Review of 'Using Spectral Methods to Obtain Particle Size Information from Optical Data: Applications to Measurements from CARES 2010', by Atkinson *et al*., 2017.

Summary and General Comments

The work presented by Atkinson *et al*. applies a spectral deconvolution algorithm (SDA) and fine mode curvature (FMC) algorithm for retrieving fine mode fraction (FMF) and effective fine mode radius ($R_{eff,f}$), respectively, from *in situ* optical measurements on aerosol particles. Although these algorithms have been applied previously to remote sensing measurements, the work reported here represents the first application to *in situ* optical measurements, allowing an assessment of the accuracy of the retrievals of FMF and $R_{eff,f}$ through comparisons with other *in situ* measurements that measure FMF and $R_{eff,f}$ in a more direct manner. The *in situ* techniques for measuring aerosol optical properties include cavity ring-down spectroscopy (extinction coefficient), nephelometry (scattering coefficient) and particle soot absorption photometry (PSAP, absorption coefficient), with measurements made at a variety of wavelengths spanning the visible and near infrared, and for aerosol ensembles using a variety of impactor cut sizes (1 µm, 2.5 µm and 10 µm). Moreover, fine mode particle size distributions are measured directly using a scanning mobility particle sizer. The reported assessments of FMF and $R_{eff,f}$ retrieval accuracies are important to those in the remote sensing community and also those seeking to characterise aerosol size distribution properties from *in situ* optical measurements. To this end, the work represents a substantial contribution and is suitable for publication in Atmospheric Chemistry and Physics. I recommend publication after the following comments have been addressed.

Specific comments

Line 160: It would be good if the authors could be more specific as to how biomass burning confounds the expectation of an anthopogenic-associated fine mode and a coarse mode associated with natural emissions. In particular, the authors reference Hamill *et al*. 2016, but it would be useful for the authors to be more specific about what this study reported that is relevant to the current argument.

Line 204: Please could the authors explain what is meant by 'polar angle representation of α_f' vs α_f'. In particular, it would be useful if this representation could be plotted using some of the extinction data later reported for the reader to visualise. Moreover, the van de Hulst parameter and how it is calculated from optical data using the polar plots referred to should be explained more clearly to provide the reader with greater clarity and tools for understanding the results later in the text. In my view, this is one change that would greatly improve understanding readability and understanding, and simply referring the reader to O'Neil et al. 2005 to get all the necessary theoretical details is not helpful. Perhaps, if such a discussion is too long for the main text, a discussion on the polar representation and example plots could be provided in the supplementary information.

Lines 322 – 326: The best-fit slope is 0.87. I'm surprised that the agreement is not better and be closer to a 1:1 relationship. Is the high noise, associated with the poorer precision in the PSU measurements, responsible for this deviation? Please could the authors describe why the PSU CRDS is less precise and state is clear terms that the data from the PSU instrument is neglected in further analysis here because of this poorer precision. Also, for the aforementioned reasons (poor slope of

0.87 and poor precision in PSU 532-nm CRDS data), I do not agree with the phrase '…the two instruments were measuring the same aerosol with comparable measurement quality…'.

Line 334: What is the basis for an inverse wavelength dependence? A reference showing that inverse wavelength dependence is a reasonable approximation would also be useful here.

Lines 401 – 403: The authors discuss errors in $R_{eff,f}$ (later in the text) that arise in part from 5% errors in cavity ring-down extinction measurements. However, no consideration is given to the uncertainties that arise in FMF or $R_{eff,f}$ from errors in the summation (scattering + absorption) data. Given the very large uncertainties and biases that exist in filter-based measurements of absorption, such as from a PSAP, can the authors comment on the corresponding uncertainties in their FMF and $R_{eff,f}$ retrievals when using the summation method. Have the authors considered the influence of absorption correction schemes for filter-based absorption measurements?

Technical comments:

Line 19: To reinforce that the ground based measurements are *in situ* opposed to ground based remote sensing, it would be effect to use the phrase 'Multi-wavelength *in situ*…' in the opening sentence.

Line 24, '*Application to in situ measurements allows for comparison…*': This is a bit ambiguous. Please can the authors specify what is being applied to the *in situ* measurements (the SDA and FMC algorithms). Also, please specify the quantities being compared when stating '…*for comparison*...'.

Line 78: Brackets are not required. In any case, full stop should be after end bracket rather than before.

Line 79: There is some ambiguity here. Please specify what is meant by 'former' and 'latter'. In part, this ambiguity is magnified by the inclusion of the preceding statement concerning the list of symbols and acronyms.

Line 99: The phrase is brackets is unclear. What does the η symbol represent? It doesn't appear in the rest of the text. What is 'ibid'? Also, full stop after the end brackets rather than before.

Line 99 – 101: This sentence is confusing on first read as it suggests that the fine mode spectral derivatives can be used with equation 1 to calculate $R_{eff,f}$. In actual fact, the authors are saying that the spectral derivatives can be used to calculate $R_{eff,f}$ using a fine mode curvature algorithm, although a strict definition of $R_{eff,f}$ in terms of the number size distribution is provided by equation 1. Perhaps, a suitable rewording would be 'The fine mode spectral derivatives can then be used to obtain the effective radius for the fine mode through a fine mode curvature algorithm. Alternatively, the fine mode effective radius can be calculated from direct measurements of size distribution (e.g. from scanning mobility particle sizer) using equation 1 (Hansen and Travis (1974)):'.

Line 108: There is some ambiguity here. Please specify that it is particle size information from SMPS data that is included in the integration.

Line 114: Please specify that the methods are 'Numerical methods'. Also, please amend text to state that these 'numerical methods' are not *for* remote sensing measurement, rather are *applied* to remote sensing data.

Line 138: For readability, the authors might want to move the sentence on lines 143 – 144 to after line 138 '…complementary *in situ* measurements.' to describe the direct measurements of $R_{eff,f}$ that the authors perform. Also, mention here that optical measurements of impactor-selected portions of the aerosol ensemble were performed to measure FMF directly.

Lines 141 – 143: Brackets not required.

Line 180 – 183: Could the authors make it clearer that α is the spectral derivative for the whole aerosol sample, while α_f is that measured when an impactor is used to remove coarse mode contributions.

Line 181, '…is combined with…': This is ambiguous. How are α and α_f combined? This is unclear to the reader at this early stage in the text. An equation to define FMF in terms of α and α_f would be useful here. Indeed, this equation is equation (2) later in the text. Could the authors move equation 2 to this point and define FMF here.

Line 189: What is meant by 'modality'? Please could the authors clarify the text here.

Line 189: What 'measurements' are the authors referring to? Size distribution measurements, perhaps.

Line 190: Could the authors give these three equations? What are the dependent variables?

Line 188 – 191: This whole sentence is vague, difficult to read and needs clarifying. What is meant by 'approximation level relative to a theoretical Mie representation' and 'limited to second order'?

Line 193: Is the set of three equations referred to here the same as the 'three succinct equations' referred to on line 190? If so, please clarify in the text.

Line 196: Please specify reference wavelength. I believe this is 500 nm, but please specify to remove any doubt.

Line 209: What is 'ibid'?

Line 210: The is ambiguity here. What is meant by 'this' in 'estimate of this purely optical parameter…'. Presumably, 'this' is referring to the van de Hulst parameter, but the authors should be more specific here to remove doubt.

Lines 225 – 226: Ambiguity; it is not clear what is meant by 'polar-coordinate system relationship'. Moreover, the phrase 'near monotonic fit' is also ambiguous; a near monotonic fit of what function?

Line 228: Brackets around reference not needed.

Line 230: Ambiguity; please specify what is being compared in 'The comparisons…'.

Line 246: What is meant by 'expensive'? Computationally expensive, or expensive in monetary terms?

Line 296: Remove brackets around reference.

Line 340: Lower case 'N' in nephelometer.

Line 425: Do the authors mean extinction, instead of scattering? For the cases of aerosols sampled here, it probably does not matter. But, with the authors preferring *extinction* throughout the manuscript, it would be good to be consistent.

Line 538: Full stop (period) required after 'fine mode distribution'.