EPA’s Review of PM FRM Oversampling Claims by the Agricultural Industry

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Background

- To protect public health against the adverse effects of exposure to airborne particulate matter (PM), the EPA has promulgated national ambient air quality standards for PM$_{2.5}$ and PM$_{10}$ and developed Federal Reference Methods for measuring these pollutant concentrations.

- Some Ag researchers (Buser et al.) have conducted their own research and developed the concept of “True” PM which contends that EPA’s PM$_{2.5}$ and PM$_{10}$ FRM samplers “oversample” agricultural aerosols. Based on this perceived oversampling, these Ag industry researchers contend that these agricultural operations are being over-regulated by EPA.
• Some from the agricultural industry conjecture that this oversampling is due to shifts in PM$_{10}$ sampler cutpoints from 10 micrometers up to 35 micrometers in diameter. PM$_{2.5}$ cutpoints estimations higher than 5 micrometers have been reported.

• EPA considers these oversampling claims to be scientifically unfounded and remains confident that over-regulation of the agricultural industry does not occur due to PM oversampling.

• Since 2010, the Ag industry, EPA, and USDA have mutually agreed to investigate and discuss the technical issues involved in PM measurements of agricultural operations.
Approach

• Faulkner et al. (TAMU): Resurrect TAMU’s aerosol wind tunnels, develop effective operating protocols for their operation, and conduct independent wind tunnel evaluation of EPA’s PM$_{10}$ inlet as a function of aerodynamic particle size and wind speed

• Vanderpool et al. (EPA): Document previous wind tunnel test results, review the “True” PM sampling approach, and conduct wind tunnel evaluation of the approach’s LVTSP sampler

• Mutual exchange of equipment, SOPs, and ideas towards reaching a consensus on key measurement issues
Ambient aerosols are bimodal in size and the relative modal concentrations can vary with site, season, and local activity. Modes are typically lognormal in shape.

**Fine Mode Formation Mechanisms**
- Combustion
- Condensation
- Gas-to-particle conversion

**Coarse Mode Formation Mechanisms**
- Mechanical breakup of larger material
- Plant pollens and debris
- Other biological material
- Dust resuspension
- Sea spray

**Graphical Representation**

- **Fine Mode**
  - MMD = 14 μm
  - GSD = 2.0

- **Coarse Mode**
Since the 1970’s, results from epidemiological studies, toxicological research, and deposition research have demonstrated that adverse health effects from exposure to airborne particles are primarily associated with those particles capable of entering the thoracic region of the human respiratory system (i.e., below the larynx).
PM$_{2.5}$ and PM$_{10}$ Method Development

PM$_{2.5}$ was developed as an indicator of ambient fine mode concentrations.

PM$_{10}$ was developed as an indicator of ambient thoracic particle concentrations.

EPA’s PM$_{10}$ and PM$_{2.5}$ method development efforts were very strongly peer reviewed and have been supported during subsequent PM NAAQS reviews.
Wind Tunnel Evaluation of Size Selective Performance

The size-selective performance of PM$_{10}$ samplers must be demonstrated in an aerosol wind tunnel at wind speeds of 2, 8, and 24 km/hr, using monodisperse aerosols from 3 to 25 μm diameter.

**Acceptance Criteria (2, 8, & 24 km/hr)**

- **Dp$_{50}$ cutpoint** = 10 ± 0.5 μm
- **Solid vs. liquid (25 μm)** = within 5%
- **Mass measurement accuracy** = ± 10%

Inlet tests are conducted under highly controlled conditions of known wind speed, particle size, particle shape, and particle density.
Recent results (2013) continue to confirm the strong historical inter-laboratory agreement during wind tunnel evaluation of PM$_{10}$ FRM performance tests.

Abstract

“Recent work at a south Texas cotton gin showed that … the cutpoint and slope of the FRM PM10 sampler shifted substantially and ranged from 13.8 to 34.5 μm and from 1.7 to 5.6, respectively, when exposed to large PM as is characteristic of agricultural sources.”

“These shifts in the cutpoint and slope of the FRM PM$_{10}$ sampler resulted in overestimation of true PM$_{10}$ concentrations by 145% to 287%.”

<table>
<thead>
<tr>
<th>MMD (μm)</th>
<th>GSD</th>
<th>Dust Conc. (μg/m$^3$)</th>
<th>“True” PM$_{10}$ (μg/m$^3$)</th>
<th>FRM PM$_{10}$ (μg/m$^3$)</th>
<th>Estimated “Oversampling”</th>
<th>Estimated PM$_{10}$ Cutpoint (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.6</td>
<td>2.3</td>
<td>1,385</td>
<td>494</td>
<td>1,099</td>
<td>122%</td>
<td>32.6</td>
</tr>
</tbody>
</table>
Two Key Questions

1. Why does the “true” PM approach of Buser et al. provides results so dramatically different than that of any other researcher?

2. Why do the “true” PM test results seem to vary during each test?

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>“True” PM$_{10}$ (µg/m$^3$)</th>
<th>FRM PM$_{10}$ (µg/m$^3$)</th>
<th>Estimated “Oversampling”</th>
<th>Estimated PM$_{10}$ Cutpoint (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>642</td>
<td>1,152</td>
<td>79%</td>
<td>23.1</td>
</tr>
<tr>
<td>2</td>
<td>294</td>
<td>687</td>
<td>134%</td>
<td>29.6</td>
</tr>
<tr>
<td>6</td>
<td>260</td>
<td>383</td>
<td>47%</td>
<td>13.8</td>
</tr>
<tr>
<td>8</td>
<td>494</td>
<td>1,099</td>
<td>122%</td>
<td>32.6</td>
</tr>
<tr>
<td>11</td>
<td>284</td>
<td>557</td>
<td>96%</td>
<td>34.5</td>
</tr>
<tr>
<td>12</td>
<td>743</td>
<td>1,708</td>
<td>130%</td>
<td>22.9</td>
</tr>
</tbody>
</table>

Definition of “True” PM

“True” PM$_{10}$ is the mass fraction of the mass less than 10 μm AED obtained from a particle size distribution of PM captured with a TSP sampler, times the measured TSP concentration.

1) Gravimetrically determine the mass concentrations of TSP and PM$_{10}$ using collocated Low-Vol TSP (LVTSP) samplers and PM$_{10}$ FRM samplers

2) Determine the particle size distribution (PSD) of collected PM on the LVTSP filter using Coulter counter analysis

3) Calculate the mass fraction of the collected LVTSP less than 10 μm AED from the measured PSD
4) Calculate the “true” PM$_{10}$ concentration by multiplying the LVTSP mass concentration by the mass fraction less than 10 microns.

\[ "True" \text{ PM}_{10} \text{ Conc.} = C_a \int_0^{D_{50}} f(D_p, MMD, GSD) \, dD_p \]

5) Calculate oversampling as:

\[ \text{Oversampling} = \left[ \frac{\text{FRM PM}_{10}}{"True" \text{ PM}_{10}} - 1 \right] \times 100\% \]

In Ag studies, the calculated “True” PM$_{10}$ concentration is typically far less than the measured FRM PM$_{10}$ concentration – thus resulting in “oversampling” estimates.
Loss of fine particles due to solubility, inefficient extraction, and incomplete counting will result in low “True” PM estimates.

There are analytical biases associated with the use of the Coulter Counter to determine ambient size distributions – particularly for fine mode particles.

**1st Problem with the “True” PM Approach**

*Oversampling = \( \frac{FRM \text{ PM}}{\text{“True” PM}} \)*
The Ag researchers’ definition of PM$_{10}$ includes modeling the performance curve as a step function – thus ignoring the influence of ambient particles to the right of the PM$_{10}$ curve. A step function (i.e., slope = 1) is not accurate and thus should not be used when predicting size-based health effects nor when estimating sampler measurement accuracy.

Oversampling = $\frac{FRM \ PM}{"True" \ PM}$

Actual Performance
3rd Problem with the “True” PM Approach

The approach of defining “True” PM$_{10}$ concentrations requires accurate sampling of large ambient particles independent of particle size and wind speed. If this does not occur, then the estimate of total mass concentration will be biased low – resulting in low “True” PM$_{10}$ concentrations estimates.

Key question: How well does the Low-Volume TSP (LVTSP) sampler provide an accurate measure of large ambient particles independent of wind speed?
Apparatus used for dispensing, aerosolizing, and charge neutralizing calibration material into the aerosol wind tunnel.

Isokinetic nozzles (114 Lpm, 90 mm filter) designed for determination of reference concentrations.

Multisizer IV Coulter Counter used for measuring the concentration and size distribution of collected test aerosols.

Polydisperse Arizona Test Dust (ATD) used during inlet evaluations.

Photograph of EPA’s wind tunnel test section during size selective evaluation of the LVTSP sampler.
Evaluation of isokinetic reference nozzles indicates that measured sampling efficiency is independent of particle size and is close to 100%, as predicted from sampling theory.
Compared to the performance of the 16.7 Lpm isokinetic sampler, the LVTSP sampler displays reduced collective efficiency with increasing particle size.
Effectiveness of the LVTSP Sampler at 8 km/h

- Avg C/R
- Avg R1/R2 = 1.00

Sampler Effectiveness vs. Aerodynamic Diameter (μm)

16.7 Lpm LVTSP sampler
Wind tunnel evaluation of the LVTSP sampler reveals that it’s incapable of measuring total ambient PM concentrations independent of particle size. The LVTSP’s performance also varies as a function of ambient wind speed.
Implications of Inefficient Large Particle Sampling

Inefficient large particle sampling by the LVTSP sampler will result in an underestimation of total mass concentration ($C_a$) of agricultural aerosols. This results in an underestimate of “True” PM$_{10}$ Concentration.

"True" PM$_{10}$ Conc. = $C_a \int_0^{D_{p50}} f(D_p, MMD, GSD) \, dD_p$

Concentration Ratio = $\frac{FRM \, Conc.}{"True" \, Conc.}$

Result: This inherent underestimation of “total” mass concentration will underestimate “true” PM$_{10}$ and overestimate “oversampling” of EPA’s PM$_{10}$ FRM sampler.
Summary and Conclusions

1. Conducted over a 30 year time period, the results of 7 independent aerosol wind tunnel studies all confirm that EPA’s PM$_{10}$ inlet performs as designed. Test results were independent of wind speed, aerosol type, and aerosol concentration.

2. For the following reasons, the Buser et al. “True” method of estimating ambient concentrations is inherently negatively biased and should not be used for evaluating the accuracy of EPA’s PM reference methods

   - Inherent biases in aerosol extraction and quantitation in liquid solution will bias measurements of fine mode (PM$_{2.5}$) concentrations

   - Modeling PM$_{2.5}$ and/or PM$_{10}$ performance curves using step-functions does not accurately reflect EPA’s definition of these metrics, and results in an underprediction of actual mass concentration
Summary and Conclusions (cont)

• EPA’s recent wind tunnel evaluation of the low-vol TSP (LVTSP) inlet revealed that it consistently under-measures total mass concentrations.

• Because the actual size distribution of the ambient aerosol during a given sampling event is unknown, the measurement bias of the LVTSP sampler cannot be mathematically corrected for during its data interpretation.

\[
\text{Oversampling} = \left[ \frac{\text{FRM PM}_{10}}{\text{"True" PM}_{10}} - 1 \right] \times 100 \%
\]
Inter-Laboratory Comparison of Effectiveness Results for 20 µm and 25 µm Particles

<table>
<thead>
<tr>
<th></th>
<th>20 Micrometer Effectiveness</th>
<th>25 Micrometer Effectiveness</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2 km/hr  8 km/hr  24 km/hr</td>
<td>2 km/hr  8 km/hr  24 km/hr</td>
</tr>
<tr>
<td>McFarland &amp; Ortiz</td>
<td>0.1%    1.0%    0.9%</td>
<td>-                   -                   -</td>
</tr>
<tr>
<td>VanOsdell &amp; Chen</td>
<td>-       -       -</td>
<td>2.3%    0.3%    3.1%</td>
</tr>
<tr>
<td>Tolocka et al.</td>
<td>0.0%    1.6%    0.0%</td>
<td>0.1%    0.2%    0.0%</td>
</tr>
<tr>
<td>Faulkner et al.</td>
<td>0.5%    3.4%    5.4%</td>
<td>0.0%    3.5%    4.0%</td>
</tr>
<tr>
<td>Mean</td>
<td>0.2%    2.0%    2.1%</td>
<td>0.8%    1.3%    2.4%</td>
</tr>
<tr>
<td>COV</td>
<td>132%    62%     138%</td>
<td>163%    141%    89%</td>
</tr>
</tbody>
</table>

There exists greater inter-laboratory variability associated with measurement of large particles (i.e., very low effectiveness values) than is observed for other size particles.
EPA's "Ideal" curve was based on results of 120 lung deposition measurements reported by Chan and Lippman (1980) using particles up to 14 μm.

"This simplistic form of the linear regression (beyond 14 μm) is probably not physically realistic... because it would imply that no particles larger than 15.3 μm would pass the extrathoracic region - which is not likely" EPA comments on proposed "Ideal" curve above 14 μm (Rodes, et al., 1982).


Missing Chan and Lippman (1980) data for "Ideal" curve above 14 μm was estimated by extrapolation which resulted in a calculated 0% effectiveness at 15.3 μm.
PM$_{2.5}$ and PM$_{10}$ Method Development

PM$_{2.5}$ was developed as an indicator of ambient fine mode concentrations

PM$_{10}$ was developed as an indicator of ambient thoracic particle concentrations