Black carbon refractive index and morphology: a Laboratory study

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1 Abstract

Global estimates of the aerosol radiative effects are generally based on models employing aerosol radiative properties derived from Mie theory and therefore limited to the assumption of spherical aerosol particles. In addition, only a very limited number of global models explicitly consider the aerosol mixing state that has a strong effect on black carbon absorption [4],[6]; as well as the UK Chemistry Aerosol community model [2]. However, such microphysical aerosol models severely lack observational constraints on both aerosol morphology and mixing state, adding to the uncertainty of the simulated radiative effects. Bond, [1] summarize the aspects of black carbon that are unknown or uncertain and affect our ability to understand their climatic effect (in order of priority):

- Understanding the variability in reported refractive index of black carbon.
- Identifying critical morphologies that effect scattering and absorption.
- Reducing these to measurable atmospheric quantities.
- How to represent this information in climate models, what are the effective parameterisations.

This project establishes via experiments a set of refractive indexes of black carbon over a range of morphologies. In addition the project quantifies the impact of the effect of BC particle morphology on global aerosol radiative forcing. Hence the study provides an essential observational constraint on the representation of black carbon in global microphysical aerosol models, such as UKCA. This study derives the refractive indices black carbon (BC) aerosol and to characterise the changes in BC optical properties as a function of particle morphology. Measurements of the extinction spectrum from the mid infrared (25 /*mum*) to the near-ultraviolet (250 nm) of black carbon aerosols are made. The measurements are carried out within the aerosol cell at the NERC Molecular Spectroscopy Facility. The method of Thomas [8] is used to derive the complex refractive index, *m* of both the measured carbon aggregates and of equivalent spheres. In addition to the black carbon refractive indices the output of this study includes a parameterisation of the effect of morphology on carbon equivalent sphere optical properties. This paper discusses the latest results of the study.

2 The problem: Wide range of reported optical properties

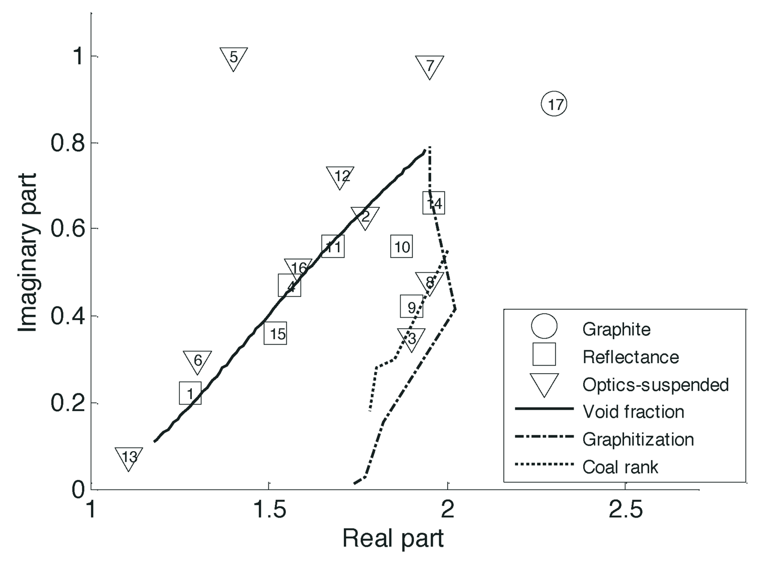

Figure 1: Spread of literature values of refractive index according to Bond[1].

3 Project outline

1. Generate representative black carbon aerosol.
2. Generate a black carbon aerosol of differing known morphologies.
3. Measure the optical properties of the aerosol (spectral transmission).
4. Measure size of aerosol during experiment.
5. Invert for refractive index assuming mie theory.
6. Show how mie derived refractive index varies with morphologies.

4 Global sources of black carbon: What is representative?

Fuel	% share
Grassland	21.2
Diesel	16.7
Tropical forest	12.4
Wood	7.9
Coal	6.9
Cooking processes	4.9
Agricultural waste	4.8
Crop residues	4
Extra-tropical forest	2.9
Dung cake	2.6
Heavy fuel oil	1.5

Figure 2: Global sources by fuel types [7].

A wide range of fuel types contribute to the global loading of black carbon. Our experiment should include the largest sources of black carbon (see Table). We intend to include some bulk samples of biomass burning. Due to the wide range of burning conditions; these cannot be simulated accurately in the laboratory. We thus intend to concentrate on mimic Diesel soot that can be produced under controlled conditions.

5 Experiential number density requirements

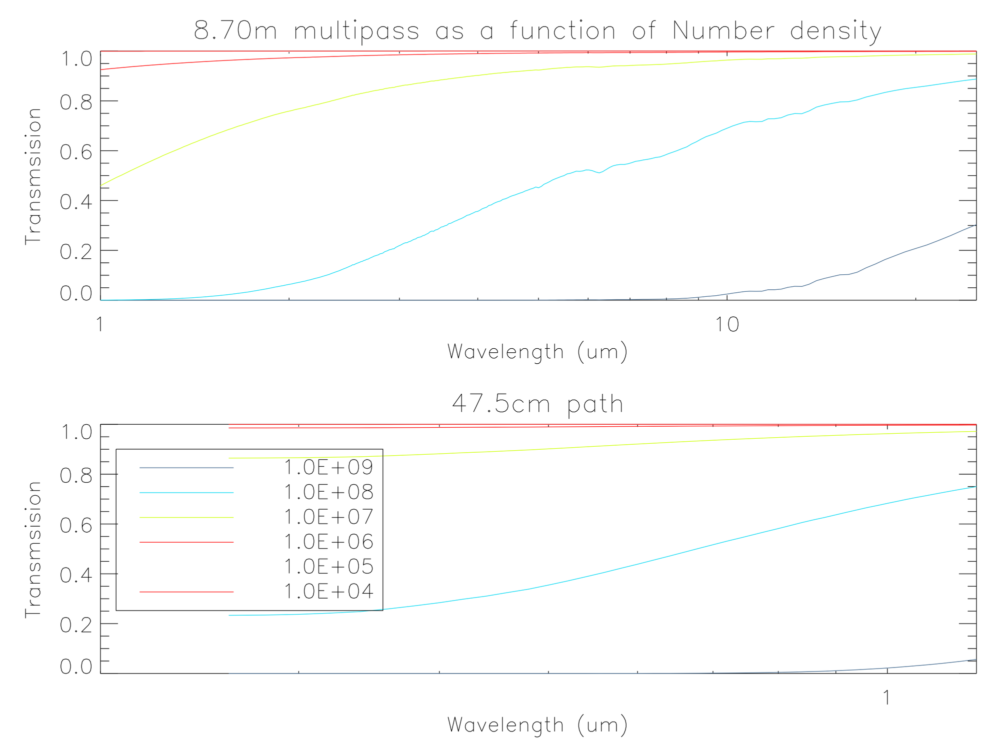

Figure 3: Transmission signal at different number densities (cm^{-1}) for a log-normal distribution of radius 150 nm and spread 1.3.

To obtain good optical measurements high number densities are required. This puts constraints on both aerosol life time in the laboratory cell and the generation methods.

6 Possible laboratory aerosol generation

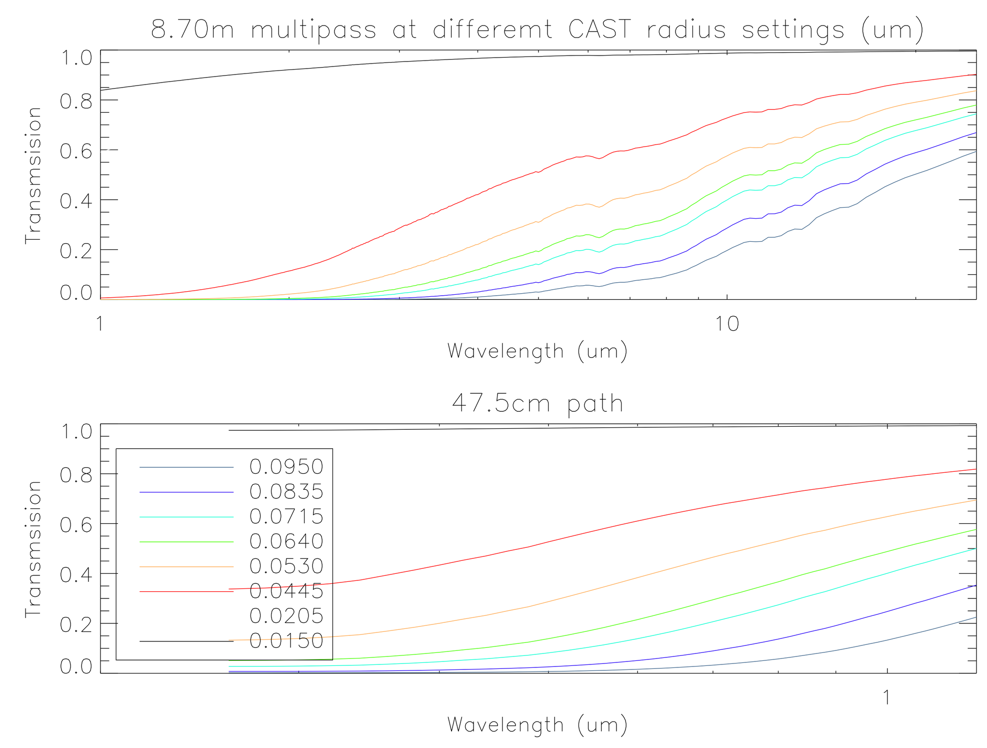

Figure 4: Expected transmission for MinCAST 5200.

Options identified include the use of a MiniCAST 5200 soot generator, or mechanical re-dispersal from a collected bulk sample. We intend to use both approaches. Other generation methods examined so far produce number density insufficient to provide on optical signal. A MiniCAST 5200 should provide a controlled repeatable aerosol of differing morphologies. With the addition of bulk samples allow exploring realistic samples of other black carbon aerosols.

7 Morphology determination

We intend to calibrate the fractal number of the MiniCAST 5200 using a combination of Differential Mobility Analyser, DMA, (giving the volume equivalent diameter) and Couette Centrifugal Particle Mass Analyser, CPMA, (giving particle mass) [5].

We will assume the fractal number is then consistent for the CAST flame settings, this will be confirmed though the experiment by the repeatability of the volume equivalent diameter measured by the DMA. TEM/SEM analysis is time consuming and costly so we wish to avoid this is possible. *Any other options?.*

8 What about organics?

The MiniCAST 5200 soot has a known repeatable organic content. Bulk dispersion of known carbon blacks may also allow control of the organic content. Other options include striping off the organics in a thermal-denuder to ensure the organic component removed or at least minimized. *Any other options?*

9 Aerosol Refractive index Archive

Our experience shows that finding and collating the refractive index's that are relevant for atmospheric modeling and remote sensing is time consuming. In addition it is difficult or impossible to obtain tabulated data for some of the older publications. A suitable electronic location for new data was also required. The Aerosol Refractive index Archive, ARIA, has been launched to reduce the effort of the community and provide a central location for refractive index data. The data base can be found at [3].

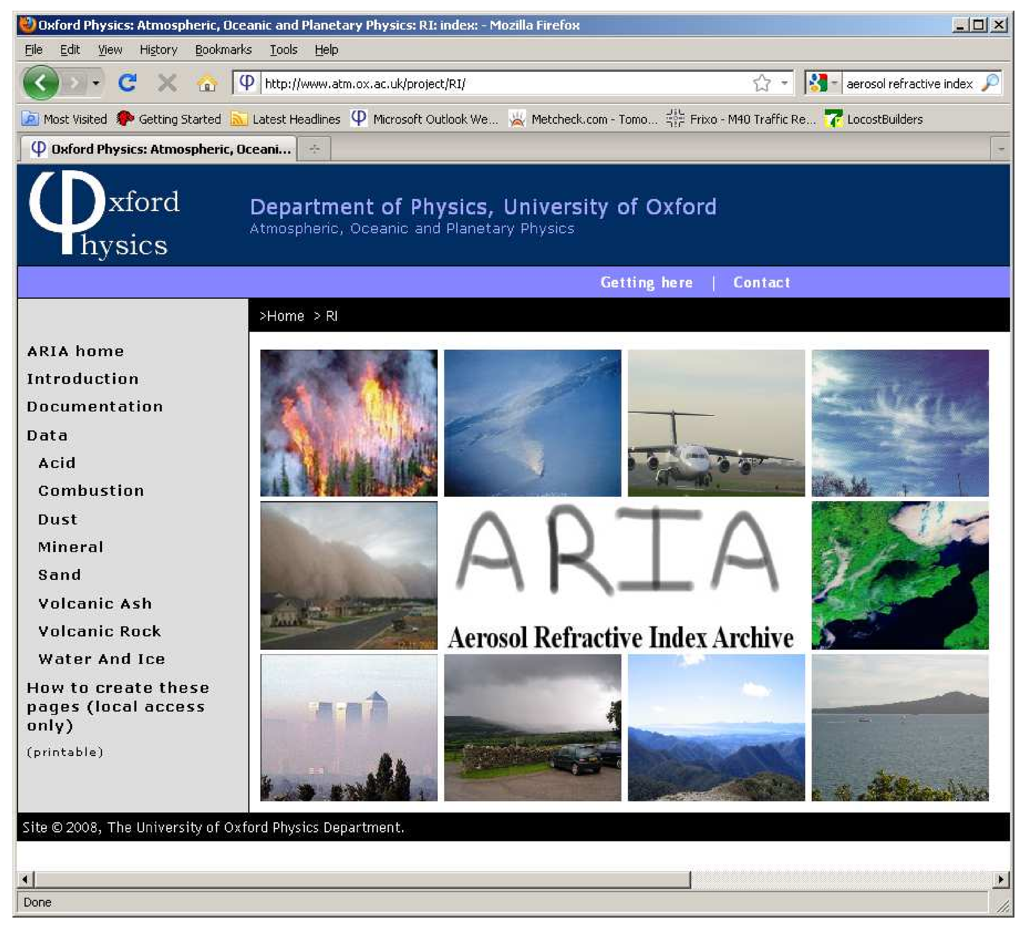

Figure 5: ARIA home page [3]

A simple human readable file format and file naming convention has been defined and documented. IDL and Matlab example read routines can be downloaded. Additional fields are available to provide information, for example the original citation in the files which is automatically displayed on the web database. We also provide fields for uncertainty of both the imaginary and real parts of the refractive index where these are available.

The database is now "live" but is still under development and we would welcome you input and comments on the current database and how it could be improved.

9.1 We need your contributions

To make the database more useful we need the *user community to contribute aerosol refractive index data.* In addition we would welcome *your comments and suggestions for improvements to the database.*

10 Acknowledgments

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