22 February 2010 Preliminary Review Comments from CASAC AAMMS on EPA's *White Paper on PM Light Extinction Measurements*

Comments received:

Mr. George Allen

These preliminary comments focus on bullets 3, 4, and 5:

3. Suitability of nephelometer and alternative approaches
4. Potential improvements to commercial nephelometers and future alternatives
5. Potential Options for FRM method

General background.

Ideally an open path method (transmissometer) would be used for B-ext measurements, since it is both an open path and an in-situ 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 closed path 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. For the present review of the PM NAAQS, I support a PM2.5 FEM sub-daily approach. I do not support a full network of b-scat and/or b-abs measurements at this time.

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. Despite its larger optical errors, the Ecotech is a good candidate for routine use in SLT monitoring networks because of its robust design. The Ecotech "Aurora" model (http://www.aurora-nephelometer.com) is the most recent version of the Ecotech model 9003 nephelometer. 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 a higher flow rate than the 5 LPM presently used. Another factor is the angular truncation of this instrument. The optical performance (truncation error) can 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 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

Changing the light source is trivial since suitable "white" LEDs are now readily available. The errors presented here for fine-mode aerosol with the Ecotech instrument are aerosol size dependent and on the order of +/- 5%, which is not large in a practical sense. The Ecotech instrument supports measurements at multiple wavelengths, so it would be possible to get scattering at 3 discrete (RGB) wavelengths as well as “white” (broad 550 nm). The hardware package is user friendly, and chamber cleaning is not difficult.

A good background paper on general issues with making b-scat measurements is:
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
This paper includes a discussion of the significant limitations of nephelometers when coarse mode particles dominate the scattering.

Nephelometer inlet considerations:

Ideally a PM10 cut would be used on a nephelometer for any secondary PM NAAQS. In practice, this is not normally done; ambient temperature nephelometers for visual range measurements are normally run without any size cut. What is essential is the use of some sort of fine mesh “bug screen” to prevent insects from entering the instrument. One consideration is to constrain the inlet to fine-mode particles; this would 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.

Absorption (B-abs) measurements:

The best commercial measurement method for b-abs is the photo-acoustic method (Moosmüller and Arnott); this could be considered for use as an FRM. For routine network use, a surrogate measurement of light absorption of atmospheric particles can be done with optical transmission (optical density) measurements. 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. This method has a substantial "spot loading effect" that biases the data low as the filter loads with aerosol; this 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

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. 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). The b-abs measurement at 880 or 670 will underestimate the b-abs at 550, since wood smoke has substantially enhanced b-abs at shorter wavelengths. A suitable "non-narrow" band light source centered at or near 550 with proper symmetry 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.

A recent and rigorous review of b-abs methods is:
Moosmüller, Chakrabartya and Arnott (2009), " Aerosol light absorption and its measurement: A review". Journal of Quantitative Spectroscopy and Radiative Transfer,
DOI: 10.1016/j.jqsrt.2009.02.035

Alternative Approaches

Charge question 3b: What are the Subcommittees thoughts on alternative instrumental approaches that should be considered to meet the light extinction goals?

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.

Dr. Judith C. Chow

PM Light Extinction Measurement Goal and Method

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.

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).

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 $PM_{10-2.5}$ 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).

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.

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.

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).

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.

Please comment on inclusion of the following additional performance specifications:

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.

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.

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.

Suitability of currently available nephelometers and filter transmission systems.

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.

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.

Instrument Improvements

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

As noted above, 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.
- 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 sample zone.
- Combining scattering and absorption measurements into a single instrument.
- Upgrading data acquisition and analysis software to better meet the needs of an urban haze standard.

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.

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

Performance vs. Design Standards

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.

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.

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.

Which aspects of a light extinction measurement could be adequately assessed in a 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.

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.

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.

Evaluation of current measurement technology

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.

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.

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.

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

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.

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.

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

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.

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

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.

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.

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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Dr. Delbert J. Eatough

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

Charge Question 1: Adequacy of the goal.

The PM light extinction goal is the precise and accurate measurement of PM light extinction at a wavelength of 550 nm.

a. Wavelength of 550 nm. I am not in total accord with the wavelength goal as stated. It is most probable that total light extinction will not be measured, but a combination of scattering and light absorption by particles will be the designated measurements. The choice of 550 nm seems to be based on assumed maximum sensitivity of a human subject to visibility degradation as outlined in the "Particulate Matter Urban-Focused Visibility Assessment, Second External Review Draft, January 2010."The draft assumes possible standard goals based on several human response studies. The human observer is most sensitive to changes in view contrast. This will be the of perceived quality of a view based on the combination of scattering and absorption which influences the observer's view. While the assumption that the effects of scattering as perceived by the viewer will be maximum at 550 nm is reasonable, I do not think that this is an important issue with respect to absorption, as absorption of light by particles results in a decrease in visual quality which is much less dependent on the actual wavelength at which the absorption measurement is made. I consider this an important question because I do not believe the wavelength of the absorption measurement will be pertinent to the observer response and the choice of 550nm for an adsorption measurement limits the use of some pieces of equipment which are well characterized, in extensive use and appropriate as a potential designated method. In particular I think of the dual wavelength Aethalometer which measures absorption at 880 and 370 nm. The two measurement results are generally very comparable and also compare well with Sunset Laboratory field measurements of "elemental carbon" (e.g. Grover, Brett D.; Eatough Norman L.; Woolwine, Woods R.; Cannon, Justin P.; Eatough, Delbert J.; Long, Russell W. "Semi-continuous Mass Closure of the Major Components of Fine Particulate Matter in Riverside, CA." *Atmos. Environ.* **2008**, **42**: 250-260). Establishing a measurement criteria which would exclude this and related instruments is unwise.

I have a few other comments not directly related to this charge question but are related to the above referenced EPA Viability Assessment which I think are relevant to the current charge to AAMMS. The Assessment includes extensive evaluation of both a possible 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, as noted above, 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) because there are no appropriate landmarks 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 studies less than persuasive, largely because of the crudeness of many of the assumptions. I think EPA should consider additional efforts to shore this area up with studies which focus on measured extinction, measured mass and, where available measure hourly average composition. When measure 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 measurements 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. The secondary standard evaluation will be immutable connected to the quality of the mass measurements to which it is compared. I urge evaluation of areas (such as SLC) where good FDMS TEOM measurements are available be focused on. Good 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.

b. Aerosol size fractionation at PM_{10}. I am not committed to the importance of a PM_{10} cut in the extinction measurement. I have commented on this further in my answer to charge question 3.

c. Operation at ambient relative humidity. Because of the strong effect of relative humidity on light scattering by fine particles, this is essential.

d. - e. All reasonable goals.

Charge Question 2: Additional Specification.

a. Measurement averaging times. The goal of the secondary standard is to provide a visual view acceptable to the human observer. We respond to changes in contrast all well as the absolute contrast. In this regard, an average day-time value would be meaningless. A one-hour average seem appropriate, particularly in light of the fact the related data to which the extinction measurement will be compared are generally available on either a 24-hr or a 1-hr average time basis.

b. and c. I need to think further on these two points.

Charge Question 3. Suitability of nephelometer and alternative approaches.

a. Suitability of currently available nephelometer light scattering and filter transmission light adsorption instruments.

I believe it is critically important that instruments be used which will give semi-continuous in-situ measurements. The 1-hr averaging is important for the reasons given above. The ability to get in-field data is important because at the current state of instrumentation, EPA should not tie itself to any method which requires additional analysis effort after sample collection, e.g. post sampling measurement of light transmission or absorption on transported filters.

Total PM extinction measurement. Total PM extinction is the measurement goal, because this is what the human observer responds to. Open path trasnmissometry is a technique used extensively by the NPS in the IMPROVE program, with good success. However, the IMPROVE program focuses on areas which generally have will have little, if any, spatial differences in the extinction measurement so that a long path length measurement is feasible. This will not be the case in an Urban environment and I do not think trasnmissometry should be considered as an option. However, the point made in Table 1 of the White Paper on the method not excluding particles greater that 10 µm is not germane, I believe. The human eye is sensitive to total extinction and not just extinction from particles smaller that 10 µm. I have commented on that point above. However, with the exception of fog and precipitation events, the point is probably not important because of the size dependant light scattering properties of particles. For this reason, I do not think the upper cut point curve of the measurement method is important. Not accurately characterizing extinction from particles, say in the range of 8 µm and higher, will generally have little influence on the accuracy of the total PM light extinction measurement. This comment also applies to the methods discussed in the following paragraphs. I would like to make comments on the cavity ring down (CRD) and cavity attenuation phase shift (CAPS) methods, but I am less familiar with these techniques. I have requested copies of the Arnott, et al., 2009, Baynard, et al., 2007 and Kebabian, et al., 2007 and Moosemuller et al., 2009 references in the White Paper and will give further input when I have seen these.

Total scattering measurement. There is much to be said for the adoption of the NGN Nephelometer used so extensively in the IMPROVE program Among other reasons, this would allow integration of the Urban and pristine extinction measurements in a nation wide evaluation.

Table 1 lists excluding ultra coarse particles as an advantage. For the reasons given in the paragraph above, I consider this a disadvantage, but not a serious one. With respect to the potential disadvantages listed in Table 1:

- Because the coarse particle loss is important only as the particle size increases, and the scattering decreases as the particle size increases, the effect on total extinction will be small, I consider this unimportant. This will generally be true because most high extinction events in Urban areas will be driven my inversion conditions with elevated fine particulate matter.
- Relative humidity and sample heating problems have already been rigorously addressed in the IMPROVE program. I do not believe any other method will do better in this regard.
- Angular truncation is again only an issue for larger coarse particles and is probably not important with respect to the scattering measurement.

Total absorption measurement. I have commented above on the desirability of allowing a broad enough definition of the absorption measurement to permit the use of instrument such as the two wavelength Aethalometer. I do not think the absorption measurement needs to be tied to a 550 nm measurement. I also think that unless there is compelling evidence for significant coarse particle absorption in Urban environments that coarse particles need to be included in this measurement.

b. Alternate instrumental approaches.

I will wait until I have had a chance to read the above mentioned references from the White Paper to comment on this issue.

Charge Question 4. Potential improvements to commercial nephelometers and future alterations.

a. and b. No comments at this time.

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

EPA is to be commended for desiring to explore new approaches to the establishment of an FRM and FEM's. In the setting of the primary standard FRM, methods were established which have made it difficult for improved methods for measuring PM mass, particularly $PM_{2.5}$ with an improved scientific basis to be used. The absolute need for the FRM for light extinction is to measure the quantity as it exists in the atmosphere. Modification of the measured aerosol to affect semi-volatile material, especially water, will produce an inferior result. I have not had a chance to read the BOSC recommendations before writing this but will do so before the Wednesday meeting. Like wise, I would be very interested in reviewing the AS/NZ Method 12.1 approach as I cannot locate a downloadable version on the web. I would be interesting in reading that to set my thinking also. With these limitations, I address the charge questions I have thought through carefully enough in this section below. There are several with no answers yet.

Charge Question 5. Potential Options for FRM method.

a. Translate the measurement goal to a performance standard.

While this is obviously desirable from the point of view of an FRM decision, it also has the possibility of boxing us into an FRM which cannot be changes and which will later be shown to not reflect extinction well. I encourage a different approach.

b. Specify the measurement principle(s). calibration procedure(s), and operational requirements and demonstration procedures.

While this approach is harder to implement, I prefer it to a. for the reasons given in previous sections.

c. Specify a particular instrument model or models as the FRM.

This is tempting, and I have made suggestions above on instruments I believe are well suited. However, I believe it is safer to use the a. approach and suggest instruments as appropriate. Certainly a harder strategy to implement, but scientifically safer.

d. a combination of b. and c.

I have no yet read the AS/NZ Method 12.1 document. But his approach is appealing.

Charge Question 7. Design standard versus performance specification.

I prefer to use performance standard throughout. I have mentioned above that I do not consider the inlet design critical for the Visibility FRM.

Charge Question 8. Data and analysis for establishing FRM.

I would encourage a study which uses the candidate instrument collocated with existing FDMS TEOM (with RH considered) and ASOS data to evaluate the performance of potential FRMs.

Charge Question 9. Alternative methods, additional questions.

a. Approaches not to consider.

b. A "Gold Standard".

Not acceptable.

c. Biases and bias correction.

This question opens a real "can of worms". The need to correctly measure extinction to allow for meaningful determination of visibility impairment is clear. I would strongly encourage that EPA avoid a standard which allows a measurement which is later shown to have bias to continue to be the basis of decisions and then allow the adjustment of data from a better measurement method to agree with the method with bias. If the standard is the correct measurement of extinction then can we adjust past data to more closely reflect the standard if flaws are discovered. A very different approach, but a scientifically better one in my opinion. Will the process allow such an approach?

d. Other factors.

The following statements are based on the assumption that the NGN nephelometer and the Aethalometer are instruments expected to give data which correctly reflect the extinction of the atmosphere. The statements also assume that resources need to install and maintain these instruments are reasonable based on past field experience.

- Cost. Any candidate method other than scattering and absorption direct measurements should be shown to be comparable in needed resources.
- If the assumptions are true, the expenditure of significant resources to replace these readily available instruments with another technique would not seem warranted unless future research clearly indicates a problem with the nephelometer and/or Aethalometer which is not currently know.
- Both the suggested instruments have a long record of successful field use.
- I will leave this question to those with more experience with other candidate instruments.

III. Questions Regarding Network Design and Probe and siting Criteria.

Charge Questions 10 and 11. Network Design Strategy.

10. Identify maximum visibility impairment. Is would be wisdom to strive for the location of maximum PM. However, the goal is human perception. Some consideration of probable impact of the site on perception (horizon view, etc.) would seem appropriate.

11. Other design considerations.

a. Site with PM characterization equipment. Such siting si obviously desirable. However, I believe that the siting with other equipment should come with strings attached. Siting with a site that has only a conventional TEOM for hourly average mass or collects only 24-h composition would not be useful. This would only encourage the proliferation of unwarranted assumptions in data analysis. I would suggest the site should have equipment which will measure semi-volatile fine particulate material and/or have equipment which measures these species on an hourly average basis correctly.

b. Intra-urban variability. Since human perception is the bottom line, the importance of several sites, as opposed to a site well chosen with respect to scenic views and time-of-day differences in perception are more important than characterizing city variability.

Charge Question 12. And 13. Probe and siting and probe height criteria.

My only current comment here is to be sure a path length measurement is not considered.

Mr. Dirk Felton

Preliminary Responses to Charge Question 5

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 will permit the greatest latitude for innovation, however 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 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 will inevitably 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.

To avoid the disadvantages of specifying a performance standard instead of a measurement principle, there are two options. The performance standards either have to be very comprehensive and include all possible monitoring environments including varying atmospheric component concentrations and environmental factors such as temperature, humidity and elevation or the performance standards have to be evaluated on an as needed basis in-situ. The site specific evaluation could be implemented in a similar fashion to the ARM program that was part of the 2006 PM-2.5 Implementation Rule.

(Can lead to black boxes that may only work where evaluated)

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.

Dr. Kazuiko Ito

Questions regarding a PM Light Extinction Measurement Goal and Method

1. Does the Subcommittee agree with the goal identified?

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

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%

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?

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?

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?

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.

Mr. Rich Poirot

Pre-Meeting Comments on PM Light Extinction Measurements White Paper and Charge Questions

Charge Questions

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 PM_{10}

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 $PM_{10\text{-}2.5}$ 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 $PM_{10\text{-}2.5}$, 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 98th percentile and 30 dv 90th 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).

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 Standard_{TM}, 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 PM_{10} 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. 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?***

   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 $PM_{2.5}$ 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 byPM 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. 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.

Suitability of Nephelometer and alternative approaches

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.

Potential improvements ot commercial nephelometers and future alternatives.

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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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).

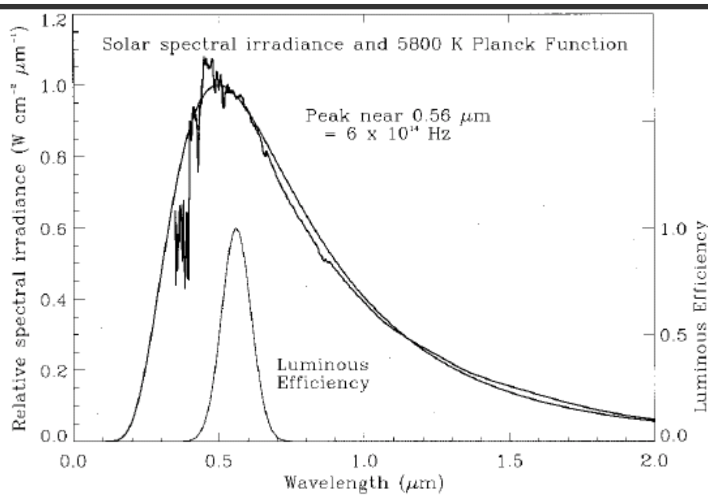

Fig. 1. The solar spectrum plotted in wavelength units peaks near 500 nm. The luminous efficiency of the eye peaks at 560 nm. All three curves appear to peak near 500–560 nm, a wavelength region generally perceived as being green.

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

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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	$11^{\pm 1}$	$16^{\pm}$ [illegible]	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.