



UNITED STATES ENVIRONMENTAL PROTECTION AGENCY  
RESEARCH TRIANGLE PARK, NC 27711

OFFICE OF  
AIR QUALITY PLANNING  
AND STANDARDS

July 27, 2023

**MEMORANDUM**

**SUBJECT:** Correction to Errors in Six Correlation Coefficients in Chapter 6 of the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft*

**FROM:** Erika N. Sasser, Director  
Health and Environmental Impacts Division  
Office of Air Quality Planning and Standards  
United States Environmental Protection Agency

**TO:** Aaron Yeow, Designated Federal Officer  
Clean Air Scientific Advisory Committee  
EPA Science Advisory Board Staff Office

In my recent memo, dated July 24, 2023, I forwarded six corrected figures for the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft* (draft PA), which had been the subject of the June 28-29, 2023 public meeting of the Clean Air Scientific Advisory Committee (CASAC) Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter Secondary National Ambient Air Quality Standards (NAAQS) Panel. To assist the Panel in its deliberations, I am forwarding additional material associated with the corrected figures in 3 attachments.

The first attachment is a table providing the correlation coefficients for the six corrected figures provided with my memo of July 24, 2023. The attached table provides the corrected coefficients for those six figures as well as correlation coefficients for the new versions of each of the corrected figures presenting only East and only West ecosystem locations. As noted in my July 24 memo, the figures showing eastern and western locations are consistent with observations noted in Chapters 2 and 6 regarding distinctions between the eastern and western U.S. in PM<sub>2.5</sub> composition. More specifically, they indicate the stronger influence of nitrogen species on PM<sub>2.5</sub> concentrations in the eastern as compared to the western U.S. This table of correlation coefficients (Attachment 1) provides additional evidence of these distinctions.

The second and third attachments are pages of the draft PA with corrections to the figures, correlation coefficients and associated text. Attachment 2 is a clean version of the affected pages and Attachment 3 is a "track changes" version of the affected pages, showing the corrections.

I am requesting that you forward this memorandum and attachments to the CASAC and CASAC Panel for their consideration ahead of the public meeting to be held on September 5-6, 2023.

Should you have any questions regarding this memo, please contact me (919-541-3889; email [sasser.erika@epa.gov](mailto:sasser.erika@epa.gov)) or Karen Wesson, of my staff (919-541-3515; email [Wesson.karen@epa.gov](mailto:Wesson.karen@epa.gov)).

cc: Tom Brennan, SAB, OA  
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Steve Dutton, ORD/CPHEA  
Steve McDow, ORD/CPHEA  
Tara Greaver, ORD/CPHEA

Attachment

## ATTACHMENT 1

| Correlations*   |  |                   |                        |                         |
|---|--|-------------------|------------------------|-------------------------|
|   |  | All<br>Ecoregions | Eastern*<br>Ecoregions | Western**<br>Ecoregions |
| <i>Nitrogen Deposition (kg N/ha-yr) with specified Air Quality Metric</i>   |  |                   |                        |                         |
| NO <sub>2</sub>   | Annual Average - Weighted Average                    | 0.06              | 0.56                   | -0.13                   |
|   | Annual average - Maximum Monitor                     | -0.05             | 0.38                   | -0.07                   |
| PM <sub>2.5</sub>   | Annual Average - Weighted Average                    | 0.52              | 0.63                   | 0.24                    |
|   | Annual average - Maximum Monitor                     | 0.03              | 0.53                   | 0.16                    |
| <i>Sulfur plus Nitrogen Deposition (m-eq/ha-yr) with specified Air Quality Metric</i>   |  |                   |                        |                         |
| PM <sub>2.5</sub>   | Annual Average - Weighted Average                    | 0.63              | 0.83                   | 0.19                    |
|   | PM <sub>2.5</sub> – Annual average - Maximum Monitor | 0.12              | 0.74                   | 0.10                    |
| <p>* Correlations derived using Correl function in Excel.</p> <p>** Eastern ecoregions are those not designated as western. Western ecoregions are any ecoregion that intersections with the states of ND, SD, CO, WY, MT, AZ, NM, UT, ID, CA, OR, WA. This is the categorization of the National Atmospheric Deposition Program's Committee on Critical Loads of Atmospheric Deposition Science.</p> |  |                   |                        |                         |

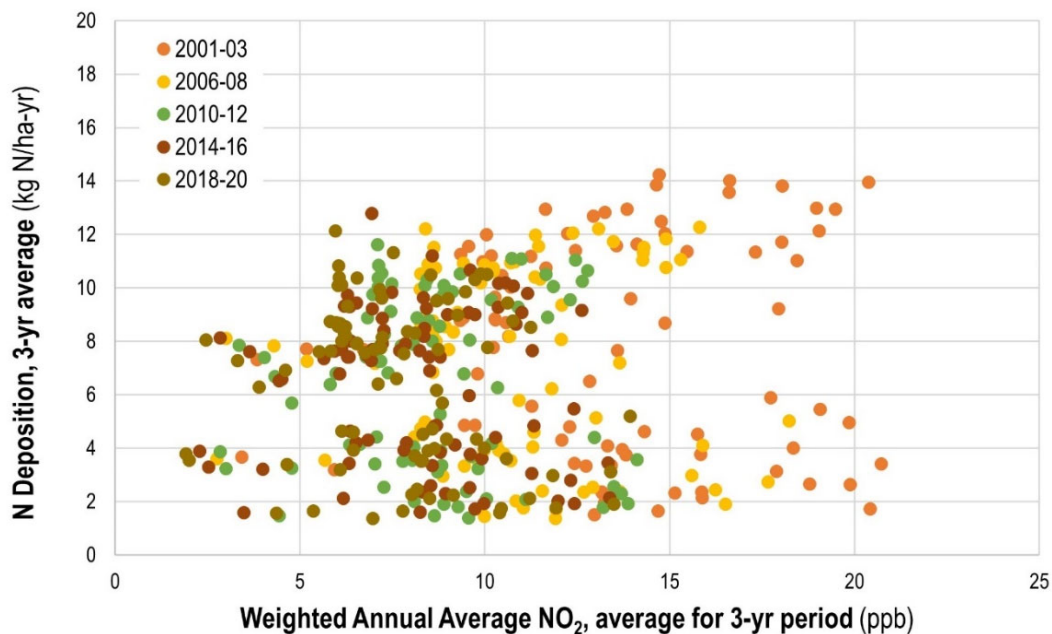
## **ATTACHMENT 2**

Corrected pages 6-30 to 6-37 and 6-41 of draft PA (*Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft*).

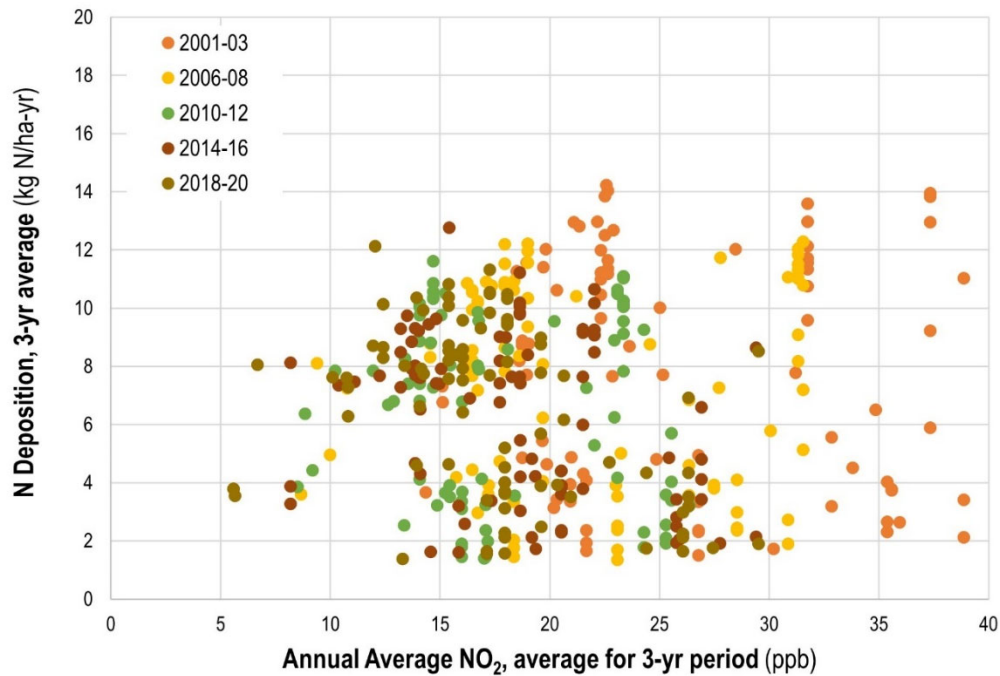
Clean Version

### 6.2.2.3 NO<sub>2</sub> Results

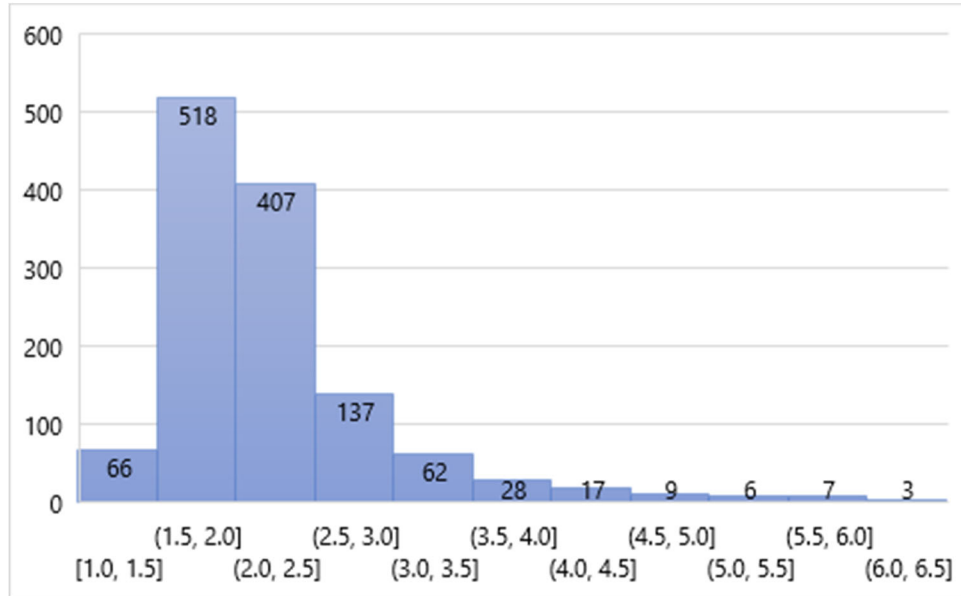
Similar analyses were completed assessing the relationship between the current secondary NO<sub>2</sub> standard (annual mean, level = 53 ppb). Based on the results of section 6.2.1, one would expect it to be less likely that the existing NO<sub>2</sub> NAAQS would be strongly correlated with N deposition (due to the multiple pathways for N deposition, including ammonia-related sources) and this expectation is confirmed. Figure 6-20 displays a comparison of 3-year average N deposition estimates (TDEP) against EAQM values for annual average NO<sub>2</sub>. While the data suggest the potential for some ecoregions with higher N depositions to be associated with higher EAQM values, the correlation coefficient is poor, particularly in comparison to what was seen for SO<sub>2</sub> ( $r = 0.06$  vs.  $r = 0.75$ ). As was also the case for SO<sub>2</sub>, Figure 6-21 illustrates that the switch to consideration of the single highest NO<sub>2</sub> DV from the set of contributing monitors, as opposed to a weighted EAQM value, further reduces the already low correlation coefficient between deposition and concentration ( $r = -0.05$  vs.  $r = 0.06$ ). The NO<sub>2</sub> ratios between maximum DVs and EAQM values typically range from 1.5 to 2.5 but can be as high as 6.5.



**Figure 6-20. Scatterplot of estimated 3-year average N deposition (ecoregion median) and the weighted secondary NO<sub>2</sub> design values from contributing upwind areas for that ecoregion (EAQM) also averaged over 3 years.**



**Figure 6-21. Scatterplot of estimated 3-year average N deposition (ecoregion median) and the secondary NO<sub>2</sub> design value over that 3-year period from the contributing monitor with the maximum value for each ecoregion.**



**Figure 6-22. Histogram of the ratio of annual average NO<sub>2</sub> concentration (ppb) averaged over a 3-year period from the contributing monitor with the maximum value for each ecoregion to the average of weighted annual average NO<sub>2</sub> design values (EAQM) over the same 3-year period.**

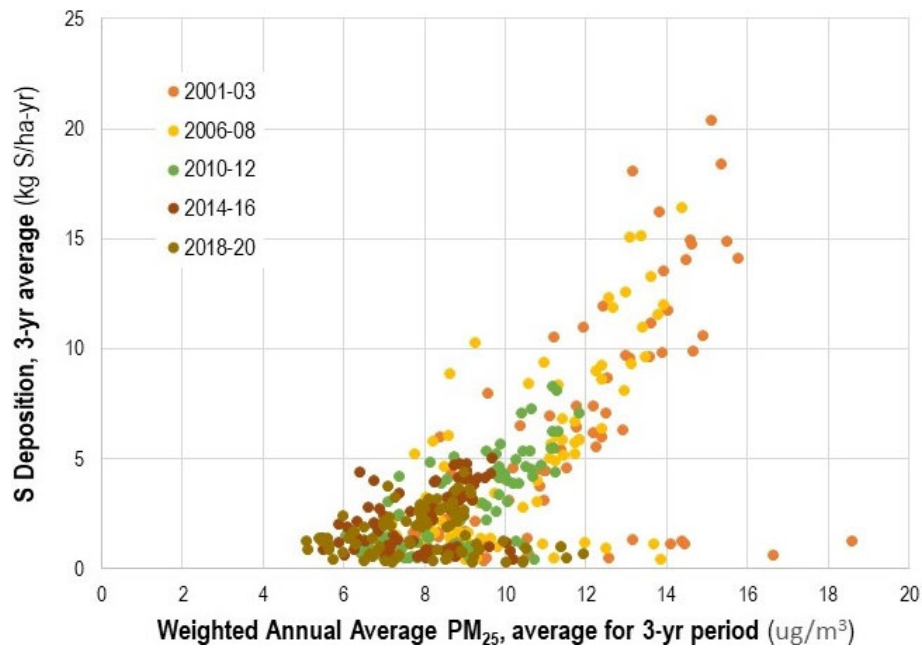
#### 6.2.2.4 PM<sub>2.5</sub> Results

Finally, similar analyses were also completed assessing the relationship between S, N, and S+N deposition and air quality design value data for the current secondary PM<sub>2.5</sub> annual standard.<sup>4</sup> Figure 6-23 shows the relationship between upwind annual average PM<sub>2.5</sub> EAQM data and S deposition levels over the usual five periods. The data points can be divided into two groups. There are a minority of data pairs where S deposition is extremely low yet PM<sub>2.5</sub> EAQM values are high. This is likely occurring in areas where the PM<sub>2.5</sub> levels are driven by components other than sulfate. Then there is a second set of data points where there is a positive association between the upwind PM<sub>2.5</sub> EAQM and downwind S deposition. Overall, the correlation for the paired data is 0.67, which falls between the range seen for the SO<sub>2</sub> and NO<sub>2</sub> EAQM data. Figure 6-24 describes the comparison between S deposition levels and the annual PM<sub>2.5</sub> DV from the highest monitor in the ecoregions' sites of influence. The correlation between these two terms is relatively low ( $r = 0.21$ ).

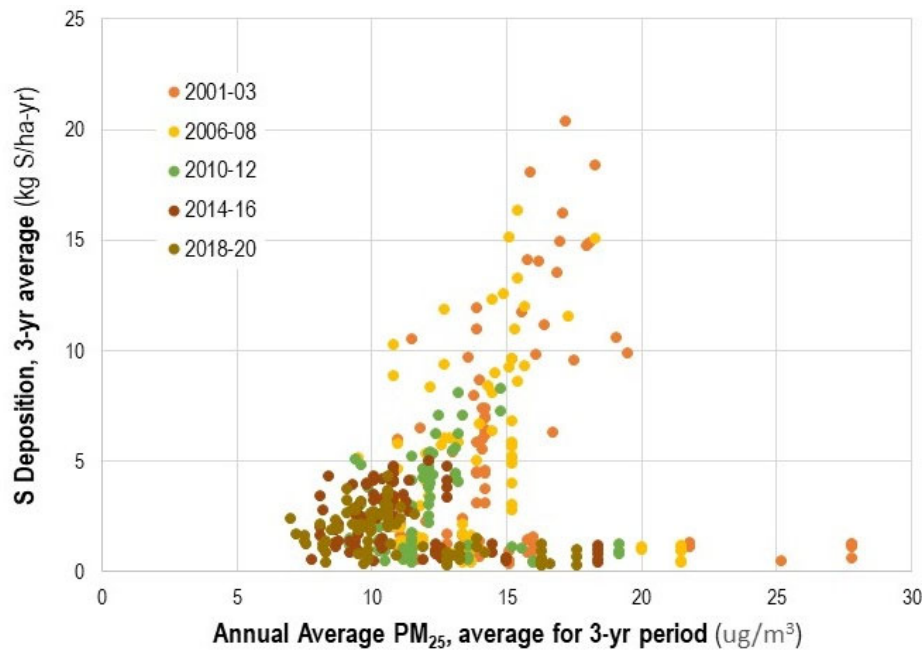
There was also an association between upwind PM<sub>2.5</sub> EAQM and downwind N deposition ( $r = 0.52$ ), as shown in Figure 6-25. This correlation was diminished ( $r = 0.03$ ) when moving from the weighted EAQM to use of the maximum PM<sub>2.5</sub> DV from the highest monitor in the ecoregions' sites of influence (Figure 6-26). As shown in Figure 6-27, the ratios between the maximum PM<sub>2.5</sub> DV in an ecoregion's sites of influence and the weighted EAQM value typically ranges from 1.11 to 1.66. Finally, Figures 6-28 and 6-29 illustrate the relationship between PM<sub>2.5</sub> design values and total S+N deposition. The data indicate correlation between PM<sub>2.5</sub> EAQM data and total S+N deposition ( $r = 0.63$ ), but less correlation with the maximum DV ( $r = 0.12$ ).

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<sup>4</sup> Given the cumulative nature of N and S deposition, it was expected that an air concentration metric with a longer averaging time would be a more appropriate potential indicator of downwind deposition, thus the EPA restricted the PM<sub>2.5</sub> analysis to the annual standard and did not include analyses for the 24-hour standard.

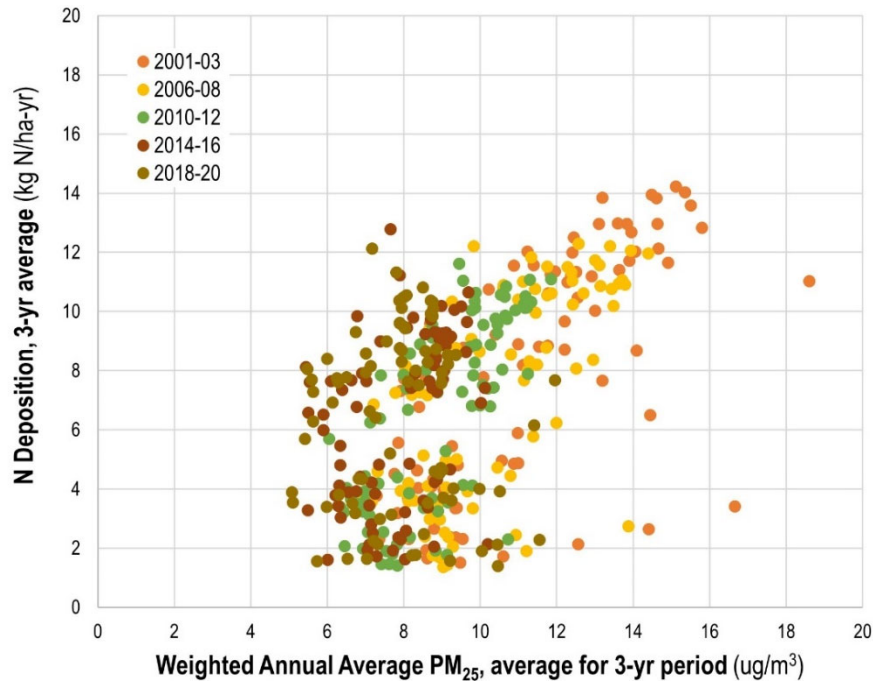


**Figure 6-23.** Scatterplot of estimated 3-year average S deposition (ecoregion median) and the weighted annual average PM<sub>2.5</sub> design values from contributing upwind areas for that ecoregion (EAQM) also averaged over 3 years.

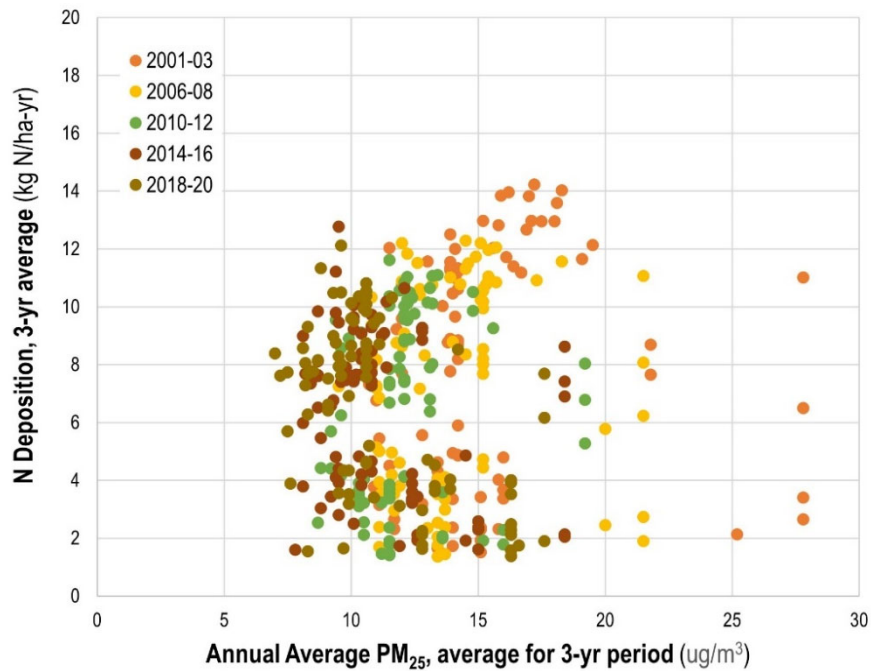


**Figure 6-24.** Scatterplot of estimated 3-year average S deposition (ecoregion median) and the average annual PM<sub>2.5</sub> design value over that 3-year period from the contributing monitor with the maximum value for each ecoregion.

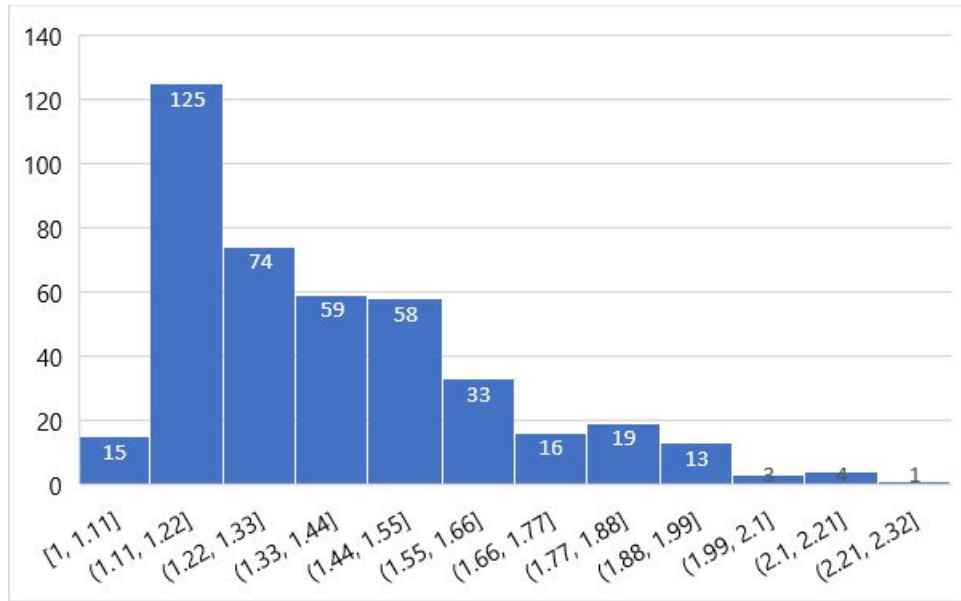




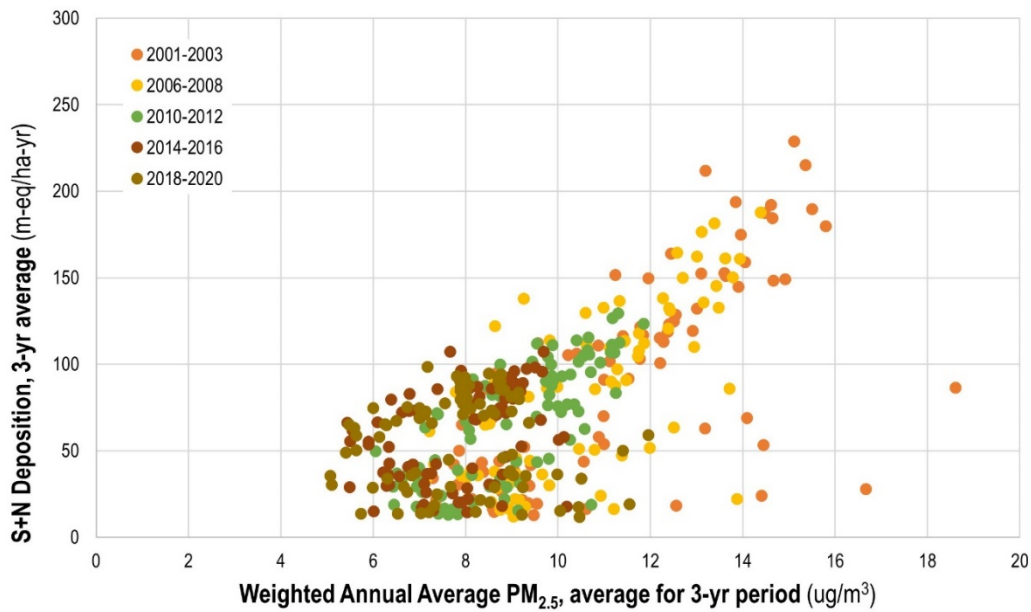
**Figure 6-25. Estimated 3-year average N deposition (ecoregion median) and average of weighted annual average PM<sub>2.5</sub> concentrations in 3-year period (EAQM) for that ecoregion.**



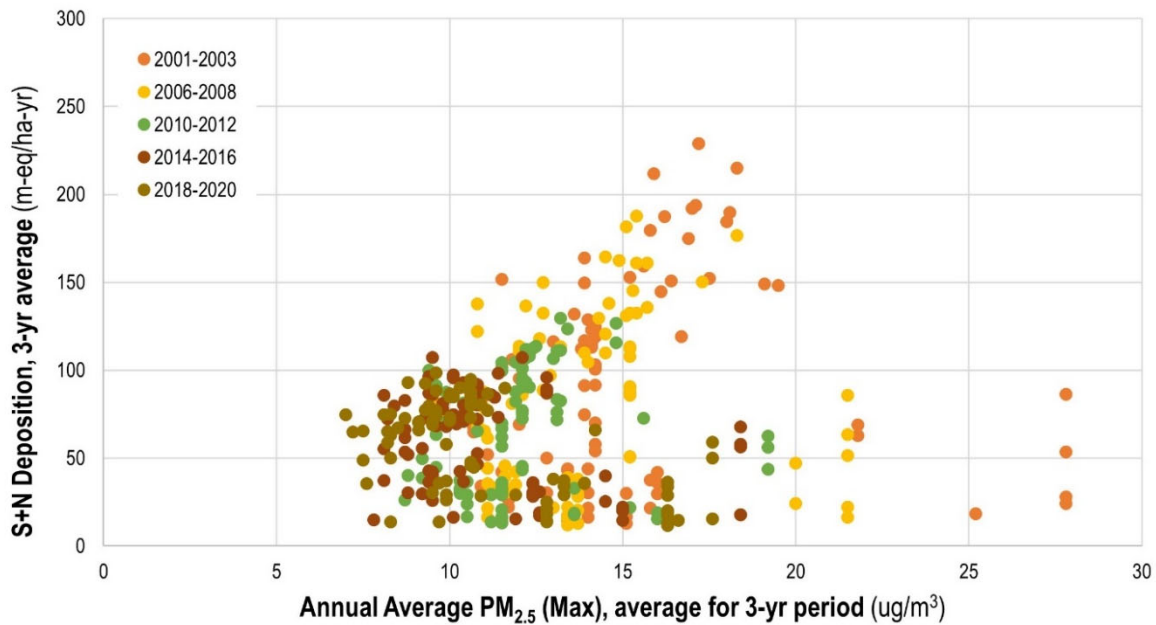
**Figure 6-26. Estimated 3-year average N deposition (ecoregion median) and annual average PM<sub>2.5</sub> concentration in 3-year period from maximum contributing monitor for that ecoregion.**



**Figure 6-27. Histogram of the ratio of average annual average PM<sub>2.5</sub> concentration (µg/m<sup>3</sup>) in 3-year period from maximum contributing monitor for that ecoregion to the average of weighted annual average PM<sub>2.5</sub> concentrations (EAQM) in 3-year period (median = 1.3).**



**Figure 6-28. Estimated 3-year average S+N deposition (ecoregion median) and average of weighted annual average PM<sub>2.5</sub> concentrations in 3-year period (EAQM) for that ecoregion.**



**Figure 6-29. Estimated 3-year average S+N deposition (ecoregion median) and average annual average PM<sub>2.5</sub> concentration in 3-year period from maximum contributing monitor for that ecoregion.**

#### 6.2.2.5 Conclusions

For SO<sub>2</sub>, we examined both the 2<sup>nd</sup> highest 3-hour maximum and an annual average metric. The results for the EAQM suggest that both metrics are correlated with S deposition, with the stronger association being for the annual average metric. There is lower correlation between the design values from the highest monitor within the ecoregion sites of influence for both the 2<sup>nd</sup> highest 3-hour maximum and an annual average SO<sub>2</sub> metrics. As shown by the ratio information, this is likely due to the large concentration gradients seen across the SO<sub>2</sub> monitors in the U.S. (for example, see Figure 2-23), with the maximum contributing monitor between generally 3 to 4 times higher than the EAQM. These figures also show that in the most recent assessed time period of 2018-2020, the median S deposition in the Ecoregion III areas was below 5 kg/ha-yr when the annual average SO<sub>2</sub> concentration, averaged over three years, at contributing monitors was less than 22 ppb and the majority of monitors were below 10 ppb. Additionally, the SO<sub>2</sub> figures indicate that there can be high measured SO<sub>2</sub> concentrations associated with low S deposition (i.e., < 5 kg S/ha-yr) and that there is generally more scatter in the data at lower deposition values. Both of these observations could be due to uncertainties in the TDEP calculations, uncertainties in our assessment methodology and/or a lack of correlation between some SO<sub>2</sub> monitor measurements and S deposition.

For NO<sub>2</sub>, the correlations between the measured annual NO<sub>2</sub> concentrations and N deposition are not as strong as they are between metrics for SO<sub>2</sub> concentrations and S deposition. This could be partially due to the fact that oxidized nitrogen only contributes to part of the total N deposition estimate, and as discussed in section 2, the contribution of reduced nitrogen to total N deposition has grown over the last few decades (*e.g.*, Li et al., 2016). The figures also show slightly less variability between the EAQM and maximum monitor concentrations for NO<sub>2</sub> (when compared to SO<sub>2</sub>), with the NO<sub>2</sub> maximum monitored values being typically about twice as high as the calculated EAQM. This result suggests less variability and smaller gradients in measured NO<sub>2</sub> concentrations across the U.S. when compared to SO<sub>2</sub>. In the most recent time period (2018-2020), median N deposition was generally maintained at 12 kg/ha-yr in Ecoregion III areas while NO<sub>2</sub> annual average, averaged over 3-years, monitored values were 30 ppb or less.

For PM<sub>2.5</sub>, the assessment looks at correlations with S deposition, N deposition and S + N deposition. The results show a correlation ( $r=0.52$ ) between measurements of annual average PM<sub>2.5</sub> and estimates of N deposition. This could be due to measurements at PM<sub>2.5</sub> monitors including both oxidized and reduced forms of N (*i.e.*, NO<sub>3</sub> and NH<sub>4</sub><sup>+</sup>), which contribute together to total N deposition. A similar correlation is observed between measurements of annual average PM<sub>2.5</sub> and estimates of S deposition ( $r = 0.67$ ). However, the results include data where the measured PM<sub>2.5</sub> mass is high when S deposition is low (*i.e.*, < 2 kg S/ha-yr). This is similar to data seen in the figures assessing S deposition and SO<sub>2</sub> air quality metrics. This could also be due to PM<sub>2.5</sub> mass at these contributing monitors having a large fraction of non-S-containing compounds, such as NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and/or organic carbon (OC). In looking at the relationship between measurements of annual average PM<sub>2.5</sub> and estimates of S+N deposition<sup>5</sup>, the results show similar correlation ( $r=0.63$ ). For measurements of annual average PM<sub>2.5</sub> there is less difference between the EAQM metric and the maximum monitor concentrations for annual average PM<sub>2.5</sub>. In the most recent time period (2018-2020), PM<sub>2.5</sub> annual average, averaged over 3-years, contributing monitored values were less than 18 µg/m<sup>3</sup> and mostly less than 15 µg/m<sup>3</sup>, corresponding to N and S deposition of approximately 6-12 kg N/ha-yr and <5 kg S/ha-yr, respectively.

### 6.3 AIR QUALITY METRICS FOR CONSIDERATION

Based on the information above, this section discusses how well various air quality metrics relate to S and N deposition. Section 6.2.1 examines this relationship in important

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<sup>5</sup> Total deposition is converted to units of milli-equivalent using the following equation: S+N deposition = (6.25\*S deposition) + (7.14\*N deposition).

assessed time period of 2018-2020, the median S deposition in the Ecoregion III areas was maintained below 5 kg/ha-yr when the annual average SO<sub>2</sub> concentration at contributing monitors, averaged over three years, was less than 22 ppb. The majority of monitors were below 10 ppb.

### 6.3.2 NO<sub>2</sub> and PM<sub>2.5</sub> Metrics

For N, the results in section 6.2.1 suggest that oxidized N deposition in rural areas is mostly from deposition of air concentrations of nitric acid and particulate nitrate, rather than NO<sub>2</sub>. Additionally, the results suggest that in some areas inorganic nitrogen (e.g., NH<sub>4</sub><sup>+</sup>) contributes to the N deposition, with higher contributions in areas near emission sources of NH<sub>3</sub>.

Section 6.2.2 examines the current form and averaging time of the NO<sub>2</sub> secondary NAAQS which is the annual average NO<sub>2</sub> concentration. As in the assessments of the other pollutants and air quality metrics, the analyses also focus on a 3-year average of NO<sub>2</sub> and N deposition and include multiple years of data to better assess more typical relationships. For NO<sub>2</sub>, the correlations between annual average NO<sub>2</sub> and N deposition were poor (r=0.06 for EAQM). In addition, the ratios between the maximum contributing monitor and the EAQM show variability, though less than was seen for SO<sub>2</sub>, across the measured annual average concentrations of NO<sub>2</sub> across the U.S., with a median ratio of 2. The correlation between annual average PM<sub>2.5</sub> and N deposition was stronger (r=0.52 for EAQM). This is likely due to HNO<sub>3</sub>, NO<sub>3</sub> and NH<sub>4</sub><sup>+</sup> being the largest contributors to N deposition and being most closely related to concentrations of PM<sub>2.5</sub>. Additionally, the ratios between the maximum contributing monitors and the EAQM are lower for PM<sub>2.5</sub> (compared to SO<sub>2</sub> and NO<sub>2</sub>) with ratios closer to 1 suggesting lower variability of annual average PM<sub>2.5</sub> across the U.S. Given this information and these relationships, the PM<sub>2.5</sub> annual average, averaged over three years, might be the better air quality metric to control N deposition. Such a metric would also provide some control over S deposition, as seen in the figures above. However, it is important to consider that this analysis focuses on PM<sub>2.5</sub> monitors that contribute to the S and N deposition across the U.S. and that these monitors (and other PM<sub>2.5</sub> monitors) also measure other non-S and N related pollutants as part of the PM<sub>2.5</sub> total mass.

### 6.3.3 Key Uncertainties and Limitations

The linkage between air concentration and deposition can vary based on site-specific conditions, including the chemical form of nitrogen and sulfur, frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity. The analyses above attempt to provide insight into these relationships and variability for multiple measured air quality metrics. As with any assessment, there are uncertainties and limitations associated with the work,

### ATTACHMENT 3

Corrected pages 6-30 to 6-37 and 6-41 of draft PA (*Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft*).

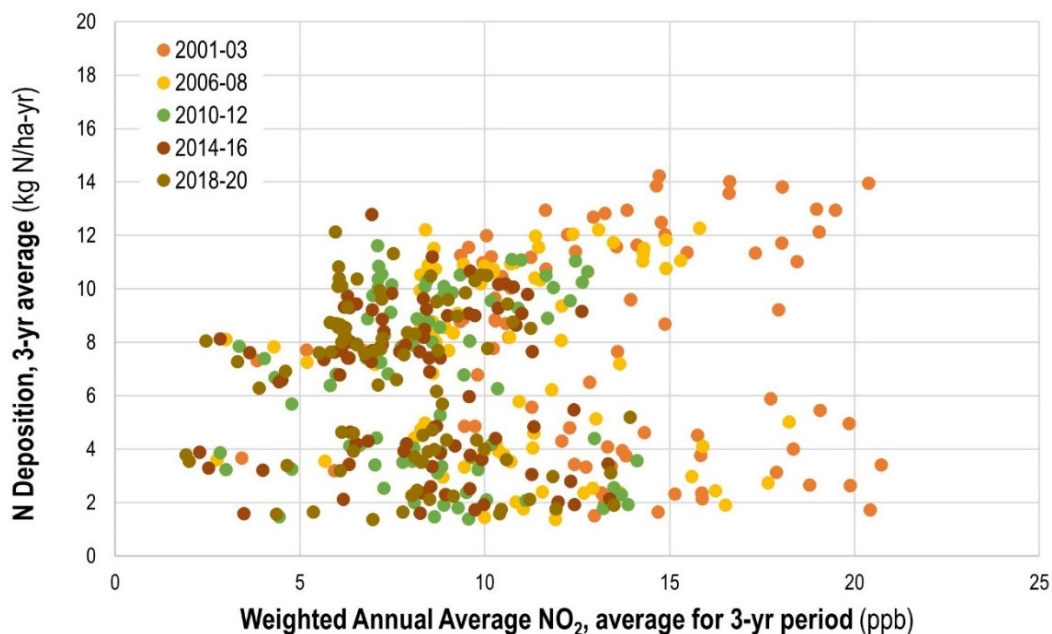
“Track Changes” Version<sup>1</sup>

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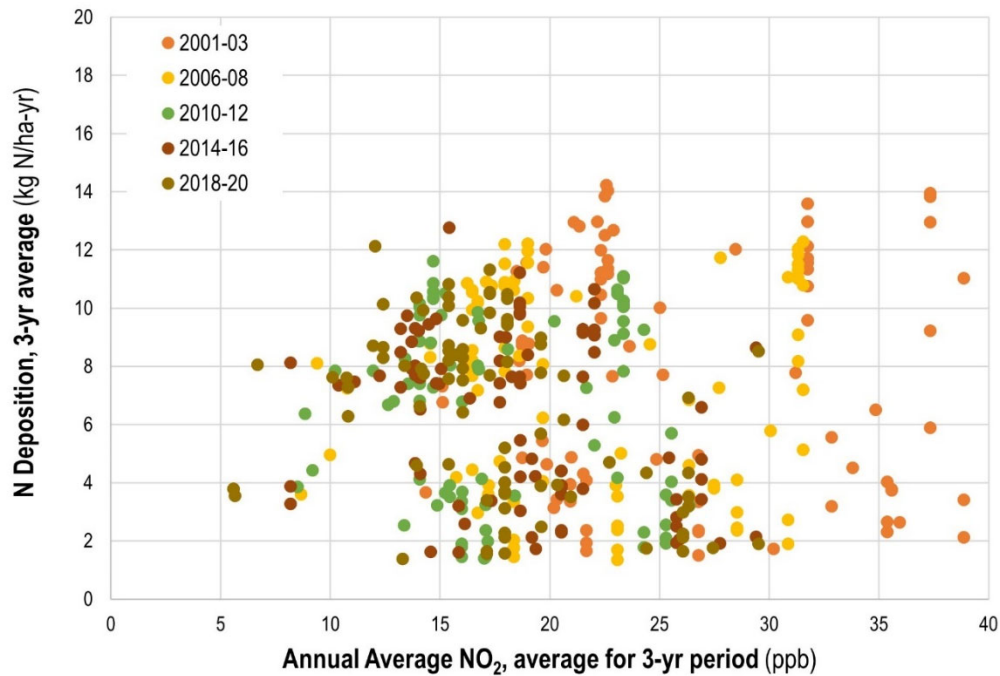
<sup>1</sup> To avoid potential for confusion with paging, this version shows, as inserted, the corrected figures and does not show the deleted incorrect figures.

### 6.2.2.3 NO<sub>2</sub> Results

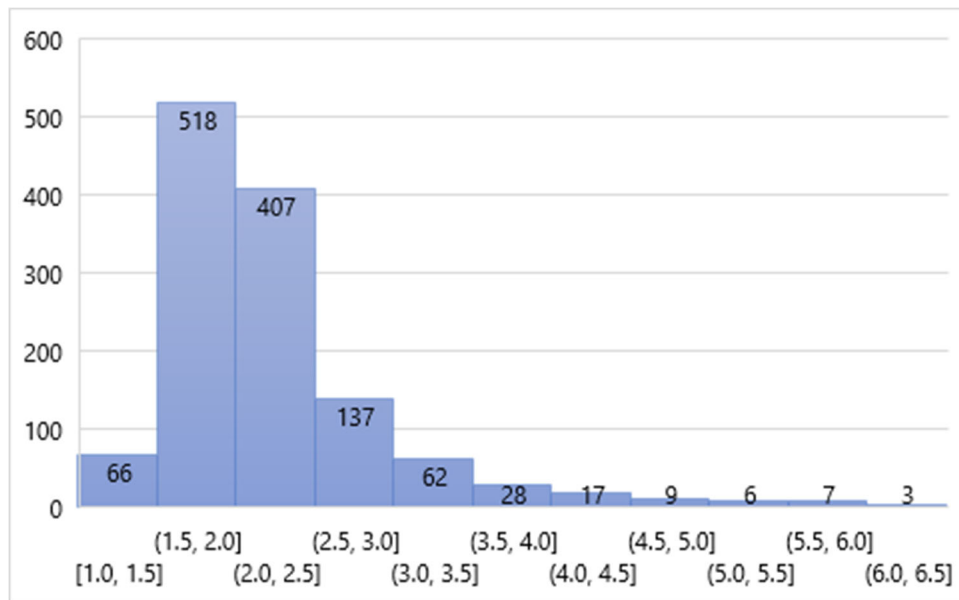
Similar analyses were completed assessing the relationship between the current secondary NO<sub>2</sub> standard (annual mean, level = 53 ppb). Based on the results of section 6.2.1, one would expect it to be less likely that the existing NO<sub>2</sub> NAAQS would be strongly correlated with N deposition (due to the multiple pathways for N deposition, including ammonia-related sources) and this expectation is confirmed. Figure 6-20 displays a comparison of 3-year average N deposition estimates (TDEP) against EAQM values for annual average NO<sub>2</sub>. While the data suggest ~~that~~ the potential for some ecoregions with higher N depositions ~~to be~~ associated with higher EAQM values, the correlation coefficient is poor, particularly in comparison to less strong than what was seen for SO<sub>2</sub> ( $r = 0.0658$  vs.  $r = 0.75$ ). ~~However, unlike SO<sub>2</sub>, the positive association appears to extend throughout the distribution of N deposition levels; that is, the correlation between deposition and EAQM is similar whether N deposition values are greater than, or less than, for example 10 kg/ha-yr.~~ As was also the case for SO<sub>2</sub>, Figure 6-21 illustrates that the switch to consideration of the single highest NO<sub>2</sub> DV from the set of contributing monitors, as opposed to a weighted EAQM value, further slightly reduces the already low correlation coefficient between deposition and concentration ( $r = -0.0535$  vs.  $r = 0.0658$ ). The NO<sub>2</sub> ratios between maximum DVs and EAQM values typically range from 1.5 to 2.5 but can be as high as 6.5.



**Figure 6-20. Scatterplot of estimated 3-year average N deposition (ecoregion median) and the weighted secondary NO<sub>2</sub> design values from contributing upwind areas for that ecoregion (EAQM) also averaged over 3 years.**



**Figure 6-21. Scatterplot of estimated 3-year average N deposition (ecoregion median) and the secondary NO<sub>2</sub> design value over that 3-year period from the contributing monitor with the maximum value for each ecoregion.**



**Figure 6-22. Histogram of the ratio of annual average NO<sub>2</sub> concentration (ppb) averaged over a 3-year period from the contributing monitor with the maximum value for each ecoregion to the average of weighted annual average NO<sub>2</sub> design values (EAQM) over the same 3-year period.**

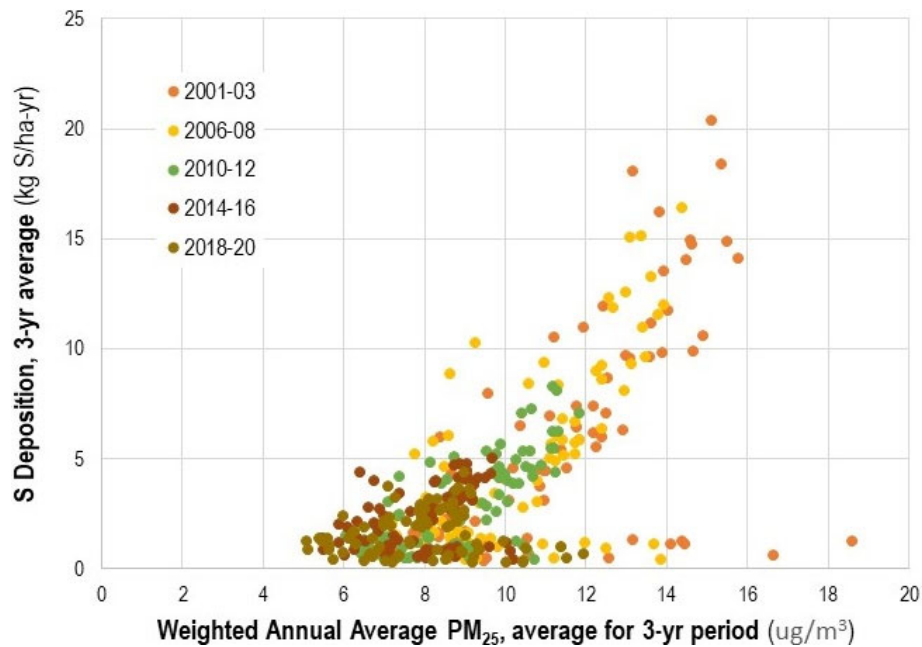


#### 6.2.2.4 PM<sub>2.5</sub> Results

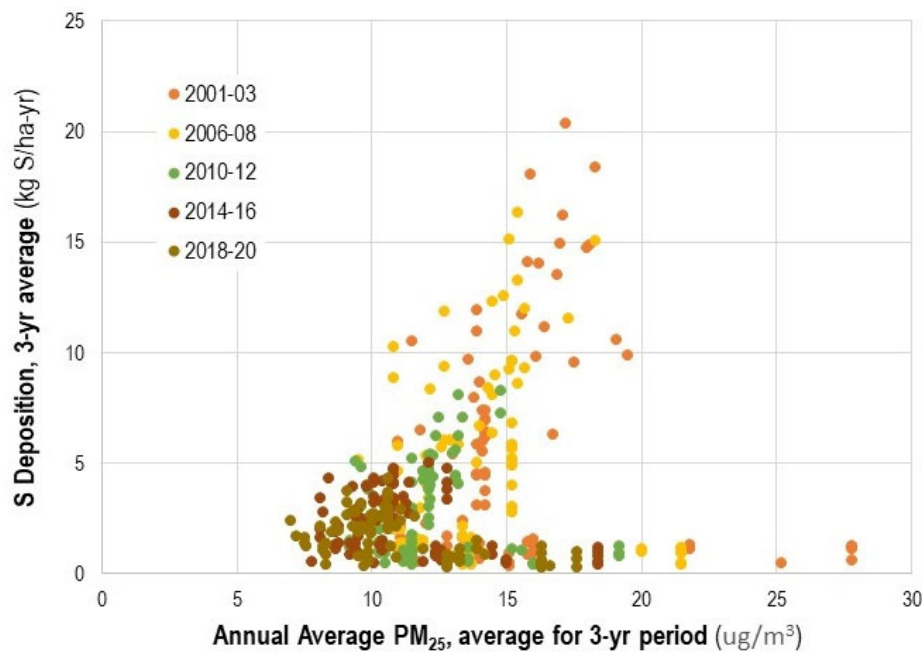
Finally, similar analyses were also completed assessing the relationship between S, N, and S+N deposition and air quality design value data for the current secondary PM<sub>2.5</sub> annual standard.<sup>4</sup> Figure 6-23 shows the relationship between upwind annual average PM<sub>2.5</sub> EAQM data and S deposition levels over the usual five periods. The data points can be divided into two groups. There are a minority of data pairs where S deposition is extremely low yet PM<sub>2.5</sub> EAQM values are high. This is likely occurring in areas where the PM<sub>2.5</sub> levels are driven by components other than sulfate. Then there is a second set of data points where there is a positive association between the upwind PM<sub>2.5</sub> EAQM and downwind S deposition. Overall, the correlation for the paired data is 0.67, which falls between the range seen for the SO<sub>2</sub> and NO<sub>2</sub> EAQM data. Figure 6-24 describes the comparison between S deposition levels and the annual PM<sub>2.5</sub> DV from the highest monitor in the ecoregions' sites of influence. The correlation between these two terms is relatively low ( $r = 0.21$ ).

~~However, there~~ There was ~~also an~~ ~~very strong correlation~~ some association between upwind PM<sub>2.5</sub> EAQM and downwind N deposition ~~throughout the entire distribution~~ ( $r = 0.5298$ ), as shown in Figure 6-25. This ~~strong~~ correlation was diminished ( $r = 0.0377$ ) ~~somewhat~~ when moving from the weighted EAQM to use of the maximum PM<sub>2.5</sub> DV from the highest monitor in the ecoregions' sites of influence (Figure 6-26). As shown in Figure 6-27, the ratios between the maximum PM<sub>2.5</sub> DV in an ecoregion's sites of influence and the weighted EAQM value typically ranges from 1.11 to 1.66. Finally, Figures 6-28 and 6-29 illustrate the relationship between PM<sub>2.5</sub> design values and total S+N deposition. The data ~~indicate~~ suggest relatively ~~strong~~ correlation between PM<sub>2.5</sub> EAQM data and total S+N deposition ( $r = 0.6388$ ), but less correlation with the maximum DV ( $r = 0.1250$ ).

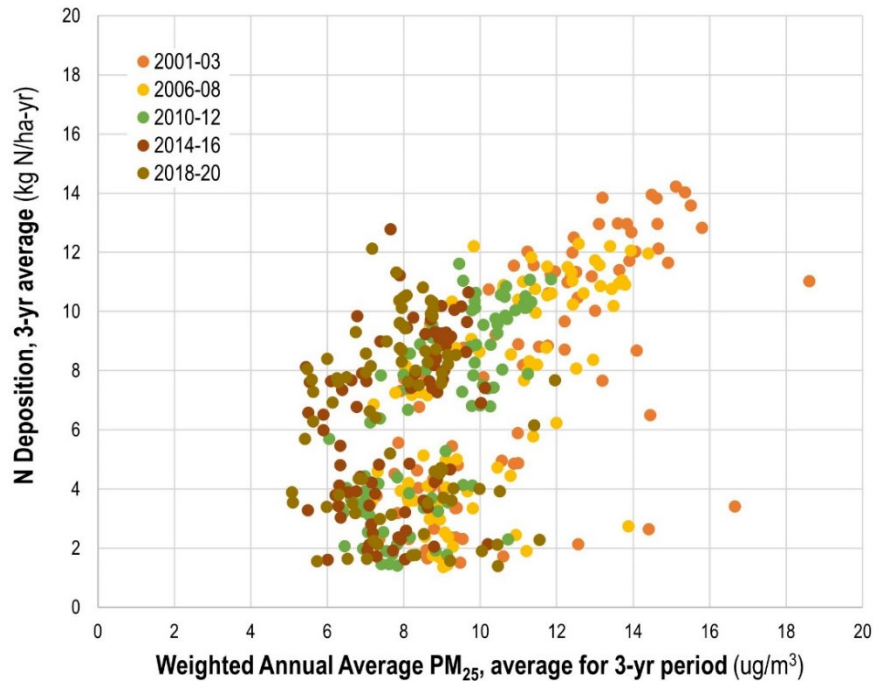
<sup>4</sup> Given the cumulative nature of N and S deposition, it was expected that an air concentration metric with a longer averaging time would be a more appropriate potential indicator of downwind deposition, thus the EPA restricted the PM<sub>2.5</sub> analysis to the annual standard and did not include analyses for the 24-hour standard.



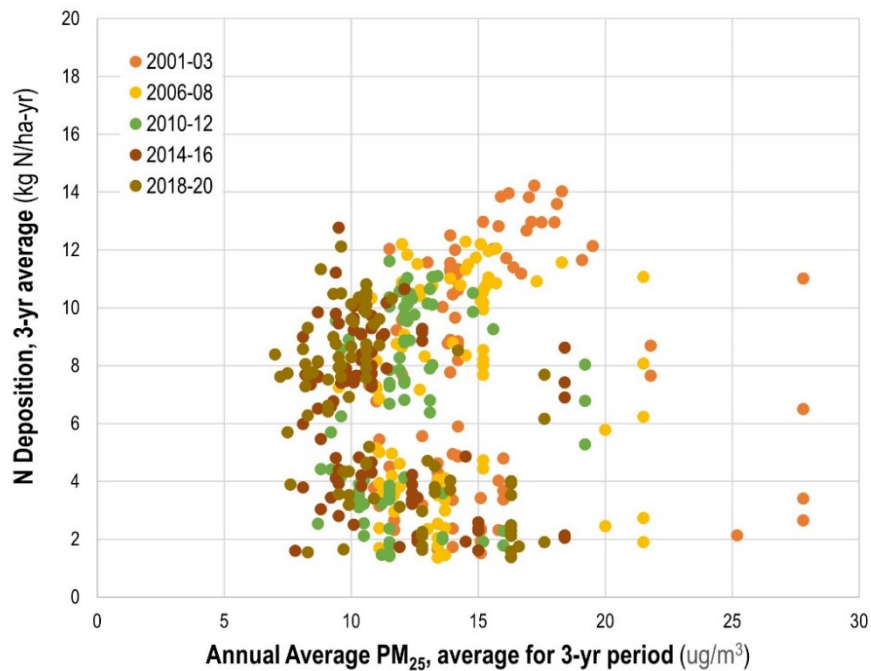
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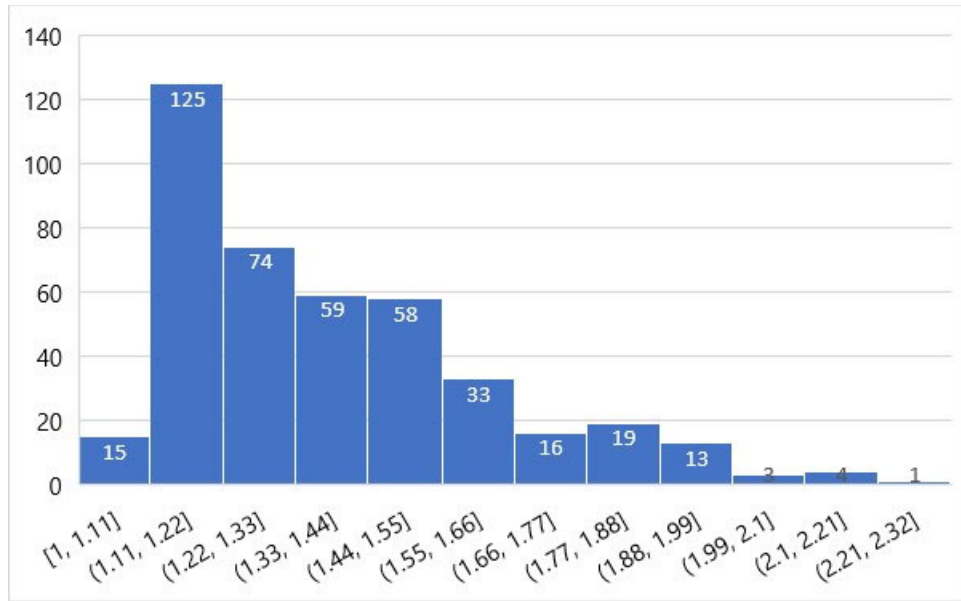
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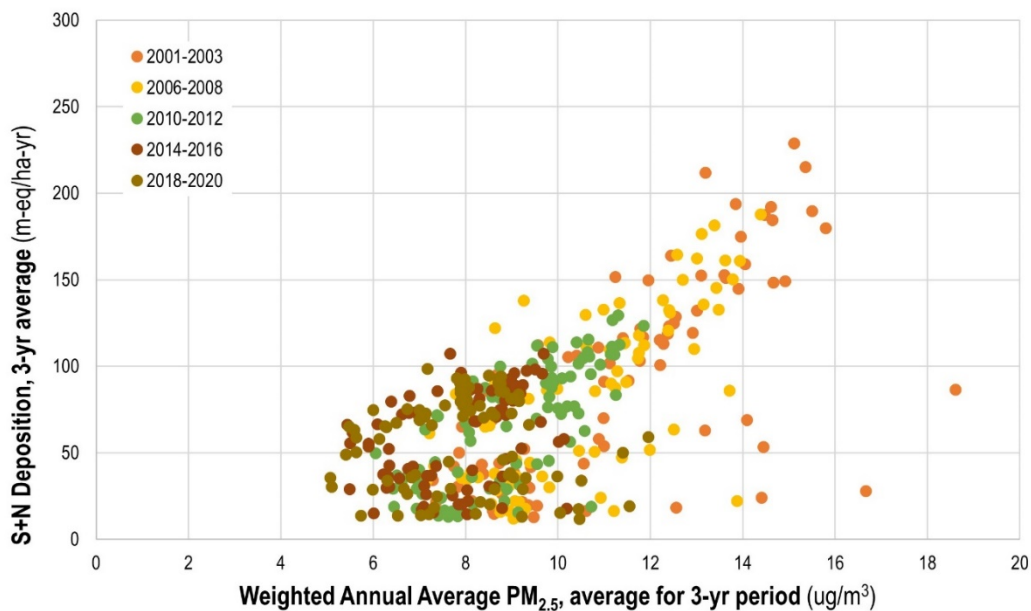
**Figure 6-25. Estimated 3-year average N deposition (ecoregion median) and average of weighted annual average PM<sub>2.5</sub> concentrations in 3-year period (EAQM) for that ecoregion.**



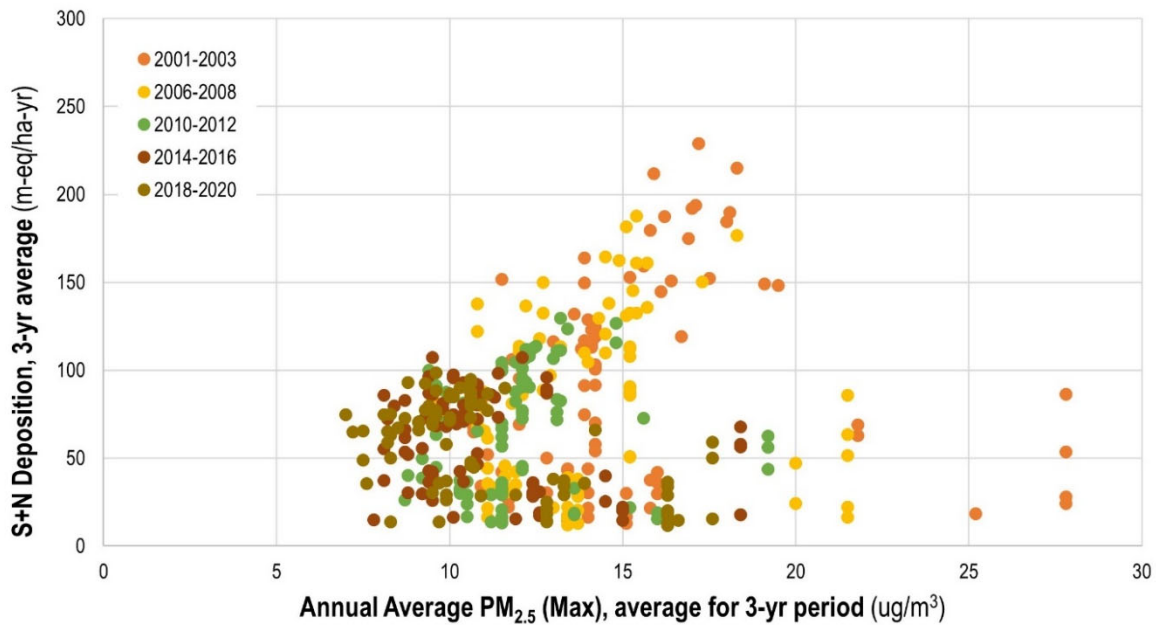
**Figure 6-26. Estimated 3-year average N deposition (ecoregion median) and annual average PM<sub>2.5</sub> concentration in 3-year period from maximum contributing monitor for that ecoregion.**



**Figure 6-27. Histogram of the ratio of average annual average PM<sub>2.5</sub> concentration (µg/m<sup>3</sup>) in 3-year period from maximum contributing monitor for that ecoregion to the average of weighted annual average PM<sub>2.5</sub> concentrations (EAQM) in 3-year period (median = 1.3).**



**Figure 6-28. Estimated 3-year average S+N deposition (ecoregion median) and average of weighted annual average PM<sub>2.5</sub> concentrations in 3-year period (EAQM) for that ecoregion.**



**Figure 6-29. Estimated 3-year average S+N deposition (ecoregion median) and average annual average PM<sub>2.5</sub> concentration in 3-year period from maximum contributing monitor for that ecoregion.**

#### 6.2.2.5 Conclusions

For SO<sub>2</sub>, we examined both the 2<sup>nd</sup> highest 3-hour maximum and an annual average metric. The results for the EAQM suggest that both metrics are correlated with S deposition, with the stronger association being for the annual average metric. There is lower correlation between the design values from the highest monitor within the ecoregion sites of influence for both the 2<sup>nd</sup> highest 3-hour maximum and an annual average SO<sub>2</sub> metrics. As shown by the ratio information, this is likely due to the large concentration gradients seen across the SO<sub>2</sub> monitors in the U.S. (for example, see Figure 2-23), with the maximum contributing monitor between generally 3 to 4 times higher than the EAQM. These figures also show that in the most recent assessed time period of 2018-2020, the median S deposition in the Ecoregion III areas was below 5 kg/ha-yr when the annual average SO<sub>2</sub> concentration, averaged over three years, at contributing monitors was less than 22 ppb and the majority of monitors were below 10 ppb. Additionally, the SO<sub>2</sub> figures indicate that there can be high measured SO<sub>2</sub> concentrations associated with low S deposition (i.e., < 5 kg S/ha-yr) and that there is generally more scatter in the data at lower deposition values. Both of these observations could be due to uncertainties in the TDEP calculations, uncertainties in our assessment methodology and/or a lack of correlation between some SO<sub>2</sub> monitor measurements and S deposition.

For NO<sub>2</sub>, the correlations between the measured annual NO<sub>2</sub> concentrations and N deposition are not as strong as they are between metrics for SO<sub>2</sub> concentrations and S deposition. This could be partially due to the fact that oxidized nitrogen only contributes to part of the total N deposition estimate, and as discussed in section 2, the contribution of reduced nitrogen to total N deposition has grown over the last few decades (*e.g.*, Li et al., 2016). The figures also show slightly less variability between the EAQM and maximum monitor concentrations for NO<sub>2</sub> (when compared to SO<sub>2</sub>), with the NO<sub>2</sub> maximum monitored values being typically about twice as high as the calculated EAQM. This result suggests less variability and smaller gradients in measured NO<sub>2</sub> concentrations across the U.S. when compared to SO<sub>2</sub>. In the most recent time period (2018-2020), median N deposition was generally maintained at 12 kg/ha-yr in Ecoregion III areas while NO<sub>2</sub> annual average, averaged over 3-years, monitored values were 30 ppb or less.

For PM<sub>2.5</sub>, the assessment looks at correlations with S deposition, N deposition and S + N deposition. The results show a ~~clear and remarkably strong~~ correlation ( $r=0.5298$ ) between measurements of annual average PM<sub>2.5</sub> and estimates of N deposition. This could be due to measurements at PM<sub>2.5</sub> monitors including both oxidized and reduced forms of N (*i.e.*, NO<sub>3</sub> and NH<sub>4</sub><sup>+</sup>), which contribute together to total N deposition. ~~While not as strong, there~~ A similar ~~There is also some~~ correlation is observed between measurements of annual average PM<sub>2.5</sub> and estimates of S deposition ( $r = 0.67$ ). However, the results include data where the measured PM<sub>2.5</sub> mass is high when S deposition is low (*i.e.*, < 2 kg S/ha-yr). This is similar to data seen in the figures assessing S deposition and SO<sub>2</sub> air quality metrics. ~~However, this~~ This could also be due to PM<sub>2.5</sub> mass at these contributing monitors having a large fraction of non-S-containing compounds, such as NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and/or organic carbon (OC). In looking at the relationship between measurements of annual average PM<sub>2.5</sub> and estimates of S+N deposition<sup>5</sup>, the results show ~~a good~~ similar correlation ( $r=0.631288$ ). For measurements of annual average PM<sub>2.5</sub> there is less difference between the EAQM metric and the maximum monitor concentrations for annual average PM<sub>2.5</sub>. In the most recent time period (2018-2020), PM<sub>2.5</sub> annual average, averaged over 3-years, contributing monitored values were less than 18 µg/m<sup>3</sup> and mostly less than 15 µg/m<sup>3</sup>, corresponding to N and S deposition of approximately 6-12 kg N/ha-yr and <5 kg S/ha-yr, respectively.

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<sup>5</sup> Total deposition is converted to units of milli-equivalent using the following equation: S+N deposition = (6.25\*S deposition) + (7.14\*N deposition).



assessed time period of 2018-2020, the median S deposition in the Ecoregion III areas was maintained below 5 kg/ha-yr when the annual average SO<sub>2</sub> concentration at contributing monitors, averaged over three years, was less than 22 ppb. The majority of monitors were below 10 ppb.

### 6.3.2 NO<sub>2</sub> and PM<sub>2.5</sub> Metrics

For N, the results in section 6.2.1 suggest that oxidized N deposition in rural areas is mostly from deposition of air concentrations of nitric acid and particulate nitrate, rather than NO<sub>2</sub>. Additionally, the results suggest that in some areas inorganic nitrogen (e.g., NH<sub>4</sub><sup>+</sup>) contributes to the N deposition, with higher contributions in areas near emission sources of NH<sub>3</sub>.

Section 6.2.2 examines the current form and averaging time of the NO<sub>2</sub> secondary NAAQS which is the annual average NO<sub>2</sub> concentration. As in the assessments of the other pollutants and air quality metrics, the analyses also focus on a 3-year average of NO<sub>2</sub> and N deposition and include multiple years of data to better assess more typical relationships. For NO<sub>2</sub>, the correlations between annual average NO<sub>2</sub> and N deposition were ~~poor~~~~somewhat low~~ (r=0.~~0658~~ for EAQM). In addition, the ratios between the maximum contributing monitor and the EAQM show variability, though less than was seen for SO<sub>2</sub>, across the measured annual average concentrations of NO<sub>2</sub> across the U.S., with a median ratio of 2. The correlation between annual average PM<sub>2.5</sub> and N deposition was ~~much~~-stronger (r=0.~~5298~~ for EAQM). This is likely due to HNO<sub>3</sub>, NO<sub>3</sub> and NH<sub>4</sub><sup>+</sup> being the largest contributors to N deposition and being most closely related to concentrations of PM<sub>2.5</sub>. Additionally, the ratios between the maximum contributing monitors and the EAQM are lower for PM<sub>2.5</sub> (compared to SO<sub>2</sub> and NO<sub>2</sub>) with ratios closer to 1 suggesting lower variability of annual average PM<sub>2.5</sub> across the U.S. Given this information and these relationships, the PM<sub>2.5</sub> annual average, averaged over three years, might be the better air quality metric to control N deposition. Such a metric would also provide some control over S deposition, as seen in the figures above. However, it is important to consider that this analysis focuses on PM<sub>2.5</sub> monitors that contribute to the S and N deposition across the U.S. and that these monitors (and other PM<sub>2.5</sub> monitors) also measure other non-S and N related pollutants as part of the PM<sub>2.5</sub> total mass.

### 6.3.3 Key Uncertainties and Limitations

The linkage between air concentration and deposition can vary based on site-specific conditions, including the chemical form of nitrogen and sulfur, frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity. The analyses above attempt to provide insight into these relationships and variability for multiple measured air quality metrics. As with any assessment, there are uncertainties and limitations associated with the work,