24-hr and annual standards would best protect populations in the Northeast and continental United States. Selecting an equivalent and stringent standards suite would ensure the broadest short- and long-term protection across the  $PM_{2.5}$  monitoring network.

16–48% and 7–17% less protection than a 30  $\mu$ g/m<sup>3</sup> standard, respectively, depending on the level of the annual standard. A 30/12  $\mu$ g/m<sup>3</sup> standard suite also provides nearly equivalent 24-hr and annual control of  $PM_{2.5}$  distributions across the United States, thereby ensuring a more uniform and consistent level of protection than unmatched or "controlling" and "backstop" standards. This could occur even within EPA staff's recommended range of standard suites, where 22– 43% of the monitors in the country could meet a controlling standard but fail to meet the combined backstop standard, resulting in inconsistent short- and long-term protection across the country. An equivalent standards combination of 30/12  $\mu$ g/m<sup>3</sup> would minimize the wide variation of protectiveness of 24-hr and annual  $PM_{2.5}$  concentrations. Furthermore, given recent associations of subdaily exposures and acute adverse health effects, in the absence of a subdaily averaging metric, a stringent 24-hr standard will more effectively control maximum hourly and multihourly peak concentrations than a weaker standard.

# **INTRODUCTION**

The Clean Air Act mandates the U.S. Environmental Protection Agency (EPA) to set health-based National Ambient Air Quality Standards (NAAQS) for particulate matter  $\leq$ 2.5  $\mu$ m (PM<sub>2.5</sub>). NAAQS provisions require EPA to establish standards stringent enough to protect public health with an adequate margin of safety, at a level that avoids unacceptable risks to both general and susceptible populations. Over the past quarter century, a growing body of scientific evidence has found associations between shortterm and long-term exposure to airborne particulate matter (PM) and cardiopulmonary health outcomes, including increased symptoms, hospital admissions, emergency department visits, and premature death. $1-4$  Population subgroups that have been identified as potentially susceptible to health effects as a result of PM exposure include children, older adults, and people with existing heart and lung diseases and diabetes. In addition, population subgroups may have increased vulnerability to pollutionrelated effects because of factors including socioeconomic status or elevated exposure levels.5 EPA is required to periodically review the PM NAAQS, last revised in 1997,6 to ensure that they provide adequate health and environmental protection reflecting the latest scientific and technical information about PM. EPA expects to propose final mass-based  $PM_{2.5}$  24-hr and annual primary standards by the end of 2005.

The primary objective of this study was to provide methodological tools to determine the degree to which the recent  $PM_{2.5}$  NAAQS recommendations of EPA staff and the PM Clean Air Scientific Advisory Committee (CASAC) will protect populations across the Northeast and continental United States. Using  $PM_{2.5}$  concentrations measured by the Federal Reference Method (FRM) monitoring network, we assessed the protectiveness of various standard levels, forms, and combination choices. EPA uses the FRM network to determine whether or not monitoring areas are in compliance (attainment) with the PM NAAQS. Areas not in compliance (nonattainment) must take steps to reduce  $PM_{2.5}$  concentrations, which presumably lowers the level of pollutants to which populations are exposed, thereby decreasing adverse health outcomes. The methodological approach of this study does not determine whether various NAAQS recommendations protect public health with an adequate margin of safety, a question beyond the capacity of this research. However, the study does assess which currently recommended EPA staff and CASAC standard levels, forms, and combinations would provide the most protection to the public by lowering fine particle concentrations across the broadest FRM monitoring network area.

EPA justification for the 1997  $PM_{2.5}$  standard level and averaging time was in large part attributable to available health effects evidence, including short- and longterm epidemiologic studies finding associations between increased PM and adverse health effects among populations living in urban areas.<sup>7-9</sup> The current PM NAAQS review also has emphasized the importance of findings of health effects associated with acute and chronic exposure to  $PM_{2.5}$  concentrations, including those characterized in time-series and cohort epidemiologic studies.10 Since 1997, multicity research has reported consistent associations of health effects across differing exposure time scales. Time-series epidemiologic studies have found associations between particulate air pollution and daily deaths, especially those using daily monitoring data.<sup>11,12</sup> Multiday effects of exposure appear to accumulate over time,13,14 and cohort studies that incorporate risk associated with longer-term exposure report even higher risk estimates.15,16 Together, these studies suggest the need for more stringent 24-hr and annual standards. But because of the inability of the majority of these studies to identify the existence or nonexistence of any justifiable threshold concentration below which effects are not detectable,<sup>17</sup> selecting primary standards that protect susceptible populations with an adequate margin of safety, as mandated by the Clean Air Act, is largely a public health policy judgment.

During both the 1997 and 2005 NAAQS review cycles, in addition to determining what 24-hr and annual PM<sub>2.5</sub> standard levels and averaging times are appropriate, a central question has been what combination of 24-hr and annual standards can best protect the entire country, given the spatial and temporal variability of concentrations in the United States. During the previous review cycle, EPA concluded that both 24-hr and annual standards could effectively control PM concentration levels

and distributions, thereby providing public health protection for short-term (from  $\leq 1$  day to  $\leq 5$  days) and longterm (seasonal to several years) exposures to  $PM_{2.5}$ . In determining optimal 24-hr and annual standard combinations, an argument was made to treat the annual standard as the generally controlling metric for lowering both short- and long-term  $PM_{2.5}$  concentrations across the monitoring network. A supplemental or backstop 24-hr standard would serve to provide protection against days with high peak  $PM_{2.5}$  concentrations, localized "hot spots," and risks arising from seasonal emissions that would not be well controlled by a national annual standard<sup>6</sup>

Since 1997, understanding of the behavior of  $PM_{2.5}$ levels in the United States has increased because of deployment of the FRM national monitoring network in 1999, providing a wealth of new data. At present, an important question is whether  $PM<sub>2.5</sub>$  standard levels, forms, and combinations other than EPA current 24-hr and annual standards would be more protective of public health. The current controlling annual standard level of 15  $\mu$ g/m<sup>3</sup> is based on the 3-yr average of annual arithmetic mean  $PM<sub>2.5</sub>$  concentrations from single or multiple community-oriented monitors. This standard is combined with a supplemental 24-hr standard level of 65  $\mu$ g/m<sup>3</sup>, which is based on the 3-yr average of the 98th percentile (form) of 24-hr  $PM<sub>2.5</sub>$  concentrations at each population-oriented monitor within an area. The current 98th percentile form represents the daily value from a year of monitoring data below which 98% of all values in the group fall. This allows the 7 highest  $PM_{2.5}$  concentration days per year to exceed the 24-hr standard level. A more stringent 99th percentile form would exclude only the 3 highest concentration days.

Both EPA staff and CASAC now recommend that EPA administrator propose more stringent  $PM<sub>2.5</sub> NAAQS<sub>10,18</sub>$ As shown in Table 1, EPA staff provides 2 alternative options to establishing more protective suites of 24-hr and annual  $PM_{2.5}$  standards. "Option A" would revise the 24-hr standard, within the range of  $30-40 \mu g/m^3$ , combined with a revised annual standard in the range of 12-14  $\mu$ g/m<sup>3</sup>, with either the 24-hr or the annual standard, or both, at the middle-to-lower end of these ranges. "Option B" would revise the 24-hr standard, within the range of 25–35  $\mu$ g/m<sup>3</sup> (based on a 98th percentile form for a standard set at the middle-to-lower end of this range, or

**Table 1.** Recommended primary PM<sub>2.5</sub> NAAQS 24-hr and annual ranges for EPA (option A and option B) and CASAC.

<b>Annual</b>	24-hr ( $\mu$ g/m <sup>3</sup> )					
$(\mu$ g/m <sup>3</sup> )	25	30	35	40	65	
12		FPA A <sup>a</sup>	FPA A	EPA A		
13			EPA A / CASAC EPA A / CASAC	EPA A		
14		EPA A / CASAC	EPA A / CASAC	FPA A		
15	FPA R <sup>b</sup>	FPA B	EPA B		EPA current <sup>c</sup>	

<sup>a</sup>EPA option A: 24-hr, annual, or both at the middle to lower end of these ranges; <sup>b</sup>EPA option B: 24-hr 98th percentile at the middle to lower end of this range or a 99th percentile at the middle to upper end of this range; <sup>c</sup>EPA current 24-hr (98th percentile)/annual standard.

a 99th percentile form for a standard set at the middleto-upper end of this range), combined with a retained annual standard of 15  $\mu$ g/m<sup>3</sup>. CASAC recommends setting a 24-hr standard at concentrations in the range of 30-35  $\mu$ g/m<sup>3</sup> with the 98th percentile form, combined with an annual standard in the range of  $13-14 \mu g/m^3$ .

The second objective of this study was to assess the extent to which current 24-hr standard averaging metrics are sufficient to control hourly and multihourly levels. During the 1997 standard setting review, EPA selected the current  $PM_{2.5}$  24-hr and annual standard averaging metrics based on epidemiologic studies using 24-hr integrated samples that reported health effects associated with shortterm and long-term exposures. Earlier health studies used 24-hr integrated samples, because most PM concentration measurements were collected in this form, often only with once every 3-day and every 6-day sampling frequency. At the time, although most reported effects had been associated with daily or longer average measures of PM, epidemiologic and toxicological evidence suggested that some effects might be associated with PM exposures  ${<}24 \text{ hr.}^{19}$ 

Since the last PM NAAQS review by EPA, advances in  $PM<sub>2.5</sub>$  monitoring have facilitated the collection of highly time-resolved fine particle data and its use in health studies. The increasing use of monitoring equipment capable of measuring PM in near-continuous time intervals has begun to improve our understanding of exposure to airborne PM as a continuous or "real-time" experience. The importance of short-duration and peak versus 24-hr exposure has been reported by recent studies finding adverse health outcomes in subdaily exposure periods.<sup>20-25</sup> Using continuous  $PM<sub>2.5</sub>$  data, clinical and epidemiologic evidence now suggests that acute cardiac health effects may be associated with PM exposures of durations with averaging times of 1 hr to several hours.26 Studies have also determined that exposures at hourly or minute scales experienced in microenvironments with elevated PM levels may lead to a significant portion of an individual's daily exposure.27–31 Such findings call into question the suitability of the current EPA 24-hr and annual standards in protecting populations from acute peak exposure periods that occur in subdaily time frames. This points to the importance of understanding the degree to which 24-hr and annual  $PM<sub>2.5</sub>$  standards can control subdaily peak levels and provide an adequate margin of safety.

In the course of both the 1997 and current PM NAAQS review, the question of peak exposures and their relation to overall risk has played a role in considerations over the selection of 24-hr/annual standard level and forms. Based on its 1996 and 2005 PM risk assessments, EPA has concluded that much, if not most, of the aggregate annual risk associated with short-term exposures results from the large number of days during which 24-hr average concentrations are in the low-to-middle range. This, in part, provided the rationale for the agency to select a controlling annual standard and a weaker backstop 24-hr standard.6,10,32,33 As noted recently by some PM CASAC members, however, another interpretation of the EPA evidence might find that 24-hr mortality per concentration day actually increases as  $PM_{2.5}$  concentrations increase. Such a finding might suggest that higher

concentration levels are important to consider for mortality-related health risks.18 Although the current EPA 24-hr average and statistical forms conceive of shortterm exposure (and thereby facilitate the assessment of health risk) in terms of low-, middle-, or high-range daily 24-hr averages, subdaily hourly averaged data may lead to a different characterization of exposure health risk. EPA does not believe enough quantitative evidence currently exists to support a subdaily standard,<sup>10</sup> but the issue likely will play a dominate role in the next PM NAAQS review cycle. Therefore, for the time being, in the absence of a shorter averaging metric, it is important to understand to what extent a 24-hr average metric can control subdaily levels. These findings have the potential to inform the selection of a 24-hr standard form, level, and combination.

## **METHODS**

FRM  $PM_{2.5}$  air pollution data from 2000, 2001, and 2002 were obtained for a Northeast dataset from EPA Air Quality System in August 2003 from 127 FRM monitors in EPA Region 1 (6 New England states) and Region 2 (New Jersey and New York), and 65 FRM monitors outside these regions in bordering states (Delaware, Washington, DC, Maryland, and Pennsylvania). Data for the same period were retrieved for three Northeast Interagency Monitoring of Protected Visual Environments sites from the Visibility Information Exchange Web System. Countrywide data for the years 2000 –2002 were obtained from EPA AirData.34 –36

Within the 2000–2002 period for the Northeast dataset, 192 PM monitoring sites had data in all 12 quarters. Data flagged with the forest fire exemption for 2002 were removed. More than 75% of the 192 sites had >50% data capture within each quarter. Data completeness affecting the remaining sites was primarily isolated to one calendar quarter. For sites with collocated monitors, the primary monitor at a site was used to determine the  $PM<sub>2.5</sub>$  concentration (27 pairs of 192 monitors). Although fewer than half of the primary monitors satisfied the 75% data completeness criteria, no substitution from collocated monitors was conducted. The relationship between the 24-hr and annual averages was not dependent on data completeness at the site, as determined by regression analysis. (The regression where y is the level of the 24-hr average and x is the level of the annual average for the subset of monitors with complete data was  $y = 1.86x$  $+10.43$  [ $n = 81$ ,  $R^2 = 0.76$ ] and for the subset of monitors with incomplete data was  $y = 1.82x + 10.90$  [ $n = 111$ ,  $R^2$  $= 0.78$ ]).

To estimate the number of persons living in counties not likely to meet different combinations of alternative 24-hr and annual  $PM_{2.5}$  standards, design values were calculated for all counties<sup>150</sup> in the eight-state study area and integrated with 2000 U.S. Census county-level population data using ArcGIS v8.2 software.37 A design value is a statistic that describes the air quality status of a given area relative to the level of the NAAQS. Design values are typically used to classify nonattainment areas, assess progress toward meeting the NAAQS, and develop control strategies. Design values were calculated in adherence with EPA criteria for determination by calculating 3-yr

averages of 24-hr 98th percentiles and annual means based on the maximum monitor within an urban area.<sup>6,38</sup>

Design values for the 70 counties with monitors were assigned from the highest monitored levels in each county for 2000 –2002. Design values for 80 counties lacking monitors were generated by interpolating countylevel monitored data from 104 monitors within the eightstate study region and 61 monitors outside the region for border counties. An interpolation scheme was employed using inverse distance squared weighting for the six nearest monitors within a 111-km radius (corresponding to 1° latitude). Massachusetts and New Hampshire had very few sites with complete data for the 3-yr period, requiring an approximation of design values for counties in those states. For the other counties in the eight-state study region, the annual design values used were generally within  $\pm 0.2$   $\mu$ g/m<sup>3</sup> of those reported by EPA using customary guidelines for data substitution and completeness determinations.38

The analysis of continuous  $PM_{2.5}$  data (50 °C Tapered Element Oscillating Microbalance [TEOM] method and Beta Attenuation Monitor) used 2001 and 2002 data collected from EPA Region 1, 2, and 3 monitoring networks. The 50 °C TEOM method daily or subdaily data are subject to large errors because of a substantial loss of semivolatile mass. Therefore,  $PM_{2.5}$  levels are likely to be underestimated on winter days with high  $PM_{2.5}$  concentrations or during hours with the highest local mobile source influence. In general, data with highest temporal resolution (e.g., 1-hr data) have the greatest potential to underestimate  $PM_{2.5}$  relative to "FRM-like" levels.

Analysis of maximum 1-, 3-, 4-, and 6-hr average and 24-hr average continuous data was conducted to assess the extent to which the 24-hr average metric controls subdaily maximum hourly averages. Cumulative frequency plots used year-round 2001 data from Regions 1 and 2 and border state monitoring networks. The analysis considered the 24-hr average of a day valid if 16 hourly values were reported. Rolling 3-, 4-, and 6-hr averages were calculated, and the maximum average for each interval was tabulated for each day. Valid averages required 3 or 4 hr for those averaging periods, respectively, whereas a valid 6-hr average required  $\geq$ 5 valid hourly values. The analysis is insensitive

to the TEOM method bias, because it relies on relationships among different averaging times rather than absolute monitored concentration.

## **RESULTS**

Northeast and continental U.S. FRM  $PM_{2.5}$  data were analyzed to assess the protectiveness of currently recommended EPA staff and CASAC  $PM_{2.5}$  24-hr and annual standard levels, forms, averaging times, and combinations. This facilitated an understanding of the various ways that different standards may reduce ambient  $PM_{2.5}$ concentrations and thereby protect populations from exposure to fine particles. Results are organized into three subsections covering the protectiveness of standard levels, forms, and combinations; 24-hr and annual standard equivalency; and 24-hr and subdaily averaging metrics.

## **Protectiveness of PM2.5 Standard Levels, Forms, and Combinations**

The study found that either 24-hr or annual standard levels can lower the entire  $PM_{2.5}$  distribution curve (including maxima), decreasing 24-hr average and annual mean concentrations. Figure 1 shows the distribution for 24-hr (98th percentile) and annual average concentrations in the Northeastern United States. Each distribution covers the entire data range (with the area under the  $curve = 1$ ) and is normalized to reflect the total number of monitored days in every grouping. This relationship also applies to the 24-hr 99th percentile form, which is 4 –5  $\mu$ g/m<sup>3</sup> more stringent than the equivalent 98th percentile form (Figure 2).

The study also found that both 24-hr 98th and 99th percentile forms control  $PM_{2.5}$  maxima. However, use of the 99th percentile form would allow fewer days above the 24-hr standard than use of the 98th percentile. Table 2 estimates the number of days that  $PM_{2.5}$  values exceed the 98th or 99th form  $\geq$ 5  $\mu$ g/m<sup>3</sup> of the 24-hr level. A 5- $\mu$ g threshold was selected because the EPA-recommended 24-hr standard levels are in  $5-\mu g$  increments. For both percentile forms, more than half the days above the standard are within 5  $\mu$ g/m<sup>3</sup>of the standard, corresponding to <3 days



Figure 1. Distribution of selected 24-hr (98th percentile) and annual PM<sub>2.5</sub> ranges (µg/m<sup>3</sup>; 2000–2002 FRM Regions 1, 2, and Delaware, Washington DC, Maryland, and Pennsylvania).



Figure 2. Distribution of selected 24-hr (98th and 99th percentile) and annual PM<sub>2.5</sub> ranges (µg/m<sup>3</sup>; 2000–2002 FRM Regions 1, 2, and Delaware, Washington DC, Maryland, and Pennsylvania).

and 1 day more than 5  $\mu$ g/m<sup>3</sup> for the 98th and 99th percentile forms, respectively. For example, at a daily concentration of 30-31  $\mu$ g/m<sup>3</sup>, for 5 of 7 days (or 71% of the time) the excluded values exceed this daily level by  $\leq$  5  $\mu$ g/m<sup>3</sup>. The remaining 2 days exceed the level by  $>$  5  $\mu$ g/m<sup>3</sup>.

Within the currently recommended EPA staff and CASAC range of standards, the final  $PM<sub>2.5</sub>$  standards proposed by the EPA Administrator may result in either modest or substantial additional protection to the Northeast, varying from 13 to 83% of the region populations living in areas that would not meet the standards. As shown in Figure 3, increasingly stringent 24-hr and annual standard levels and forms result in a greater percentage of the total population living in nonattainment areas that would require increased control measures to lower  $PM_{2.5}$  concentrations, thereby benefiting public health by reducing exposure levels. By meeting the current 24-hr (98th percentile)/annual standard of  $65/15 \mu g/m^3$ , 16% of the region population benefits from  $PM_{2.5}$  emission control

Table 2. Number of days PM<sub>2.5</sub> values exceed 98th or 99th percentile form  $\geq$ 5  $\mu$ g/m $^3$  of the 24-hr level (2000–2002 FRM Regions 1, 2, and Delaware, Washington DC, Maryland, and Pennsylvania).

	<b>Number of Days Above 24-hr Standard</b>				
		<b>98th Percentile Form</b>	99th Percentile Form		
24-h Average <b>Concentration</b> $(\mu$ g/m <sup>3</sup> )	No. Days $\leq$ 5 µg/m <sup>3</sup> of Level	No. Days $>5 \mu$ g/m <sup>3</sup> of Level	No. Days $\leq$ 5 µg/m <sup>3</sup> of Level	No. Days $>5 \mu$ g/m <sup>3</sup> of Level	
$25 - 26$	3.9	3.1			
$27 - 29$	4.3	2.7			
$30 - 31$	5.0	2.0	1.9	1.1	
$32 - 33$	4.3	2.7	2.0	1.0	
$34 - 35$	4.8	2.2	2.1	0.9	
$35 - 36$	4.6	2.4			
$36 - 37$	4.1	2.9	1.9	1.1	
$38 - 39$	3.9	3.1	1.7	1.3	
$40 - 41$	4.4	2.6	1.7	1.3	
Average # days	4.4	2.6	1.9	1.1	

strategies. Regarding the EPA staff "Option A" recommendation, the least-stringent  $40/14 \mu g/m^3$  standard (98th percentile) would result in 29% of the population of the region being in nonattainment, whereas the most-stringent  $30/12 \mu g/m^3$  standard (98th percentile) would result in 84% of the population in nonattainment, or 68% more than afforded by the current standard. For the EPA staff "Option B" recommendation, the least-stringent 35/15  $\mu$ g/m<sup>3</sup> (99th percentile) would result in 84% nonattainment, whereas the most-stringent  $25/15 \mu g/m^3$  (98th percentile) would result in 99% nonattainment. Overall, the range of protection within both options is 29 –99%, or 13– 83% more than the current standard. Within the CASAC recommendation, the overall range of protection is 36 – 84%, or 20 – 68% beyond the current standard. (The 99th percentile levels are 5  $\mu$ g/m<sup>3</sup> approximations based on the average relationship between the 98th and 99th percentiles from Figure 2.)

Figure 3 also illustrates the levels at which currently recommended 24-hr and annual standard combinations become controlling in the eight-state Northeast study area. Because most Northeast monitoring site 24-hr (98th percentile) averages cluster in the  $30-35 \mu g/m^3$  range, a sharp increase in protection occurs below a 24-hr level of 35  $\mu$ g/m<sup>3</sup> in combination with annual levels ranging from 12 to 15  $\mu$ g/m<sup>3</sup>. A 24-hr (98th percentile) standard level of 30  $\mu$ g/m<sup>3</sup> behaves as a controlling standard, resulting in a 68% increase in nonattainment protection for Northeast populations compared with the current standard, regardless of whether the combined annual level is 12-15  $\mu$ g/m<sup>3</sup>. Conversely, 24-hr (98th percentile) levels set at  $\geq$ 35  $\mu$ g/m<sup>3</sup> would result in an 18–52% increase in protection, depending where the  $12-15 \mu g/m^3$  annual level was set. This finding makes clear the implications of selecting a 24-hr standard  $\geq$ 35 µg/m<sup>3</sup>, especially in combination with less-stringent annual standards. For example, were the current 24-hr 98th percentile standard reduced from 65 to 40  $\mu$ g/m<sup>3</sup> in combination with an annual standard of 15  $\mu$ g/m<sup>3</sup>, no additional protection would be realized in the Northeast study area. Of interest, an  $11-\mu g/m^3$  annual standard level, although not under



Figure 3. Estimated percent total population in New England, New Jersey, and New York that would benefit from compliance with alternative 24-hr (98th and 99th percentile) and annual PM<sub>2.5</sub> standards ( $\mu$ g/m<sup>3</sup>; 2000–2002 FRM Regions 1, 2).

consideration, would control  $PM_{2.5}$  levels over the lower range of 24-hr levels considered.

Expanding the preceding Northeast analysis to the entire U.S. monitoring network (using comparatively similar EPA calculations; ref. 10) finds that within the recommended primary 24-hr/annual health-based ranges of EPA staff and CASAC, the difference between a 24-hr (98th percentile) standard of 30 and 35  $\mu$ g/m<sup>3</sup> would have a disproportionate effect on the protectiveness of Northeast versus U.S. populations. As shown in Figure 4, a 24-hr (98th percentile)/annual standard combination of 30/14  $\mu$ g/m<sup>3</sup> would protect 48% more of the Northeast population than a combination of  $35/14 \mu g/m^3$ . A combination of 30/13  $\mu$ g/m<sup>3</sup> would protect 37% more of the Northeast population than a  $35/13 \mu g/m^3$  pairing. This 48% and 37% Northeast difference compares to a 17% and 12% respective difference for the entire United States. Thus, the Northeast region will not benefit as widely as the nation as a whole unless  $PM_{2.5}$  standards are set at or below a 24-hr (98th percentile)/annual  $30/12 \mu g/m^3$  level. Within EPA staff recommended ranges were the annual standard set between 12 and 14  $\mu$ g/m<sup>3</sup>, the difference between selecting a 24-hr (98th percentile) standard of 30

 $\mu$ g/m<sup>3</sup> and 35  $\mu$ g/m<sup>3</sup> amounts to a 16–48% difference in the Northeast and 7–17% difference in the United States. However, a  $30/12 \mu g/m^3$  98th percentile standard would result in more even countrywide protectiveness, with 84% of the Northeast and 78% of the U.S. populations living in areas that would not meet the new standards.

#### **24-hr and Annual Standard Equivalency**

Although the preceding findings show that both 24-hr (98th and 99th percentile) and annual standards recommended by EPA staff and CASAC can control the distribution of  $PM_{2.5}$  levels, the study has also found that neither standard in isolation is sufficient to ensure maximum protection across the United States for both 24-hr short-term and annual long-term exposure time scales. As shown in the following figures, the spatial and temporal variability of  $PM_{2.5}$  concentrations across the U.S. FRM monitoring network results in a wide variation of 24-hr and annual levels. Unequal standards providing either weak 24-hr or annual protection in the form of lessstringent standards may lead to inadequate protection of populations across either averaging metric. When set at an appropriately stringent level, equivalent or matching



Figure 4. Estimated percent total population in New England, New Jersey, and New York (Northeast) vs. total U.S. population that would benefit from compliance with alternative EPA staff and CASAC recommended 24-hr (98th and 99th percentile) and annual PM<sub>2.5</sub> standard ranges (µg/m<sup>3</sup>; 2000–2002 FRM Regions 1, 2 for Northeast; 2001–2003 FRM country-wide for total U.S.; ref. 10).

combinations of 24-hr and annual standards would, together, provide more uniform and consistent protection across the country than unequal standard combinations by minimizing the variation of short- and long-term exposures.

Figure 5 illustrates the short- and long-term variability of  $PM<sub>2.5</sub>$  concentrations across the U.S. FRM network. The figure plots the site-by-site relationship between 3-yr average 24-hr (98th percentile) and annual average levels for selected U.S. urban areas, showing that many monitoring areas will experience a wide range of  $PM_{2.5}$  concentrations on a 24-hr or annual basis when satisfying one or the other standard. Using an example 24-hr/annual standard of 14/35  $\mu$ g/m<sup>3</sup> (as provided by vertical and horizontal lines on the figure), data in Figure 5 fall into four categories of monitor levels that do the following: (1) meet both 24-hr/annual standards; (2) miss the annual standard; (3) miss the 24-hr standard; and (4) miss both standards. Monitoring sites in the upper right quadrant would miss both standards. Sites in the lower left quadrant would meet both standards. Sites in the upper left quadrant would miss the 24-hr standard but would meet the annual standard. Sites in the lower right quadrant would miss the annual standard but meet the 24-hr standard. For example, at monitoring sites in Seattle, a stringent controlling annual standard of 11.8  $\mu$ g/m<sup>3</sup> would experience gradually less stringent backstop 24-hr levels ranging from 31 to  $42 \mu g/m^3$ . If a stringent annual standard of 12  $\mu$ g/m<sup>3</sup> were combined with a less-stringent 24-hr standard of 40  $\mu$ g/m<sup>3</sup>, the annual standard would effectively protect populations from long-term exposure, but the weaker 24-hr standard would allow exposures  $\leq 40 \mu g/m^3$ .

Findings presented in Figure 6 suggest that the optimal  $PM_{2.5}$  standard selection would use both 24-hr and annual standard levels to provide consistent and uniform protection by minimizing short- and long-term  $PM_{2.5}$ variability, thereby maximizing protection across the broadest FRM monitoring network area. The figure aggregates 24-hr (98th percentile) and annual  $PM_{2.5}$  levels for the entire U.S. network (1137 monitors) to show the magnitude of various standard combinations falling into the



**Figure 5.** Relationship between 24-hr (98th percentile) and annual  $PM_{2.5}$  levels ( $\mu$ g/m<sup>3</sup>; 2000-2002 selected FRM country-wide monitors). Using an example 24-hr/annual standard of  $14/35 \mu q/m^3$  (as provided by vertical and horizontal lines on the figure), data in the figure fall into four categories: monitor levels that (1) meet both 24-hr/annual standards; (2) miss the annual standard; (3) miss the 24-hr standard; and (4) miss both standards.

same four categories (monitors that meet both standards, either the 24-hr or annual standard, or neither standard). Results are presented across a range of five annual levels (11-15  $\mu$ g/m<sup>3</sup>), within which are nested a range of five 24-hr levels (25–45  $\mu$ g/m<sup>3</sup>). Annual and 24-hr ranges were purposely selected outside of EPA staff and CASAC recommended ranges to extend the analysis beyond the bounds of the recommended ranges. The lower portion or first segment of the bars represents the percentage of monitors that meet both 24-hr and annual standards. The next segment represents monitors that miss the annual but meet the 24-hr standard. The third segment represents monitors that miss both standards. The top portion or fourth segment of the bars represents monitors that miss the 24-hr but meet the annual standard.

Across all five of the 24-hr/annual groupings, Figure 6 shows that as 24-hr and annual standard levels increase in stringency (i.e., move from 15 to 11  $\mu$ g/m<sup>3</sup> or from 45 to 25  $\mu$ g/m<sup>3</sup>, respectively), more monitors miss either the annual or 24-hr standards or miss both standards. Within each grouping, as the 24-hr standard becomes less stringent (i.e., moves from 25 to 45  $\mu$ g/m<sup>3</sup>), fewer monitors miss the 24-hr standard, with a greater percentage missing only the annual standard. Thus, more "control" is ceded to the annual standard within the 11–13  $\mu$ g/m<sup>3</sup> range when combined with a 24-hr standard ranging from 35 to  $45 \mu g/m<sup>3</sup>$ . Conversely, as the 24-hr standard becomes more stringent, more control is ceded to the 24-hr standard as a higher percentage of monitors miss only the 24-hr standard. Across the five groupings, a 25–30  $\mu$ g/m<sup>3</sup> 24-hr standard range would control nearly all of the monitors regardless of the annual standard level.

Figure 6 suggests that an optimal standard combination would occur when the number of sites that miss the 24-hr standard equals the number of sites that miss the annual standard. Such matching of 24-hr and annual standards would minimize the occurrence of monitored areas experiencing elevated backstop 24-hr or annual levels relative to a controlling standard. This analysis should not be construed as implying that matching standards are acceptable regardless of the level at which they are set. We assume that both standards levels would be set at a defensible level of health protection as established by EPA staff and CASAC. The more stringent the standards, the more health protection they will afford.

Table 3 shows a subset of data graphed in Figure 6 and illustrates the concepts of controlling, backstop, and equivalent standards encompassing the CASAC recommended  $PM_{2.5}$  standard range, which lies in the middle of the EPA staff range. The table demonstrates the contrast between a controlling/backstop combination versus a matched pair of standards. In this case, the controlling annual level,  $12 \mu g/m^3$ , is paired with a backstop 24-hr level of 40  $\mu$ g/m<sup>3</sup>. Alternatively, a controlling 24-hr of 30  $\mu$ g/m<sup>3</sup> is paired with a backstop annual of 14  $\mu$ g/m<sup>3</sup>. These two combinations seem to offer similar levels of protection, with 58% and 59% of the monitors failing to meet either standard pairing. However, under the matching standard scenario, an additional 9% or 8% of monitors would fail to meet the paired standards, thus providing more extensive protection.



Figure 6. Estimated percent of U.S. FRM monitors ( $n = 1137$ ) that attain or miss alternative 24-hr 98th percentile (25–45 µg/m<sup>3</sup>) and annual (11-15  $\mu$ g/m<sup>3</sup>) PM<sub>2.5</sub> standards ( $\mu$ g/m<sup>3</sup>; 2000-2002 FRM country-wide monitors).

In addition, a significant percentage of monitors that do not meet the controlling standard but that do meet the less-stringent backstop standard would potentially realize additional health benefits by complying with a morestringent equivalent standard. For example, within the  $40/12 \mu g/m^3$  standards combination, there is a 43% difference between those monitors that miss the backstop 24-hr (98th percentile; 2%) and those that miss the controlling annual (45%); within the  $30/14 \mu g/m^3$  combination, there is a 22% difference between the controlling 24-hr (98th percentile; 24%) and backstop annual (2%). Alternatively, the difference between equivalent 30/12  $\mu$ g/m<sup>3</sup> standards is only 1%. In this manner, equivalent or matching standards have the effect of minimizing the wide variation of short- and long-term  $PM_{2.5}$  concentrations within the EPA staff recommended standard range on both 24-hr and annual time scales from 22 to 43% to 1% across the FRM  $PM_{2.5}$  network, thus ensuring consistent and uniform protection for both standard time scales.

Figures 7 and 8 show how unmatched 24-hr and annual standards may lead to inadequate protection of populations. Whereas a controlling standard can ensure protection across its respective 24-hr or annual time scale, the companion noncontrolling or backstop standard will allow a wide variation of either short- or long-term exposures to occur. In Figure 7, the *x*-axis represents 24-hr ranges of 5  $\mu$ g/m<sup>3</sup> centered about integer mass values from 23 to 52  $\mu$ g/m<sup>3</sup>. Six annual average range categories are used to create the bar chart; each bin is centered around annual levels (11–15) in  $1-\mu g/m^3$  intervals. The *y*-axis gives the percentage of monitors in each annual

range that fall in each 24-hr range on the *x*-axis. In Figure 8, the *x*-axis represents annual ranges of 1  $\mu$ g/m<sup>3</sup> centered about integer mass values from 8 to 20. Six 24-hr average range categories are used to create the bar chart; five of the bins are centered around 24-hr levels in  $5$ -µg intervals (25, 30, 35, 40, and 45) with a sixth bin representing values  $\geq$ 48 µg/m<sup>3</sup>. The *y*-axis gives the percentage of monitors in each 24-hr range that fall in each annual range on the *x* axis.

With respect to EPA staff–recommended  $PM_{2.5}$  standard ranges, about two-thirds of U.S. sites in Figure 7 with an annual range of 11.5–12.49  $\mu$ g/m<sup>3</sup> experience 24-hr averages between 28 and 42  $\mu$ g/m<sup>3</sup>. An additional 6% of U.S. sites are  $>42 \mu g/m^3$ . In Figure 8, about one-half of U.S. sites with a 24-hr range of  $28-32 \mu g/m^3$  experience annual averages between 11.5 and  $14.49 \mu g/m^3$ . An additional 11% of U.S. sites are  $>$ 14.5  $\mu$ g/m<sup>3</sup>. This indicates that within EPA staff-recommended 24-hr and annual standard combinations, neither standard alone is sufficient to constrain both short- and long-term  $PM_{2.5}$  concentrations across a substantial percentage of the monitoring network. Matching 24-hr and annual standard levels, however, would effectively constrain the upper distributions of 24-hr and annual ranges, thereby providing more-uniform protection across the country.

The preceding figures suggest that within EPA staff and CASAC recommended standard ranges, an optimal pairing occurs with a 24-hr (98th percentile)/annual standard combination of  $30/12 \mu$ g/m<sup>3</sup>. This analysis finds that an appropriately stringent 24-hr standard and an appropriately stringent annual standard, when combined with

Table 3. Estimated percentage of U.S. FRM monitors ( $n = 1137$ ) that attain or miss alternative 24-hr 98th percentile (30, 40  $\mu$ g/m<sup>3</sup>) and annual (12, 14  $\mu$ g/m<sup>3</sup>) PM<sub>2.5</sub> standards ( $\mu$ g/m<sup>3</sup>) (2000–2002 FRM country-wide monitors).

<b>Standard Metric</b>	Annual $(\mu g/m^3)$	24-hr $(\mu q/m^3)$	Attains Both (%)	Misses Annual (%)	Misses Both (%)	Misses 24-hr $(\%)$
Controlling annual		40	42	45		
Matching standards		30	33	10	46	
Controlling 24-hr		30	41		33	24



Figure 7. Frequency of alternative 24-hr (98th percentile) and annual PM<sub>2.5</sub> levels for monitoring sites in United States (µg/m<sup>3</sup>; 2000–2002 FRM country-wide monitors).

equivalence, appear to provide superior protection compared with other standard levels and combinations throughout the entire distribution of the  $PM_{2.5}$  FRM monitoring network.

#### **24-hr and Subdaily Averaging Metrics**

Although current  $PM<sub>2.5</sub>$  standards are intended to protect populations from both short-term and long-term exposures, the use of 24-hr average and annual mean metrics may require reevaluation because of the growing body of studies finding effects associated with exposure periods -24 hr (e.g., 1 hr to several hours) and characterizing high subdaily excursions.39,40,41,42 At this time, the EPA NAAQS review is not considering a primary  $PM_{2.5}$  subdaily standard. Therefore, in the absence of a subdaily standard option, the question arises as to what extent a 24-hr or annual  $PM_{2.5}$  standard will provide protection against peak excursions experienced by populations on subdaily hourly scales.

We conducted an exploratory analysis of  $PM_{2.5}$  continuous data from Region 1, 2, and bordering states continuous monitoring sites (2001 and 2002) to assess the relationship between 24-hr concentrations and subdaily concentrations, assuming that the annual metric is less effective at controlling the distribution of maximum

 $PM_{2.5}$  levels.<sup>43</sup> The study found that increasingly stringent 24-hr average standards will lower subdaily maximum hourly average levels, as depicted in Figure 9. The figure shows the distribution of maximum 3-hr averages associated with 24-hr averages within a discrete range for year-round 2001 values. The cumulative frequency of the 3-hr maximum values is plotted for each of the 24-hr average bins centered around  $5-\mu g/m^3$  breakpoints of 15, 20, 25, 30, 35, and 40, with the number of days about each of these values in parentheses in the figure legend. The solid horizontal line demarks a 1-day/week frequency. An estimate of the 3-hr maximum level experienced at a monitor once per week can be read from the graph by dropping a vertical line from the intersection of the horizontal solid line with the 24-hr average cumulative curve. For example, the line that represents days around a 19 –21  $\mu$ g/m<sup>3</sup> 24-hr average will experience a 3-hr maximum level of  $\geq$ 38  $\mu$ g/m<sup>3</sup> once per week. The analysis was also conducted for 1-, 4-, and 6-hr averages (data not presented), finding structurally similar behavior.

The analysis of continuous  $PM<sub>2.5</sub>$  data also found that the 24-hr average metric smoothes subdaily peak  $PM_{2.5}$ levels across the entire distribution of 24-hr levels, thereby masking exposure variability during low, moderate, and high 24-hr average time periods. Figure 10 plots



Figure 8. Frequency of alternative annual and 24-hr (98th percentile) PM<sub>2.5</sub> levels for monitoring sites in United States (µg/m<sup>3</sup>; 2000–2002 FRM country-wide monitors).



**Figure 9.** Relationship of maximum 3-hr and 24-hr (98th percentile) PM<sub>2.5</sub> averages (µg/m<sup>3</sup>; 2001 Region 1, 2, and border state continuous monitoring sites).

ascending 24-hr averages relative to corresponding 1-hr and 6-hr maximum averages between May and September 2002 from 39 sites in the Northeast and adjacent states, illustrating the limitations of conceiving of peak exposures in terms of 24-hr averaging periods. Only summertime values were plotted to enhance visual resolution, although similar behavior was observed for wintertime data. The figure suggests that a more realistic conception of exposure might characterize peak exposures by minutes or hours, because receptors may experience a series of episodic bursts throughout a 24-hr period depending on their activity patterns and proximity to sources.

Figures 9 and 10 indicate that subdaily peak concentrations across individual days are frequent across the entire range of 24-hr average concentration days at Northeast urban sites. This finding is in contrast to the conventional characterization of 24-hr peak concentrations as being limited and infrequent across the total distribution of low, medium, and high concentration days.32,33 Given these results, a 24-hr averaging metric, although capable of reducing maximum hourly averages, may not be the most effective and efficient way to control subdaily peaking.

## **DISCUSSION**

The current PM NAAQS review process charged to select  $PM<sub>2.5</sub>$  primary standards that are adequate to protect public health delineates a range of 24-hr and annual standards recommended by EPA staff and CASAC. This study has attempted to contribute to the understanding of how the combination and the stringency of the level and form of various 24-hr and annual standards can be selected to protect exposed populations. The analysis also assesses the extent to which current standard 24-hr averaging metrics can protect populations from subdaily exposures.

With respect to the Northeast study area (New England, New Jersey, New York, and border state monitors), a central study finding is that the final selection of  $PM_{2.5}$ NAAQS could result in a modest or substantial percentage of the Northeast population benefiting from revised standards through increased nonattainment designations triggering more stringent pollution control measures. Whether the size of populations protected is modest or substantial depends on how stringent the 24-hr and annual standard levels are. Because 24-hr  $PM_{2.5}$  values in the Northeast cluster within the 30–35  $\mu$ g/m<sup>3</sup> range, the population of the region would receive minimal additional protection unless a 24-hr 98th percentile standard were



Figure 10. Maximum 1-, 6-, and 24-hr PM<sub>2.5</sub> concentrations (µg/m<sup>3</sup>; 2002 summertime from 39 Northeast monitoring sites in Region 1, 2, and border states).

tightened from the current 65  $\mu$ g/m<sup>3</sup> to <40  $\mu$ g/m<sup>3</sup>, were the 15  $\mu$ g/m<sup>3</sup> annual standard retained. As shown in Figures 3 and 4, across the current recommended annual standard range  $(12-15 \text{ }\mu\text{g/m}^3)$  the most substantial impact on Northeast nonattainment status would occur were the 24-hr (98th percentile) standard lowered to  $\leq 30$  $\mu$ g/m<sup>3</sup> and annual concentration lowered to 12  $\mu$ g/m<sup>3</sup>.

If standards are selected at the less-stringent end of the EPA staff and CASAC recommended range, the Northeast region will not benefit as widely as the nation as a whole. Were the annual standard set between the EPA staff 12–14  $\mu$ g/m<sup>3</sup> range, the difference between selecting a 24-hr (98th percentile) standard of 30  $\mu$ g/m<sup>3</sup> and 35  $\mu$ g/m<sup>3</sup> in the Northeast amounts to a 16 –48% difference in population living in areas that would not meet the new standards. This difference is less pronounced for the United States, where only 7–17% of the population live in areas that would not meet the new standards. Within the more narrowly recommended standard ranges of the CASAC, the difference between a 24-hr (98th percentile) standard of 30  $\mu$ g/m<sup>3</sup> and 35  $\mu$ g/m<sup>3</sup> within the annual standard range of 13–14  $\mu$ g/m<sup>3</sup> is a 37–48% increase in the Northeast and 12–17% in the United States of populations living in areas that would not meet the new standards. The consequences of this disparity in protection are of public health concern to the Northeast, because the majority of the populations of the region that would benefit from more-stringent standards live in the most densely populated region of the United States, an urban corridor that experiences the highest  $PM_{2.5}$  concentrations of the region.<sup>5</sup> A 30/12  $\mu$ g/m<sup>3</sup> 24-hr (98th percentile)/annual standard would result in more congruent protection across the country, with 84% of the Northeast and 78% of the U.S. populations living in areas that would not meet the new standards.

This study also found that a standard-setting approach that selects matching or equivalent standards would ensure the broadest possible protective coverage in most U.S. areas given the substantial variability of concentrations across the country where 24-hr and annual averages are not well-correlated. Within the recommended range of standards, findings indicate that an appropriately stringent 24-hr standard and an appropriately stringent annual standard, such as  $30/12 \mu g/m^3$  (98th percentile), when combined, together provide superior protection throughout the entire U.S. distribution of PM<sub>2.5</sub> 24-hr and annual levels.

Conversely, the former 1997 NAAQS decision by the EPA setting the current  $65/15 \mu g/m^3$  24-hr (98th percentile)/annual  $PM_{2.5}$  standard used a controlling and backstop approach, wherein the annual standard controlled the distribution of measured concentrations while the 24-hr standard served as a weaker or backstop standard to limit peak 24-hr average concentrations. Although both standards can effectively shift the low- and middle-range  $PM_{2.5}$  levels within the total distribution curve, these mismatched standards have permitted areas with high 24-hr-to-annual mean  $PM_{2.5}$  ratios to experience levels at which health effects occur when the backstop standard fails to constrain  $PM_{2.5}$  concentrations.

The selection of 24-hr standard percentile forms also has bearing on the level of public health protection afforded by recommended  $PM_{2.5}$  standards. During the 1997 and current PM NAAQS review, two competing factors were considered when deciding whether to choose a 98th percentile form or a 99th form. The first factor relates to the importance of a more stable metric in minimizing year-to-year exceedances as they pertain to determining the attainment status of an area, which a 98th percentile form evidently offers. The second factor relates to the importance of providing public health protection from peak  $PM_{2.5}$  concentrations, especially at sites with periodic high seasonal peaks, source-oriented peaks, and localized hot spots. Presumably, reducing the number of excluded peak 24-hr average days that populations are exposed to would benefit public health, such that a 99th percentile form would be more protective than a 98th percentile form.

As shown in Table 2, in the Northeast United States, the majority ( $\sim$ 65%) of excluded days above both 24-hr average 98th and 99th percentile form levels are within 5  $\mu$ g/m<sup>3</sup> of this level. This finding indicates that peak 24-hr concentrations typically lie close to the standard cut point and suggests that either percentile form can control  $PM_{2.5}$  24-hr maxima. However, in Figures 2 and 3, the 99th percentile form generates a 24-hr PM<sub>2.5</sub> standard  $\sim$ 5  $\mu$ g/m<sup>3</sup> lower or more stringent than a 98th form by removing four additional peak days. Thus, to achieve an equivalent 24-hr average, a 98th percentile form 24-hr standard would need to be  $\sim$ 5  $\mu$ g/m<sup>3</sup> more stringent than a comparable 99th percentile form standard. Even if removing the 99th percentile form from consideration effectively decreases the number of 24-hr level alternatives available to decision-makers, a 98th form can offer the same level of public health protection, in terms of exposure to 24-hr levels, assuming it is comparably stringent and the range of 24-hr 98th percentile levels encompasses an absolute level of stringency provided by the 99th percentile range.

Although the current 98th percentile form is intended to balance the dual needs of limiting periodic peak values and increasing stability in 24-hr standard nonattainment designations, it is worth noting that exempted natural event peak value days have the potential to contribute significantly to  $PM<sub>2.5</sub>$  concentrations. Populations experience these real-world exposures, which are not reflected in design value calculations used to determine compliance with  $PM_{2.5}$  standards. As shown in Table 4, the impact of high peak day exemptions because of forest fires on  $PM_{2.5}$  levels was found to be significant in some areas in the Northeast study area during 2002, a year with heavy upwind forest fire activity in Canada. The table

**Table 4.** Reduction in 24-hr and annual PM<sub>2.5</sub> from peak concentration forest fire exemptions (2002 FRM Regions 1, 2, and Delaware, Washington DC, Maryland, and Pennsylvania).

<b>Reduction</b>	24-hr ( $\mu$ g/m <sup>3</sup> )	Annual $(\mu g/m^3)$	
Maximum	23.60	1.03	
95th percentile	11.56	0.74	
75th percentile	4.50	0.58	
Median	1.90	0.51	
Average	3.36	0.48	
25th percentile	0.70	0.38	
5th percentile	0.00	0.17	
Minimum	0.00	0.08	

shows the potential impact of peak  $PM_{2.5}$  concentration exemptions in reporting 24-hr and annual levels. For 129 of 192 sites that exempted  $PM_{2.5}$  data, annual means were as much as 1  $\mu$ g/m<sup>3</sup> lower and on average ~0.5  $\mu$ g/m<sup>3</sup> lower. For the 24-hr average, data removal resulted in an average change of  $\sim$ 4 µg/m<sup>3</sup>. The maximum change was  $\sim$  24  $\mu$ g/m<sup>3</sup>.

With respect to recent scientific evidence indicating that adverse health effects are associated with subdaily exposures, in the absence of a new subdaily PM NAAQS averaging metric, this study set out to determine to what extent a 24-hr metric can control subdaily excursions. The study found that suitably stringent 24-hr levels, such as a  $30-\mu g/m^3$  standard, are more effective at constraining subdaily hourly and multihourly averages than weaker 24-hr levels, as shown in Figure 9. Figure 10, however, shows how current 24-hr standard averaging metrics reduce the distribution of continuous excursions into one composite 24-hr average, leveling peak variability across all of the 24-hr average periods, regardless of concentration. These findings suggest that populations are exposed to peak subdaily levels that may contribute to aggregate health risk across much of the 24-hr average distribution, including more frequently occurring "typical" days, as well as less-regular high days. This indicates that populations could receive relatively high subdaily peak exposures even on low 24-hr average days. In addition, highrisk scenarios could occur wherein physically active outdoor populations are exposed to nearby high-source environments (e.g., roadways) during peak excursion periods (e.g., morning rush hour).

These considerations suggest that different scales of exposure should be taken into account when selecting future averaging periods for PM standards. The extent to which multiplicative subdaily peak exposures occur across the entire range of 24-hr averages over the course of days, weeks, and years may inform the toxicological and epidemiologic study of acute health events and help to connect specific activity patterns and exposure events with emissions sources. Additional research into continuous exposure variability should be conducted to determine whether a subdaily standard is more effective in protecting populations from short-term exposures. For the time being, until additional health studies based on continuous  $PM_{2.5}$  data additionally inform these initial findings, the most effective way to limit subdaily exposures is to set the most stringent 24-hr standard possible.

This analysis of  $PM_{2.5}$  standard metrics was subject to analytical limitations. With respect to the use of monitoring data, the assessment followed EPA methods by assigning the highest annual or 24-hr design values as the design values for the entire county. Likewise, for those counties without monitors, the highest annual or 24-hr interpolated levels from counties with monitors were used. This method could result in an overestimation of the number of persons exposed to  $PM_{2.5}$  concentrations at the county level. However, the study applied countylevel population estimates to include all persons in the study region. EPA currently defines attainment/nonattainment areas by consolidated metropolitan statistical areas that aggregate counties. The study did not take into account upwind areas designated as nonattainment when

estimating the percentage of populations living in counties with PM levels above standard combinations.

Application of a 3-yr dataset (2000 –2002) incorporating a wide range of monitoring sites and concentration values allowed us to establish the relationship between various  $PM_{2.5}$  standard metrics. The inclusion of additional years to the analysis likely would not materially change this relationship, unless factors driving PM concentrations across the Northeastern region were suddenly to change. Since 2002, this has not happened. Nonetheless, the percentage estimates of nonattainment areas in various 24-hr/annual standard combinations pertain to 2000 –2002 only and may not be identical to estimates generated using more recent monitoring data. Recognizing the difficulty in determining absolute population numbers or pollution levels, the study focused on establishing data structure and inherent relationships between the 24-hr and annual metric and the potential impact that these standards and their relative stringency have on the level of public health.

In conclusion, study findings show that within the EPA staff and CASAC recommended range of primary  $PM_{2.5}$  standards, the most appropriate 24-hr (98th percentile)/annual standards would be  $30/12 \mu g/m^3$ . The standard is low enough to provide a stringent level of shortand long-term protection for a substantial percentage of both the Northeast and the U.S. populations. This level of protection is justifiable, because it recognizes current unresolved issues concerning the existence or nonexistence of a  $PM<sub>2</sub>$ , health effects threshold, as well as the extent to which protection of all populations, including susceptible groups, can be protected with an adequate margin of safety. The standard also provides nearly equivalent 24-hr and annual coverage across the most monitoring areas, thereby providing a more uniform level of short- and long-term protection across the largest area possible. This finding contrasts with the current PM NAAQS controlling and backstop approach, where neither standard alone is sufficient to ensure maximum protection across broad areas of the United States. Furthermore, given recent associations of subdaily exposures and acute adverse health effects, in the absence of a subdaily averaging metric, a stringent 24-hr standard will more effectively control maximum hourly and multihourly peak  $PM_{2.5}$  levels than a weaker standard.

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