U.S. EPA's Advisory on PM Light Extinction Measurements

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

<u>Questions regarding a PM Light Extinction Measurement Goal and Method</u>

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

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

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

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

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

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

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

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

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

a. Averaging times are very important in using a point measurement to represent an areally dispersed phenomenon. Short averaging times would make the measurement unduly sensitive to local "puff" emissions or short term variations in PM composition or concentration. In the context of using this measurement to supplement health protective PM mass concentration regulations, the averaging time should be set to approximate the relevant exposure time (e.g., 24-hr light attenuation to supplement 24-hr PM mass regulations). If the goal is to provide a welfare benefit of good regional

visibility, then the averaging time should reflect human perception of “good visibility” based on survey or laboratory studies of human responses to short term visibility degradation. The latter would probably use shorter averaging time, on the order of one or a few hours, rather than the 24-hr criterion derived from current health-based PM NAAQS.

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

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

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

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

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

be used for transmission absorption measurements as they violate the physical assumptions of the measurement.

b. Alternative measurements of light absorption are available. Although subject to some siting constraints, subtraction of nephelometer scattering from long path light extinction can yield light absorption. Within the context of keeping the measurement compatible with traditional monitoring site operations, two alternatives are available. Switching to a reflection measurement with the filter set against a white background (and measuring the base transmission of an unexposed portion of the filter as I_0) is a viable method, compatible with existing FRM samplers (and assuming that the protocol takes account of the problem of heavily loaded filters – a problem also present in the transmission measurement). Alternatively, the nephelometer can be replaced with a cavity ring-down optical measurement[1] which can give both total extinction (on-axis decay) and scattering (off-axis intensity), and thus absorption by subtraction, all in a single instrument. Such a machine has been built by Dr. Anthony Strawa for use by NASA in airborne atmospheric research, and could be easily commercialized if EPA elected to pursue this method. The cavity ring-down instrument would eliminate the dual instrument problem and facilitate unified calibration.

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

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

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1 Strawa, A.W., R. Castaneda, T. Owano, D. Baer, B. Paldus, The Measurement of Aerosol Optical Properties Using Continuous Wave Cavity Ring-Down Techniques, *Journal of Atmospheric and Oceanic Technology* 20, 454-465, 2002.
DOI: 10.1175/1520-0426(2003)20<454:TMOAOP>2.0.CO;2

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