Aerosol Research at the Rutherford Appleton Laboratory

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Introduction

The Molecular Spectroscopy Facility (MSF) provides laboratories for high-resolution, broadband optical spectroscopy covering 10 to 50,000 cm^{-1} (1 mm to 200 nm). The spectrometers available include Fourier transform (FTS), CCD and diode-array instruments providing spectral resolution up to one part in one million, high-sensitivity and sub micro-second time-resolution. The spectrometers may be interfaced to a range of temperature-variable spectroscopic cells for optical absorption, emission or scattering measurements on gases, vapours, liquids and aerosols at path-lengths from 1 mm to 1 km and temperatures from 175 to 340 K.

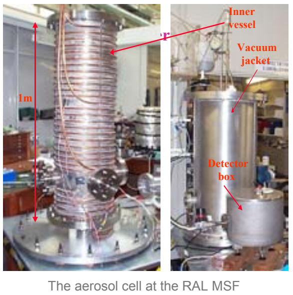
The Bruker IFS 66v/S FTS for measurements requiring moderate spectral resolution

UV photochemistry using the high-resolution Bruker IFS 120HR FTS

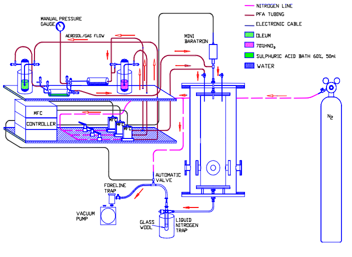
Long path-length absorption cell for broadband quantitative measurements from 32 m to over 1 km optical path

Aerosol Cell

The aerosol cell[1,2,3] has a 75 litre volume and is interfaced to a FTS. The cell comprises an inner vessel that contains the sample and an outer jacket that envelopes the inner vessel in a vacuum for thermal insulation. The inner vessel is stainless-steel, and can be cooled. There are a number of inlets / outlets for the addition of gases or for sampling composition or size distribution. There are four ports for optical measurement. The cell has 20 platinum resistance thermometers (PRT), located at various places on the exterior and interior of the inner vessel.



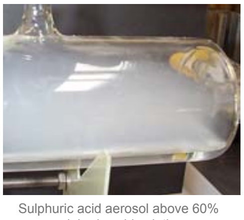
The aerosol cell at the RAL MSF



The aerosol generation system for STS aerosol

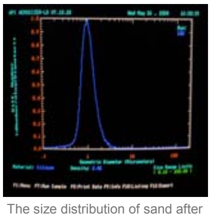
Aerosol Generation

A flexible system for generating stable flows of aerosols such as STS and sulphate has been developed. The flow rate of components, the carrier gas and the cell pressure can be varied to give the required composition. Systems for generating solid aerosols with known size distributions are currently under development, and preliminary tests using crushed sand are encouraging. This new system will be used to study volcanic and Saharan dust samples.



Sulphuric acid aerosol above 60% sulphuric acid solution

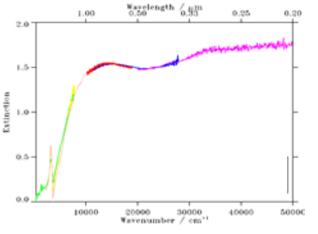
Solid aerosol generation and size distribution measurement



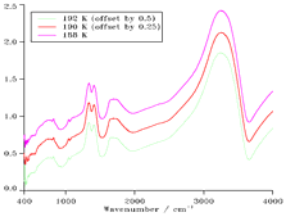
The size distribution of sand after passing through a 20 cm cell

Results

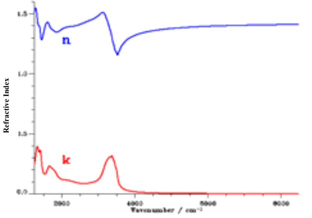
The IR and visible/UV extinction spectra of STS aerosol are shown below. The spectrum was constructed by combining data from FTS and CCD measurements. The temperature dependence of the IR extinction of STS aerosol with a nitric acid content of 25% is also shown. The STS data (currently consisting of over 1000 different spectral measurements) is being analysed by The University of Oxford to generate complex refractive indices. A new model that determines the complex dielectric constant from a damped harmonic oscillator is being used.



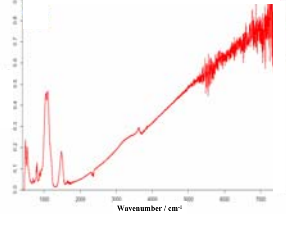
IR and visible/UV extinction spectra of STS aerosol recorded using the Bruker IFS66v/S FTS and fibre-optic CCD spectrometers



Temperature-dependent IR extinction spectra of STS aerosol



Refractive indices of STS aerosol retrieved from extinction spectra at 189 K



The IR extinction spectrum of crushed sand at room temperature using a 20 cm path cell

Future Work

A new programme of measurements on other types of PSC, e.g. NAT, will begin shortly. Preliminary measurements have shown the system's ability to generate aerosols of nitric acid hydrates. Previously the number density and particle size distribution was calculated during the derivation of complex refractive indices. Simultaneous measurements by a scanning mobility particle sizer and an optical particle counter will allow these quantities to known independently. The integration of a mass spectrometer with the aerosol cell will allow the measurement of aerosol chemical composition.

In the longer term the addition of a tuneable laser spectrometer will allow the concentrations of gas phase components to be measured simultaneously at high-resolution and at stratospheric temperatures. The MSF is currently developing optical multi-pass systems for measurements on aerosols at lower number densities for use with FTS and laser systems.

References

[1] R. A. McPheat, D. A. Newnham, R. G. Williams and J. Ballard, Large-volume, coolable spectroscopic cell for aerosol studies, *Appl. Optics*, 40 (36) 6581-6586, 2001.
[2] R. A. McPheat, S. F. Bass, D. A. Newnham, J. Ballard and J. J. Remedios, Comparison of aerosol and thin film spectra of supercooled ternary solution aerosol, *J. Geophys. Res. – Atmospheres,* 107 (D19), 4371, 2002.
[3] Suzanne B. Couling, John Fletcher, Andrew B. Horn, David A. Newnham, Robert A. McPheat and R. Gary Williams, First detection of molecular hydrate complexes in sulfuric acid aerosols, *Phys. Chem. Chem. Phys.*, 5, 4108-4113, 2003.

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