Aerosol optical properties

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1 Abstract

Traditionally the atmospheric physics department at the University of Oxford has applied optimal estimation techniques for the retrieval of atmospheric properties of gases; temperature, pressure and volume mixing ratio from satellite measurements. This poster describes the latest novel application of these techniques in deriving aerosol optical properties in laboratory experiments. Two examples are given; a spectral resolved technique allows an aerosol refractive index to be derived over a wide wavelength range and a method of deriving single particle refractive index and size from a novel aerosol instrument.

2 Optimal estimation; a generic method

- Measurements **y** are related to the state **x**, by physics. This is represented by the forward model **F**(**x**):

$$\mathbf{y} = \mathbf{F}(\mathbf{x})$$

- We need to invert our measurements to find the state, (i.e. refractive index, and particle size). We also us the best knowledge of the solution $\mathbf{x_a}$ before the measurement was made:

$$\mathbf{x} = \mathbf{F}^{\mathbf{-1}}(\mathbf{y}, \mathbf{x_a})$$

- To do this we minimise the cost function to find the state **x**:

$$\mathbf{\Phi}(\mathbf{x}) = (\mathbf{x}-\mathbf{x_a})^{\mathbf{T}}\mathbf{S_{xa}}(\mathbf{x}-\mathbf{x_a}) + (\mathbf{y}-\mathbf{F}(\mathbf{x})^{\mathbf{T}})\mathbf{S_y}^{\mathbf{-1}}(\mathbf{y}-\mathbf{F}(\mathbf{x}))$$

- And we find:

$$\hat{\mathbf{x}} = \mathbf{x_a} + \mathbf{S_{x_a}}\mathbf{K_x^T}\mathbf{S_y^{-1}}(\mathbf{y}-\mathbf{F}(\hat{\mathbf{x}}))$$

where $\mathbf{K_x} = \frac{\partial \mathbf{F}}{\partial \mathbf{x}}$

- Which is solved by the Levenberg-Marquardt algorithm (Gauss-Newton and the gradient descent iteration methods).

3 Aerosol refractive index

3.1 Method

The extinction cross section is related to the optical transmission by:

$$T(\lambda) = \exp^{-\beta(\lambda)x}$$

Where:
T Transmission.
β Volume extinction coefficient.
x Measurement path length.

The volume extinction coefficient is given by:

$$\beta(\lambda) = \int_0^\infty \sigma_{ext}(r, m, \lambda)n(r)dr$$

Where:

σ_{ext}	Extinction coefficient.
r	Particle radii.
m	Particle complex refractive index.
λ	Wavelength.
$n(r)dr$	Number of particles between radii r and $r + dr$.

Hence to obtain the extinction coefficient, σ_{ext} we require the measurements of the optical transmission, T as well as the particle distribution, $n(r)$. The forward model **F**(**x**) represents the transmission of the cell, and the size distribution and refractive index the state **x**.

3.2 Experimental setup

Figure 1 outlines the basic configuration of the experiments undertaken. The aerosol is generated (the generation method is chosen based on the aerosol type) then introduced to the small aerosol cell. The aerosol cell has optical windows fitted, allowing the aerosol absorption to be measured via the Fourier Transform Spectrometer, FTS. Particle size distribution of the aerosol is then determined using techniques insensitive to particle refractive index, and the aerosol vented into a fume cupboard. The configuration also included a water bath to allow the relative humidity to be controlled.

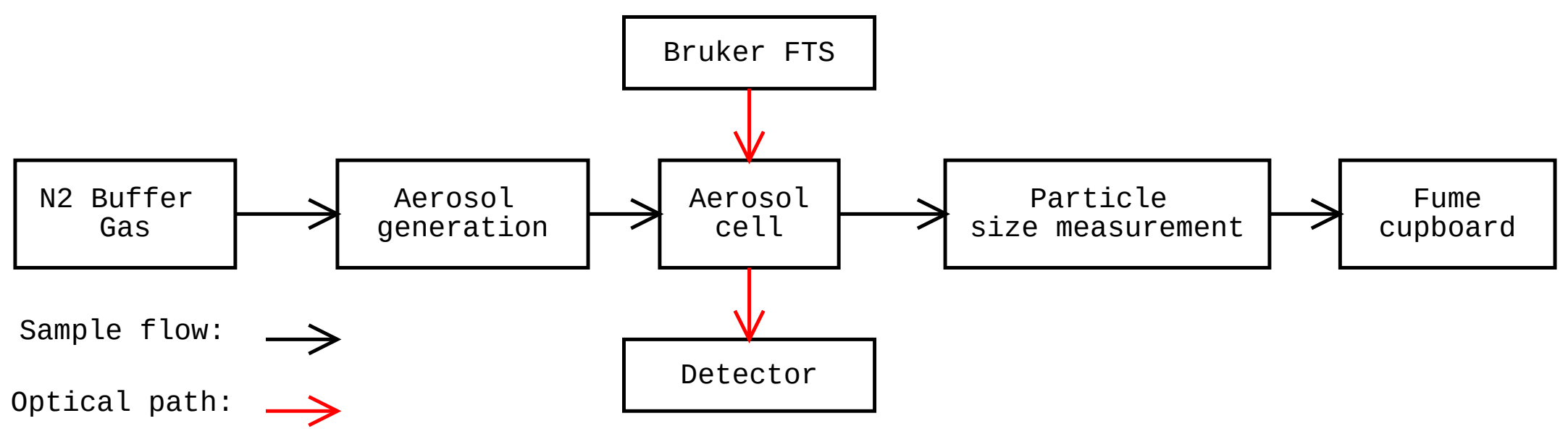

Figure 1: Simplified diagram of experimental configuration.

The MSF small aerosol cell has an optical path length of 30 cm. Future work will use a multi-pass cell. Spectral intensity measurements are made using a Bruker FTS. Measurements of the detected intensity are obtained with and without the aerosol to calculate the transmission spectrum, $T(\lambda)$.

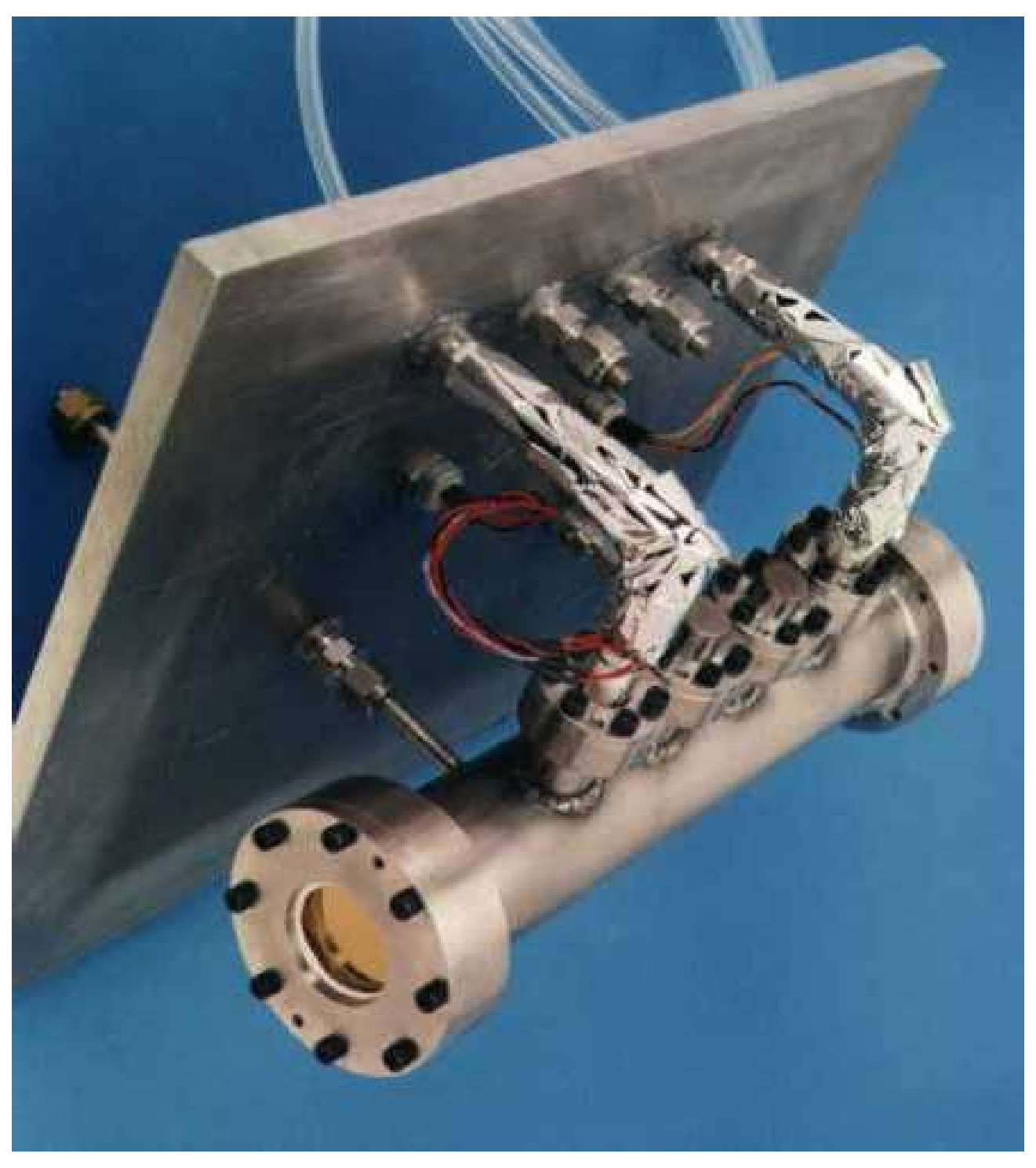

Figure 2: The small aerosol cell.

3.3 Results: Refractive index

The method describe by Thomas [4][3] has been used to retrieval the refractive index of the aerosol from the measured absorption spectra. An example fit is shown in figure 3, complete with the associated refractive index in figure 4.

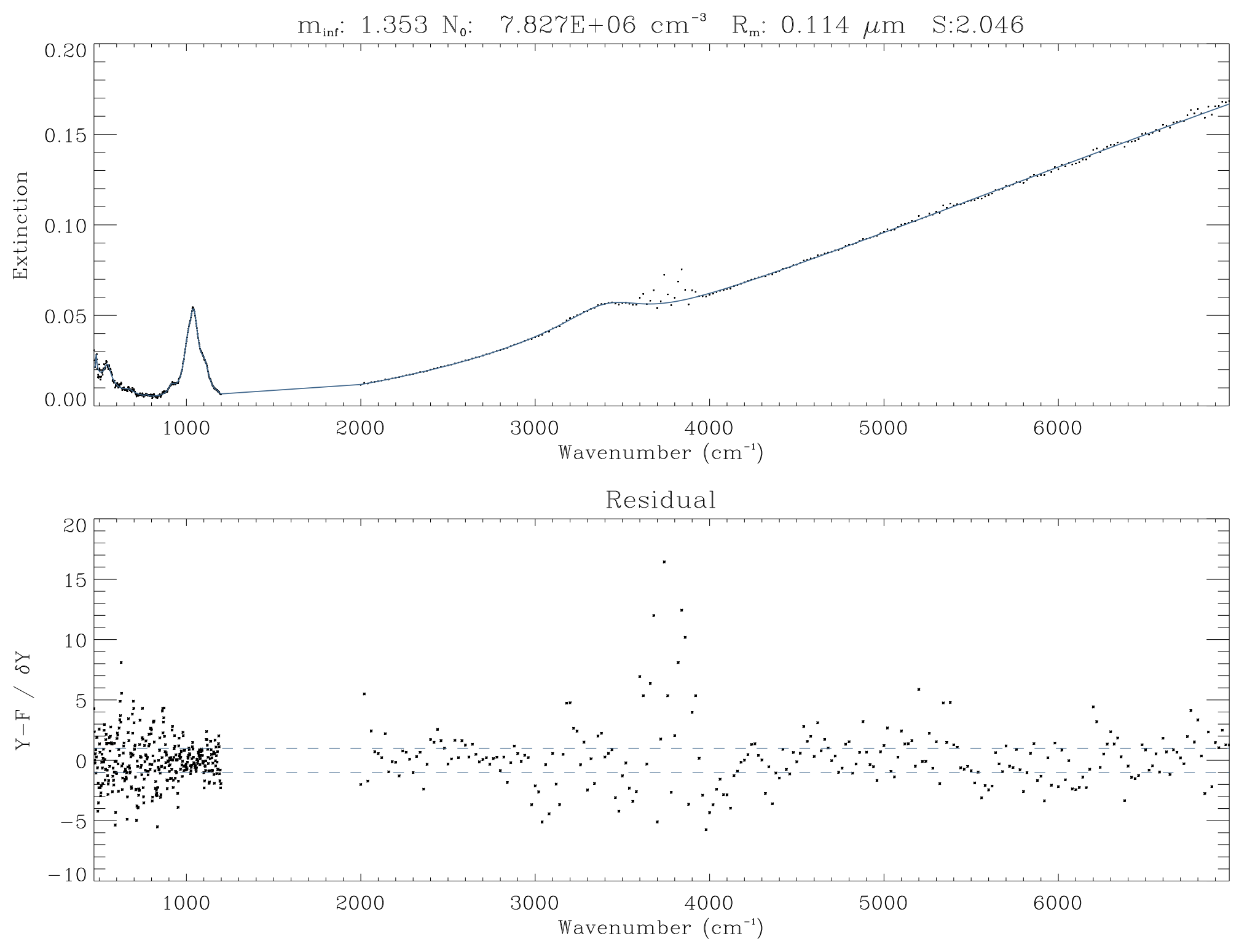

Figure 3: Top: Measured spectra (dots) and fitted spectra (lines). Bottom: residual.

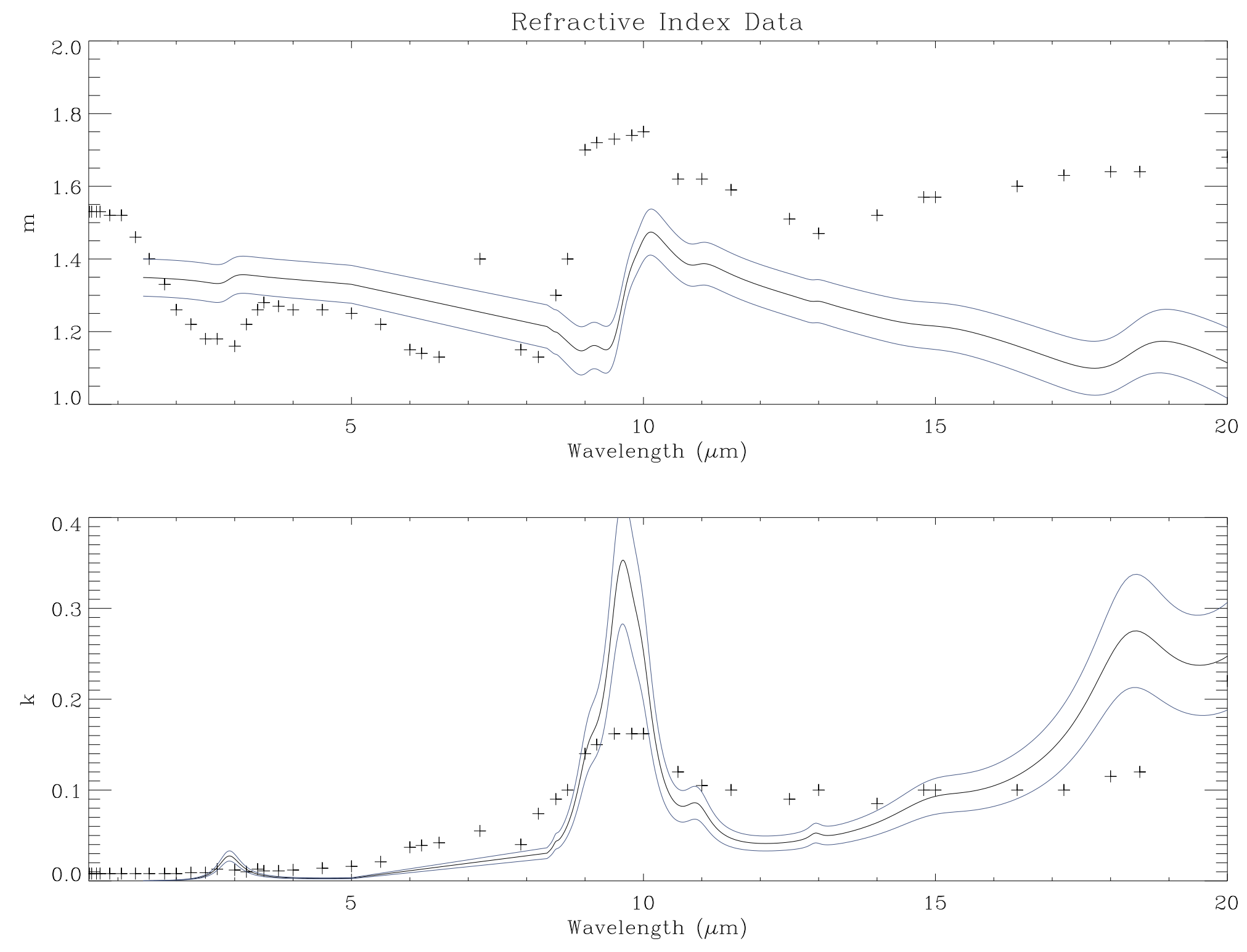

Figure 4: Inverted complex refractive example for Cape Verde dust (black line). Blue line indicates estimated uncertainties. Crosses show the existing published data [2]

4 A novel instrument

4.1 Method

The scattering of radiation by a particle is not only dependent on its size and shape, but also on its refractive index. Traditionally, this dependence has been viewed as a hindrance to the performance of optical particle counters, as it requires assumptions to be made about the refractive index to allow size estimates to be calculated. Instruments such as the Wyoming OPC use white light and carefully positioned detectors to minimize the sensitivity of the instrument to particle refractive index [1]. The SPARCLE instrument represents a shift in measurement principle to actively using this dependence to gain more information about the aerosol particles. Additionally, data analysis is performed on a particle-by-particle basis in the SPARCLE system. This enables the attribution of particles' sources (by their refractive index) to different size ranges this cannot be done using the Wyoming OPC system, where the returned data are simply the number of counts per channel.

There are two key innovations that form the basis of the SPARCLE instrument:

- The use of a solid state detector array within a small, autonomous in situ instrument to provide a high resolution measurement of particle scattering on single particles.
- The analysis scheme employed by the instrument is completely original. This is the first numerical retrieval scheme which provides both the particle radius and the complex refractive index from a measurement of light in the Mie scattering regime. The development of such an algorithm and its application to actual measurements represents a new state-of-the-art in optical particle measurement.

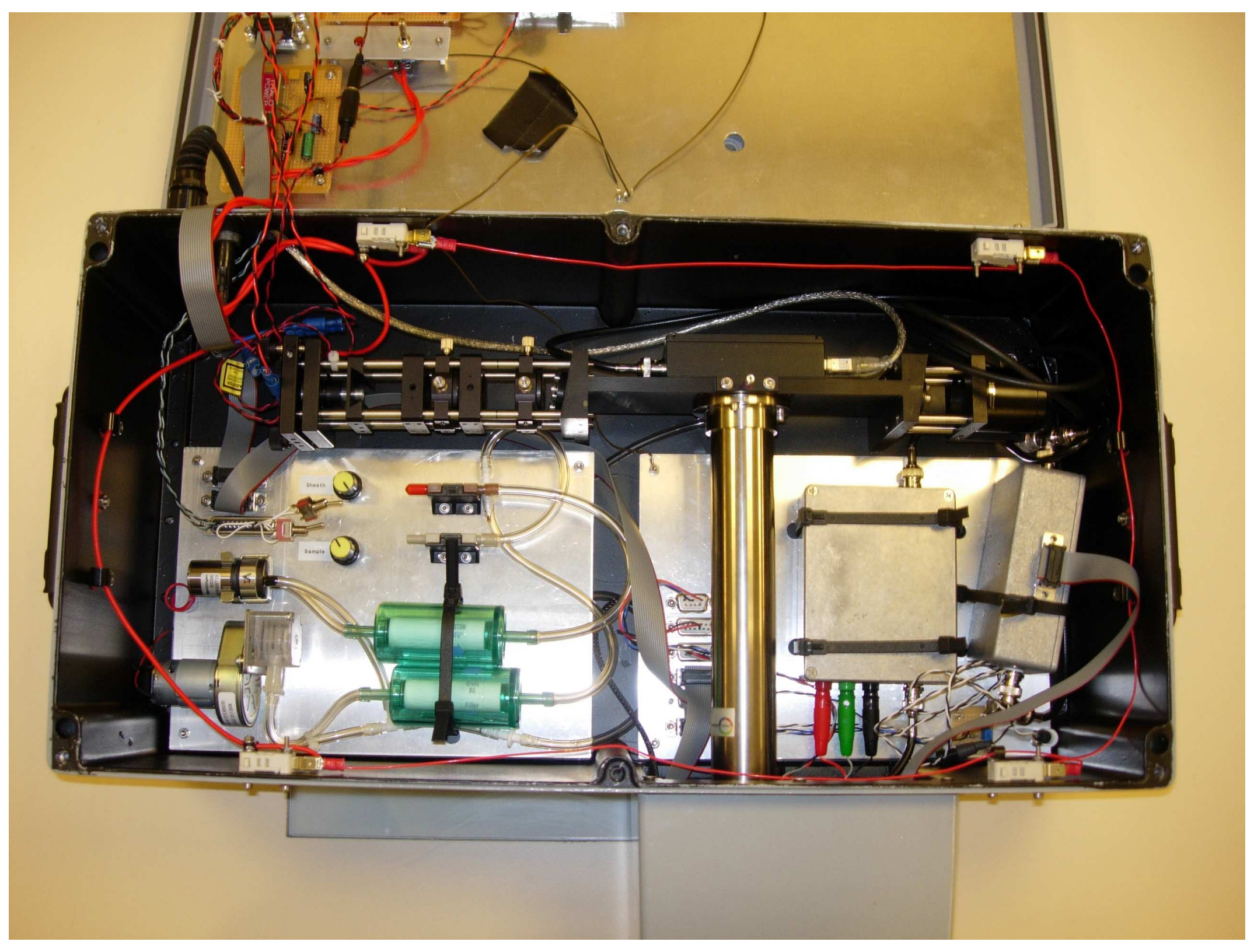

Figure 5: Prototype SPARCLE as built for the Omega partnership.

Sensitivity to both refractive index and size allows measurement of the single scatter albedo on each particle (by assuming Mie theory). SPARCLE is the first instrument to be able to make this measurement on single particles in the atmosphere.

4.2 Results: Novel instrument

The instrument uses two detectors; a fast sensitive photomultiplier tube (PMT) and a linear detector array (LDA). The LDA records individual particle phase functions and the PMT measures over a wide angular range to provide sensitivity to the smallest particles. Thus the forward model; **F**(**x**) relates the detectors measured phase function via mie theory to the particle size and refractive index, the state vector **x**.

The optimal position of the detectors has been determined to allow the highest instrument performance to be obtained (see figure 6).

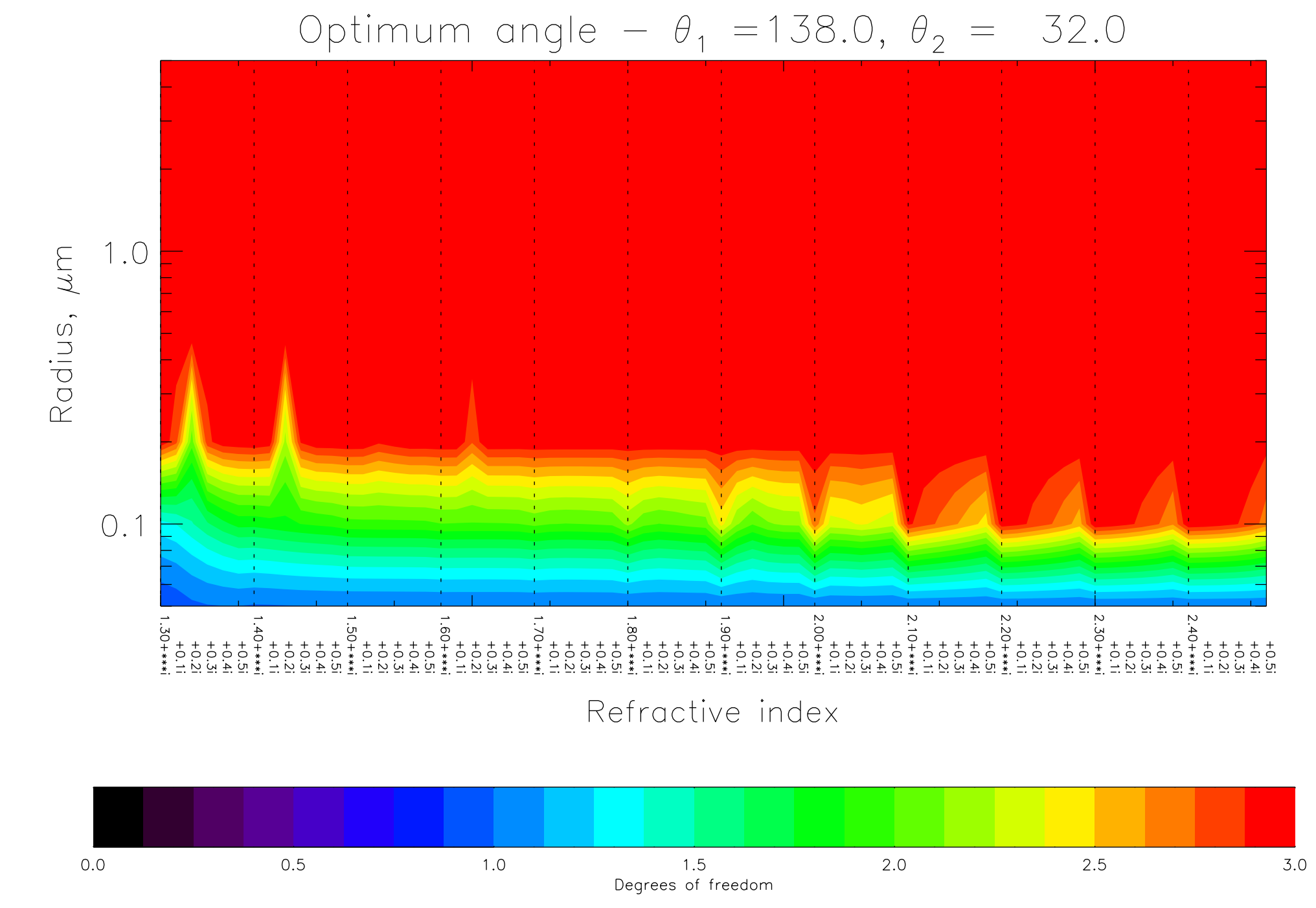

Figure 6: Optimized instrument performance.

The current prototype instrument is able to detect and measure the phase function of test particles and has been field tested. Further work is required to increase the LDA sensitivity. The current set up is limited by digitization noise. Low noise pre-amplification is required to ensure the detectors are dark current limited hence obtaining the highest instrument sensitivity. The increased instrument sensitivity will allow the instrument to reach it's full potential in determining single particle optical properties.

5 Conclusions

Two examples of the application of optimal estimation our given. The Aerosol refractive index of Cape Verde dust has been successfully retrieved. A novel method to determine individual particle refractive index has been describe, and this is very much work in progress (subject to funding) and shows potential to determine single scatter albedo, particle refractive index and size on individual particles for the first time.

References

[1] T. Dashler and et al. Thirty years of in situ stratospheric aerosol size distribution measurements from laramie, wyoming (41°n), using balloonborne instruments. *J. Geophys. Res*, 2003.

[2] Eric P. Shettle and Robert W. Fenn. Models for the aerosols of the lower atmosphere and the effects of humitdity variations on their optical properties. Technical Report AFGL-TR-79-0214, Air Force Geophysics Laboratory, September 1979.

[3] G. E. Thomas, R. G. Grainger, and R. A. McPheat. A new method of retrieving aerosol optical properties form IR extinction measurements. *COSMAS annual meeting Bristol*, 2004.

[4] Gareth E. Thomas, Stephen F. Bass, Roy G. Grainger, and Alyn Lambert. Retrieval of aerosol refractive index from extinction spectra with a damped harmonic-oscillator band model. *Applied Optics*, 2005.