Using Spectral Methods to Obtain Particle Size Information from Optical Data:
Applications to Measurements from CARES 2010

Dean B. Atkinson¹, Mikhail Pekour², Duli Chand², James G. Radney¹**, Katheryn R. Kolesar⁵**, Qi Zhang⁴, Ari Setyan³**, Norman T. O’Neill⁹, Christopher D. Cappa⁵

[1] [Department of Chemistry, Portland State University, Portland, OR, USA, 97207]
[2] [Pacific Northwest National Laboratory, Richland, WA, USA, 99352]
[3] [Department of Environmental Toxicology, University of California, Davis, CA, USA, 95616]
[4] [Centre d’Applications et de Recherches en Télédétection, Université de Sherbrooke, Sherbrooke, Canada]
[5] [Department of Civil and Environmental Engineering, University of California, Davis, CA, USA, 95616]

* Now at: Air Sciences, Inc., Portland, OR, 97214, USA
** Now at: Empa, Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf, Switzerland
*** Now at: Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland, 20899, USA

Correspondence to: D. B. Atkinson (atkinsond@pdx.edu)

Abstract

Multi-wavelength aerosol extinction, absorption and scattering measurements made at two ground sites during the 2010 Carbonaceous Aerosols and Radiative Effects Study (CARES) are analyzed using a spectral deconvolution method that allows extraction of particle size-related information, including the fraction of extinction produced by the fine mode particles and the effective radius of the fine mode. The spectral deconvolution method is typically applied to analysis of remote sensing measurements. Application to in situ measurements allows for comparison with more direct measurement methods and validation of the retrieval approach. Here, the retrieved fine mode fraction and effective radius generally compare well with other in situ measurements, including size distribution measurements and scattering and absorption measurements made separately for PM₁ and PM₁₀, but some limitations are also identified. These results indicate that for campaigns where size, composition, and multi-wavelength optical property measurements are made, comparison of the results can result in closure or can identify unusual circumstances. The comparison here also demonstrates that in situ multi-wavelength optical property measurements can be used to determine information about particle size distributions in situations where direct size distribution measurements are not available.
Introduction

Aerosols remain a substantial source of uncertainty in climate models, despite considerable progress in scientific understanding of their chemical, physical and optical properties in the last few decades (IPCC, 2013). As greater understanding has developed in each of these areas, new complexity is also uncovered and the interconnectedness of the various properties becomes even more evident. Light scattering by atmospheric particles has a net cooling effect on climate that is one major offset to greenhouse gas induced climate warming (Charlson et al., 2005; Bond et al., 2011). The efficiency with which the atmospheric aerosol interacts with electromagnetic radiation (e.g. sunlight) is dependent upon the size, composition, shape and morphology of the particles. These properties are not static in time, instead evolving as particles are transported through the atmosphere as a result of chemical processing, scavenging and changes in the environmental conditions (e.g. relative humidity and temperature) (Doran et al., 2007; George and Abbatt, 2010; Lack and Cappa, 2010).

Characterization of the spatial distribution of aerosol particle concentrations and properties is important to assessing their impact on the atmospheric radiation budget through direct aerosol-radiation and indirect aerosol-cloud interactions. Aerosol optical properties can be measured directly in the laboratory and in the field using both in situ methods (Andrews et al., 2004; Moosmuller et al., 2009; Coen et al., 2013) and remote sensing instruments/platforms, such as sunphotometers and satellites (Holben et al., 1998; Anderson et al., 2005). Alternatively, aerosol optical properties can be inferred from measurements of particle composition, abundance and size distributions (Atkinson et al., 2015). One particular advantage of the remote sensing instruments is that they allow for characterization of column-average atmospheric particle burdens and properties over a large spatial scale and are free from sampling biases as the particles are characterized as they exist in the atmosphere. However, they can only reliably retrieve aerosol properties under cloud-free conditions, and determination of properties beyond the aerosol optical depth (such as the single scatter albedo or the aerosol size distribution) typically requires a data ‘inversion’ process that relies on an assessment of the wavelength-dependent light attenuation and scattering (Dubovik and King, 2000). In situ methods can allow for more detailed characterization of aerosols, including the relationships between size, composition and optical properties, but typically at the expense of reduced spatial coverage and with long-term measurements typically restricted to the surface (Andrews et al., 2004). Given the wide-spread use of aerosol remote sensing and the extensive availability of the data (in particular from ground-based sunphotometer networks such as AERONET and AEROCAN (Holben et al., 1998; Bokoye et al., 2001)), continued
assessment and validation of the inversion methods by comparison with measurements by *in situ*
methods is important.

Multi-wavelength optical measurements can yield information about the aerosol size distribution, a
principle that dates back to Ångström’s observation that the wavelength-dependence of light
attenuation by particles was weaker for larger particles (diameters of hundreds of nanometers to
micrometers) than for smaller particles (Ångström, 1929). One of the simplest ways of characterizing the
wavelength-dependence of optical measurements (whether extinction, scattering or absorption) is
through the Ångström exponent. For a pair of optical measurements at different wavelengths, $\hat{\alpha}$
$= \log(b_{\lambda_1}/b_{\lambda_2})/\log(\lambda_1/\lambda_2)$, where $b_{\lambda}$ is the optical coefficient at one of the wavelengths $\lambda$; for
scattering and extinction $\hat{\alpha}$ typically increases as particle size decreases. The dependence of $b_\alpha$ on
wavelength can alternatively be obtained from a log($b_\alpha$) vs. log($\lambda$) plot using two or more wavelengths;
if the dependence is linear, a regression would obtain the same value as the pair-wise treatment, but
non-linearity can be accommodated by using the continuous derivative $\alpha = \frac{\mathrm{d}\ln(b_\alpha)}{\mathrm{d}\ln(\lambda)}$ at a
specified wavelength. (A list of the symbols and acronyms used in this work is provided in Appendix A.)
The former will be referred to here as the Ångström exponent and the latter as the spectral derivative.

Particle size classification schemes have been proposed (Clarke and Kapustin, 2010) and
supported/validated (Eck et al., 2008; Massoli et al., 2009; Cappa et al., 2016) based on the Ångström
exponent of extinction or scattering. When observations are made at more than two wavelengths
(ideally, widely spaced), further information regarding the nature of the particle size distribution can be
extracted. For example, an additional level of refinement of wavelength-dependent measurements of
aerosol optical depth (path integrated extinction) was introduced by O’Neill et al. (2005) to aid in the
interpretation of the data obtained by the ground-based sunphotometer networks AERONET and
AEROCAN. Specifically, O’Neill et al. (2003; 2005) showed that the fine mode fraction (FMF) of extinction
and the fine mode effective radius, $R_{\text{eff},f}$ could be extracted directly from the multi-wavelength optical
depth or extinction measurements available from remote sensing. The FMF provides for an approximate
discrimination between what are typically naturally produced coarse mode particles (dust or sea spray)
and what are often anthropogenically associated fine mode particles. Thus, parameters such as the FMF
can provide a nominal indication of the relative contributions of natural versus anthropogenic particles
to the atmospheric AOD. Variations in $R_{\text{eff},f}$ provide information on the sources of the fine mode particles
- as different sources yield fine mode particles with different size distributions - or the extent to which
particles have undergone atmospheric processing, which can change the size distribution (and chemical composition) in systematic ways.

In the spectral curvature approach of O’Neill et al. (2003), the fine mode spectral derivatives ($\alpha_1$ = first derivative and $\alpha'_1$ = second) and the FMF are first extracted from multi-wavelength extinction data using a process described as Spectral Deconvolution ($\eta$ was used for FMF in ibid.) The fine mode spectral derivatives are then used to obtain the effective radius for the fine mode, defined by Hansen and Travis (1974) as:

$$R_{eff} = \frac{\int_0^\infty R^2 \frac{dN}{dlnR} dlnR}{\int_0^\infty \pi R^2 \frac{dN}{dlnR} dlnR}$$ (1)

where $R$ is the particle geometrical radius and $dN/dlnR$ is a number weighted size distribution for which $R_{eff}$, is the first moment (average radius) of the surface-area weighted size distribution. $R_{eff}$ is an effective radius that characterizes, approximately, the average size of particles in the fine mode that scatter solar radiation. In this work, we compare the optically obtained $R_{eff}$ retrievals to those calculated from the observed size distributions (produced by scanning mobility particle sizes) by numerically evaluating the integrals of Equation 1 to produce comparator values. A single log-translatable particle size distribution (i.e., a PSD that can be translated along the log-transformed particle size axis without changing the form of the distribution function) is, in many cases, a reasonable representation of the size distribution of observed aerosol fine modes (O’Neill et al., 2005). In these cases, the fine mode can be characterized by the single parameter $R_{eff}$ facilitating comparisons and examination of trends in sources and/or atmospheric processing.

Methods such as those developed by O’Neill et al. (2003) for remote sensing measurements can also be applied to in situ extinction measurements. Beyond adding to the utility of the in situ optical measurements, this provides an opportunity to test the methods against other, complementary measures of particle size and size-dependent scattering and extinction. For example, Atkinson et al. (2010) used the approach of O’Neill et al. (2003) to analyze in situ, three-wavelength aerosol extinction measurements made during the 2006 TexAQS II campaign near Houston, TX. More recently, Kaku et al. (2014) showed, for a range of marine atmospheres, that the application of this spectral approach to obtain FMF from three-wavelength scattering coefficient measurements was largely coherent with the sub-micron fraction of scattering (SMF), obtained from scattering coefficient measurements of the fine and coarse mode components using impactor-based separation of the aerosol. These studies, and others, provide a useful basis for understanding the accuracy and applicability of the parameters.
retrieved from remote sensing data. However, further assessment in a wide range of environments is necessary given that networks employing such spectral remote sensing algorithms (AERONET and some surface based sites) represent locations impacted by particles from diverse sources.

In this work, measurements of aerosol optical properties (extinction, scattering and absorption coefficients) made at multiple wavelengths during the 2010 Carbonaceous Aerosols and Radiative Effects Study (Fast et al., 2012; Zaveri et al., 2012) are reported and analyzed using the O'Neill et al. (2003) and the O'Neill et al. (2008b) methods. The measurements were made at two locations near Sacramento: a more urban site in Granite Bay, CA (T0) and a more rural site in Cool, CA (T1) that were often linked by direct atmospheric transport. The multi-wavelength measurements were made using three types of optical instruments (specifically seven separate instruments at the two locations). The multi-wavelength measurements of the extinction coefficients (either measured directly or produced from the sum of scattering and absorption coefficients) are used to retrieve the fine mode fraction of extinction and fine mode effective radius. These results from the retrieval, described in more detail in the next section, are compared to other, complementary in situ measurements. Scattering and absorption coefficients were measured after aerodynamic separation into the PM$_1$ and PM$_{10}$ fractions, which allowed the sub-micron fraction (SMF) of extinction to be directly determined. The in situ SMF can be compared with the FMF from the spectral retrieval method. (In this work, sub-micron particles are those with nominal aerodynamic diameters ($d_{pa}$) smaller than 1 μm, likely resulting in geometric diameters below 800 nm.) Also, size distribution measurements allowed for determination of the fine-mode effective radii (via Eqn. 1), which are compared with that obtained from the spectral retrieval.

**Theoretical Approach**

*The Spectral Deconvolution Algorithm with Fine Mode Curvature (SDA-FMC) Approach*

This section provides a qualitative description of the fine and coarse mode AOD (or extinction) deconvolution (SDA) algorithm and fine mode optical sizing (FMC or fine mode curvature) method developed by O’Neill. The details of the derivation and application of the SDA are provided in previous publications (O’Neill et al., 2005; Atkinson et al., 2010; Kaku et al., 2014). The MATLAB code that implements the approach is available from O’Neill upon request. Application of both approaches requires a robust set of measurements of aerosol optical extinction or scattering (or optical depth) at a minimum of three wavelengths that should be widely spread across the optical region of the spectrum (near UV through the visible to the near IR; see, for example, O’Neill et al. (2008a)).
The fundamental assumption of the SDA approach is that most ambient aerosol size distributions are composed of two optically-relevant modes: a fine mode having an effective radius (and to a lesser extent, geometric standard deviation) that is a function of atmospheric processing, and a separate coarse mode, largely in the supermicron \(d_{pa} > 1 \mu m\) size range. A common assumption is that the fine mode is more closely associated with anthropogenic activities and the coarse mode with natural sources, although this can be somewhat confounded by smoke from biomass burning (Hamill et al., 2016). The FMC (Fine Mode Curvature) algorithm employs the fine mode optical parameters retrieved using the SDA to estimate both a fundamental indicator of optical particle size (the fine mode van de Hulst parameter) and from this, an indicator of microphysical particle size (the fine mode effective radius); these are both defined below.

**Spectral deconvolution of the fine and coarse mode extinction and derivation of the fine mode spectral derivatives (SDA)**

The spectral deconvolution algorithm begins by isolating the fraction of total extinction due to particles in the fine mode, based on the stronger dependence of the extinction (scattering)\(^1\) on wavelength for smaller particles. Current applications of the method start by fitting \(\ln(b_{ext})\) (or \(\ln(b_{scat})\) or \(\ln(AOD)\)) versus \(\ln(\lambda)\) to a second order polynomial, where \(b_{ext}\) is the measured wavelength-dependent extinction coefficient (see Atkinson et al. (2010) and Kaku et al. (2014) for scattering and extinction coefficient applications, Saha et al. (2010) for a sunphotometry AOD application and Baibakov et al. (2015) for a starphotometry AOD application). The extinction and its first and second derivatives at a reference wavelength of 500 nm are determined from the fit. The first derivative (i.e. slope) is denoted \(\alpha\) in analogy to the Ångström exponent, but in this non-linear, second order approach it is a function of wavelength. The second derivative \(\alpha'\) (i.e. spectral curvature) may, in principle, be wavelength dependent over the observed range, but using a second order polynomial fit yields a wavelength-invariant version. Values of \(\alpha\) and \(\alpha'\) associated with the fine mode and the coarse mode are indicated using subscript \(f\) or \(c\), respectively. In this work, only a second order fit is possible because only three measurements are used to define the wavelength dependence. In the SDA-FMC approach, the observed spectral derivative \(\alpha\) is combined with the spectral derivative of the fine modes \(\alpha_f\) to produce the fine

\(^1\) We will stop inserting “(scattering)” at this point although all references below should be understood to apply to both scattering and extinction.
mode fraction of extinction while the fine mode slope and curvature are both used in determining the
fine mode effective radius.

One reason for choosing a mid-visible reference wavelength of 500 nm for assessing curvature and
slopes is that the variation of the extinction for coarse mode aerosols is minimal in this spectral region
(O’Neill et al., 2001). The algorithm assumes constant values of the spectral slope and curvature for all
coarse mode aerosols at this wavelength (500 nm), specifically $\alpha_c = -0.15$ and $\alpha'_c = 0.0$ (with an assumed
uncertainty, as per O’Neill et al. (2003), of ±0.15 and ±0.15 respectively). An assumption of aerosol
bimodality (at least as far as measurements in the visible and near-IR are concerned) yields a series of
three succinct equations if the approximation level relative to a theoretical Mie representation (O’Neill
et al., 2001) is limited to second order in $\ln \lambda$ space. These three equations express the relationships
between the observed parameters (AOD or extinction coefficient, $\alpha$, $\alpha'$) and their fine and coarse mode
analogues (O’Neill et al., 2001). A set of three equations can be then inverted to yield the fine mode
spectral derivative, the fine mode curvature ($\omega'$) and the fine and coarse mode AOD or $b_{ext}$ values. The
observationally determined total and fine mode spectral derivative and prescribed coarse mode spectral
derivative are then used to calculate the fine mode fraction of extinction at the reference wavelength
as:

$$FMF = \frac{\alpha - \alpha_c}{\alpha_f + \alpha_c}$$  \hspace{1cm} (2)

Estimation of the Fine Mode Effective Radius – the Fine Mode Curvature (FMC) approach

Using the spectral derivatives for the fine mode obtained from the SDA portion of the approach, an
estimate of the fine mode effective radius is obtained. The basis for this approach is a parameterization
of a strong relationship between the effective van de Hulst phase shift parameter for fine mode
aerosols and a polar angle representation of $\alpha'$ vs. $\alpha$ (O’Neill et al., 2005). The fundamental van de Hulst
parameter for the fine mode, $\rho_{eff,f}$, is given by:

$$\rho_{eff,f} = 2 \times \frac{2\pi R_{eff,f}}{\lambda} |m - 1|$$  \hspace{1cm} (3)

\hspace{1cm} 7
where $\lambda$ is the reference wavelength and $m$ is the complex refractive index at that wavelength (ibid.) An estimate of this purely optical parameter derived from the $\alpha'$ vs. $\alpha$ polar relationship allows extraction of an effective radius for the fine mode from the SDA-obtained slope and curvature, if the refractive index of the particles is known. Since the refractive index is generally unknown for the situations we consider here, the information provided by this approach is actually a combination of size and composition. In many cases, an average, constant value for the real portion of the refractive index can be assumed and the imaginary part neglected to provide an estimate of the effective radius; this is, in part, because the imaginary component is typically much smaller than the real component of the refractive index, and thus the $D_{\text{eff},f}$ value is relatively insensitive to variations in the imaginary component. This treatment is questionable if strong changes in the average composition that lead to changes in $m$ are suspected, for example if the composition shifted from pure sulfate aerosol ($m = 1.53 + 0i$) to a brown carbon organic ($m = 1.4 + 0.03i$) this would introduce a 33% shift in the derived radius with no change in actual size; the majority of this shift in the derived radius would result from the change in the real component of the refractive index.

The FMC method has been less rigorously validated than the SDA portion and is expected to be more susceptible to problems related to measurement errors and a decreasing sensitivity with decreasing fine mode fraction of extinction. The polar-coordinate system relationship is a strong, near monotonic fit based on Mie simulations over a variety of aerosol types and sizes (O’Neill et al., 2005; O’Neill et al., 2008a); its validation is largely confined to comparisons with the more comprehensive AERONET inversions of (Dubovik and King, 2000). These inversions, which require the combination of AOD and sky radiance data, are of a significantly lower frequency than simple AOD measurements (nominally once per hour versus once every 3 minutes respectively). The comparisons (for the limited data set of O’Neill et al. (2005) and confirmed by more recently unpublished AERONET-wide comparisons) show averaged AERONET‐SDA differences of $10\% \pm 30\%$ for large FMF values $> 0.5$.

Application of the SDA-FMC method to in situ extinction measurements

This paper seeks to address the following two key questions pertaining to the use of the SDA-FMC algorithm with extinction measurements, especially those produced by the cavity ring-down instruments, to extract information about aerosol size, both the partitioning of the extinction between the fine and coarse modes and the extraction of a single parameter size characterization of the fine mode.
1.) Can the approach be used reliably to extract the fine and coarse mode fractions of the extinction in situations where only a single optical instrument is used?

and,

2.) In situations where complementary measurements (mobility-based sizers, parallel or switching nephelometers, etc.) are available, what information can be determined from the comparison of the products of the SDA-FMC approach to comparable information obtained in other ways?

It has been suggested that a single multi-wavelength optical measurement of the fine mode fraction could be less expensive than derivation of the sub-micron fraction of scattering using parallel nephelometers (Kaku et al., 2014). The use of two size-selected inlets (e.g., 1 and 10 μm cyclones) and parallel nephelometers is not prohibitively expensive, but the typical concerns regarding calibration maintenance and careful and consistent application of correction factors for truncation angle and non-Lambertian illumination can be magnified when measurements are combined (either as differences or ratios) since systematic errors may not undergo partial cancellation like random errors.

In principle, the use of two parallel CRD extinction measurements could mitigate some of the possible errors with parallel nephelometers. Cavity ring-down measurements do not (in principle) need to be calibrated and have very small truncation errors (Smith and Atkinson, 2001; Brown, 2003). In practice, two types of “calibrations” are applied to CRD measurements: a zeroing procedure that is usually a measurement of filtered air for aerosol measurements and a cavity path length correction because the complete mirror-to-mirror distance of the optical cavity is typically not filled with aerosols (to keep the mirrors clean) (Langridge et al., 2011). The former (zeroing) limits instrument precision and sometimes accuracy while the latter (path length) limits instrument accuracy. In general these procedures are identical for the two parallel instruments and are very stable in time, so they would only be expected to produce a small and consistent bias. To our knowledge, currently no single-package, multi-wavelength direct extinction (cavity-enhanced) instruments are commercially available. Multiple single-wavelength instruments operating at different wavelengths could be deployed, but might be prohibitively expensive.

For detailed knowledge of the fine mode size distribution, the use of scanning mobility analyzer-based sizing instruments is preferable since the full mobility size distribution is obtained, as opposed to only the effective radius provided by the FMC procedure. However, scanning mobility sizer instruments typically have maximum diameters of only 700 to 800 nm, and both scanning and multi-channel variants are of comparable expense and complexity as CRD instruments. In order to obtain additional
information about the coarse mode size distribution and contribution to the optical effects, an aerosol particle spectrometer (APS) is generally added to the measurement suite.

The purely spectrally-based mode separation inherent in the SDA obviates the need for a physical cut point selection, such as that required to measure the PM$_2.5$ scattering product used in this work. This can be advantageous, since selection and maintenance of a size cut-point is a possible source of differences between some measurements (and variability of all measurements using physical separation) of the sub-micron fraction (SMF) of scattering, absorption or extinction. The SMF is fundamentally different from the FMF, although both provide an indication of the fractional optical contribution of smaller particles. In fact, there are fundamental differences between many of the SMF or FMF data products that are currently available. For example, the Dubovik and King (2000) SMF data product tries to locate the separation radius (called the inflection point) at a minimum of the particle size distribution obtained from the inversion procedure. This results in a variable cut point that can be interpreted as assigning a portion of the coarse mode to the fine mode (O’Neill et al., 2003). The aerodynamic diameter selected for the physical separation used in the SMF presented in this work might result in some mis-assignment of fine mode extinction to the coarse mode, since (i) the aerodynamic separation results in a cut point that is less than 1 μm geometric diameter and (ii) the cut point might not correspond to a local minimum of the size distribution. These definitional differences should be kept in mind when comparing fine mode apportionments (SMF or FMF) from different measurements/data treatments. And all of these data products will usually differ significantly from the optical properties of the PM$_{2.5}$ fraction used to define the fine mode for air quality regulations and to exclude larger particles in the CRD instruments at T0. The latter allowed a significant fraction, but not all of the optically coarse particles into the instruments, as shown in the Results section. For the comparisons presented in this work, in cases where there is significant penetration of one of the modes into the size regime defined by the physical cut-point as the other mode (or significant overlap of two or more size modes) there are noticeable differences between the physically-defined SMF and the FMF produced by the SDA.

**Experimental**

The instrument suites used, sampling conditions and methodology and goals of the CARES study have been summarized by (Zaveri et al., 2012). A summary of the instrumentation used to make the light extinction, scattering and absorption measurements is provided in Table 1. Extinction was measured either directly (using cavity ringdown spectroscopy) or as the sum of scattering and absorption. A brief description of the key instruments used in the current analyses is given below.
Table 1: Summary of optical instruments used at the T0 and T1 sites

<table>
<thead>
<tr>
<th>Property</th>
<th>Instrument</th>
<th>Wavelength</th>
<th>Size Cut*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extinction</td>
<td>UCD CRD</td>
<td>405, 532 nm</td>
<td>2.5 µm</td>
</tr>
<tr>
<td></td>
<td>PSU CRD</td>
<td>1064 nm</td>
<td>2.5 µm</td>
</tr>
<tr>
<td>Scattering</td>
<td>PNNL Nephelometer</td>
<td>450, 550, 700 nm</td>
<td>1 µm, 10 µm</td>
</tr>
<tr>
<td>Absorption</td>
<td>PNNL PSAP</td>
<td>470, 522, 660 nm</td>
<td>1 µm, 10 µm</td>
</tr>
<tr>
<td>Extinction</td>
<td>PSU CRD</td>
<td>355, 532, 1064 nm</td>
<td>None applied</td>
</tr>
<tr>
<td>Scattering</td>
<td>PNNL Nephelometer</td>
<td>450, 550, 700 nm</td>
<td>1 µm, 10 µm</td>
</tr>
<tr>
<td>Absorption</td>
<td>PNNL PSAP</td>
<td>470, 522, 660 nm</td>
<td>1 µm, 10 µm</td>
</tr>
</tbody>
</table>

*For the entries with two size cuts listed, the sampling system switched between the two on a 6 minute cycle.

Instruments used at the T0 site (American River College, Granite Bay, CA USA)

Cavity Ring-down Extinction: The $b_{ext}$ measurements at 405 nm and 532 nm were made using the UCD Davis two-wavelength Cavity Ring Down-Photoacoustic Spectrometer (CRD-PAS) instrument (Langridge et al., 2011; Lack et al., 2012). Full details of these measurements are available in Cappa et al. (2016) and Atkinson et al. (2015). These measurements were only made for a subset of the CARES campaign, from 20:00 PDT on 16 June through 09:00 PDT on 29 June. At 532 nm, $b_{ext}$ was measured at low (~25%), mid (~75%) and high (~85%) relatively humidity. At 405 nm only low RH measurements were made, and so only the low RH 532 nm measurements are used in this study. The CRD-PAS sampled behind a PM$_{2.5}$ (aerodynamic diameter <2.5 µm) URG Teflon-coated aluminum cyclone. A separate CRD instrument deployed by the PSU group at T0 used a single optical cavity to measure the sub-2.5 µm (sampled through a similar URG cyclone) aerosol extinction coefficient at 532 and 1064 nm simultaneously (Radney et al., 2009). This instrument did not incorporate intentional RH control, but efforts were made to maintain nearly ambient conditions, resulting in low RH (25 - 40 %) throughout most of the campaign, as measured by an integrated RH/T sensor (Vaisala HMP70). Daytime ambient RH was similar to the low RH value during the CARES campaign (Fast et al., 2012).

To obtain three-wavelength $b_{ext}$ measurements for use in the SDA-FMC analysis, we combined the measurements from the two CRD instruments. First, the 532 nm time series data were overlaid and examined for differences: temporal correspondence between the data was demonstrated (except for a clear difference in precision, the UCD CRD having approximately 3 times better precision than the PSU...
instrument at comparable integration times). A scatterplot (Figure S1) between the two data sets showed good correlation with a best fit line having a slope = 0.87 and an intercept that was statistically indistinguishable from zero. With this assurance that the two instruments were measuring the same aerosol with comparable measurement quality, the PSU 1064 nm data are used with the UCD 532 nm and 405 nm low RH data for the SDA-FMC analysis.

Size-selected absorption and scattering (Nephelometer and PSAP): The low RH scattering and absorption coefficients were alternatively measured for PM10 and PM1 aerodynamic size selected aerosol using the PNNL Aerosol Monitoring System, a clone of NOAA/CMDL’s Aerosol Monitoring System (detailed description at http://www.esrl.noaa.gov/gmd/aero/instrumentation/instrum.html and in Zaveri et al. (2012)). The relevant wavelengths are: light absorption coefficients at three-wavelengths (Radiance Research Particle Soot Absorption Photometer [PSAP]) and total scattering coefficients (three-wavelength nephelometer, TSI 3563). The absorption coefficients were adjusted to the nephelometer wavelengths using an inverse wavelength dependence. The absorption and scattering coefficients for PM1 or PM10 are then summed after averaging to one-hour intervals and using the mean of the 450 and 550 nm values to obtain $b_{\text{ext}}(500 \text{ nm})$. The extinction fraction of the PM1 (herein, the SMF) at the visible wavelength (500 nm) is then calculated from their ratio

$$SMF_{\text{ext}} = \frac{b_{\text{ext,PM1}}}{b_{\text{ext,PM10}}} \quad (4)$$

Particle size control was effected by 2 impactors (1 μm and 10 μm) upstream of the PSAP and Nephelometer. The 10-μm impactor was always present in the sampling line, and the flow was switched to run through the 1-μm impactor on 6-min intervals, yielding alternating 6-min measurements of submicron and coarse (<10 μm) particle modes.

Fine particle size distribution: The submicron dry particle mobility diameter ($d_{\text{p,m}}$) size distribution (12 nm to 737 nm) was measured using a scanning mobility particle sizer (SMPS) comprised of a charge neutralizer, differential mobility analyzer and condensation particle counter (TSI 3081 DMA column and model 3775 CPC). The SMPS data were corrected for multiply-charged particles and diffusional losses. These size distribution measurements are used to calculate $R_{\text{eff,}}$ values from Eqn. 1, which will be referred to as $R_{\text{eff, size}}$. It should be noted that a mobility diameter of 737 nm corresponds to an aerodynamic diameter of 919 nm (assuming a density of 1.5 g cm$^{-3}$, a reasonable value for the campaign based on the observed particle composition (Atkinson et al., 2015)).
Instruments used at the T1 site (Evergreen School, Cool, CA USA)

Cavity Ring-down Extinction: The PSU group deployed a custom CRD instrument that used separate optical cavities to measure $b_{\text{ext}}$ at 355 nm, 532 nm, and 1064 nm simultaneously in each of four separate flow systems that were intended to measure total and submicron aerosol and submicron aerosol that had been conditioned to have elevated and suppressed RH. Only the total aerosol flow results are used here as this prototype system suffered from signal to noise problems and RH/temperature control issues. As with the T0 PSU instrument, the total aerosol system attempts to measure particle extinction at nearly ambient conditions, resulting in low RH (25 – 40 %) throughout most of the campaign, as measured by an integrated RH/T sensor (Vaisala HMP70). No intentional size cut was applied to these measurements, although the system was not optimized for transmission of coarse mode particles.

Size-selected absorption and scattering (Nephelometer and PSAP): An identical instrument suite to that used at T0 was deployed and the same data analysis was conducted.

Fine particle size distribution: The SMPS used at T1 is a similar design described in (Setyan et al., 2012) and it measured low RH particle sizes from 10 nm to 858 nm. The SMPS data were corrected to take into account the DMA transfer function, the bipolar charge distribution, the CPC efficiency and the internal diffusion losses. (Setyan et al., 2014)

Results and Discussion

Fine mode fraction of extinction

The CRD-based extinction measurements were used to derive the FMF$_{\text{ext}}$ using the SDA. This will be referred to as the FMF$_{\text{ext,CRD}}$. For the T0 site, the FMF$_{\text{ext,CRD}}$ is for PM$_{2.5}$ while at T1 no physical cut point was introduced, so PM$_{10}$ is a reasonable expectation. The time series of the CRD-based $b_{\text{ext}}$ values and of the derived FMF$_{\text{ext,CRD}}$ at the T0 site are shown in Figure 1 (all times in PDT – local time during the study). The FMF$_{\text{ext,CRD}}$ varies from 0.55 to 1, with a mean of 0.78 ± 0.1 (1 σ).
Figure 1 – Time series of CRD extinction coefficient observations (left axis) and the derived FMF\textsubscript{ext,CRD} (right axis) at T0 during the time period analyzed in this work. The blue, green and red traces are the 405 nm, 532 nm and 1064 nm $b_{\text{ext}}$ (respectively) and the black points show the 1 h average FMF\textsubscript{ext,CRD} from the SDA analysis. A PM$_{2.5}$ size cut was applied during the sampling. The fine mode fraction of extinction at T0 was alternatively determined from the PM$_{10}$ $b_{\text{ext}}$ measurements from the nephelometer and PSAP, referred to as FMF\textsubscript{ext,sum}. The SDA-derived FMF\textsubscript{ext,CRD} and FMF\textsubscript{ext,sum} values are compared with the sub-micron fraction of extinction determined from the combined PM$_{1}$ and PM$_{10}$ nephelometer and PSAP measurements (from the latter part of the campaign) at T0 (Fig. 2). The FMF\textsubscript{ext,CRD}, FMF\textsubscript{ext,sum} and SMF\textsubscript{ext,sum} all exhibit the same general temporal dependence. In general, the FMF\textsubscript{ext,CRD} $>$ FMF\textsubscript{ext,sum} $\sim$ SMF\textsubscript{ext,sum} although the specific relationships vary with time. For example, there are periods when the FMF\textsubscript{ext,sum} and SMF\textsubscript{ext,sum} are nearly identical (e.g. 20 June – 23 June) and periods when the SMF\textsubscript{ext,sum} is somewhat lower than the FMF\textsubscript{ext,sum} (e.g. 24 June – 25 June).

Figure 2 – Time series of the fine mode fractions and sub-micron fraction of extinction at T0. The red trace is the SMF\textsubscript{ext,sum} determined from the $b_{\text{ext}}$(PM$_{1}$) / $b_{\text{ext}}$(PM$_{10}$) ratio. The black and blue traces are the FMF\textsubscript{ext} from the SDA analysis of the CRD extinction (black) and nephelometer + PSAP extinction (blue). The FMF\textsubscript{ext,CRD} values are the same as those of Fig. 1 for the latter half of the campaign.

The FMF\textsubscript{ext,CRD} was determined for PM$_{2.5}$ while the FMF\textsubscript{ext,sum} was determined for PM$_{10}$. If a substantial fraction of the scattering was contributed by particles with diameters $>$ 2.5 μm, then the FMF\textsubscript{ext,CRD}...
should be larger than the $\text{FMF}_{\text{ext,sum}}$, as was observed. Kassianov et al. (2012) used measured particle size distributions from CARES to show that supermicron particles contributed significantly to the total scattering, consistent with the observation that $\text{FMF}_{\text{ext,CRD}} > \text{FMF}_{\text{ext,sum}}$. Variability in the difference between the $\text{FMF}_{\text{ext,CRD}}$ and $\text{FMF}_{\text{ext,sum}}$ likely reflects variability in the contribution of these larger particles to the total scattering.

The $\text{FMF}_{\text{ext,CRD}}$, $\text{FMF}_{\text{ext,sum}}$ and $\text{SMF}_{\text{ext,sum}}$ were similarly determined from the measurements at the T1 site (Figure 3). For T1, the CRD measurements were made for particles without any intentional size cut applied, as opposed to the PM$_{2.5}$ size cut used for the T0 measurements. At this downwind site the $\text{SMF}_{\text{ext,sum}}$, $\text{FMF}_{\text{ext,CRD}}$ and $\text{FMF}_{\text{ext,sum}}$ were all very similar, both in terms of the absolute magnitude and the temporal variability. The $\text{FMF}_{\text{ext,CRD}}$ ranged from 0.3 to 0.85, with a mean of 0.66 ± 0.19. That the $\text{FMF}_{\text{ext,CRD}}$ and $\text{FMF}_{\text{ext,sum}}$ are very similar in absolute magnitude for T1 but differ at T0 (while still exhibiting similar temporal variability) is likely related to the application of an intentional size cut for the CRD measurements at T0 but not at T1. The observations suggest that the T1 CRD without the size cut samples coarse-mode particles with a similar efficiency as the nephelometer and PSAP having the PM$_{10}$ size cut.

Overall, these results indicate that the use of the spectral deconvolution algorithm on optical data can robustly provide information on the fine mode fraction of extinction. Moreover, since the $\text{FMF}_{\text{ext}}$ results at T1 are similar for the two types of extinction measurements, it seems that the narrower wavelength range of the nephelometer (450, 550, 700 nm) and PSAP (470, 522, 660 nm) compared to the CRD instruments used here is still adequate to define the spectral dependence of extinction for extraction of the slope and curvature parameters. However, the differences observed at both sites highlight the fact that there is not a precise definition of “fine” and “coarse” in terms of a specific size cut in the optical method. The effective size cut is dependent on the shapes of the size distributions in the “fine” and “coarse” size regimes and the extent of overlap between them, which is dependent on the size range of particles sampled (e.g. PM$_{2.5}$ versus PM$_{10}$). Nonetheless, since the major sources of fine and coarse mode particles are likely to be reasonably distinct in many environments, the $\text{FMF}_{\text{ext,CRD}}$ can provide a characterization of the variability in the contributions of such sources to the total scattering.
Figure 3 – the fine mode fraction of extinction (SMF and \( \text{FMF}_{\text{ext}} \)) for the latter half of the campaign at T1. Here, the \( \text{FMF}_{\text{ext,CRD}} \) is determined for particles sampled without a size cut applied.

Effective fine mode radius product of SDA-FMC

The SDA-FMC analysis also allows for derivation of the fine mode effective radius, \( R_{\text{eff,f}} \), via Eq. 3. Determination of \( R_{\text{eff,f}} \) requires knowledge of the real and imaginary parts of the refractive index. Here, an average value of \( m_r = 1.55 \) is used, based on Atkinson et al. (2015), and absorption is assumed to be negligible. The latter is a reasonable assumption given the relatively high single scatter albedo values at the two sites (Cappa et al., 2016), and because assuming the particles to be slightly absorbing has minimal influence on the results. Values of \( R_{\text{eff,f}} \) are determined using both the CRD-measured \( b_{\text{ext}} \) and the PM10 \( b_{\text{ext}} \) from the nephelometer + PSAP measurements for both T0 and T1 (Figure 4). \( R_{\text{eff,f}} \) values are also determined from the PM1 nephelometer + PSAP measurements at both sites. Comparison of the \( R_{\text{eff,f}} \) values between the PM10 and PM1 measurements provides a test of the robustness of the overall retrieval method. The \( R_{\text{eff,f}} \) from the CRD measurements will be referred to as \( R_{\text{eff,f,CRD}} \) and from the nephelometer + PSAP as \( R_{\text{eff,f,sum}} \). Comparator values of \( R_{\text{eff,f}} \) were also calculated from the observed mobility size distributions using Eqn. 1, and are referred to as \( R_{\text{eff,f,size}} \).
Figure 4 – Time series of the effective fine mode radii, $R_{\text{eff}, f}$, produced by the SDA-FMC analysis of the CRD data (black) and the nephelometer + PSAP data (blue) from T0 (top) and T1 (bottom). For the nephelometer + PSAP observations, separate results are shown using either the PM$_{10}$ (dark blue) or PM$_{1}$ (light blue) observations. The $R_{\text{eff}, f}$ values determined from the size distribution measurements (i.e. from Eqn. 1) are shown in red.

The SDA-FMC-derived $R_{\text{eff}, f}$ values from the CRD and from the nephelometer + PSAP exhibit reasonably good agreement in terms of the absolute values and the temporal variability at both the T0 and T1 sites (Table 2, Fig. 4). Notably, there is good agreement between the $R_{\text{eff}, f, \text{sum}}$ values obtained from the PM$_{10}$ and PM$_{1}$ measurements. This provides an important validation of the SDA-FMC procedure, since the coarse mode contribution to the PM$_{10}$ extinction is substantial and highly variable (Figure 2 and Figure 3).

At T0, the derived $R_{\text{eff}, f}$ values range from approximately 70 nm to 140 nm (Table 2), with a few short-duration periods when $R_{\text{eff}, f}$ is outside this range, reflecting short-duration variability in the particle sources. At T1 the derived $R_{\text{eff}, f}$ are generally less variable, ranging from approximately 65 nm to 110 nm, with fewer particularly low or high periods. The mean $R_{\text{eff}, f}$ values between the two sites are similar (Table 2). At T0, there is a fair degree of temporal coherence of the SDA-FMC results and those obtained
from integration of the size distributions. The generally good temporal agreement between the optically- and size-derived $R_{\text{eff}, \text{size}}$ values are even observed during periods where the changes in radius happened rapidly, for example near midnight between June 21-22. On that night there is some evidence that paving operations near the T0 site produced a strong local source of asphalt particles in the coarse mode with a long tail into the sub-micron regime (Zaveri et al., 2012; Cappa et al., 2016). This short-duration source of large particles pushed the $R_{\text{eff}, \text{f}}$ temporarily towards larger values. (The $R_{\text{eff}, \text{f}}$ changes from the nephelometer + PSAP at this time were smaller than from the CRD or size distribution observations. Most likely this reflects the alternating 6-min sampling of the nephelometer and the very short duration of the event leading to discrepancies in the 1 h average.)

Despite the generally good correspondence between $R_{\text{eff}, \text{f}, \text{size}}$ and the optically derived values, the $R_{\text{eff}, \text{f}, \text{size}}$ values were often (but not always) smaller (Table 2). This is most clearly seen when comparing the average diurnal profiles of the $R_{\text{eff}, \text{f}}$ values from the different methods, as shown in Figure 5. All three $R_{\text{eff}, \text{f}}$ estimates exhibit similar diurnal behavior at T0, even though the $R_{\text{eff}, \text{f}}$ from the SDA-FMC method are larger than $R_{\text{eff}, \text{f}, \text{size}}$. The diurnal variability in the $R_{\text{eff}, \text{f}}$ is more pronounced at T0 than at T1. The diurnal trend in the effective radius of the fine mode at T0 from all methods exhibits a minimum at around mid-day and then an increase to a maximum right near daybreak. Particle number and sizes at both sites were influenced by frequent regional new particle formation and growth events during CARES (see Figure S2). The events tended to start in the morning with a sharp increase of 10 - 20 nm particles followed by growth of these particles to 50 – 100 nm in the afternoon as discussed in Setyan et al. (2014). The next day the cycle repeats (on average) with the introduction of the new small particles which has the effect of decreasing the average particle radius (Setyan et al., 2014). Although observed at both sites, the new particle formation events had a greater impact on the size distributions at T0, especially in terms of surface area-weighted size distributions that determine $R_{\text{eff}, \text{f}}$. In part, this is likely because of continued growth of the new particle mode as it transits from T0 to T1.
Figure 5 – The diurnal dependence of $R_{\text{eff},f}$ for the period shown in Fig. 4 for the (a) T0 and (b) T1 sites. The box and whisker plot (bottom and top of box are 5% and 95% of data range, bar is mean, and whiskers extend to full range) shows the results from the direct size distribution measurement ($R_{\text{eff},f,\text{size}}$), while the lines show the mean diurnal dependence of the optically derived $R_{\text{eff},f}$ using the CRD (black) and nephelometer + PSAP (blue) measurements.

One possible explanation for the differences between the optically and size-derived $R_{\text{eff},f}$ in particular at T0, may be inaccurate specification of the refractive index. Temporal variations in or an overall offset of the real refractive index used here from the true value would lead to errors in the optically derived $R_{\text{eff},f}$. The refractive index is used to convert the derived van de Hulst parameter to $R_{\text{eff},f}$ (Eqn. 3). Given the form of the relationship, an absolute error in the real RI of 0.1—likely an upper limit—corresponds to an error in the derived $R_{\text{eff},f}$ of 20%, with larger values of the real RI leading to smaller derived $R_{\text{eff},f}$. The imaginary component was assumed zero. The effective imaginary RI is likely ≤ 0.01, given the range of
single scatter albedo values observed (Cappa et al., 2016). Thus, the assumption of zero for the
imaginary RI introduces negligible error. The actual real RI depends on the particle composition since
different chemical components (e.g. sulfate, organics, dust) have different RI values. Here, the RI values
used were determined based only on measurements of the non-refractory PM composition and only an
average value was used (Atkinson et al., 2015). To the extent that refractory components, in particular
dust or sea salt, contributed to the fine mode scattering, their influence on the real RI would not be
accounted for. However, dust and sea salt contributions are most likely confined primarily to the coarse
mode. Thus, the fine mode real refractive index is unlikely to be strongly affected by their presence and
the real RI can probably be constrained to a fairly narrow range around 1.5. The relative uncertainty of
the $R_{eff,f}$ derived from the SDA-FMC method has been estimated as ranging from 40% to 70%. This range
of values was computed from a quadrature combination of the estimated errors (20-50%) in the SDA-
FMC retrieval (O'Neill et al., 2003), the CRD measurements (< 5% for the UCD and T0 PSU instrument
and 20% for the T1 PSU instrument) and the refractive index term above (estimated maximum of 20%).
In this context, the agreement shown in Fig. 4 is acceptable and may suggest that the above error
estimates are overly conservative.

**Table 2**: Summary statistics for $R_{eff,f}$ values (nm)

<table>
<thead>
<tr>
<th>Site</th>
<th>Method</th>
<th>Maximum (nm)</th>
<th>Minimum (nm)</th>
<th>Mean (nm)</th>
<th>Standard Deviation (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>T0</td>
<td>SDA-FMC + CRD (PM$_{2.5}$)</td>
<td>189</td>
<td>35</td>
<td>100</td>
<td>21</td>
</tr>
<tr>
<td>T0</td>
<td>SDA-FMC + Neph. &amp; PSAP (PM$_{10}$)</td>
<td>139</td>
<td>62</td>
<td>97</td>
<td>14</td>
</tr>
<tr>
<td>T0</td>
<td>Size Distribution Integration</td>
<td>133</td>
<td>54</td>
<td>85</td>
<td>14</td>
</tr>
<tr>
<td>T1</td>
<td>SDA-FMC + CRD (no size cut)</td>
<td>160</td>
<td>42</td>
<td>93</td>
<td>18</td>
</tr>
<tr>
<td>T1</td>
<td>SDA-FMC + Neph. &amp; PSAP (PM$_{10}$)</td>
<td>101</td>
<td>69</td>
<td>83</td>
<td>6</td>
</tr>
<tr>
<td>T1</td>
<td>Size Distribution Integration</td>
<td>118</td>
<td>52</td>
<td>88</td>
<td>11</td>
</tr>
</tbody>
</table>

**Conclusions**

This work demonstrates that the use of a non-size-selected, three wavelength CRD measurement in
continuous field monitoring, coupled with the SDA-FMC analysis, can provide information about the
relative contribution of the fine mode to the observed total particle extinction. The retrieved value of
the fine mode fraction of extinction is dependent upon the size range of particles sampled and the
overall nature of the particle size distribution. The relationship between the $F_{ext}$ and the $F_{ext}$,
determined from near-coincident measurement of extinction by PM$_1$ and PM$_{10}$, provides insights into
the effective $F_{ext}$ split size. For one of the sites considered here the split point size is around 1 μm
while for the other it is somewhat larger than 1 μm and perhaps more variable. In many environments,
variability in aerosol properties on short (<10 min) timescales is relatively minimal. In such cases, a single instrument can be used to sequentially sample PM$_1$ and PM$_{10}$, allowing for in situ measurement of both the FMF$_{ext}$ and SMF$_{ext}$. However, remote sensing measurements characterize only the FMF$_{ext}$, (or at best, an optically influenced size cut as is done in the AERONET retrievals of Dubovik & King, 2000). Thus, further consideration of in situ measurement results, such as those investigated in this study, can provide insights into the interpretation of the FMF$_{ext}$ determined from remote sensing in different environments.

The SDA-FMC approach also allows for determination of the effective fine mode radius. The $R_{eff,f}$ characterizes the surface-area weighted size of the particles within the fine mode distribution. The similarity of the results in Figure 4 for application of the SDA-FMC to both size-selected and non-size-selected aerosol as well as the comparison with results derived from the PSD measurements verify that “whole air” measurements (i.e., no imposed size-selection) can provide reliable fine mode radii at least for large FMF values.

Acknowledgements

This work was supported by the Atmospheric System Research (ASR) program sponsored by the US Department of Energy (DOE), Office of Biological and Environmental Research (OBER), including Grant No. DE-SC0008937. Funding for data collection was provided by the US DOE’s Atmospheric Radiation Measurement (ARM) Program. All data used in this study are available from the ARM data archive at: http://www.arm.gov/campaigns/aaf2009carbonaerosol. The views expressed in this document are solely those of the authors and the funding agencies do not endorse any products or commercial services mentioned in this publication.
**Appendix A – Glossary of Symbols and Acronyms used**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>å</td>
<td>Ångström exponent (from wavelength pair)</td>
</tr>
<tr>
<td>α</td>
<td>Spectral derivative of optical property</td>
</tr>
<tr>
<td>α'</td>
<td>Curvature (second derivative of optical property in log-log space)</td>
</tr>
<tr>
<td>αᵢ or αᵢ'</td>
<td>Fine mode version of properties (also coarse mode properties αᵢ)</td>
</tr>
<tr>
<td>AOD</td>
<td>Aerosol optical depth</td>
</tr>
<tr>
<td>bₑₓᵗ, bₛᶜᵃᵗ, bₐₜᵇ</td>
<td>Optical coefficient for extinction, scattering, absorption (inverse length units)</td>
</tr>
<tr>
<td>CRD</td>
<td>Cavity ring down</td>
</tr>
<tr>
<td>Rₑᶠₛ</td>
<td>Effective radius for fine mode</td>
</tr>
<tr>
<td>FMF (aka η)</td>
<td>Fine mode fraction of an optical property, usually extinction</td>
</tr>
<tr>
<td>SMF</td>
<td>Sub-micron fraction (particle mode with radius or diameter smaller than 1 μm)</td>
</tr>
<tr>
<td>ρᵢ</td>
<td>Fine mode van de Hulst parameter (product of refractive index and effective radius)</td>
</tr>
<tr>
<td>SDA</td>
<td>Spectral Deconvolution Algorithm</td>
</tr>
<tr>
<td>FMC</td>
<td>Fine Mode Curvature approach</td>
</tr>
<tr>
<td>PM₁</td>
<td>Particulate matter with diameter (or radius) smaller than 1 μm (also PM₂·₅, PM₁₀)</td>
</tr>
<tr>
<td>PSAP</td>
<td>Particle soot absorption photometer instrument</td>
</tr>
</tbody>
</table>
References:

Anderson, T. L., Charlson, R. J., Bellouin, N., Boucher, O., Chin, M., Christopher, S. A., Haywood, J.,
Plains cloud and radiation test bed site: 1. Aerosol optical properties, J. Geophys. Res.-Atmos., 109,
Ángström, A.: On the atmospheric transmission of sun radiation and on dust in the air, Geografika Ann.,
and columnar aerosol spectral measurements during TexAQS-GoMACCS 2006: testing parameterizations
Atkinson, D. B., Radney, J. G., Lum, J., Kolesar, K. R., Cziczo, D. J., Pekour, M. S., Zhang, Q., Setyan, A.,
Zelenyuk, A., and Cappa, C. D.: Aerosol optical hygroscopicity measurements during the 2010 CARES
Synchronous polar winter starphotometry and lidar measurements at a High Arctic station, Atmos.
Bokoye, A. I., Royer, A., O'Neill, N. T., Cliche, P., Fedosejevs, G., Teillet, P. M., and MacArthur, L. J. B.:
Characterization of atmospheric aerosols across Canada from a ground-based sunphotometer network:
black carbon and organic matter with the Specific Forcing Pulse, Atmos. Chem. Phys., 11, 1505-1525,
Brown, S. S.: Absorption Spectroscopy in High-Finesse Cavities for Atmospheric Studies, Chemical
Zhang, Q.: Understanding the optical properties of ambient sub- and supermicron particulate matter:
results from the CARES 2010 field study in northern California, Atmos. Chem. Phys., 16, 6511-6535,
doi:10.5194/acp-16-6511-2016, 2016.
Charlson, R. J., Valero, F. P. J., and Seinfeld, J. H.: In search of balance, Science, 308, 806-807,

Flentje, H., Hyvarinen, A., Jefferson, A., Jennings, S. G., Kouvarakis, G., Lihavainen, H., Myhre, C. L.,

Malm, W. C., Mihapopoulos, N., Molenar, J. V., O'Dowd, C., Ogren, J. A., Schichtel, B. A., Sheridan, P.,


Dubovik, O. and King, M. D.: A flexible inversion algorithm for retrieval of aerosol optical properties from

Sun and sky radiance measurements, J. Geophys. Res.-Atmos., 105, 20673-20696, 


Eck, T. F., Holben, B. N., Reid, J. S., Sinyuk, A., Dubovik, O., Smirnov, A., Giles, D., O’Neill, N. T., Tsay, S. C.,


Gulf and United Arab Emirates in summer, J. Geophys. Res.-Atmos., 113, D01204, 


George, I. J. and Abbatt, J. P. D.: Heterogeneous oxidation of atmospheric aerosol particles by gas-phase


using the Mahalanobis distance, Atmos. Environ., 140, 213-233, doi:10.1016/j.atmosenv.2016.06.002, 

2016.

Hansen, J. E. and Travis, L. D.: Light-Scattering in Planetary Atmospheres, Space Sci. Rev., 16, 527-610, 


Y. J., Nakajima, T., Lavenu, F., Jankowiak, I., and Smirnov, A.: AERONET - A federated instrument network


IPCC: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth

Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press,

Cambridge, United Kingdom and New York, NY, USA, 2013.

Kaku, K. C., Reid, J. S., O’Neill, N. T., Quinn, P. K., Coffman, D. J., and Eck, T. F.: Verification and

application of the extended spectral deconvolution algorithm (SDA plus ) methodology to estimate


