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April 29, 2010

EPA-CASAC-10-010

The Honorable Lisa P. Jackson
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, N.W.
Washington, D.C. 20460

Subject: Review of the White Paper on Particulate Matter (PM) Light Extinction Measurements

Dear Administrator Jackson:

The Clear Air Scientific Advisory Committee (CASAC) Ambient Air Monitoring and Methods Subcommittee (AAMMS, or the Subcommittee) met February 24-25, 2010, in Washington, D.C. to review EPA's white paper on Particulate Matter (PM) Light Extinction Measurements. The Chartered CASAC held a public teleconference on March 26, 2010, to review and approve the report. This letter provides CASAC's overall comments and evaluation.

The white paper was developed to assess possible technologies that could be used to measure light extinction. The assessment supports the on-going PM NAAQS review where EPA is considering a secondary standard that is protective of visibility. The white paper presented options for direct measurement of light extinction as an indicator for a secondary standard and included a brief discussion of potential instruments. The white paper focused primarily on the use of a nephelometer to measure the scattering portion of the total light extinction due to PM and a filter transmission-based instrument for the absorption portion. The Subcommittee was asked to comment on the white paper and respond to the Agency's charge.

The Subcommittee thanks EPA for requesting input at this relatively early stage in the process. The Subcommittee viewed the white paper as a good assessment of the potential approaches to measuring light extinction due to atmospheric PM. The white paper identified instruments in contention for use in a network were and tabulated their respective strengths and weaknesses, though the Subcommittee recommended a few additional approaches for consideration. If the Agency chooses to restrict the discussion of instruments to those whose performance in routine monitoring applications has already been demonstrated, then the choice of a nephelometer and filter transmission-based instrument is logical. The Subcommittee notes that two approaches to measure light extinction directly may soon become available and might be preferable. Both of the promising alternatives are "Cavity" technologies, with direct "closed path" extinction measurement. As these devices are not currently commercially available, costs are less certain and they would have to be thoroughly tested for use in a network. Data from existing, continuous $PM_{2.5}$ Federal Equivalent Method (FEM) monitors could be used as a surrogate for fine-mode visibility assessment until data from extinction monitoring methods are more widely available. Subcommittee members view such mass-based methods as a practical interim approach, or bridge, to extinction monitoring. These options are further discussed in our responses to the charge questions.

The CASAC and AAMMS memberships are listed in Enclosure A. The Subcommittee's consensus responses to the Agency's charge questions are presented in Enclosure B. Individual review comments from the Panel are compiled in Enclosure C.

Sincerely,

/Signed/

Dr. Armistead (Ted) Russell, Chair
CASAC Ambient Air Monitoring &
Methods Committee

/Signed/

Dr. Jonathan M. Samet, Chair
Clean Air Scientific Advisory Committee

Enclosures

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Enclosure A

Rosters of the Ambient Air Monitoring & Methods Subcommittee and CASAC

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Ambient Air Monitoring and Methods Subcommittee (AAMMS)

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Enclosure B

CASAC Responses to the Agency Charge Questions

Questions regarding a PM Light Extinction Measurement Goal and Method

> *The accompanying white paper proposes an overall PM light extinction measurement goal. This goal would provide for measuring daylight hourly PM light extinction at a wavelength of 550nm with an aerosol size fractionation of PM_{10} under ambient relative humidity conditions with overall accuracy and precision $\leq$ 10% in a range of condition from 10 Mm^{-1} to 1000 Mm^{-1} for relative humidity conditions $\leq$ 90%. EPA staff believe that such a goal would be reasonable starting point for establishing performance specifications to support light extinction measurements for a PM visibility standard.*

1. *Does the Subcommittee agree with the goal identified?*

Overall the Subcommittee agrees that using the measurement of light extinction as the indicator for specifying a secondary PM NAAQS is technically feasible. Although the white paper is a good starting point, further documentation of studies and evaluations performed with current and emerging technologies need to be considered. The measurement methods should not be dictated by the status quo, but should promote innovation and continued improvement in promising emerging technologies that meet desired operational measurement attributes. Comments on the specific goals stated above follow.

> *Please comment on each of the specifications for the goal, the adequacy of each specification, and whether each specification is attainable. If applicable, please explain other useful options for the specifications and a rationale for why a different specification should be considered.*

a. *Wavelength of 550 nm*

The specification of 550 nm wavelength in measuring PM light extinction is too restrictive and not justified given wavelengths associated with visual perception. The selection of wavelengths, chosen appropriately within the visible range, should be driven by the overall precision, accuracy, performance and costs of the instruments to make the desired measurement. Multiple wavelength approaches which enhance the information provided by scattering and absorption measurements should be encouraged.

b. *Aerosol size fractionation at PM_{10}*

The consensus is that $PM_{2.5}$ is a better choice as it responsible for the great majority of the scattering in typical urban conditions, and the measurement of coarse particle extinction presents significant challenges requiring considerable resources to resolve, while providing minimal contribution to the total extinction. Finally, the spatial distribution of $PM_{10-2.5}$ in urban areas is likely locally-generated and not uniformly distributed along the sight path, minimizing the visibility-relevance of $PM_{10-2.5}$ measurements made at any specific location.

c. Operation at ambient relative humidity (RH)

The consensus is that relative humidity critically effects scattering measurements and therefore tracking the effect of ambient relative humidity on PM light extinction will be essential. Unfortunately PM light extinction and RH measurements are prone to significant errors at humidity >90%. It remains an open issue as to how we might accommodate PM light extinction measurements at high humidity levels (>90%) and if the application of a humidity cutoff is an appropriate resolution. It may be advisable to use a smart heater or inlet dryer to bring RH down to 90% or suitable drier to reduce the potential for fogging of optical surfaces of the instruments during cold and damp conditions, and to reduce the frequency of required cleaning and maintenance.

d. Overall accuracy and precision $\leq$ 10%

Overall accuracy and precision is very much dependent on the PM size fraction, humidity cutoff and the base PM light extinction to be considered. The 10% accuracy and precision in laboratory settings appear to be reasonable goals, as well as 10% precision in the field. However, the accuracy of ambient extinction measurements will be difficult to ascertain, and a 10% goal is overly ambitions.

e. Range of conditions from 10 Mm^{-1} to 1000 Mm^{-1}

This seems like a reasonable range, but should be reviewed once the specification of the secondary PM light extinction standard is set.

f. Valid measurements (with all other appropriate checks) when sampled at $\leq$ 90% relative humidity

The goal of 95% valid measurement data (excluding span and zero checks) when the RH is less than 90% is a good target.

2. *Based on the method selected there may be additional specifications that should be considered for a PM light extinction measurement goal. Please comment on inclusion of the following additional performance specifications:*

a. Measurement averaging times

The measurement should be capable of producing 1-minute or 5-minute averages to address RH relationships and data validation. Most technologies already report data at 1-minute intervals, or better. For reporting purposes, agencies would include averages, plus min, max, s.d. and count for each hour. The same statistics should be reported for relative humidity. Consideration needs to given to the RH requirement and aggregation rules for data management. For example: Do you exclude an hour of b_{ext} if RH exceeds 90% for 1 minute? Alternatively, do you make the determination on a minute by minute basis? RH and temperature should be reported.

b. *Instrument specific parameters such as angular integration for nephelometers?*

If we accept the idea of correctable bias (Question 9c), then the only *a priori* specifications would be for precision and accuracy for fine particle extinction measurements. Once accepted as an FRM or FEM, manufacturers would provide their design specifications, including, for example: light spectra, truncation error, flow rate, inlet configuration. Instrument specs for RH could/should be posed up front.

c. *Calibration with a gas that has known Rayleigh scattering properties.*

In general, the light scattering properties of gases and particles and filters are different. Therefore, gases and neutral density filters should be used only for "ranging" instruments and for continuing verification, but not for calibration per se. Fundamental calibration of measurement techniques should be done with particles of known size (or distribution) and well-defined composition (sulfate, nitrate or carbon dominated). This is an essential component of laboratory testing and demonstration. Ideally, an "aerosol in a can" approach would be developed for use as a transfer standard in the field.

If applicable, please explain the parameter(s), whether the parameter applies to one or more types of instruments, the purpose of the parameter(s) and an appropriate goal to support a PM light extinction measurement.

3. *As summarized in the white paper, EPA staff believe that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals.*

   a. *To what extent does the Subcommittee support the staff's position that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals?*

Light Scattering by "wet" nephelometer (b_{scat})
There is at least one commercially available nephelometer that is practical for operation in a routine monitoring network. It does not utilize best available technologies and designs, but with modest improvements and additional characterization tests it would be suitable. EPA may need to work with vendors to clearly define performance goals such as minimal change in sample conditions within the instrument and optimized optics to minimize sensitivity to different particle sizes and scattering properties. Appropriate caveats with regard to limitations of the data, especially for coarse mode particles, are needed.

Filter transmission light absorption (b_{abs} surrogate)
There are at least two instruments potentially suitable for this measurement. Acknowledging the limitations of this filter-based approach to b_{abs}, either instrument could be used with the caveat that the measurement is only appropriate for sub-micron aerosols. Both instruments are expected to be upgraded to new hardware platforms within the next year or two, improving performance and reliability. The Aethalometer is widely used in existing monitoring networks, but (at least for the current model) there may be need for a correction of bias (both at seasonal and sub-daily

time-scales) resulting from sample aerosol matrix effects. Next-generation aethalometers may address these artifacts, and should be evaluated when they are they become available. The MAAP design should not need this correction, but there are insufficient published data to properly evaluate its performance in routine networks at this time. Neither instrument measures at or near 550 nm, but this could be implemented. The aethalometer can measure at multiple wavelengths, a potential advantage for non-visibility related use of the data such as identifying contributions from biomass burning.

b. *What are the Subcommittee's thoughts on alternative instrumental approaches that should be considered to meet the light extinction goals?*

Two promising alternatives not yet commercially available are "Cavity" technologies that are direct "closed path" extinction measurement (b_{scat} plus b_{abs}). These methods should be evaluated when stable commercial versions are available. The Droplet Technologies Photo-Acoustic Soot Spectrometer is a commercial instrument that measures b_{abs} at 780 nm. Another approach to b_{abs} measurement is by difference. If robust b_{ext} and b_{scat} measurements are available, b_{abs} can be calculated without the data quality issues inherent in filter-based b_{abs} methods.

An indirect alternative visibility metric that could be considered is to use $PM_{2.5}$ Class III FEM hourly data from the existing national network, but the Subcommittee views this would be much less accurate as a measure of light extinction. This approach does not reflect the enhanced scattering at high humidities but is a practical alternative that could be rapidly implemented for this revision of the NAAQS with relatively few additional resources. It is possible to apply a generic RH correction to these data (between ~60 and 90%) to better approximate visibility under humid conditions. If this route were chosen, use of an instrument that may lose substantial mass of semivolatile species (e.g., due to heating) should be avoided, and averaging times of greater than 1 hour should be considered.

4. *Considering the potential need to deploy nephelometer light scattering and filter transmission light absorption instruments in routine monitoring applications, EPA solicits the Subcommittee's input on:*

   a. *Suggestions for improvement to the commercial versions of these technologies for optimization in future routine monitoring applications for light extinction. Note: please offer any suggestion for improvement either generically for all types of instruments or for specific makes and models. A good starting point for existing makes and models might include both light scattering nephelometers correlated to PM mass already used in routine monitoring programs as well as filter-based absorption methods used in support of characterizing black carbon PM.*

When FRMs and FEMs are defined based only on technology available at the time of designation, practical experience over a wider range of environmental conditions than were evaluated before designation, development of new technology and methods, and more efficient manufacturing methods reveal deficiencies in the FRM or FEM. Where design criteria are necessary, they should consider the extent to which components are commonly available or must be custom produced. Non-standard components can increase production costs with no improvement in quantification. The ideal performance criteria should be stated, with uncertainty allowances to accommodate current technologies, but with periodic tightening of the specifications. Co-funded development opportunities like the Small Business Innovative Research (SBIR) and Science to Achieve Results (STAR) programs are good models, which should be considered as a means to speed up the development process. The vendor specification lists for different instruments are not always in agreement with published independent tests.

Changes to some instruments might include:

- Measuring at additional wavelengths with specified bandwidth.
- Using more energy-efficient and cooler solid-state illumination sources to minimize heating of the nephelometer scattering chamber and to more precisely define the wavelength and bandwidth.
- Adding temperature and RH sensing in the optical sensing zone.
- Combining scattering and absorption measurements into a single instrument (e.g., Photo-Acoustic, Soot, and aerosol Sensor - three wavelength [PASS-3]).
- Acquiring less than five minute averages for the measurements, with stable hourly measurements (e.g., including on-line data processing to minimize post processing for hourly data).
- Upgrading data acquisition and analysis software to better meet the needs of a visibility standard, and to allow real-time quality assurance and data reporting.

b. *If applicable, what are the Subcommittee's suggestions for improvement of alternative instrumental approaches for use in future routine monitoring applications?*

EPA should not dictate the measurement principles, designs, or manufacturers. It should set out the extinction measurement goals as specifically as possible based on performance standards as opposed to design standards and allow ingenuity to rise to the challenge.

Tests to be considered include:

- Effects of water and light-induced absorption on measurements.
- Equivalency and comparability between cavity-based spectrometers and other particle light scattering and absorption methods.
- Characterization of nephelometer truncation angle over the relevant range of fine mode aerosol (~ 0.15 to 1 μm).

Questions Regarding the Establishment of Specifications and Procedures for Approval of Federal Reference Methods (FRM's) and Federal Equivalent Methods (FEM's).

If a traditional approach to designation of light extinction measurements is taken, EPA will need to define how FRM's are to be approved so that a reference method is available for approval of potentially subsequent FEM's and/or deployment in routine monitoring networks. Considering the need to establish FRM's and performance criteria for FEM's to meet the light extinction measurement goal and also considering the recommendation above from the BOSC review, please address the following questions:

5. *Identify the advantages and disadvantages of the following potential options for approval of a light extinction method as a FRM. Please provide specific advice on how to best address scientific questions on interferences, precision, accuracy, and operability; degree of data needed to support decisions; who could perform the work; what kind of peer review would be appropriate, and whether the approach would potentially lead to more innovation in the measurements system or not. Note: if an option could lead to more or less innovation, depending on other factors, please explain.*

    a. *Translate the measurement goal to a performance standard(s) plus procedures for demonstrating that the performance standard is met, without specifying any particular measurement principle. What aspects of performance should the standards cover?*

This option is preferred over the others because it will permit the greatest latitude for innovation, however if not properly implemented, it also has a greater potential to lead to the approval of methods that do not work well in environments other than where the method was evaluated. Specifying a performance standard alone allows instrument manufacturers to propose methods that are unrelated to the conventional methods in use today. This can lead to truly innovative technologies and may also lead to the approval of methods that have complimentary co-benefits for the monitoring communities and other stakeholder groups such as health effects researchers. The disadvantage of this option is that the use of methods that are not uniformly used throughout a monitoring network can lead to bias between different approved methods. Due to the nature of how different monitoring methods work, this bias is most likely going to be manifest unevenly across the country. Methods that are more sensitive to humidity or the concentration of one atmospheric component over another will tend to have a regional bias. This type of non-uniformity is not ideal for a FRM or FEM and must be reduced to the extent possible through the development of very precise performance standards.

The performance standards must include acceptable ranges for instrument response in any environment where the regulatory standard is expected to apply. These environments include widely varying atmospheric component concentrations and environmental factors such as temperature, humidity, direct sunlight and elevation. The starting point for the performance standards must be based on the response to laboratory generated aerosols that are generated with component concentrations of specific interest that exceed what is likely to be found in the environment. These laboratory evaluations should include as many known interferences as can be accommodated with synthetic aerosol. Manufacturers can also take advantage of models that can estimate or interpolate instrument responses across other particle size fractions not evaluated in the laboratory.

The performance standards must include specifications for instrument maintenance, data availability and calibration. The equipment manufacturers are of course free to choose how this is done as long as the recommended procedures ensure that once in an operational environment, the instrument users will be able to demonstrate that the instrument can reliably meet precision and accuracy goals over a multi-year period. To demonstrate that candidate instruments work reliably, instrument manufacturers are encouraged to operate their instruments in several areas of the country that represent different mixes of aerosols over at least a one year period. This is necessary in order to determine if the instrument has a significant seasonal bias in relation to other methods. Costs could be reduced for long term field evaluations by partnering with state and local monitoring agencies and research organizations. The EPA should be notified of proposed field work before it is initiated so they can comment on the suitability of the location and also visit the site while the candidate method is being operated.

The use of performance standards for the selection of FRMs and FEMs will lead to a situation where new methods could be developed that are vastly superior to methods currently designated. In expectation of this occurrence, the EPA must have a procedures and appropriate resources to review the acceptable methods quickly. Periodically, performance standards should be reviewed and adjusted as appropriate. These decisions could be handled internally by the EPA, however, it would be advantageous to have input on these decisions from representatives of state and local monitoring agencies and the data user community.

The disadvantage of the performance approach is that EPA would need to develop three or four separate sets of performance standards: one each for b_{scat}, b_{abs}, and b_{ext}, and possibly one for b_{abs} by difference. This is awkward, and would require more resources and provides more possibilities for important specifications to be omitted.

b. Specify the measurement principle(s), calibration procedure(s), and operational performance requirements and demonstration procedures? What aspects of performance should the standards cover?

This option is easier than option (a) because limiting the method to one measurement principle reduces the number of variables that have to be considered in the method. The simplicity stems from the assumption that a single measurement principle built into analyzers from any acceptable instrument vendor will respond similarly in a variety of monitoring locations and atmospheric conditions. The end result of this approach is a fairly uniform and consistent database, however, it is still possible that the database will suffer from inaccuracies in certain regions or environmental conditions due to biases in the specified measurement principle.

This option encourages innovation but only as related to the specific measurement principle. This kind of "linear" innovation is helpful and will likely result in future instrumental improvements but only as related to the specific measurement principle. This approach will still eventually result in an obsolete method.

c. Specify a particular instrument model or models as the Federal Reference Method, and rely on the equivalent method process to allow for approval of other models. What side-by-side performance testing requirements would be appropriate under this approach?

This approach will stifle innovation because the manufacturer of the specified instrument model has no incentive to improve upon an accepted method. Manufacturers of potential FEMs will also be stifled because even if a newly proposed method is superior to the designated instrument, the FEM equivalency evaluation process forces the new technology to emulate the old method.

d. Provide the specification for the measurement principle(s), calibration procedure(s), and operational performance requirements and demonstration procedures as in b. above; but also specify one or more specific makes and models that would serve as already approved reference methods. Note this would be similar in practice to the Australian/New Zealand Standard™, Methods for sampling and analysis of ambient air, Method 12.1: Determination of light scattering – Integrating nephelometer method. In that method, a generic approach for the method is provided with an appendix that describes the calibration and response of specific integrating nephelometers.

This option is not recommended because it suffers from the disadvantages of both option b and c. The specification of measurement principle limits the number of candidate methods and the designation of specific makes and models will force proponents of new instruments to compare their data to the designated models.

6. Which aspects of a light extinction measurement could be adequately assessed in a laboratory and which require field studies (perhaps across multiple air sheds). For example, are laboratory challenges for a calibration gas and other similar test sufficient to test an instrument, or are experimental studies needed to ascertain the sensitivity of (or effects of humidity on) the instruments and are field challenges required to evaluate different real world aspects of the performance standard (e.g., aerosols varying geographically and interferences)? If a combination of both, please explain which aspects of an instrument are best suited for laboratory challenges and which in the field.

The proposed phased approach outlined in the white paper presents a logical sequence in which a careful assessment of currently available information would help identify and prioritize those aspects of "aerosol light extinction" measurements that can and should be most effectively assessed in laboratory vs. field evaluations.

Certain aspects of instrumental response, such as effects of varying temperature, RH, aerosol size distribution and chemical composition and consistent responses to calibration gases or aerosol mixtures can and should be evaluated in rigorous laboratory testing. For b_{abs} by filter methods, "spot loading" effects need to be evaluated using a sooty (black) "worst case" aerosol. A serious application of existing modeling technologies should be applied to define and determine theoretical compliance with performance specifications. Several laboratory-generated aerosol mixtures could be presented to each candidate to determine how the instrument will respond when compared to a primary standard. Nephelometer truncation errors should be characterized as a function of aerosol size.

Following laboratory testing, a limited number of field or laboratory intercomparison studies could help assess the performance and operational characteristics of various candidate methods under a range of challenging conditions. Then finally, a small pilot network deploying the most promising methods, directly operated by a limited number of state/local agencies, in areas with varied aerosol composition, size distribution, temperature, and/or humidity conditions would provide the most realistic operational and performance feedback.

7. *Would some aspects of performance be better addressed through a design standard, e.g., for the flow rate and the geometry of the PM_{10} inlet, rather than a performance specification and demonstration requirement?*

In the past, design specification for FRM has resulted in considerable implementation problems and impeded the development and use of more suitable measurement methods. Performance specification is a more flexible approach if the performance for scattering and absorption can be specified and adequately evaluated. Therefore, the consensus of the Subcommittee is that performance specification is recommended over design specification.

8. *What data and analysis does the Subcommittee believe EPA staff should have studied or performed in establishing some kind of FRM (5.a-d) for use in regulatory decisions and to help inform the public?*

The data collection and analysis for establishing an FRM should include systematic observations that determine the detection limit, interferences and precision in different environments. A critical set of closure measurements should be focused on establishing consistency between direct extinction measurement and scattering plus absorption. The relative humidity cutoff should be a special focus of the measurement design and analyses. A subset of the pre-FRM studies should include the role of the chosen wavelength, both for the scattering and absorption measurements.

The pre-FRM data analysis should include the examination of the available data from both routine and special study monitoring (e.g. EPA Supersite program, MANE-VU RAIN, IMPROVE and SEARCH). Field studies should include other candidate instruments and continuous measurements and their respective performance for providing a light-extinction indicator should be evaluated and compared. For informing the public, the use of visibility-cams, WinHaze images, and airport visibility (e.g., ASOS) should be included as an augmentation of the chosen instrumental visibility indicator.

As recommended in the white paper, a more detailed assessment of available information should be conducted. The preparation of the FRM should be supported by the development of a guidance document on visibility measurements.

9. *9. As detailed in the white paper, there are a number of instrumental approaches that could be used for making these measurements, including single instruments that measure total light extinction or instrument combinations that measure light scattering and light absorption separately. Some of the methods have inherent limitations that require data adjustments for known biases. While we have already solicited advice on a method to meet the light extinction measurement goal, we would like to explore this topic further as it relates to options for FRM's and FEM's and their eventual deployment in routine monitoring networks.*

a. *Of the available or soon to be available approaches, are any sufficiently limited so that EPA should not further consider them as FRM candidates, need not ensure that the FEM provisions provide a path to their approval as FEMs, and should not consider them when offering advice to or procuring equipment for state, local, and tribal agencies?*

We believe that performance based standards are preferable given that there are multiple approaches that could provide adequate measurements of extinction or scattering and extinction. As noted in the white paper, each method has strengths and weaknesses, but it is likely that in many cases, the identified weaknesses can be addressed by the vendors sufficient to meet appropriate performance standards. The ability to meet performance standards will be easier for some techniques if the range of particle sizes is restricted to $PM_{2.5}$.

b. *Are any of the methods clearly superior in operation and also meet the measurement goal, such that they should be adopted as the FRM and thus serve as the "gold standard" for approval of FEMs (under one of the three FRM approaches listed in question 5(c or d)), and/or for possible widespread deployment?*

Again, there are methods that are currently better developed and implemented than others, but that should not preclude the vendors from working to meet the appropriate standards to become an FRM or FEM. There is certainly no instrument that has been demonstrated as the "gold standard", but there are several very promising *in situ* methods on the horizon for extinction and absorption measurements.

c. *What does EPA staff need to know about the biases of various instruments and should the FRM and FEM require methods to adjust for these biases to ensure data of known quality?*

At this time, there is some literature on particular instruments, but without a "gold standard" biases are undefined. Some methods can be tested for "internal consistency" to evaluate reproducibility of short-term data (spot loading artifacts for filter-based b_{abs} for example). The critical issue is the development of some basic calibration methods (gases, known composition and concentration aerosol with known particle size distribution) that can be used to test the instruments with respect to their basic operating principles. There is a particular problem with respect to coarse particles though the Subcommittee has recommended that the measurement of the indicator be limited to $PM_{2.5}$.

d. *What weight should EPA give to other factors in establishing a reference method for routine PM light extinction monitoring? Please comment on each of the following:*
    i. *resources needed to acquire and fully support routine operation;*
    ii. *current availability;*
    iii. *record of successful field experience; and*

iv. *ability to generate supplemental information (e.g. multiwavelength scattering/absorption, albedo, forward/backscattering, scattering polarization, etc.)?*

Unique advantages of a PM light extinction indicator include the facts that light extinction is an actual characteristic of the ambient atmosphere, and that its relationships to human visual perception, as well as to the physical and chemical characteristics of ambient PM and associated meteorological variables are reasonably well known. For these reasons, a secondary standard based on a PM light extinction indicator can confidently be established to protect against adverse visibility effects, without advance knowledge of the exact method or methods by which PM light extinction would be measured. Currently available methods could provide reasonable measurements or estimates in the near future, with performance improvements from promising new methods forthcoming within a few years.

The Subcommittee commends EPA for looking at ways to enhance the scientific value of the instruments that would be deployed and the measurements thereby obtained. In general, the approach should be to obtain sound basic instruments that meet the performance standards. Current availability of the instrument is not critical. Clearly all instruments should be robust in the field, easy to calibrate, and easy to monitor and troubleshoot through the internet. Having the instruments available as needed in the development and implementation of the new PM secondary NAAQS process is the critical criterion. Instrument need to be field tested to ascertain their robustness and ease of operation under realistic conditions. All else equal, instruments that provide additional information are preferred.

Questions Regarding Network Design and Probe and Siting Criteria

EPA anticipates that a network design strategy would focus on sites that are well suited to characterize visibility impairment on an area-wide basis such as neighborhood and larger scales that have the highest levels of PM. Probe and siting criteria should include specifications that minimize ground effects and other positive and negative interferences (e.g., an HVAC vent), and are consistent with the intent of the NAAQS.

10. *To what extent does the Subcommittee concur that it would be appropriate to focus a network design strategy on sites that can characterize the maximum visibility impairment across an urban area? What other considerations should EPA include in setting a network design strategy?*

11. *EPA and the State monitoring programs have an extensive historical dataset of $PM_{2.5}$ mass and speciation measurements. In the Visibility Assessment Document,[1] EPA used existing PM speciation and mass data to evaluate visibility impairment at a single site in each of 15 cities. However, the selection of sites used in this evaluation was severely constrained by the availability of sites with the necessary types of collocated measurement, and in several cases the site used was not the site with the highest concentrations of PM in the respective city. EPA expects that a review of available data*

[1] Available at http://www.epa.gov/ttn/naaqs/standards/pm/data/20100121UFVAforCASAC.pdf

within each city combined with information from networks assessments[2] would be appropriate to identify likely candidate locations for light extinction measurements. Such measurements are likely to be in the area of expected maximum PM concentration that are also at neighborhood or urban scale and would complement and be complemented by PM mass and speciation measurements.

a. *To what extent does the Subcommittee support collocation of PM mass and light extinction measurements to complement each of the measurements systems while also achieving the purpose of both the primary NAAQS and potential secondary NAAQS? Please offer specifics as to the advantages and disadvantages of collocating both types of measurements systems in an area-wide location of expected maximum concentration.*

b. *Considering the intra-urban variability of PM in any city, what additional factors (e.g., population, expected poor visibility, scenic views, etc.) should be considered to prescribe monitoring locations? Under what circumstances would multiple sites be appropriate to characterize the maximum area-wide visibility impairment across an urban area?*

Some members made the point that multiple sites in an urban area would allow better characterization of area-wide visibility. The consensus of the Subcommittee was that the spatial averaging inherent in vision through the atmosphere typically would allow a single extinction measurement site to adequately characterize visibility in urban centers. The Subcommittee strongly favored collocation of extinction measurements with PM mass, PM speciation, and precursor gas measurements, identifying continuous PM mass and speciation measurements as being of particular value. National Core (NCore) multi-pollutant monitoring sites were identified as worth considering even though these would not necessarily capture maximum concentrations and visibility impairment in an urban area. There was general support for making public communication an important consideration in network design, for example by selecting a monitoring site that can be associated with a vista that is recognized by a significant fraction of the local population.

12. *12. What aspects of probe and siting criteria should be emphasized to ensure that the placement of a PM light extinction instrument is not in a local "heat island" which could also be a "dry spot" with respect to relative humidity?*

The Subcommittee members thought that the relative humidity issue with local surface conditions might not be limited to thermal effects. Any deviation from the prevailing surface characteristics of the site in question (e.g., grass surface, proximity to a large body of water, near a vent outlet of a large building HVAC system) may create local relative humidity conditions that produce light extinction data not representative of the city. Clearly, these aspects need to be taken into consideration when developing siting criteria and choosing a site.

[2] Network Assessments are required of each State or delegated monitoring agency every five years with the next assessment due to EPA Regional Offices by July 1, 2010.

13. *What aspects of probe and siting criteria should be emphasized to ensure that the placement of a PM light extinction instrument is not in a local "heat island" which could also be a "dry spot" with respect to relative humidity?*

The Subcommittee members identified several factors that may need to be considered in selecting probe height including: (1) avoiding the influence of unrepresentative emissions of particles in the immediate vicinity; (2) heights that represent aerosol mixing representative of the city (e.g., sulfate vs. carbon); (3) heights relevant to viewing the scenery of the city (e.g., on a higher floor of a building); and (4) using NCore sites where possible. The Subcommittee thought that the probe height should be at least four meters above the ground. For b_{scat} measurements in the IMPROVE network using a nephelometer, the entire instrument along with ambient temperature and RH sensors are typically mounted on a meteorological tower approximately four meters above any surface. The four-meter height is to insure that surface solar heat does not unduly influence the b_{scat} measurement by drying it out, which may be a factor in this application. This issue should be further examined for the suite of instrument(s) under consideration for assessing attainment of a secondary visibility standard.

Enclosure C

Compendium of Individual Review Comments from the AAMMS on EPA's Light Extinction Measurement white paper

Comments received:

Mr. George Allen

General background

Ideally an open path method (transmissometer) would be used for b-ext measurements (requiring subtracting out b-ext from gases), since it is an in-situ direct measurement. But this method is not practical for use in routine networks. It may have a role as an FRM, but even that limited use presents many challenges, both operational and budget related. Thus I conditionally support the use of some combination of closed path b-ext, b-scat, and b-abs measurements for any urban visibility characterization network. Currently available b-scat and b-abs instruments that are practical for routine network deployment are not ideal but could be useable with modest improvements and appropriate caveats with regard to limitations of the data. Single instruments that measure b-ext are not yet commercially available but may be candidate methods in the future.

For the present review of the PM NAAQS, I support a PM2.5 FEM sub-daily approach, perhaps with some generic RH adjustment factor when RH is in the range of ~60 to 90%. I do not support a full network of b-scat and/or b-abs measurements at this time; the length and complexity of these charge questions reflect the wide range of complex and unresolved issues for a visibility network using these measurements.

CQ1: "Does the Subcommittee agree with the goal identified?"
(where the goal is to use PM light extinction...to support light extinction measurements for a PM visibility standard).

I do not support this goal for the present revision process of the secondary PM NAAQS. As noted above, I support a simple PM2.5 FEM measurement, limited to daytime or mid-day hours. I do not support a full network of b-scat and/or b-abs measurements at this time. I do support and encourage a limited pilot program, both laboratory and field based, to better understand and assess possible technologies for use in a future "true" visibility network.

CQ3a: Available Technology

Scattering measurements (nephelometry)

There are three commercial nephelometers that could be considered for this work: NGN, TSI, and Ecotech (the Radiance Research M903 nephelometer is not a practical candidate). Of these, the NGN and TSI are more research oriented, have better optics (e.g., smaller truncation errors), and have been more intensively characterized. The Ecotech is a good candidate for routine use in SLT monitoring networks because of its robust design. The Ecotech "Aurora" model is the most recent version of the Ecotech model 9003 nephelometer (http://www.aurora-nephelometer.com). This instrument is well designed, but needs modest changes to be suitable for use outdoors at ambient T and RH, such as ambient and in-chamber temperature and RH measurements, and the ability to be "tower mounted" with appropriate solar and rain shields.

Characterization of the optical performance (truncation error) over a range of relevant fine-particle mode sizes is critical for any nephelometer. Truncation error may be improved somewhat by using a broader-band light source ("white", centered at 550 nm) instead of the near-monochromatic sources now used. For a detailed discussion of performance issues with the TSI and the older 9003 Ecotech nephelometers, see: Müller et al. (2009), "Angular Illumination and Truncation of Three Different Integrating Nephelometers: Implications for Empirical, Size-Based Corrections".
Aerosol Science and Technology, 43:581–586, DOI: 10.1080/02786820902798484

If a broad-band light source is shown to improve performance, "white" LEDs are now readily available. The Ecotech instrument supports measurements at multiple wavelengths, so it would be possible to get scattering at discrete wavelengths as well as “white” (broad ~550 nm). The hardware package is user friendly, and chamber cleaning is not difficult.

<u>Nephelometer inlet considerations:</u>

Measuring coarse mode particle scattering is problematic with nephelometery, as discussed in: Massoli et al. (2009), "Uncertainty in Light Scattering Measurements by TSI Nephelometer: Results from Laboratory Studies and Implications for Ambient Measurements", Aerosol Science and Technology, 43:1064–1074, DOI: 10.1080/02786820903156542

One consideration is to constrain the inlet to fine-mode particles; this would also keep the instrument chamber cleaner. In the eastern US, coarse mode aerosols usually make only a minor contribution to b-scat, and the larger uncertainty of coarse mode aerosol b-scat measurements by nephelometer decreases the value of measuring this size fraction. One option for areas with a substantial coarse mode contribution to visibility impairment might be to use on-line PM-coarse measurements to estimate the b-scat from this particle mode; composition and relative humidity growth are not significant factors for coarse particles.

<u>Absorption (b-abs) measurements:</u>

The best commercial measurement method for b-abs is the photo-acoustic method (Moosmüller and Arnott) from Droplet Technologies; this method could be considered for use as an FRM but may be too expensive and complex for wide deployment. For routine network use, a surrogate measurement of light absorption of atmospheric particles can be done with optical transmission (optical density) measurements. However, care must be taken in interpreting these data for visibility use, since the optical extinction of the aerosol is modified by the filter and sample matrix.

There are several commercial methods for b-abs by filter optical transmission. Two are practical instruments for network use: the Thermo Scientific MAAP and the Magee Scientific Aethalometer. In the U.S., the Aethalometer has been used widely as a surrogate for BC or soot mass concentration, in the NATTS and other measurement programs. The Aethalometer uses multiple wavelengths; the 2-channel (880 and 370 nm) configuration is the most common, but other wavelengths such as 520 and/or 565 nm could be used. The current version of this method has a substantial "spot loading effect" that biases the data low as the filter loads with aerosol; the error is a strong function of the aerosol matrix and is largest when the aerosol is dominated by black aerosols. Compensation methods have been developed that can remove the worst of the error, but only on a time scale of many days to weeks. See:

Virkkula et al. (2007), "A Simple Procedure for Correcting Loading Effects of Aethalometer Data", J. Air & Waste Manage. Assoc. 57:1214–1222, DOI:10.3155/1047-3289.57.10.1214

Kirchstetter and Novakov (2007). "Controlled generation of black carbon particles from a diffusion flame and applications in evaluating black carbon measurement methods." Atmospheric Environment 41, 1874–1888, doi:10.1016/j.atmosenv.2006.10.067

Turner, Hansen, Allen (2007). "Methodologies to Compensate for Optical Saturation and Scattering in Aethalometer Black Carbon Measurements". Paper No. 37, Symposium on Air Quality Measurement Methods and Technology, San Francisco, CA, April 30 – May 2, 2007.

Coen et al., 2009. "Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms." Atmos. Meas. Tech. Discuss., 2, 1725-1770. http://www.atmos-meas-tech-discuss.net /2/1725/2009/amtd-2-1725-2009.html

It remains to be seen if the next version of the Aethalometer (the "next-gen" instrument) will properly measure (e.g., without significant filter loading and aerosol composition matrix effects) b-abs at a sub-daily time-scale without this bias; this needs to be re-evaluated when the instrument is available, perhaps later in 2010.
http://mageesci.com/products/upcoming_products.htm

The Thermo MAAP is a more sophisticated measurement method, incorporating scatter from the filter media into the measurement. This should (in theory) minimize the variability of b-abs measurements from filter spot loading aerosol matrix effects. However, there is only limited published ambient data that demonstrates that the MAAP achieves this goal: Petzold et al. (2005), " Evaluation of Multiangle Absorption Photometry for Measuring Aerosol Light Absorption". Aerosol Science and Technology, 39:40–51, DOI: 10.1080/027868290901945
The MAAP is a single wavelength instrument, using a 670 nm source.
http://www.thermo.com/com/cda/product/detail/1,,19884,00.html

To be used for b-abs, the wavelength would need to be changed to ~550 nm. It is important to note the strong b-abs spectral dependance of biomass combustion (wood smoke). A b-abs measurement at 880 or 670 will underestimate the b-abs at 550nm, since wood smoke has substantially enhanced b-abs at shorter wavelengths. A suitable light source near 550 is needed for proper b-abs measurements, since wood smoke is a significant component of urban aerosols in areas with cold winters, making up approximately 20% of cold-month PM2.5.

The current production versions of both the MAAP and the Aethalometer are based on old hardware designs. Both instruments are expected to be updated in the near future, using current technologies. This will improve reliability, but it is unclear at this time what changes in performance may result from these updated methods.

CQ3b: Alternative Approaches

For the current review of the PM NAAQS, PM2.5 from a FEM continuous monitor is an adequate indicator for the secondary standard even though it does not reflect the effects of humidity or aerosol composition. A daytime (mid-day) 4- to 8-hour mean could be used instead of the 24-hour average used for the primary NAAQS. This approach was suggested during the last review of the PM-NAAQS. It has several advantages over a wide-deployment of b-scat and/or b-abs measurements in a new network for the present PM NAAQS review; the FEM PM2.5 network is or soon will be widely deployed, the technology is reasonably mature, and air agencies are familiar with the operation of these methods. I do not support an averaging time of less than 4 hours both in terms of a stable and relevant design value and limitations of the precision of 1-hour data from FEM PM2.5 instruments.

This FEM PM2.5 network could be supplemented with a pilot network of b-scat and b-abs measurements at a few sites. This would provide a comparison with the FEM estimates of visual range. It would also allow a field evaluation of routine use for these methods in the context of routine state/local monitoring networks, and allow refinements of these methods to make them more appropriate for this use, potentially under future revisions of the secondary PM NAAQS. It is not advisable to proceed with wide deployment of a b-scat and/or b-abs measurement network at this time, both for technical and resource limitations.

Two promising alternatives not yet commercially available are the "Cavity" technologies that are direct "closed path" extinction measurement (b-scat plus b-abs). These methods should be evaluated when a stable commercial version is available. The Droplet Technologies Photo-Acoustic Soot Spectrometer is a commercial instrument that measures b-abs at 780 nm. Another approach to b-abs measurement is by difference. If robust b-ext and b-scat measurements are available, b-abs can be calculated without the data quality issues inherent in filter-based b-abs methods.

CQ5: Issues with development of FRMs for b-ext measurements

I agree with the panel’s sentiment that a performance-based standard should be used. However, EPA may find itself in the position of needing to define multiple standards given the options of various combinations of methods and measurements. It should be noted that although a direct PM2.5 b-ext measurement would satisfy the PM Light Extinction Measurement Goal, it may be desirable to have some information on the relative contributions of b-scat and b-abs to support control measures. Given that the NAAQS and thus the FRM metric is b-ext, if that is measured by a combination of instruments for b-scat and b-abs, would an FRM or FEM need to be defined as a matched pair of these methods, or could b-scat and b-abs methods be defined separately even though neither is a b-ext method?

Questions Regarding Network Design and Probe and Siting Criteria

Siting for urban visibility monitoring should be neighborhood or larger scale, within the core urban area. Visibility sites should be collocated with NCore monitoring if at all possible. Probe height is problematic for proper b-scat or b-ext measurements because of the strong influence of chamber temperature on RH and thus b-scat measurements. To avoid local surface heating effects, b-ext or b-scat measurements must be made several meters (10?) above any surface. While this is not easy to implement, it is a critical siting aspect.

A related topic is the proper assessment of the instrument chamber temperature and/or RH for b-ext or b-scat instruments. The ambient temperature used to generate a chamber deltatemperature (above ambient) measurement must be at 10 meters or higher to be reasonably free from local surface heating effects. This is especially critical given that the metric will be a 4 to 8-hour daytime measurement, emphasizing the effects of solar radiation on these measurements.

Dr. Judith C. Chow

PM Light Extinction Measurement Goal and Method

CQ1: Does the Subcommittee agree with the goal identified?

The concepts presented in the white paper are a good starting point. The white paper recognizes the need to take better advantage of previous studies, to more thoroughly evaluate currently available instruments, and to identify emerging technologies that might better achieve measurement objectives.

The measurement method goal should be more ambitious than dictated by current technology. An ambitious goal would encourage more innovation and continued improvement in monitoring technology. On the other hand, the goal should not sacrifice the good in pursuit of the perfect. The sole focus on compliance hinders with the utility of data for a wider range of applications, such as climate assessments, source zones of influence (including hot-spots), and source and receptor modeling (Scheffe et al., 2009; U.S.EPA, 2008).

When Federal Reference Methods (FRMs) and Federal Equivalent Methods (FEMs) are defined based on technology available at the time of designation, practical experience over a wider range of environmental conditions than available before designation, development of new technology and methods, and more efficient manufacturing methods reveal deficiencies in the FRM or FEM. Examples are the freezing of oil and rapid overloading of the WINS impactor in the $PM_{2.5}$ FRM (Kenny et al., 2004; Pitchford et al., 1997), the heated PM_{10} TEOM that underestimates PM_{10} mass for semi-volatile aerosols (Allen et al., 1997; Chow et al., 2006b), and changes in PM_{10} inlet cut-points with inlet loading (Rodes et al., 1985a; Rodes et al., 1985b; Wedding et al., 1985a; Wedding et al., 1985b; Wedding et al., 1985c). Where design criteria are necessary, they should consider the extent to which components are commonly available or must be custom produced, thereby increasing production costs with no improvement in quantification. An example of this is the $PM_{2.5}$ inlet tube (U.S.EPA, 1997a) that specifies dimensions are not available as common tubing stock, thereby increasing the complexity of manufacture and the cost of the instrument.

A possible way to address this is to set performance standards that approach an ideal, but that also allow for fairly large deviations around these standards with reductions in these deviations at ~5 year intervals (a reasonable lifetime for most monitors). This type of performance standard (Watson et al., 1995) would encourage innovation and improvement, as opposed to the current motivation to degrade new instrument performance so that it mimics the older FRMs. More specifics are given in the answers to the questions.

a. Wavelength of 550 nm

The 550 nm wavelength is specified because it is near the peak (555 nm) International Commission on Illumination's (CEI) photopic response curve for a "standard observer (Fairman et al., 1997; Smith and Guild, 1931)." Visual perception is more complex and depends on a melding of the different wavelengths perceived, usually in the red, green, and blue regions of the spectrum (Fairman, 1995; Fulton, 2009; Viénot, 1980). The goal should be to acquire extinction at several wavelengths that might be better related to what people see than extinction at a single wavelength. Scattering and absorption at several wavelengths would provide information on particle size (Thielke et al., 1972) and black vs. brown carbon (Andreae and Gelencser, 2006) that would be useful to determine the causes of haze episodes.

A wavelength centerpoint of 520 to 530 nm would be more practical as a starting point since there are several light emitting diodes (LEDs) and laser diodes available within this region. A bandwidth needs to be specified, as it is for the TSI 3563 three-color nephelometer. The narrower the bandwidth, the better for estimating scattering properties (Ruby and Waggoner, 1981). Wavelengths used for currently available light scattering and absorption instruments include:

- 450±40, 550±40, and 700±40 nm for the TSI 3563 nephelometer (http://www.tsi.com/en-1033/models/3158/3563.aspx). Wavelength specifications are close to those reported by (Anderson et al., 1996)
- Broad band peaking at ~620 nm for the OPTEC NGN-2 and NGN-3 open air nephelometer (Molenar, 1997).
- 450, 525, and 635 nm for the Ecotech nephelometer (http://www.ecotech.com.au/ecotech/nenav.nsf/LinkView/A2619E971A03E075CA2572720010F11FD82C0114BA147F41CA25715600207006.
- 530 nm for the Radiance M903 nephelometer (Richards et al., 2001).
- 655 nm for the TSI DustTrak II and DRX nephelometer/optical particle counter combinations (http://www.tsi.com/en-1033/products/14000/dusttrak%C3%A2%E2%80%9E%C2%A2_aerosol_monitors.aspx)
- 405, 532 and 781 nm for the DMT PASS-3 Photoacoustic instrument (www.dropletmeasurement.com/products/carbon-sensing-instruments/55). PASS-1 uses the 781 nm wavelength.
- 370, 470, 520, 590, 660, 880 and 950 nm for the AE31 aethalometer (mageesci.com/products/rack_mount_aethalometer.htm). The AE22 and OT21 use 370 and 880 nm, and the AE51 uses 880 nm.
- 670 nm for the Thermo Scientific Model 5012 Multi Angle Absorption Photometer (MAAP) (http://www.thermo.com/com/cda/product/detail/0,1055,19884,00.html).
- 567 nm for the Radiance Research Particle Soot Absorption Photometer (PSAP) (Bond et al., 1999).

There should be a near correspondence between the scattering and absorption measurements, which seems to be possible at 450-470 nm, 520-550 nm, and 630-700 nm. The value of the 370 nm absorption wavelength should not be discounted, as this has been found useful for separating biomass burning smoldering soot from higher temperature fuel combustion soot (Kirchstetter et al., 2004; Sandradewi et al., 2008a; Sandradewi et al., 2008b).

b. Aerosol Size Fractionation at PM_{10}.

A $PM_{2.5}$ size cut is a better choice than PM_{10}. The rationale for a PM_{10} size fraction to measure urban haze is not given in the white paper. Under most urban circumstances (i.e., $PM_{2.5}$ mass as half of PM_{10}), $PM_{2.5}$ will cause >90% of the scattering at 550 nm. If the particles are on the large side of the $PM_{10\text{-}2.5}$ fraction in urban areas, they are probably locally-generated and are unlikely to be as uniformly distributed along the sight path as $PM_{2.5}$ (Burton et al., 1996; Chow et al., 1992; Chow et al., 1999; Chow et al., 2000; Chow et al., 2002a; Magliano et al., 1999;

Monn et al., 1997; Wilson et al., 2005). This will be aggravated by slanted sight paths that probably experience a stronger decrease of $PM_{10-2.5}$ relative to $PM_{2.5}$ (Chan and Kwok, 2000).

c. Operation at ambient relative humidity (RH) for RH<90%.

This is a good idea for the scattering measurement, but it is not necessary for the absorption measurement when these are measured separately. In photoacoustic measurements the particle heating evaporates water, thereby decreasing the acoustic intensity that corresponds with light absorption (Arnott et al., 2003; Murphy, 2009). An acceptable interval needs to be specified (e.g. ambient ± 5% RH, similar to the specification for $PM_{2.5}$ FRM filter equilibration). It may be advisable to use a smart heater to bring higher humidity down to 90% to reduce the potential for fogging of optical surfaces during cold and damp conditions.

d. Overall accuracy and precision < 10%

Accuracy and precision should be defined separately. There should be separate requirements for the scattering and absorption measurements. A ±10% interval seems reasonable for precision. A ±10% accuracy could be attained for consistent primary standards (e.g., light scattering or absorbing gases, neutral density filters, and particles generated with a known composition and size distribution), but would probably experience higher deviations among instruments for more complex urban aerosols.

Methods for precision estimation should be specified, possibly following the collocated sampling in different environments currently in use for $PM_{2.5}$ FRMs or with respect to a variety of laboratory-generated aerosols (Sheridan et al., 2005) that cover a broad range of conditions.

e. Range of conditions from 10 Mm^{-1} to 1000 Mm^{-1}

This range is reasonable and has been attained by current technology. There may be some non-linearity in the concentration response at high concentrations that needs to be evaluated for a specific configuration. This has been observed for filter light transmission measurements (Lin et al., 1973; Watson and Chow, 2002).

f. Valid measurements (with all other appropriate checks) when sampled at < 90% relative humidity

A more complete validation procedure is needed to elaborate on this. U.S. EPA (2008) is a good starting point. It requires at least 45 minutes of data to represent an hourly average, as well as specifying frequencies for performance tests (e.g., zero and span), re-calibrations, and audits. Other checks could include extreme values and sudden increases in measurements that might be from electronic noise rather than a change in particle extinction, runs tests to determine that there is some change over a period of time, sensing chamber temperature and RH variability, and correlations or lack thereof with collocated readings.

CQ2: Please comment on inclusion of the following additional performance specifications:

a. Measurement averaging times

The one hour averaging time is reasonable, but data should be acquired for <5 min averages. Sharp spikes of short duration probably represent localized emitters (Watson and Chow, 2001) that should not be included in a longer average intended to represent the uniform distribution along a sight path.

b. Instrument specific parameters such as angular integration for nephelometers?

Nephelometer truncation errors have been evaluated for various configurations (Abu-Rahmah et al., 2006; Bond et al., 2009; Ensor and Pilat, 1971; Ensor and Waggoner, 1970; Guyon et al., 2003; Heintzenberg et al., 2006; Jonasz, 1990; Moosmüller and Arnott, 2003; Muller et al., 2009; Penaloza, 1999; Quenzel et al., 1975; Quirantes et al., 2008; Rabinoff and Herman, 1973; Reed and Howser, 1995; Rosen et al., 1997; Shkuratov et al., 2007; Varma et al., 2003), and their results indicate that unmeasured forward and backward scattering is not important for the 2-4 µm end of the $PM_{10-2.5}$ distribution expected along a sight path. Nevertheless, an evaluation of truncation biases should be part of the FRM certification process. This might use the same urban size distribution required for evaluation of PM_{10} inlet sampling efficiencies (U.S.EPA, 1987) and one or more of the methods described in the previously cited articles.

c. Calibration with a gas that has known Rayleigh scattering properties. If applicable, please explain the parameter(s), whether the parameter applies to one or more types of instruments, the purpose of the parameter(s) and an appropriate goal to support a PM light extinction measurement.

Except for ultrafine particles, scattering by gases differs from scattering by particles, especially when the scattered wavelength is about the size of the particle circumference (Moosmüller and Arnott, 2009). The goal for transfer standards should be a consistently generated ambient aerosol that mimics one or more urban aerosols and size distribution. A high sulfate content might be specified for the eastern U.S. with a high organic carbon content specified for the western U.S. to reflect these obvious differences in PM composition (DeBell et al., 2006). Several aerosol generation systems have demonstrated the ability to do this (Evans et al., 2003a; Evans et al., 2003b; Gerde et al., 2004; Gill et al., 2006; Guo and Kennedy, 2007; Horvath and Gangl, 2003; Kim et al., 2006; Kirchstetter and Novakov, 2007; Mikhailov et al., 2006; Sheridan et al., 2005; Teague et al., 2005; Veranth et al., 2000; Vlasenko et al., 2005; Widmann et al., 2005). This standard would be applicable to light scattering, light absorption, and light extinction instruments.

Refrigerant gases and CO_2 are often used for nephelometer calibration, but these gases do not mimic PM characteristics (Horvath and Kaller, 1994). Neutral density filters (Macleod, 2001) are a long-accepted standard for densitometry (i.e., filter light transmission), but they do not separate the optical properties of particles on a filter surface from scattering and absorption of the filter media. The wavelength dependence of gases (λ^{-4}) and neutral density filters does not follow the relationship for particles of different particle sizes, shapes, and compositions.

Primary standard instruments could be established using more advanced, but not necessarily the commercially-available or cost-effective technologies needed for a widespread network, to characterize these transfer standards that could be used for field calibration of more practical, cost-effective, and commonly available instruments.

CQ3: Suitability of currently available nephelometers and filter transmission systems.

a. To what extent does the Subcommittee support the staff's position that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals?

Currently available nephelometer and filter transmission light absorption instruments are sub-optimal for a robust urban visibility measurement goal. Current commercial technology should be considered as the starting point, not the end-point, for a more ambitious goal.

Currently available commercial instruments have been designed and marketed by small businesses with limited markets (e.g., researchers), limited research and development budgets, and limited testing in a variety of environments. With the prospect for orders of magnitude increases in sales resulting from a FRM designation, these and other manufacturers would attract the investments needed to develop better, more versatile, and more cost-effective instruments. An opportunity is lost if the bar isn't raised to encourage the next generation, rather than locking users into old technology for decades to come. It would be worth a few years of Small Business Innovative Research (SBIR) initiatives or research solicitation to encourage the development and commercialization of the next generation of instrumentation.

b. What are the Subcommittees thoughts on alternative instrumental approaches that should be considered to meet the light extinction goals?

Wang et al. (2009) describe a nephelometer for $PM_{2.5}$ combined with an optical particle counter (OPC) for larger particles that allows a better measure of scattering, the potential effect of $PM_{10-2.5}$ on scattering, and the distribution of large (i.e., locally-emitted, >10 µm) vs. smaller (more widely dispersed, 2.5-4 µm) coarse particles.

Adaptations of nephelometers in commercial smoke detectors (Edwards et al., 2006; Litton et al., 2004) and portable AE 51 aethalometers could be located along a sight path at low cost offer the opportunity to obtain a more representative measure than quantification at a single location.

The National Weather Service's Automated Surface Observing System (ASOS) (ASOS, 2002; Powell, 1993; SAO, 2002) that replaced human observer airport measurements should be considered. These are currently truncated at 10 miles visual range, but they are valid over longer distances that are not well defined.

Currently-expensive multiwavelength photoacoustic instruments for light absorption, as noted in the white paper, might be used initially as a primary standard at a few regionally distributed locations to certify aerosol generation transfer standards, then used to replace filter transmission methods in the future as their size and costs are reduced with the advent of newer technologies.

CQ4: Instrument Improvements

a. Suggestions for improvement to the commercial versions of these technologies for optimization in future routine monitoring applications for light extinction.

When FRMs and FEMs are defined based on technology available at the time of designation, practical experience over a wider range of environmental conditions than available before designation, development of new technology and methods, and more efficient manufacturing methods reveal deficiencies in the FRM or FEM. Where design criteria are necessary, they should consider the extent to which components are commonly available or must be custom produced. Non-standard components increase production costs with no improvement in quantification. The ideal performance criteria should be stated, with uncertainty allowances to accommodate current technologies, but with periodic tightening of the specifications. Co-funded development opportunities, with funds allocated by a competitive proposal process (the SBIR and STAR programs are good models) should be planned to speed up the development process. A small initial investment will result in large long-term savings in terms of field operation, data processing, data validation, data dissemination, and legal expenses.

The vendor specification lists for different instruments are not always in agreement with published independent tests. Changes to some instruments might include:

- Measuring at additional wavelengths with specified bandwidth.
- Using more energy-efficient and cooler solid-state illumination sources to minimize heating of the nephelometer scattering chamber and to more precisely define the wavelength and bandwidth.
- Adding temperature and RH sensing in the sensing zone.
- Combining scattering and absorption measurements into a single instrument (e.g., Photo-Acoustic, Soot, and aerosol Sensor - three wavelength [PASS-3]).
- Acquire less than five minute averages for the measurements, with stable hourly measurements (e.g., including on-line data processing to minimize post processing for hourly data).
- Upgrading data acquisition and analysis software to better meet the needs of an urban visibility standard.

b. If applicable, what are the Subcommittees suggestions for improvement of alternative instrumental approaches for use in future routine monitoring applications?

EPA should not dictate the measurement principles, designs, or manufacturers. It should set out the visibility characterization goals as specifically as possible based on performance standards as opposed to design standards and allow American ingenuity to rise to the challenge. Tests to be considered include:

- Effects of water on absorption measurements.
- Equivalency and comparability between cavity ring-down spectrometers and other particle light scattering methods.

Pros and Cons of Different Procedures for Approval of Federal Reference Methods (FRM's) and Federal Equivalent Methods (FEM's).

CQ5: Performance vs. Design Standards

a. Translate measurement goal to performance standards and methods to demonstrate performance.

This should be the preferred alternative. The white paper and the time available for these comments are insufficient to define these standards and the ways to attain them. Demonstration methods might include:

- Theoretical analyses of size cuts, particle transmission, and changes in particle size and composition: Computerized fluid dynamics (CFD) (Chen et al., 2005; Gimbun et al., 2005; Hari et al., 2007; Hu et al., 2007; Wang and McMurry, 2006), non-spherical optics (Fuller et al., 1999; Kalashnikova and Sokolik, 2004; Mishra and Tripathi, 2008; Wind et al., 2004), particle/filter interactions (Chen et al., 2004), and equilibrium (Nenes et al.,

1998) models are now accurate enough to estimate the performance of instrumentation under a variety of different conditions.

- Replication of values from primary and transfer standards: Several suggestions are given above.
- Temperature and RH deviations between the sample measurement zone and ambient air. Tolerances could be set based on findings from the previous analyses.
- Fast averaging times. < 5 min averages should be attained to separate local from urban-scale contributions that would better represent extinction along the sight path.

b. Specify a particular instrument model or models as the Federal Reference Method, and rely on the equivalent method process to allow for approval of other models.

This approach should be avoided. It will lead to difficulties and controversies, as it has done in the past.

d. Provide the specification for the measurement principle(s), calibration procedure(s), and operational performance requirements and demonstration procedures as in b. above; but also specify one or more specific makes and models that would serve as already approved reference methods.

This approach should be avoided. If adopted, it should be only on an interim basis, to be terminated within 5 years in favor of methods that achieve a more ambitious, but attainable, goal.

CQ6: Which aspects of a light extinction measurement could be adequately assessed in laboratory and which require field studies (perhaps across multiple air sheds).

As noted above, a serious application of existing modeling technologies should be applied to determine theoretical compliance with performance specifications. Several laboratory-generated aerosol mixtures could be presented to each candidate to determine how the instrument will respond when compared to a primary standard.

CQ7: Would some aspects of performance be better addressed through a design standard, e.g., for the flow rate and the geometry of the PM_{10} inlet, rather than a performance specification and demonstration requirement?

No. Several examples of the failure of this approach have been cited above, and many more examples could be assembled with some effort.

CQ8: What data and analysis does the Subcommittee believe EPA staff should have studies or performed in establishing some kind of FRM for use in regulatory decisions and to help inform the public?

There is a wide literature on this subject that has not yet been completely reviewed and evaluated (Andreae and Gelencser, 2006; Bond and Bergstrom, 2006; Chow, 1995; Chow et al., 2002c; Chow et al., 2008; Hand and Malm, 2007; Heintzenberg and Charlson, 1996; Horvath, 1993a; Hyslop, 2009; Kerker, 1997; Kokhanovsky and Zege, 1997; Liou and Takano, 1994; Moosmüller et al., 2009; Moosmüller and Arnott, 2009; Sorensen, 2001; Watson, 2002; Watson et al., 2005; Watson and Chow, 1994; Wilson et al., 2002). A substantial expansion of the white paper or a guidance document on urban visibility measurement should be commissioned from researchers with broad experience in these measurements to document and evaluate what is already known. This would evaluate detection limits, interferences, collocated precision in different environments, calibration methods, data validation techniques, and data analysis approaches applied in prior studies.

There is substantial potential for additional analyses from the supersites data bases (CARB, 2009; NARSTO, 2009) and the IMPROVE filter and continuous measurement sites (VIEWS, 2009) that can look at relationships between aerosol size and composition as well as scattering and absorption under different emissions and environmental conditions.

CQ9: Evaluation of current measurement technology

a. Of the available or soon to be available approaches, are any sufficiently limited so that EPA should not further consider them as FRM candidates, need not ensure that the FEM provisions provide a path to their approval as FEMs, and should not consider them when offering advice to or procuring equipment for state, local, and tribal agencies?

Each of the currently available instruments has advantages and limitations that need to be more explicitly stated and referenced in the white paper. Much of this information can be extracted from published reports and articles and from interviews with users. It is beyond the scope of these comments or the time available to prepare them to do this here.

b. Are any of the methods clearly superior in operation and also meet the measurement goal, such that they should be adopted as the FRM and thus serve as the "gold standard" for approval of FEMs?

There is no "gold standard" as yet. Certainly newer technologies are superior to older technologies. The aethalometer is much more stable and better referenced than its Coefficient of Haze (COH) predecessor. The earlier MRI/Belfort nephelometers were often better indicators of temperature fluctuations than light scattering.

There are several published comparison studies for light scattering, absorption, and extinction (Adams et al., 1989; Allen et al., 1999; Arnott et al., 2003; Arnott et al., 2005a; Arnott et al., 2005b; Arnott et al., 2006; Bennett, Jr. and Patty, 1982; Bond et al., 1999; Bundke et al., 2002; Cappa et al., 2008; CARB, 2003; Chakrabarti et al., 2004; Chow et al., 2006a; Chow et al., 2006b; Clarke et al., 1987; Edwards et al., 1983; Fischer and Koshland, 2007; Foot and Kilsby, 1989; Heintzenberg et al., 2006; Hitzenberger et al., 1984; Horvath, 1993b; Japar et al., 1990; Kashuba and Scheff, 2008; Lack et al., 2008; Liu et al., 2002; Malm et al., 2000a; Malm et al., 2000b; Mertes et al., 2003; Moosmüller et al., 1998; Park et al., 2006; Petzold et al., 2005; Reid et al., 1998; Ruoss et al., 1991; Ruoss et al., 1992; Ruoss et al., 1993; Saathoff et al., 2003; Sioutas et al., 2000; Slowik et al., 2007; Snyder and Schauer, 2007; Turpin et al., 1990; Virkkula et al., 2005; Wallace, 2005; Watson et al., 1989; Watson et al., 2005; Watson and Chow, 2002; Weingartner et al., 2003; Weiss and Waggoner, 1984; Wu et al., 2005) that have been insufficiently evaluated. Many of these have insights and suggestions for improvement that have not yet been catalogued and pursued.

c. What does EPA staff need to know about the biases of various instruments and should the FRM and FEM require methods to adjust for these biases to ensure data of known quality?

Many of these have been identified above or in the literature cited. A comprehensive list of potential issues needs to be assembled from a careful review of published articles. This information should be used to adapt some of the CFD, optical, and equilibrium technologies cited above into a practical model that can be used to evaluate different instrument designs. As noted, primary standards and transfer aerosol generation systems are needed to truly evaluate the measurement accuracy.

d. What weight should EPA give to other factors in establishing a reference method for routine PM light extinction monitoring?

i. Current availability

Zero weight. Current technology is more than a decade old and was not designed for this purpose. Commercially available instruments should only be used as a stop-gap measure and should not dictate the desired performance standard goals.

ii. Record of successful field experience

Zero weight. There is no quantitative record. The only data base is anecdotal, so there is no basis on which to quantify such a record. One cannot compare an older technology, which is sub-optimal but has gone through several iterations, with a newer and better technology that is in the improvement process.

iv. Ability to generate supplemental information (e.g. multiwavelength scattering/absorption, albedo, forward/backscattering, scattering polarization, etc.)?

High weight. As noted above, the measurements should go “beyond compliance” (Chow and Watson, 2008) in addressing issues beyond a secondary urban visibility standard.

Network Design and Probe and Siting Criteria

CQ10: To what extent should network design characterize maximum visibility impairment across an urban area? What other considerations should EPA include in setting a network design strategy?

The goal should be to determine average extinction along a sight path. Valued views (and their accompanying sight paths) will vary with location, so any value will be imperfect. $PM_{2.5}$ network design guidance (Chow et al., 2002b; U.S.EPA, 1997b) should be adaptable to this application, as it discusses spatial averaging, special purpose monitors at hot-spots, and setback distances from nearby sources. It would be advisable to locate monitors along a valued sight path, possibly with one in a maximum $PM_{2.5}$ concentration area (neighborhood-scale), one in a suburban area (urban-scale), and one in a rural area (regional-scale). Subtraction of high-frequency signals and of neighborhood-scale contributions as described by Watson and Chow (2001) might be considered.

CQ11

a. To what extent does the Subcommittee support collocation of PM mass and light extinction measurements to complement each of the measurements systems

while also achieving the purpose of both the primary NAAQS and potential secondary NAAQS? Please offer specifics as to the advantages and disadvantages of collocating both types of measurements systems in an area-wide location of expected maximum concentration.

Continuous visibility measurements should be collocated with $PM_{2.5}$ sites, especially those with speciation measurements, wherever the siting criteria are attained. Even at hotspot sites, the high time resolution will allow nearby source contributions to be subtracted and will allow for better understanding of local contributions to the 24-hour $PM_{2.5}$ sample. Site- and season-specific relationships can be established between $PM_{2.5}$ mass and light scattering (Chow et al., 2006a) and between elemental carbon and light absorption (Park et al., 2006) that can determine what is happening within a 24-hour period and between the 3- to 6-day filter samples (U.S.EPA, 2010) acquired at many locations

b. Considering the intra-urban variability of PM in any city, what additional factors (e.g., population, expected poor visibility, scenic views, etc.) should be considered to prescribe monitoring locations? Under what circumstances would multiple sites be appropriate to characterize the maximum area-wide visibility impairment across an urban area?

Multiple sites along a sight path are essential, as described above. Some precision and accuracy for a single point measurement might be sacrificed in favor of lower cost and greater portability for several instruments that can be located along a sight path.

CQ12. What aspects of probe and siting criteria should be emphasized to ensure that the placement of a PM light extinction instrument is not in a local "heat island" which could also be a "dry spot" with respect to relative humidity?

This is a minor consideration compared to other uncertainties. Other considerations are more important, such as surface moisture, snow cover, low inversion pockets that might trap pollutants in a small region around the monitor.

CQ13. Considering site path, aerosol mixing, the goal of PM light extinction measurements, site logistics, and the location of other air monitoring equipment inlets, what should be the acceptable range for probe height?

Inlets should be 3 to 10 m above ground level, on the rooftops of 1 to 3 story buildings and at least 1 m above the rooftop. $PM_{2.5}$ network design and continuous monitoring guidance documents (U.S.EPA, 1997b; U.S.EPA, 1998) provide a good starting point for sampler siting criteria.

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Mr. Bart Croes

These comments also reflect input from California Air Resources Board (ARB) staff responsible for implementing U.S. EPA monitoring requirements and using the data in source apportionment and health studies.

Questions regarding a PM Light Extinction Measurement Goal and Method

1. *Does the Subcommittee agree with the goal identified? Please comment on each of the specifications for the goal, the adequacy of each specification, and whether each specification is attainable. If applicable, please explain other useful options for the specifications and a rationale for why a different specification should be considered.*
    a. *Wavelength of 550 nm*
    b. *Aerosol size fractionation at PM10*
    c. *Operation at ambient relative humidity*
    d. *Overall accuracy and precision < 10%*
    e. *Range of conditions from 10 Mm-1 to 1000 Mm-1*
    f. *Valid measurements (with all other appropriate checks) when sampled at < 90% relative humidity.*

a. The 550 nm wavelength is the peak of the solar visible spectrum (seen as green light), and is often used as a monochromatic surrogate for all visible light. This specification is reasonable, but it needs to be refined by adding a defined spectral range and sensitivity, so that photometric instruments used to make this measurement are comparable.

b. Fractionation to PM10 is appropriate. Although most combustion-derived light attenuation is due to particles in the range of 0.5 to 2.5 μm, a significant fraction of PM optical effects is due to larger particles, particularly in the case of soil dust or mechanically produced anthropogenic particles. A smaller cutpoint would be inappropriate as it ignores a major contributor to reduced visibility in many industrial and rural settings.

d. Accuracy and precision of 10% is reasonable in light of the necessity that a point measurement will be used to represent a phenomenon (atmospheric turbidity) that is only meaningful (in a public perception sense) over distances of multiple kilometers and which is also variable across viewing environments. A 10% uncertainty is acceptable, so long as the difference among observing systems (multiples of the same instrument, or between competing instruments) is unbiased. Any adopted method must be defined so as to prevent “cherry picking” between instruments to bias monitoring statistics. Striving for higher performance, per se, would be a waste of resources.

e(1). The upper range of conditions for accurate measurement is appropriate, as it is higher than the expected range of optical conditions possible due to variation of aerosol composition when concentrations approach the current health-based PM NAAQS. If the measurement is to be applied in situations outside the constraints of PM NAAQS, then the upper limit of the range should be considered in light of other legal or physical constraints.

e(2). The lower limit, which approximates Rayleigh scattering for green light in particle free air at about 2 km above sea level (about 20% clearer than “clean” dry air at sea level) is appropriate for most urban areas in the U.S. If the measurement is to be deployed at high altitude, especially in remote areas, such as Western National Parks or Wilderness Areas, a more sensitive lower limit should be required to insure good measurement performance in clean conditions. This, however, would require a fairly sophisticated approach to field calibration, as merely filtering air would not allow a lower limit test.

f. Establishing a humidity cutoff is appropriate to prevent condensation on particles from turning otherwise acceptable air quality into an exceedance of PM optical criteria, when they are in effect. The relative humidity limit of 90% is acceptable, so long as EPA approaches this with appropriate understanding of the consequences. At very high humidity, particle growth is dominated by water, and it may be inappropriate to “penalize” wet conditions. California uses a 70% cutoff, which may be too low for the more humid conditions found in the rest of the U.S., but a compromise (say, 85%?) may be more appropriate. EPA should examine the number of hours that would be exempted in some very humid locations, and make a determination based on practicality and measurement reliability.

2. *Based on the method selected there may be additional specifications that should be considered for a PM light extinction measurement goal. Please comment on inclusion of the following additional performance specifications:*
    a. *Measurement averaging times*
    b. *Instrument specific parameters such as angular integration for nephelometers?*
    c. *Calibration with a gas that has known Rayleigh scattering properties.*

If applicable, please explain the parameter(s), whether the parameter applies to one or more types of instruments, the purpose of the parameter(s) and an appropriate goal to support a PM light extinction measurement.

a. Averaging times are very important in using a point measurement to represent an areally dispersed phenomenon. Short averaging times would make the measurement unduly sensitive to local “puff” emissions or short term variations in PM composition or concentration. In the context of using this measurement to supplement health protective PM mass concentration regulations, the averaging time should be set to approximate the relevant exposure time (e.g., 24-hr light attenuation to supplement 24-hr PM mass regulations). If the goal is to provide a welfare benefit of good regional visibility, then the averaging time should reflect human perception of “good visibility” based on survey or laboratory studies of human responses to short term visibility degradation. The latter would probably use shorter averaging time, on the order of one or a few hours, rather than the 24-hr criterion derived from current health-based PM NAAQS.

b. The angular integration of a nephelometer is strongly sensitive to particle size. Since real aerosols may exhibit anything from strong backscatter to strong forward scatter, or may approximate isotropic scattering, a wide field of view of the nephelometer is the best way to measure light scattering without introducing unintentional particle size or humidity weighting into the measurement.

c. Calibration with a highly scattering gas is desirable as it is both simple and repeatable with limited technical sophistication. The historic practice of calibrating with Freon (CFC-12), a strong greenhouse gas and stratospheric ozone depleter, should be explicitly banned in any new measurement specification; any gases proposed for this use should be carefully reviewed for their suitability for use over multiple decades. "Milk glass" standards have been used in the past as an alternative to gas calibration. This approach, using a solid scattering medium, should be considered, but caution is needed to prevent creating an opportunity to down-bias instruments if used improperly.

3. *As summarized in the white paper, EPA staff believe that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals.*
    a. *To what extent does the Subcommittee support the staff's position that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals?*
    b. *What are the Subcommittees thoughts on alternative instrumental approaches that should be considered to meet the light extinction goals?*

a(1). Existing nephelometers, such as those used in the IMPROVE network are quite suitable to the task, and offer the benefits of an existing installed basis for a network for those agencies which currently use them.

a(2). Filter transmission measurements of light absorption need tightly defined protocols and specification of the filter medium to be reliable. The principle, as applied by the IMPROVE network, is workable, but EPA should be cognizant of the critical role of the filter medium in this measurement. In order to measure only absorption, light scattering by material on the filter needs to be overwhelmed by scattering by the filter itself, and filter loadings need to be modest (little more than a mono-layer). This last constraint is a weakness of the commonly used aethalometer. EPA should be wary of accepting existing aethalometers for this purpose; at minimum, instrument operations protocols should be reviewed, and careful laboratory and field studies done to quantify the uncertainty, bias, and environmental (temperature and humidity) responses of current production models. Thin, non-filamentous filter substrates, such as Teflon, should not be used for transmission absorption measurements as they violate the physical assumptions of the measurement.

b. Alternative measurements of light absorption are available. Although subject to some siting constraints, subtraction of nephelometer scattering from long path light extinction can yield light absorption. Within the context of keeping the measurement compatible with traditional monitoring site operations, two alternatives are available. Switching to a reflection measurement with the filter set against a white background (and measuring the base transmission of an unexposed portion of the filter as I_0) is a viable method, compatible with existing FRM samplers (and assuming that the protocol takes account of the problem of heavily loaded filters – a problem also present in the transmission measurement). Alternatively, the nephelometer can be replaced with a cavity ring-down optical measurement[3] which can give both total extinction (on-axis decay) and scattering (off-axis intensity), and thus absorption by subtraction, all in a single instrument. Such a machine has been built by Dr. Anthony Strawa for use by NASA in airborne atmospheric research, and could be easily commercialized if EPA elected to pursue this method. The cavity ring-down instrument would eliminate the dual instrument problem and facilitate unified calibration.

4. *Considering the potential need to deploy nephelometer light scattering and filter ransmission light absorption instruments in routine monitoring applications, EPA solicits the Subcommittee's input on:*
    a. *Suggestions for improvement to the commercial versions of these technologies for optimization in future routine monitoring applications for light extinction. Note: please offer any suggestion for improvement either generically for all types of instruments or for specific makes and models. A good starting point for existing makes and models might include both light scattering nephelometers correlated to PM mass already used in routine monitoring programs as well as filter-based absorption methods used in support of characterizing black carbon PM.*
    b. *If applicable, what are the Subcommittees suggestions for improvement of alternative instrumental approaches for use in future routine monitoring applications?*

a. Existing nephelometers are adequate, but calibration methods should be reviewed. Existing aethalometers are inadequate and unreliable, especially as they respond to temperature and humidity variation; these should be viewed with suspicion for regulatory applications. Absorption measured by integrating sphere, as developed for the IMPROVE program is suitable, so long as relatively open weave filamentous filter substrates are used.

b. EPA should explore alternatives, especially the unified measurement of both scattering and total extinction possible with cavity ringdown technology.

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[3] Strawa, A.W., R. Castaneda, T. Owano, D. Baer, B. Paldus, The Measurement of Aerosol Optical Properties Using Continuous Wave Cavity Ring-Down Techniques, *Journal of Atmospheric and Oceanic Technology* 20, 454-465, 2002.

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Dr. Kenneth L. Demerjian

Questions regarding a PM Light Extinction Measurement Goal and Method

The accompanying white paper proposes an overall PM light extinction measurement goal. This goal would provide for measuring daylight hourly PM light extinction at a wavelength of 550nm with an aerosol size fractionation of PM_{10} under ambient relative humidity conditions with overall accuracy and precision < 10% in a range of condition from 10 Mm^{-1} to 1000 Mm^{-1} for relative humidity conditions <90%. EPA staff believes that such a goal would be reasonable starting point for establishing performance specifications to support light extinction measurements for a PM visibility standard.

Adequacy of the goal (1)

1. Does the Subcommittee agree with the goal identified? Please comment on each of the specifications for the goal, the adequacy of each specification, and whether each specification is attainable. If applicable, please explain other useful options for the specifications and a rationale for why a different specification should be considered.

a. Wavelength of 550 nm – ***There is nothing magic about the 550nm wavelength in measuring PM light extinction. The choice of wavelength (within the visible range) should be driven by the overall precision, accuracy, performance and costs of the instruments to make the desired measurement. If multiple wavelengths are to be considered, justification should be provided with respect to the value added information and it's utility in supporting mitigation strategies.***

b. Aerosol size fractionation at PM_{10} – ***Choice of PM10 needs to be further assessed in terms of its robustness in attributing PM light extinction. Further documentation of PM2.5 and PM10 contributions to PM extinction by region and season is needed to determine the optimal PM size cut. Measuring PM course particle extinction contributions will be challenging.***

c. Operation at ambient relative humidity - ***Tracking the effect of ambient relative humidity on PM light extinction is essential in development of management strategies***.

d. Overall accuracy and precision < 10% - ***Overall accuracy and precision will be very much dependent on the PM size fraction, humidity cutoff and base PM light extinction to be considered. The 10% accuracy and precision seems a very ambitious goal.***

e. Range of conditions from 10 Mm-1 to 1000 Mm-1 – ***This seems like a reasonable range, but should be reviewed once the specification of the secondary PM light extinction standard is set.***

f. Valid measurements (with all other appropriate checks) when sampled at < 90% relative humidity ***– I would think 95% valid measurement data (excluding span and zero checks) is a good target for routine monitoring systems and is obtainable for many of the systems identified.***

2. Based on the method selected there may be additional specifications that should be considered for a PM light extinction measurement goal. Please comment on inclusion of the following additional performance specifications:

a. Measurement averaging times – ***High time resolution (e.g. minute average) data should be collected and stored if available. High time resolution data will support in reconciling PM extinction through consideration of with PM component species, relative humidity and absorptive gases.***

b. Instrument specific parameters such as angular integration for nephelometers?

c. Calibration with a gas that has known Rayleigh scattering properties. ***Instrument manufacturers should provide standard calibration procedures for which Rayleigh scattering of gases is likely necessary, but not sufficient calibration.***

If applicable, please explain the parameter(s), whether the parameter applies to one or more types of instruments, the purpose of the parameter(s) and an appropriate goal to support a PM light extinction measurement.

3. As summarized in the white paper, EPA staff believes that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals.

a. To what extent does the Subcommittee support the staff's position that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals? – ***This may be true, but seems to be reverting to the lowest common denominator from a technology point of view. New technologies as mentioned in section b) look promising, have gotten significant SBIR support and will likely be commercially available within the next year.***

b. What are the Subcommittees thoughts on alternative instrumental approaches that should be considered to meet the light extinction goals? ***– New technologies such as Cavity Ring Down CRD and Cavity Attenuation Phase Shift (CAPS) look extremely promising and should be evaluated and considered as a possible alternative to nephelometer***.

6. Which aspects of a light extinction measurement could be adequately assessed in a laboratory and which require field studies (perhaps across multiple air sheds). For example, are laboratory challenges for a calibration gas and other similar test sufficient to test an instrument, or are experimental studies needed to ascertain the sensitivity of (or effects of humidity on) the instruments and are field challenges required to evaluate different real world aspects of the performance standard (e.g., aerosols varying geographically and interferences)?
If a combination of both, please explain which aspects of an instrument are best suited for laboratory challenges and which in the field. ***– Laboratory aerosol chamber systems exist which can generate and characterize primary and secondary aerosols to evaluate light extinction measurement technologies. These aerosol environments can also be perturbed by changes in temperature and/or relative humidity to test the measurement systems sensitivity to these factors***.

Questions Regarding Network Design and Probe and Siting Criteria

EPA anticipates that a network design strategy would focus on sites that are well suited to characterize visibility impairment on an area-wide basis such as neighborhood and larger scales that have the highest levels of PM. Probe and siting criteria should include specifications that minimize ground effects and other positive and negative interferences (e.g., an HVAC vent), and are consistent with the intent of the NAAQS.

10. To what extent does the Subcommittee concur that it would be appropriate to focus a network design strategy on sites that can characterize the maximum visibility impairment across an urban area? What other considerations should EPA include in setting a network design strategy? ***– Site selection should characterize representative (area-wide) visual impairment of a vista recognized by a significant fraction of the local population.***

11. EPA and the State monitoring programs have an extensive historical dataset of $PM_{2.5}$ mass and speciation measurements. In the Visibility Assessment Document, EPA used existing PM speciation and mass data to evaluate visibility impairment at a single site in each of 15 cities. However, the selection of sites used in this evaluation was severely constrained by the availability of sites with the necessary types of collocated measurement, and in several cases the site used was not the site with the highest concentrations of PM in the respective city. EPA expects that a review of available data within each city combined with information from networks assessments would be appropriate to identify likely candidate locations for light extinction measurements. Such measurements are likely to be in the area of expected maximum PM concentration that are also at neighborhood or urban scale and would complement and be complemented by PM mass and speciation measurements.

a. To what extent does the Subcommittee support collocation of PM mass and light extinction measurements to complement each of the measurements systems while also achieving the purpose of both the primary NAAQS and potential secondary NAAQS? Please offer specifics as to the advantages and disadvantages of co-locating both types of measurements systems in an area-wide location of expected maximum concentration. ***Co-location light extinction measurements with PM mass, PM species composition and precursor gases is essential in attributing primary PM sources and understanding processes and the attribution of secondary PM production sources. As mentioned above site selection should characterize representative (area-wide) visual impairment of a vista recognized by a significant fraction of the local population.***

b. Considering the intra-urban variability of PM in any city, what additional factors (e.g., population, expected poor visibility, scenic views, etc.) should be considered to prescribe monitoring locations? Under what circumstances would multiple sites be appropriate to characterize the maximum area-wide visibility impairment across an urban area?
It is unlikely that intra-urban variability of PM is sufficiently large to warrant multiple PM extinction measurements with any urban center.

Dr. Delbert Eatough

I am agreement with AAMMS response to the White Paper. This documents provides additional comments on issues related to the developing EPA document on the consideration of measurement of light extinction by both an extinction measurement and a PM mass measurement as they relate to the AAMMS subcommittee charge questions from EPA.

I. Questions Regarding a PM Light Extinction Measurement Goal and Method.

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I have a few comments in connection with this charge question in connection with the EPA Viability Assessment referenced in the White Paper which I think are relevant to the current charge to AAMMS. The Assessment includes extensive evaluation of both the magnitude of a possible secondary PM standard based on human response studies and an evaluation of current visibility conditions in several urban areas.

With respect to the observer studies, a couple of comments. In general the studies are well suited to contribute to the Urban Visibility Goal decisions. However the observer is sensitive to contrast and not to absolute extinction. In this regard, I think the Washington D.C. studies are outliers (as they appear in Figure 2-16, page 2-26 of the Assessment) because there are no appropriate landmarks in the Washington study from which the observer can judge contrast in the same context as the other cities. This results in the larger suggested threshold observed in these studies. Thus, I think they should be discounted when the standard is set. I also wonder to what extent time of day versus location has been evaluated in connection with the difference in perceived visibility quality as influenced by observation of a scene in the forward scattering and the back-scattering mode. The response of a human subject will be dependent on the location of the observer to the observed distant scene in the morning and the afternoon.

Finally, I found the evaluation of current extinction in the urban areas studied in the Assessment less than persuasive, largely because of the crudeness of many of the assumptions. I understand these assumptions were necessary because of the data sets that EPA chose to evaluate and that EPA is fully aware of the limitations of the approach used in the Assessment. I think EPA should consider additional efforts to shore this area up with studies which focus on measured extinction, measured hourly mass and, where available, measured hourly average composition. When measured mass is included in the evaluation, I strongly urge that conventional TEOM data not be used in this evaluation because of the serious problems with the measurement of semi-volatile ammonium nitrate and organic material.

We have worked ourselves into a difficult hole with respect to the primary standard by the use of inferior mass measurements for the establishment of the standard. Now that much better methods are available for semi-continuous measurement, we cannot use them easily for the primary standard because they do not reproduce the mass measurements as measured by the FRM method for the standard. The secondary standard evaluation will be immutable connected to the quality of the mass measurements to which it is compared. We do currently have several instruments which will measure fine particulate mass with minimization of the loss of semi-volatile material such as ammonium nitrate and sem-volatile organic material. These include the FDMS TEOM, the BAM and the GRIMM PM monitors. Generally these instruments do not measure total aerosol water content. However, we have shown that the GRIMM can either measure aerosol water (Grimm, H. and Eatough D.J. "Aerosol Measurement: The Use of Optical Light Scattering for the Determination of Particulate Size Distribution, and Particulate Mass, Including the Semi-volatile Fraction," *J. Air & Waste Manage. Assoc.* **2009, 59**; 101-107) or exclude aerosol water (Hansen J., et al., "Semi-continuous Particulate Matter ($PM_{2.5}$ and PM_{10}) Mass and Composition Measurements in Lindon Utah During Winter 2007." *J. Air & Waste Manage. Assoc.* **2010, 60**: 346-355), depending on the instrument configuration chosen. I urge evaluation of areas (such as SLC) where good FDMS TEOM (or comparable techniques) measurements are available be focused on. Reasonable extinction measurements on an hourly basis are available at essentially all urban areas as airport ASOS data for this evaluation. We have demonstrated the use of the ASOS data in evaluation of sources of visibility degradation in urban areas. (Eatough, D.J. and Farber R. "Apportioning Visibility Degradation to Sources of $PM_{2.5}$ Using Positive Matrix Factorization," *J. Air & Waste Manage. Assoc.* **2009, 59**; 1092-1110). At a minimum, the use of ASOS extinction, FDMS TEOM fine particulate mass and any measurement of coarse mass would give a better evaluation of the current status of visibility impairment in an Urban area in my opinion. Adding fine particulate composition which measures semi-volatile ammonium nitrate and organic material would further add. This would allow the better assumption of an f(RH) factor to the analysis

The evaluation of such data would be a great assist when a choice is made of a potential mass measurement which could also be considered for an FRM for a future PM secondary standard. It is important as EPA moves forward in this evaluation that a performance based approach be taken in the choice of the mass monitor, as the committee has urged for the identification of a method for the measurement of extinction.

Mr. Dirk Felton

General Comments:

Visibility Based Secondary PM NAAQS:

It is premature to institute a secondary PM NAAQS for visibility at this time. The EPA should conduct further investigation of other welfare based effects that ambient PM has on all areas, not just urban areas. Two of the more significant aspects of ambient PM are the contribution to climate change and the impact from wet and dry deposition. The impacts of climate change and deposition could easily be more significant than visibility for both human well being and for the health of the ecosystem. The timing is also not right for a secondary standard based on visibility. Many other PM and visibility related NAAQS are currently in the midst of their biggest change in the past 30 years. NO_2 is directly related to visibility and the new NAAQS will likely lead to future controls on NO_2 which will lead to improvements in visibility. Other recently proposed NAAQS including SO_2 and the upcoming proposals for CO and PM will also have both direct and indirect effects on visibility. It is more appropriate to determine if a secondary visibility related standard is necessary after these NAAQS have been fully implemented.

If it is determined that the measurement of visibility is necessary at this time, other simpler options should be considered. Visibility can be calculated from PM component concentration data and these calculations can be compared to existing airport visual range instruments and air pollution cameras. The accuracy of a light extinction measurement is not necessary for the rather subjective goal of determining when 50% of the population determines that a specific view is impacted. Light extinction measurements are also of limited value to health effects researchers and to agencies that must design PM control strategies. The visibility data will not be well correlated with data collected for the primary PM NAAQS because the determination of light extinction, as laid out in the assessment paper includes the effects of relative humidity.

Urban Focused Visibility Standard:

The EPA has proposed PM standards in the past that attempted to apply different air quality standards to areas of the country that were classified as either urban or rural. This approach did not sit well with the public at the time and the proposal was not implemented. An urban focused visibility standard is likely to suffer a similar fate. State and local air monitoring agencies do not want to be put in the awkward position of explaining to the public that a national secondary air quality standard was written specifically for people living in urban areas but does not equally cover people living in rural areas. If the EPA wants to implement a secondary visibility standard that covers all areas of the country, the proposal will have to include a monitoring network design that includes non-urban areas or include a plausible explanation of why rural areas do not need to be covered by this standard.

Specific Comment for Charge Question 3a:

Instruments Potentially suitable for filter transmission light absorption (b-abs surrogate)

This list should include the Sunset EC/OC analyzer and the Magee OT21 Transmissometer. The Sunset instrument provides a thermal as well as a laser transmission based measurement of EC on an interval from 30 minutes to 3 hours. This instrument is specifically designed to differentiate OC fractions from EC fractions which is not necessary for the determination of b-abs. The Sunset is usually operated by state and local agencies interested in source attribution research, however the data should be used for as many purposes as possible.

The Magee Transmissometer is a simple instrument that determines the amount of light absorption through manually loaded sample filters. This type of instrument is not suitable for continuous or short interval data but its advantage is that it can use archived filters from multiple sampling locations to estimate spatial gradients of b-abs.

Dr. Kazuhiko Ito

Questions regarding a PM Light Extinction Measurement Goal and Method

1. Does the Subcommittee agree with the goal identified?

General Comment: It is good to have a concrete set of goals identified, but, given the potential uncertainties regarding the definition of the visual impairment of sight path to "valued urban scenes" across cities, it may be premature to set specific numerical values with these goals.

c. Operation at ambient relative humidity

Comment: Frequency and its diurnal profile of ambient relative humidity likely vary across regions, cities, and even within a city. Therefore, such information needs to be characterized to determine what the feasible range of the ambient relative humidity is.

d. Overall accuracy and precision < 10%

Comment: Does this goal incorporate the range of "acceptable" visibility found in the urban visibility preference studies across cities? I am not sure if it is essential to have a numerical value set now.

Questions Regarding Network Design and Probe and Siting Criteria

10. To what extent does the Subcommittee concur that it would be appropriate to focus a network design strategy on sites that can characterize the maximum visibility impairment across an urban area? What other considerations should EPA include in setting a network design strategy?

Comment: EPA talks about the visual impairment of sight path to "valued urban scenes." I wonder if EPA can come up with a concrete list of such valued urban scenes.

12. What aspects of probe and siting criteria should be emphasized to ensure that the placement of a PM light extinction instrument is not in a local "heat island" which could also be a "dry spot" with respect to relative humidity?

Comment: I am not sure how big a problem this is, but satellite surface temperature data and land use data (e.g., imperviousness) may help to identify such "dry spots". However, the extent of "a local heat island" effects may vary across cities, depending on what fraction of the city is considered a "heat island" (e.g., Manhattan).

13. In an urban area the average height of the typical sight path is likely well above the inlet height of most current air quality monitoring; however, the mixing of aerosols impacting light extinction occurs throughout the boundary layer. Considering site path, aerosol mixing, the goal of PM light extinction measurements, site logistics, and the location of other air monitoring equipment inlets, what should be the acceptable range for probe height?

Comment: Ideally, monitoring probes should be placed so that the sampled air represents the air along the sight path to the "valued urban scene". However, logistically, setting the probe height at such sight path may be difficult. The answer also depends on whether EPA will use a "closed path" method or an "open path" method.

For a "closed path" method, as long as the light extinction measured at a location is highly correlated with the actual visibility impairment occurring at the sight path, I think it is acceptable. Therefore, it may not necessarily be a range of probe height that is important. What is important may be the lack of too strong or too local source impacts, or "dry spots" mentioned above, around the monitor so that the light extinction at the site is still highly correlated with the visibility impairment relevant along the sight path to the valued urban scene. Thus, it is possible that we may still be able to use existing air quality sites for light extinction measurements. We just need assurance (data) that they correlate well with the relevant visual impairment. The issue then becomes how high correlation is acceptable.

For an "open path" method, there is less excuse for the location to be away from the actual sight path to the "valued urban scene". In this case, the probe height can be in any height where people actually observe the urban scenes.

Dr. Donna Kenski

These comments on the White Paper are more general than EPA requested in its charge questions. Regretfully, I lack expertise in the specifics of visibility monitoring and equipment. However, there were aspects of the White Paper that raised some concerns that I want to express.

The White Paper did a fine job laying out some of the issues that need to be discussed at the upcoming meeting. Nevertheless, the discussion on FRMs/FEMs and measurement goals seems quite premature and I fear that EPA is rushing to establish a secondary standard and accompanying monitoring method without laying the appropriate groundwork or considering reasonable alternatives. While the Visibility Assessment made a solid case for determining what visibility conditions are acceptable to the public, it did not provide any discussion on the level, form, or averaging time of a potential standard. The White Paper seems to presume that a standard would be set in terms of light extinction and measured at hourly intervals, and that those measurements should be made with a combination of nephelometers and aethalometers, which are to be operated in ways not currently accepted as standard practice (i.e., without drying the sample stream before measurement). While that MAY be the optimum way of determining light extinction, I believe it is possible to propose a standard that is protective of visibility and yet does not require rolling out hundreds of untested monitors at a time when states are struggling to maintain existing criteria networks and meet new monitoring regulations that are inadequately funded. EPA needs to more carefully examine the alternatives, and take a more inclusive view of methods. Specifically, methods based on mass measurements should be included for consideration – both total PM2.5 and speciated PM2.5. Long path instruments and photographic methods also seem to have been dismissed without adequate discussion of their potential advantages and disadvantages. The measurement goals, as laid out in the White Paper, are much too restrictive at this point in the process. Our need to measure visibility with accuracy and precision should not supersede a common sense approach that tolerates greater measurement variability but yields significant benefits through ease of use, dependability, and economy. A standard that protects visibility could be posed in a number of ways that take advantage of existing networks and data—just one of which might be light extinction as calculated from the IMPROVE equation from speciated PM2.5 data (as done in the Visibility Assessment) or from hourly PM2.5 mass measurements and RH. The hourly PM mass measurements could be incorporated into a sub-24-hour but more than 1-hour standard to help smooth out the greater variability in these measurements. Along those lines, the White Paper and Visibility Assessment seem to put undue weight on the coarse mass contribution to visibility impairment. From the data presented there (with the possible exception of Phoenix) coarse mass contributes little to visibility impairment and its presence could generally be ignored, or perhaps incorporated only when it is a significant fraction of total PM as established by historic data.

I urge EPA and CASAC to carefully consider the pragmatic aspects of a visibility standard. Visibility has been determined for years from speciated and mass measurements of PM. Neither the Visibility Assessment document nor the White Paper make a strong case for discarding this time-tested and practical method. It simply is not feasible, in our current economic climate, to consider requiring states to implement a new network of monitors without first showing decisively that such equipment is vastly superior to alternatives. By all means, new technology should be encouraged, but it needs to be thoroughly vetted in the field. It has been my experience that technologies which appear promising in the lab almost inevitably exhibit significant flaws when deployed under real-world monitoring conditions. Thus any proposed FRM/FEM technology must be first be demonstrated in a pilot study that compares its performance with these older, time- and field-tested technologies. It seems unlikely that such a study could be completed in this review cycle. Consequently any secondary standard proposed as part of this review should not require measurements that meet the very tightly prescribed goals in the White Paper, but rather allow for extinction to be measured or estimated from the data being collected now as part of the PM2.5 network.

Dr. Peter McMurry

EPA staff are recommending that extinction (scattering + absorption) be measured by using integrating nephelometry to measure scattering and filter light transmission to measure absorption. I am writing to provide my opinions on those recommendations.

<u>Suitability of Nephelometer and alternative approaches</u>

1. I support the idea of independently measuring scattering and absorption, since this provides information on types of species that contribute to extinction.

2. I support the use of point measurements rather than the use of long path transmissometers, especially if they are co-located at speciation measurement sites. This will allow more detailed analyses on the contributions of different species to extinction. Also, point measurement methods allow for the use of PM10 inlets, so that the optical properties of particles smaller than 10 µm aerodynamic diameter can be measured. Long path measurements do not allow for that possibility.

3. I support the use of integrating nephelometers. They have been used for more than 30 years, and have provided valuable information on aerosol scattering coefficients. Their limitations are quite well understood, although not always easy to overcome.

4. I have reservations about the use of filter transmission measurements to measure absorption coefficients. On the positive side, commercial instruments are available and have been used extensively. Furthermore, they provide data with high time resolution, enabling analyses in concert with scattering data from integrating nephelometers. Concerns include:

(i) The optical properties of particles deposited on filter substrates are different from the optical properties of airborne particles because morphology and mixing characteristics are altered by deposition onto filter surfaces. Also, filter transmission is affected by multiple scattering within the filters. For example, Cappa et al (2008) found "the presence of this OA [organic aerosol] in an external mixture of absorbing aerosol and OA can cause an increase in the light absorption measured by the PSAP, relative to that measured by the PAS [photoacoustic aerosol spectrometer], by more than a factor of two."

(ii) Laboratory studies (Zhang et al. 2008; Khalizov et al. 2009; Lack et al. 2009; Murphy 2009), field studies (Lack et al. 2009) and modeling work (Nessler et al. 2005) have shown that the absorption coefficients of soot are altered by transparent coatings of materials such as sulfuric acid and oily organic compounds. This sensitivity arises in three ways: (i) transparent liquid coatings lead to enhanced absorption, (ii) hygroscopic coatings absorb water, thereby affecting the amount of transparent material condensed on the absorbing particles, and (iii) changes in morphology due to the evaporation/condensation cycles of water onto hygroscopic particles lead to more compact structures that are more absorptive. These observations are based primarily on measurements that have been carried out on gasborne particles using cavity ring down or photoacoustic spectrometry. It is highly unlikely that accurate information on relative humidity-dependent absorption could be measured with by filter light absorption, yet the proposed standard requires accurate measurements in the 10%-90% RH range.

The types of measurement errors mentioned above are the source of my reservations regarding the use of filter transmission methods for a secondary standard. I think it is likely that filter transmission instruments would eventually be replaced with instruments designed for in-situ measurements. It would be unfortunate to make a major investment in instrumentation that will be replaced. An alternative might be to use measurements of BC or EC to estimate absorption coefficients. This would not be a good long-term solution since those are not optical measurements and they do not provide adequate time resolution. However, EC/BC data are now available at speciation sites.

<u>Potential improvements ot commercial nephelometers and future alternatives.</u>

1. The White Paper discusses uncertainties regarding sampling efficiencies of instruments such as nephelometers for particles up to 10 µm. This is an important question that needs to be resolved.
2. In-situ measurement methods that include cavity ring down spectrometers and photoacoustic spectrometers (PAS) have been studied extensively in recent years. Cavity ring down spectrometers measure extinction, while PAS measure absorption. In-situ measurements of aerosol optical properties are likely to be more accurate than filter transmission methods. However, in-situ techniques are also prone to measurement artifacts. For example, Moosmuller et al (2009) state that "these [in situ] methods may suffer from some interference due to light-induced particle evaporation." Murphy (2009) found that evaporation of water from coated soot particles reduces the photoacoustic effect, leading to measured values of absorption coefficients that are below true values. To avoid such errors, Murphy recommends that water be removed prior to measuring absorption coefficients, which is inconsistent with EPAs recommendation that optical properties be measured at ambient RH. In ambient aircraft measurements, Strawa and coworkers (2006) reported good agreement (2%) for extinction measured by cavity ring down and by nephelometry and filter transmission. Use of their "reciprical nephelometer" also enabled them to obtain scattering coefficients that agreed with values measured with the TSI nephelometer to within 2%.

Given the fundamental limitations of filter transmission methods for determining absorption coefficients, the long-term goal should be to adopt in-situ measurement methods. I would prefer to see the agency focus on supporting the development of such methods rather than investing in methods that we know to be fundamentally flawed. Because single scattering albedos are typically in the 0.8-0.9 range, scattering dominates extinction. Delaying the measurement of absorption until a better instrument is available might not be a bad tactic. A great deal of research is underway to develop better in-situ measurements of absorption. This work is driven primarily by the need of understand absorption so as to better quantify the effects of aerosols on the earth's radiation balance and on local and regional precipitation patterns. Hopefully, those efforts should lead to better commercial instrumentation in the near future.

The White Paper discusses the possibility of sponsoring an "invitational measurement intercomparison study." The cost would be limited if "instrument manufacturers covered their own cost to participate." *It is important that EPA support the participation of researchers with prototype instruments in such a workshop, especially for in situ measurements of absorption coefficients.*

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Mr. Rich Poirot

1. Does the Subcommittee agree with the goal identified? Please comment on each of the specifications for the goal, the adequacy of each specification, and whether each specification is attainable. If applicable, please explain other useful options for the specifications and a rationale for why a different specification should be considered.

a. **Wavelength of 550 nm**
This (wavelength of maximum human visual sensitivity) is a reasonable choice, but it may not be critically important to focus exclusively on one wavelength, if instrumental responses to other wavelengths or ranges of wavelengths can be obtained and reasonably scaled to represent human perceptual responses. Scattering and absorption Information for other wavelengths could also provide useful information on particle size distributions and/or composition.

b. **Aerosol size fractionation at PM10**
This is not an unreasonable choice, although off-hand I can't think of any logical reason to specify 10 microns as an upper size limit. Also, I think the arguments to include (or attempt to approximate) coarse particle light scattering (and/or coarse particle adsorption) in this indicator may be overstated. Coarse particles will make relatively minor contributions to light extinction at most locations – and especially at locations likely to exceed a secondary standard (which are not likely to be Phoenix). For example, a PM extinction level of 100 Mm^{-1}, about mid-range of what's been proposed for a new secondary standard, would be exceeded by 25 ug/m^3 of non-hygroscopic $PM_{2.5}$ organic matter (30% below the level of the current $PM_{2.5}$ standard). But it would require a $PM_{10-2.5}$ concentration of 170 ug/m^3 – well above the level of the current PM_{10} standard - to contribute a similar level of extinction. If the 25 ug/m^3 of $PM_{2.5}$ was hygroscopic ammonium sulfate at 80% RH, the PM extinction would exceed the upper end of the range being considered, contributing over 200 Mm^{-1}, which would require over 325 ug/m^3 of $PM_{10-2.5}$, more than double the current PM_{10} standard, to produce a similar optical effect.

Attempts to include coarse particle scattering &/or adsorption in the indicator also add substantial measurement challenges and will cause instrument maintenance problems. Neither nephelometers nor aethalometers respond with the same efficiency to coarse particles than to fine ones, so simply employing a PM_{10} inlet is not an ideal approach, and doubling the number of samplers or employing periodically switching, size-fractionating heads adds substantial costs and/or complexity. There will also be added maintenance costs if PM_{10} heads are used on nephelometers, especially in humid, urban environments. Conceivably, methods measuring scattering or scattering plus adsorption from fine particles only could be specified except for locations where the coarse/fine ratio exceeds X. Possibly also, continuous PM_{10} (&$PM_{2.5}$) mass measurements could be used if/where needed to estimate coarse scattering, with an assumption of no hygroscopic growth, as most coarse particles are hydrophobic, except (natural) sea salt.

c. **Operation at ambient relative humidity**
This is a reasonable choice if measuring optical effects at under ambient air conditions is the objective. However, a PM extinction indicator does not necessarily need to be constrained to ambient RH conditions, and might well be defined in terms of "at 70% RH", at "70% RH or less", or "at ambient conditions but only considering hours of less than 70% RH", etc. As with coarse particles, some maintenance issues might be avoided if highly humidified aerosols (droplets) were excluded from samplers. If it were feasible, valuable added information content would be provided if both wet & dry scattering – at various RH levels below ambient – could be obtained.

d. **Overall accuracy and precision < 10%**
This is not unreasonable, but seems difficult to justify given the very wide range of combined levels + forms currently being considered as appropriate for a secondary NAAQS – from as low as 64 Mm^{-1}, 98^{th} percentile to as high as 191 Mm^{-1} 90^{th} percentile. That seems like a very wide range for which to require both accuracy and precision of <10%. Accuracy will be impossible to determine for "ambient aerosols" and will most likely be limited to laboratory testing using surrogates.

e. **Range of conditions from 10 Mm^{-1} to 1000 Mm^{-1}**
This is not unreasonable, but may be overly restrictive as it requires a low end well below the 64 Mm^{-1} to 191 Mm^{-1} range currently being considered. Also, if a very lenient form like 90^{th} %tile is employed, the standard is more based on a "counting" metric than on a precise minimum threshold.

f. **Valid measurements (with all other appropriate checks) when sampled at < 90% relative humidity**
This is reasonable, although, as indicated above, 90% RH may be an unnecessarily high upper RH bound, and alternative and possibly more effective regulatory metrics might be considered for visibility effects at (lower) RH levels "below XX% RH" or even "standardized to YY% RH". Such lower RH limits might also reduce maintenance problems and increase data capture efficiency (within more narrowly constrained RH limits). A lower RH limit will also reduce effects of measurement errors or occurrences of spatially varying RH, where higher RH or even fog may occur within the sight path but not at the monitor.

2. Based on the method selected there may be additional specifications that should be considered for a PM light extinction measurement goal. Please comment on inclusion of the following additional performance specifications:

a. **Measurement averaging times**
An hourly averaging time is not unreasonable, given the nearly instantaneous nature of human perception of impaired visibility. However, longer averaging times – such as 4 to 8-hour daylight averages, might make for a much more stable regulatory metric, place less emphasis on early morning water, and allow use of a wider variety of instruments for which 1-hour data can be noisy.

b. **Instrument specific parameters such as angular integration for nephelometers?**
This may be necessary, but my preference would be to avoid instrument-specific specifications, if possible, and as indicated above, I think coarse particles and their related angular truncation issues might more efficiently be eliminated from the regulatory metric, rather than compromising the method to accommodate a relatively unimportant influence.

c. **Calibration with a gas that has known Rayleigh scattering properties.**
I don't know of viable alternatives, although it would be desirable (for this and other uses) to have a standard "aerosol in a can" for calibration and audits. Also, a scattering gas tells nothing about light absorption.

3. As summarized in the white paper, EPA staff believe that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals.

a. **To what extent does the Subcommittee support the staff's position that currently available nephelometer light scattering and filter transmission light absorption measurement instruments are suitable to meet the light extinction goals?**
A nephelometer or combination of nephelometer + aethalometer would seem to be the methods that come closest to being considered both readily available and also suitable for use in routine network operations. That being said, I don't think either method is fully ready for deployment in routine network operations and that both methods would have problems associated with the proposed PM_{10} size limit. It may be a very costly undertaking to require both methods and also to accommodate differences in fine and coarse particle responses. While the potential use of these semi-standard methods should be evaluated, I would not like to see these methods pushed to the exclusion of other optional approaches, especially as there appear to be several promising techniques on the horizon (and lingering bugs in the "standard methods").

b. **What are the Subcommittees thoughts on alternative instrumental approaches that should be considered to meet the light extinction goals?**
The currently-stated "light extinction goals" should not be viewed as set in concrete, and it might be productive to consider measurement goals and potential methods together. For example, an indicator based on fine particle scattering and absorption or just fine particle scattering could be very protective of visibility most times and places (especially where/when impairment is greatest), would avoid coarse particle measurement and maintenance issues and substantially reduce costs. Along similar lines, a sub-daily 4 to 8-hr $PM_{2.5}$ mass indicator such as was recommended by both EPA staff and CASAC in the last review cycle could certainly be protective of visibility, and could be measured by existing continuous instrumentation, with little or no added cost. If need be, a generic "mixed aerosol f(RH) function" could be developed (perhaps on a regional basis) and used to "humidify" the fine mass to make it more extinction-like . There would be some uncertainty in such an adjustment, but I think it would be rather small compared to the difference between 20 dv 98^{th} percentile and 30 dv 90^{th} percentile, which is currently being considered as the range within which a standard might be selected. Conversely, a generic mixed aerosol f(RH) adjustment might be applied (backward) to extinction levels within the 20 to 30 dv range of

extinction considered adverse to select a reasonably protective level of fine particle mass – perhaps averaged over 4 to 8 daylight hours – that would be beneficial for visibility, measurable by current networks, and which would substantially reduce the large East/West differences that would characterize a wet extinction indicator – if the Agency finds id necessary to require a "threshold-based form", rather than a "progress-based form" of a secondary standard as was recommended by the CASAC PM panel.

I would also like to see the existing, widely deployed Belfort 6230a (or other) foreword scatter meters – widely employed at nearly 1000 FAA, NWS or DOD sites in the US (and many more worldwide) – at least considered in this pilot evaluation process, and use of the extensive existing measurement network (though there are many limitations) should be given some consideration. Indeed there could be substantial benefits for both aviation safety and aerosol visibility protection if EPA and NOAA could cooperate more closely in the collection, processing, archival and redistribution of these valuable data. At a minimum, an effort should be made to make a substantial subset of these data available in their raw uncensored and un-binned form. In the recent past (2003-05?), Sonoma Technology, with modest funding support from EPA, used to provide such raw ASOS data (complete with an inverse f(RH) function to estimate $PM_{2.5}$ mass) as part of the AIRNOW program. It would be very useful to resurrect something like this as a component of or complement to an urban visibility pilot network.

Lastly, some consideration should be given to automated digital camera techniques, from which optical extinction estimates can be extracted, which may be crude compared to more precise (point) measures, but which can provide integrated long path information on combined effects from scattering & absorption, fine and coarse (over many different parts of a scene) and would have obvious huge advantages as a public communication (and future assessment) tool. Conceivably, photo-derived extinction estimates might initially be used to determine compliance with a relatively lenient secondary standard, and then support establishment of refined "local" target visibility goals or rates of progress in the implementation phase.

4. Considering the potential need to deploy nephelometer light scattering and filter transmission light absorption instruments in routine monitoring applications, EPA solicits the Subcommittee's input on:

a. Suggestions for improvement to the commercial versions of these technologies for optimization in future routine monitoring applications for light extinction. Note: please offer any suggestion for improvement either generically for all types of instruments or for specific makes and models. A good starting point for existing makes and models might include both light scattering nephelometers correlated to PM mass already used in routine monitoring programs as well as filter-based absorption methods used in support of characterizing black carbon PM.

Others on the panel will have much better suggestions than I can offer here. Generally, nephelometers currently used as PM mass monitors are heated and/or cut (at 2.5 um) and are not necessarily likely to be the best starting points for measuring fine and coarse scattering under ambient conditions. Currently deployed aethalometers would benefit from improved guidance for more standardized operating and data processing procedures.

b. If applicable, what are the Subcommittees suggestions for improvement of alternative instrumental approaches for use in future routine monitoring applications?
Others on the panel will have much better suggestions than I can offer here. Although transmissometers do not seem to be advocated in the white paper, they have for whatever reasons been utilized for light extinction measurements to determine compliance with visibility standards in the few locations (Denver and Phoenix) where standards have been developed. As such, a more careful consideration of this approach, and possible ways to improve upon it (or to rule it out) seems warranted. The various photoacoustic, CDR and CAPS methods all seem promising but (absent EPA encouragement) there doesn't seem to be much incentive to develop them further in the near-term. Perhaps one of the sites in a pilot exploratory urban visibility network (or a few small laboratory research grants) could be used to enhance and further evaluate these evolving methods. It would also be useful to evaluate the extent to which the presumably superior approach of nephelometer + aethalometer with PM_{10} heads approach can be demonstrated to provide a superior, visibility-relevant regulatory metric to that which might be provided by using existing hourly $PM_{2.5}$ mass, airport ASOS or camera techniques.

Questions Regarding the Establishment of Specifications and Procedures for Approval of Federal Reference Methods (FRM's) and Federal Equivalent Methods (FEM's).

Considering the need to establish FRM's and performance criteria for FEM's to meet the light extinction measurement goal and also considering the recommendation above from the BOSC review, please address the following questions:

5. Identify the advantages and disadvantages of the following potential options for approval of a light extinction method as a FRM. Please provide specific advice on how to best address scientific questions on interferences, precision, accuracy, and operability; degree of data needed to support decisions; who could perform the work; what kind of peer review would be appropriate, and whether the approach would potentially lead to more innovation in the measurements system or not. Note: if an option could lead to more or less innovation, depending on other factors, please explain.

a. **Translate the measurement goal to a performance standard(s) plus procedures for demonstrating that the performance standard is met, without specifying any particular measurement principle. What aspects of performance should the standards cover?**
   See below.

b. **Specify the measurement principle(s), calibration procedure(s), and operational performance requirements and demonstration procedures? What aspects of performance should the standards cover?**
   See below.

c. **Specify a particular instrument model or models as the Federal Reference Method, and rely on the equivalent method process to allow for approval of other models. What side-by-side performance testing requirements would be appropriate under this approach?**
See below.

d. **Provide the specification for the measurement principle(s), calibration procedure(s), and operational performance requirements and demonstration procedures as in b. above; but also specify one or more specific makes and models that would serve as already approved reference methods. Note this would be similar in practice to the Australian/New Zealand StandardTM, Methods for sampling and analysis of ambient air, Method 12.1: Determination of light scattering – Integrating nephelometer method. In that method, a generic approach for the method is provided with an appendix that describes the calibration and response of specific integrating nephelometers.**
Other panel members will have more informed opinions on these questions. As I see it, it does not make sense to propose a specific secondary PM light extinction NAAQS without knowing clearly in advance and specifying a method by which it could be widely measured (at urban, suburban and rural – all non-class 1 areas) throughout the country. At best, a small, pilot exploratory urban visibility monitoring network may be in place at the time the NAAQS will need to be promulgated. So unless some form of existing measurements – such as continuous $PM_{2.5}$ mass or ASOS visibility is employed it seems like a requirement for a strictly defined PM light extinction indicator essentially pushes any secondary NAAQS decision into the next PM review cycle, and renders many of these detailed questions on performance standards, calibration methods and equivalent methods to be premature.

6. Which aspects of a light extinction measurement could be adequately assessed in a laboratory and which require field studies (perhaps across multiple air sheds). For example, are laboratory challenges for a calibration gas and other similar test sufficient to test an instrument, or are experimental studies needed to ascertain the sensitivity of (or effects of humidity on) the instruments and are field challenges required to evaluate different real world aspects of the performance standard (e.g., aerosols varying geographically and interferences)? If a combination of both, please explain which aspects of an instrument are best suited for laboratory challenges and which in the field.
Certain aspects of instrumental response, such as effects of varying temperature, RH, aerosol size distribution and chemical composition and consistent responses to calibration gases or aerosol mixtures can and should be evaluated in laboratory testing. However, since there has been very limited experience in the consistent, long-term application of the proposed methods in the field (outside of relatively clean Class 1 areas), there is a need to rigorously confirm that any proposed methods will perform as expected under challenging and varied field conditions.

7. Would some aspects of performance be better addressed through a design standard, e.g., for the flow rate and the geometry of the PM10 inlet, rather than a performance specification and demonstration requirement?
As indicated above, I don't necessarily agree that a PM10 inlet is desirable. Attempting to include coarse particle effects has several disadvantages and offers little payback. I would prefer limiting to PM 2.5 or developing methods that might periodically switch back and forth from PM 2.5 to 10 inlets.

8. What data and analysis does the Subcommittee believe EPA staff should have studies or performed in establishing some kind of FRM (5.a-d) for use in regulatory decisions and to help inform the public?

As indicated above, I think a $PM_{2.5}$ mass indicator, existing ASOS measurement or camera techniques are all worth considering. Impaired visibility is/should be the most readily and publicly perceptible effect of air pollution, and care should be taken to make any regulatory metric based on it to be very simply and clearly communicable to the public. Conveying (processed, uncensored) ASOS data and camera views of impaired visibility to the public in n3ear-real-time via AIRNOW would be very useful.

9. While we have already solicited advice on a method to meet the light extinction measurement goal, we would like to explore this topic further as it relates to options for FRM's and FEM's and their eventual deployment in routine monitoring networks.

a. **Of the available or soon to be available approaches, are any sufficiently limited so that EPA should not further consider them as FRM candidates, need not ensure that the FEM provisions provide a path to their approval as FEMs, and should not consider them when offering advice to or procuring equipment for state, local, and tribal agencies?**
   Other panel members will have more informed opinions on this question. I think it is way too early to be thinking about any FEM procurement advice to SLTs – if there were money or methods – of which there are neither.

b. **Are any of the methods clearly superior in operation and also meet the measurement goal, such that they should be adopted as the FRM and thus serve as the "gold standard" for approval of FEMs (under one of the three FRM approaches listed in question 5(c or d)), and/or for possible widespread deployment?**
   No.

c. **What does EPA staff need to know about the biases of various instruments and should the FRM and FEM require methods to adjust for these biases to ensure data of known quality?**
   No opinion.

d. **What weight should EPA give to other factors in establishing a reference method for routine PM light extinction monitoring? Please comment on each of the following:**

   i. **resources needed to acquire and fully support routine operation;**
      This is obviously important, especially given the many new (unfunded) monitoring requirements (for source-specific Pb, source-specific SO_2, roadside NO_2, rural ozone, etc. that EPA has recently imposed, and current state budget crises and hiring freezes, and total lack of current extinction measurements. Consequently any approach like PM2.5 mass or ASOS that could utilize existing measurements should be carefully considered.

ii. current availability;

Since it doesn't seem likely a new PM light extinction standard can actually be implemented in this review cycle, current availability may not be critical. Conversely, if a protective secondary standard is mandated by the court decision, then priority should be given to using currently available measurements.

iii. record of successful field experience; and

Obviously more important for near-term deployment and less important for more future applications.

iv. ability to generate supplemental information (e.g. multiwavelength scattering/absorption, albedo, forward/backscattering, scattering polarization, etc.)?

Assuming a more distant future application of new methods and increasing future importance for climate forcing analyses, such supplemental information is always desirable, pending costs.

Questions Regarding Network Design and Probe and Siting Criteria

10. To what extent does the Subcommittee concur that it would be appropriate to focus a network design strategy on sites that can characterize the maximum visibility impairment across an urban area? What other considerations should EPA include in setting a network design strategy?

I'm not sure the "maximum" impairment is necessarily the best/only focus. Maybe consider the "most typically perceived" impairment, or that which most impacts scenic vistas, etc.

11. EPA and the State monitoring programs have an extensive historical dataset of PM2.5 mass and speciation measurements. In the Visibility Assessment Document, EPA used existing PM speciation and mass data to evaluate visibility impairment at a single site in each of 15 cities. However, the selection of sites used in this evaluation was severely constrained by the availability of sites with the necessary types of collocated measurement, and in several cases the site used was not the site with the highest concentrations of PM in the respective city. EPA expects that a review of available data within each city combined with information from networks assessments would be appropriate to identify likely candidate locations for light extinction measurements. Such measurements are likely to be in the area of expected maximum PM concentration that are also at neighborhood or urban scale and would complement and be complemented by PM mass and speciation measurements.

a. To what extent does the Subcommittee support collocation of PM mass and light extinction measurements to complement each of the measurements systems while also achieving the purpose of both the primary NAAQS and potential secondary NAAQS? Please offer specifics as to the advantages and disadvantages of collocating both types of measurements systems in an area-wide location of expected maximum concentration.

Collocated PM mass and visibility measurements are highly desirable for assessing causality, as a form of quality assurance and to facilitate public communication.

b. Considering the intra-urban variability of PM in any city, what additional factors (e.g., population, expected poor visibility, scenic views, etc.) should be considered to prescribe monitoring locations? Under what circumstances would multiple sites be appropriate to characterize the maximum area-wide visibility impairment across an urban area?
It seems likely that intra-urban variability should be greater for light absorption than for fine particle scattering. To the extent that such variability is caused by individual sources or coarse particles, I think it should be avoided rather than sought after, its spatial variability is likely too complex to capture in routine monitoring networks. Conversely, perhaps those are the kinds of situations where long (integrating) path measurements would be most appropriate.

12. What aspects of probe and siting criteria should be emphasized to ensure that the placement of a PM light extinction instrument is not in a local "heat island" which could also be a "dry spot" with respect to relative humidity?
The least of our problems…

13. In an urban area the average height of the typical sight path is likely well above the inlet height of most current air quality monitoring; however, the mixing of aerosols impacting light extinction occurs throughout the boundary layer. Considering site path, aerosol mixing, the goal of PM light extinction measurements, site logistics, and the location of other air monitoring equipment inlets, what should be the acceptable range for probe height?
Good question. I don't know, but another reason some sort of path measurement might be desirable.

Dr. Warren H. White

Measurement goal for light extinction

The details of the measurement goal – the 550 nm wavelength and the 10 um size cut – create unnecessary difficulties for the measurement.

The wavelength is the easier of the two to dismiss as an arbitrary choice. The specified 550 nm is, indeed, the approximate wavelength to which "humans are most sensitive". But it is not the wavelength to which the eye is most exposed, which is shorter (box A). Nor is it the wavelength carrying the most information from distant objects, which is longer (box B).

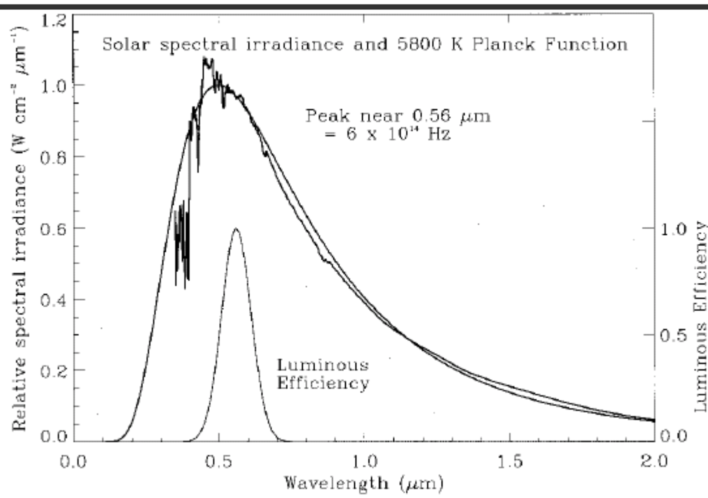

Fig. 1. The solar spectrum plotted in wavelength units peaks near 500 nm. The luminous efficiency of the eye peaks at 560 nm. All three curves appear to peak near 500–560 nm, a wavelength region generally perceived as being green.

A. B.H. Soffer and D.K. Lynch (1999) American Journal of Physics 67, 946-953.

B. H. Horvath (1981) Atmospheric Environment 15, 1785-1796.

> Any instrument that is used for indirect visibility determinations has a spectral sensitivity different to the human eye. In order to compare readings by one of the instruments and visibility observations, the wavelength of 550 nm is generally used, since this is the wavelength of maximum sensitivity of the human eye. In the atmosphere the brightness difference of a distant target is highest in the red, and smaller in the green. Therefore the most important wavelength for visibility is not at 550 nm, but approximately at 580 nm, because here the higher contrast of the target is more important than the slightly smaller sensitivity of the eye (Horvath, 1975). This wavelength should be used for converting telephotometer and nephelometer data into visibilities.

It would be no more arbitrary, in other words, to pick any other wavelength between, say, 520 nm and 580 as our measurement goal. But such flexibility would then allow the use of lasers operating at 531 nm, as noted in Table 1 of the white paper. This, in turn, would open the door to cavity ring-down and photo-acoustic instruments, which are directly based on physical principles and many view as the preferred techniques of the future. Sure, Agency guidance can always specify empirical Angstrom exponents with which data can later be translated to different wavelengths, but why create the need for such "corrections" if we don't have to?

Like the 550 nm wavelength, the PM_{10} size fraction is another arbitrary choice. Particles larger than 10 um contribute very little extinction, but particles larger than 2.5 um don't contribute all *that* much either. Consider the size-resolved extinction data in Box C, from measurements in the arid Southwest: even within the dust mode, composed overwhelmingly of particles larger than 2.5 um ("coarse"), half or more of the PM_{15} extinction was from the sub-2.5-um ("fine") tail.

C. W.H. White, E.S. Macias, R.C. Nininger and D Schorran (1994) Atmospheric Environment 28, 909-921.

Table 3. Mass and scattering budgets for fine and coarse haze and dust

Spirit Mountain, NV				Meadview, A			
Mass (%)				*Mass* (%)			
	Haze	Dust	Sum		Haze	Dust	Sum
Fine	$29^{\pm 2}$	$9^{\pm 2}$	38	Fine	$22^{\pm 2}$	$10^{\pm 2}$	32
Coarse	$7^{\pm 2}$	$56^{\pm 2}$	62	Coarse	$5^{\pm 2}$	$63^{\pm 2}$	68
Sum	$35^{\pm 4}$	$65^{\pm 4}$	100	Sum	$27^{\pm 3}$	$73^{\pm 3}$	100
Scattering (%)				*Scattering* (%)			
	Haze	Dust	Sum		Haze	Dust	Sum
Fine	$51^{\pm 3}$	$23^{\pm 3}$	74	Fine	$46^{\pm 4}$	$22^{\pm 4}$	68
Coarse	$11^{\pm 1}$	$16^{\pm 1}$	26	Coarse	$8^{\pm 1}$	$24^{\pm 1}$	32
Sum	$62^{\pm 4}$	$38^{\pm 4}$	100	Sum	$54^{\pm 5}$	$46^{\pm 5}$	100

Values represent daytime (1100–1900) averages from April to September, 1989. Superscripts indicate the variability introduced by varying estimates of the haze and dust size distributions.

Fine and coarse mass concentrations were determined by filter sampling behind fine ($D_{aero} < 2.5\ \mu m$) and total ($D_{aero} < 15\ \mu m$) inlets. Haze and dust mass concentrations are inferred from measured fine and coarse mass concentrations, based on estimated size distributions of haze and dust.

Fine and coarse scattering coefficients were determined by nephelometry behind fine ($D_{aero} < 2.5\ \mu m$) and total (unrestricted) inlets. Measured coarse scattering is doubled to correct for angular truncation error. Haze and dust contributions are derived by regression of size-resolved scattering on haze and dust mass.

Much of the extinction by particles greater than 2.5 um in diameter takes the form of extreme-forward scattering. As the white paper notes, this is under-measured by all nephelometers. (Just as forward-scattered light is less evident to nephelometers, it has less impact on visibility under most viewing conditions.) The angular truncation error of nephelometers will be well over the 10% target for any aerosol dominated by coarse particles, and will be sensitive not only to the particles' size distribution, but also to their almost certainly non-spherical shape. It is hard to see how the effects of minor design differences between instruments could be accounted for by any manageable inter-calibration procedure.

The 10 um cut-point was originally designed to reflect aerodynamic characteristics of the upper respiratory system, a rationalization relevant to a health standard but irrelevant to visibility. The other PM NAAQS size cut, at 2.5 um, is also grounded in a health rationale, but is much better suited to optical monitoring. It delivers a fraction accounting for most of the extinction but much less affected by sampling losses and measurement artifacts.

Dr. Yousheng Zeng

General Comments:

EPA is considering either light extinction or a mass concentration based metric as the indicator for the PM Secondary Standard. Although I agree that light extinction is a more direct measurement of visibility degradation than a mass concentration metric, using light extinction may bring more challenges in implementing the standard, particularly as it relates to the New Source Review (NSR) program under the Clean Air Act.

One of the reasons to set an ambient air quality standard is to manage air pollution sources, especially anthropologenic sources that can be managed. If EPA plans to implement the PM Secondary Standard in the same way as current SO_2 Secondary Standard, a proposed source subject to the NSR in attainment area will undergo a Prevention of Significant Deterioration (PSD) review. As part of PSD review, the source will perform an air quality modeling analysis. The modeling analysis will limit the mass emission rate so that the new source will not exceed or contribute to exceedance of the NAAQS and PSD Increment. This linkage between the emission limit and the NAAQS and PSD Increment will have to be on mass basis. If the PM Secondary Standard (and presumably corresponding PSD Increment) is based on light extinction, EPA presumably will establish an algorithm to convert the initial model output, which is mass concentration (*e.g.*, $\mu g/m^3$), to light extinction.

The algorithm to convert modeled PM concentration to light extinction must be part of the regulations and enforceable. If this conversion is required by regulations anyway, it will be easier to just set the PM Secondary Standard in $\mu g/m^3$ rather than light extinction. Although using light extinction as the indicator makes scientific sense, using PM mass concentration as the indicator leads to the same policy and regulatory endpoint and brings multiple practical benefits. These benefits include: (1) the same PM monitoring instruments (maybe with relative humidity instrument added) can be used for both PM Primary and Secondary Standards – a significant cost saving, (2) data needed for attainment designation may be derived from existing PM monitoring instruments – shorten the designation time, and (3) both regulating and regulated communities are already familiar with the PM mass concentration in $\mu g/m^3$ – minimizing confusion and training effort.

Using light extinction as the indicator may be advantageous if EPA allows use of different algorithms in different geographic areas or under different environmental conditions. In that case, there will be one light extinction standard across the country, but different corresponding mass concentration levels depending on local conditions. However, this will further increase complexity of the implementation related regulations.

CQ 8 – What data and analysis does the Subcommittee believe EPA staff should have studied or performed in establishing some kind of FRM (5.a-d) for use in regulatory decisions and to help inform the public?

As part of the effort to establish FRM for light extinction measurement, EPA should conduct a study on candidate methods at a broad wavelength band centered around 550 nm vs. the monolithic wavelength of 550 nm (or 531 nm), or multi-wavelength vs. 550 nm. The study should include not only instrument measurement, but also corresponding images for a sample of population to view, similar to the urban visibility preference studies.

The wavelength is one of the multiple aspects in evaluating candidate methods for FRM. Other aspects should also be studied such as relative humidity, aerosol size fraction, precision, accuracy, and operability. In field studies, PM mass concentrations should be measured side-by-side with candidate light extinction measurement methods. The comparative study between light extinction measurement and PM mass concentration measurement is very important in addressing the implementation issues discussed in the General Comments section above.

CQ 12 – What aspects of probe and siting criteria should be emphasized to ensure that the placement of a PM light extinction instrument is not in a local "heat island" which could also be a "dry spot" with respect to relative humidity?

If there is an urban scale heat island, the scale is actually consistent with the objective of light extinction measurement, *i.e*., how people living in the city feel about visibility. In this case, there is no need to avoid urban heat island.

As long as the monitoring site is not in close proximity of a large heat source, this does not seem to be a major concern. Typical monitoring sites have sufficient distance from major sources. Localized hot air moves upward and have less impact to the measurement. The ground surface or surface of the structure where light extinction instrument stands may have some influence on localized relative humidity change, *e.g*., a concrete pad below the instrument may cause localized heating and reduction in relative humidity. This type of surface should be avoided, especially if the sample inlet is low. Other significant heat sources, such as discharge vent of a large building ventilation system.

A site near water body may not be representative. Some industrial stationary sources also emit a lot of water vapor. EPA may consider some dispersion modeling study to establish distance guideline, heat output/distance guideline, or even heat output/moisture output/distance guideline.

CQ 13 – In an urban area the average height of the typical sight path is likely well above the inlet height of most current air quality monitoring; however, the mixing of aerosols impacting light extinction occurs throughout the boundary layer. Considering sight path, aerosol mixing, the goal of PM light extinction measurements, site logistics, and the location of other air monitoring equipment inlets, what should be the acceptable range for probe height?

From the viewpoint of the purpose of light extinction measurement, sight path should be the most important factor in probe height consideration. For this reason, the probe height should be higher than typical PM mass concentration monitors. A higher probe will also minimize near ground impact of large particles. A height comparable to a typical met tower at a monitoring station (e..g., 10 m) seems a good starting point. To minimize aerosol composition change, the distance from the probe intake to actual measurement chamber or filter should be as short as possible and as straight as possible.