Journal of Quantitative Spectroscopy & Radiative Transfer 199 (2017) 52–65

Contents lists available at ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

Mass extinction spectra and size distribution measurements of quartz and amorphous silica aerosol at 0.33–19 µm compared to modelled extinction using Mie, CDE, and *T*-matrix theories

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ARTICLE INFO

Article history:
Received 23 March 2017
Revised 11 May 2017
Accepted 11 May 2017
Available online 19 May 2017

Keywords:
Mass extinction coefficient
Aerosols
Quartz
Amorphous silica
Mie theory
Non-spherical scattering
T-matrix theory
Rayleigh CDE model

ABSTRACT

Simultaneous measurements were made of the spectral extinction (from 0.33–19 µm) and particle size distribution of silica aerosol dispersed in nitrogen gas. Two optical systems were used to measure the extinction spectra over a wide spectral range: a Fourier transform spectrometer in the infrared and two diffraction grating spectrometers covering visible and ultraviolet wavelengths. The particle size distribution was measured using a scanning mobility particle sizer and an optical particle counter. The measurements were applied to one amorphous and two crsystalline silica (quartz) samples. In the infrared peak values of the mass extinction coefficient (MEC) of the crystalline samples were 1.63 ± 0.23 $\mathrm{m^2 g^{-1}}$ at 9.06 µm and 1.53 ± 0.26 $\mathrm{m^2 g^{-1}}$ at 9.14 µm with corresponding effective radii of 0.267 and 0.331 µm, respectively. For the amorphous sample the peak MEC value was 1.37 ± 0.18 $\mathrm{m^2 g^{-1}}$ at 8.98 µm and the effective radius of the particles was 0.374 µm. Using the measured size distribution and literature values of the complex refractive index as inputs, three scattering models were evaluated for modelling the extinction: Mie theory, the Rayleigh continuous distribution of ellipsoids (CDE) model, and *T*-matrix modelling of a distribution of spheroids. Mie theory provided poor fits to the infrared extinction of quartz ($R^2 < 0.19$), although the discrepancies were significantly lower for Mie theory and the amorphous silica sample ($R^2 = 0.86$). The CDE model provided improved fits in the infrared compared to Mie theory, with $R^2 > 0.82$ for crsytalline sillica and $R^2 = 0.98$ for amorphous silica. The *T*-matrix approach was able to fit the amorphous infrared extinction data with an R^2 value of 0.995. Allowing for the possibility of reduced crystallinity in the milled crystal samples, using a mixture of amorphous and crystalline *T*-matrix cross-sections provided fits with R^2 values greater than 0.97 for the infrared extinction of the crystalline samples.

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1. Introduction

Atmospheric aerosols directly affect the Earth's radiative balance, across the electromagnetic spectrum from the infrared (IR) to the ultraviolet (UV), by scattering and absorbing solar radiation as well as absorbing and emitting infrared radiation [1,2]. Atmospheric aerosols also affect the climate indirectly by acting as ice and cloud condensation nuclei [3,4] and by providing catalytic surfaces for heterogeneous chemical reactions [5]. Uncertainties in quantifying the direct and indirect effects of aerosols on radiative forcing are two of the largest contributions to the uncertainty in predicting future climate change [6].

Atmospheric mineral dust has a significant impact on the Earth's climate [6]. Between 1000–3000 Mt of wind blown dust enters the atmosphere annually [7], largely from arid and semi-arid regions located in a broad band that stretches from the west coast of North Africa, through the Middle East, to Central Asia [8]. The Sahara is the largest emission source, at an estimated 670 Mt $\mathrm{yr^{-1}}$, generating the largest atmospheric mass loading of mineral dust aerosol [9]. Strong winds and convection from surface heating uplift mineral dust particles into the troposphere, at

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http://dx.doi.org/10.1016/j.jqsrt.2017.05.011
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which point atmospheric circulation can transport the dust on intercontinental scales [10–13], reaching as far as Northern Europe and South America [14,15].

Silicon dioxide (silica) is the most abundant oxide in the Earth's crust comprising an estimated 12% by volume [16]. Naturally occurring silica is found in the form of crystalline alpha-quartz which has a regular structure of SiO_4 tetrahedra. Quartz is a major externally mixed constituent of wind blown mineral dust, although its percentage by mass in atmospheric dust samples has been found to vary significantly by location [17,18].

Silica is also a major internally mixed constituent of volcanic ash, which has silica content varying from 50–75% [19,20]. Even moderately sized volcanic eruptions can inject many hundreds of megatonnes of volcanic ash into the troposphere and stratosphere, following which the plume can be transported thousands of kilometres posing significant risk to aviation [21–23]. The 2010 Eyjafjallajökull volcanic eruption injected approximately 380 Mt of ash into the atmosphere [24] and is estimated to have cost the global economy US $ 5 bn [25,26] resulting from six days of European airspace closure as the ash plume propagated downwind. The silica within volcanic ash is mainly quenched into fragments of amorphous volcanic glass, which consists of disordered networks of SiO_4 tetrahedra interconnected with other minerals [27]. The strong infrared absorption feature of volcanic ash at 8–12μm, attributable to $T{-}O^-$ vibrations (where T indicates fourfold coordinated cations, predominately Si^{4+}, Al^{4+} and Fe^{3+}) [28], is widely exploited for rapid ash detection using satellites and other remote sensing instruments [29,30]. For example, large negative brightness temperature differences between 10 and 12μm channels are used by nadir imaging instruments to indicate the presence of ash [31].

The scattering and absorption of radiation produced by aerosol particles depend on the particles' size distribution, complex refractive index and shape distribution [32]. A large component of the uncertainty in modelling the effect of aerosols on atmospheric radiative transfer comes from uncertainty in assumptions about these parameters and the untangling of their individual effect on bulk optical properties such as the volume extinction coefficient and phase function; these uncertainties propagate to uncertainties in remote sensing of aerosols and radiative forcing calculations [33–35].

A scattering theory is required to determine the bulk optical properties from the size distribution, complex refractive index and shape distribution of the particles. In many applications the particles are assumed to be spheres, and Mie theory is used. The extinction produced by irregularly shaped mineral dust particles, including near to their infrared absorption features, is not well modelled by assuming spheres [36]. Significant errors are introduced when Mie scattering is assumed for remote sensing retrievals and climate forcing calculations [37,38]. In the Rayleigh approximation, simple analytic expressions can be derived for the scattering and extinction cross sections of randomly oriented idealized particle shapes (such as disks, needles and ellipsoids). The extinction predicted by assuming a continuous distribution of ellipsoids (CDE), in which all geometric shape factors are equally probable, provides a much improved fit to the infrared extinction of silica particles than Mie theory [32].

Substantial theoretical progress has been made in modelling the light scattering of non-spherical atmospheric aerosols. The discrete dipole approximation [39,40] and the finite difference time domain [41,42] methods have been applied to non-spherical, inhomogeneous particles. However, these techniques are highly computationally expensive, limiting their applicability to relatively small particles. *T*-matrix methods are widely used to model the scattering of irregularly shaped atmospheric aerosols [43,44]. The spheroid approximation is commonly applied within the *T*-matrix calculations because the orientational averaging is greatly simplified, reducing computational expense. The spheroid approach has been shown to provide more flexibility in matching the scattering of mineral dust compared to polyhedral prisms, and performs well compared to many other scattering models at matching light scattering data [45]. *T*-matrix methods, using a distribution of spheroids, have been applied to model climate forcing and within mineral dust remote sensing retrievals [46–48].

Despite the importance of the optical properties of solid atmospheric aerosol particles there are relatively few experimental measurements of these properties over the full spectrum relevant to atmospheric radiative transfer. This paper details a newly developed experimental technique and apparatus capable of accurately measuring the spectral extinction and size distribution of solid aerosol particles suspended in nitrogen gas. Two optical systems were used, covering wavelengths from the UV through to the IR (0.33–19 μm) and providing high spectral resolution (1.9 cm^{-1} in the IR and $<$ 1.5 nm in the shortwave). Simultaneously, the size distribution of particles was measured and filter samples suitable for scanning electron microscope (SEM) imaging were taken, allowing a full microphysical analysis of the particles. Previous high-quality laboratory measurements of the spectral extinction and size distribution of solid aerosol particles have been performed by [49–51]. These earlier measurements were limited to infrared wavelengths and were performed at the lower resolution of 8 cm^{-1}. Previous broadband extinction measurements of quartz aerosol have been performed by [52], however the particles' size distribution was not measured concurrently in this work.

This paper presents measurements applied to three high purity silica samples. The experimental apparatus and method have been significantly adapted from previous experiments on black carbon, presented in [53]. The motivation for the silica measurements is twofold. Firstly, silica is a major component of mineral dust and volcanic ash, both of which are important atmospheric aerosols. Secondly, the complex refractive indices of amorphous and crystalline silica are well documented in the literature, allowing the experimental extinction data to be evaluated against various scattering models (using the measured size distribution as an input), and allowing an assessment of the accuracy of the experimental method.

The paper is organised as follows. In Section 2 the experimental method and apparatus are outlined, and details about the three silica samples are provided. In Section 3 the analysis of the experimental data is detailed and results for the size distribution and the spectral mass extinction coefficient, with uncertainty, of the samples are presented. In Section 4, three scattering models — Mie theory, the Rayleigh CDE model, and *T*-matrix methods applying a distribution of spheroids are described. The modelled extinction using each theory, taking literature values of the complex refractive index of silica and the measured size distribution as inputs, are compared to the experimental extinction data. In Section 5 conclusions and planned future work are described.

2. Method

2.1. Overview of experimental setup

Experiments were performed at Rutherford Appleton Laboratory's Molecular Spectroscopy Facility (RAL-MSF). The samples were dispersed in nitrogen gas and passed into a 75 l aerosol cell chamber; the aerosol cell has been described in detail in [54]. Inside the cell, the optical extinction of the aerosol was measured at infrared wavelengths using a Fourier transform spectrometer, concurrent to ultraviolet and visible (VIS) extinction measurements made by two diffraction grating spectrometers. Upon leaving the cell, the size distribution of particles was measured by a scanning

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