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New approach for the determination of aerosol refractive indices – Part II: Experimental set-up and application to amorphous silica particles

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ABSTRACT

This article is the Part II of a work aimed at proposing a new method for determining the optical constants of aerosols. The Part I detailed the theoretical and numerical basis of an algorithm devoted to retrieve the imaginary and the real part of complex refractive indices from extinction spectra of aerosols. This algorithm associates the Mie theory, the single subtractive Kramers-Kronig relation, and an optimal estimation method in an iterative process. This Part II presents the experimental set-up developed to record simultaneously high spectral resolution extinction spectra and size distributions of airborne silica particles. Extinction spectra are measured with a high spectral resolution on a broad spectral range, including both infrared ($650-2{,}500\ cm^{-1}$) and UV-visible ($9{,}000-32{,}500\ cm^{-1}$) spectral regions. Experimental data were used to retrieve the complex refractive indices of aerosol particles. By associating the numerical procedure presented in the first paper and this experimental set-up, complex refractive indices of silica spherical aerosol particles have been determined under controlled experimental conditions. Additional comparison between experimental and simulated extinction spectra from retrieved complex refractive indices shows that this new methodology provides optical properties representative of the material.

1. Introduction

Due to their ability to absorb and scatter solar and terrestrial radiations, aerosols play an important role in the Earth's radiative balance [1]. Depending on their size, shape and mineralogical composition, aerosols contribute to a positive or negative radiative forcing [2] and, more generally, have direct and indirect effects on the climate [3], which still remain quantitatively misestimated [4].

Estimating and modeling the effect of aerosols on atmospheric processes requires accurate information about physico-chemical composition [5–7], size and shape distributions of the particles [5,7–9], and a complete understanding of aerosol absorption and scattering properties on a broad spectral range.

While still challenging, remote sensing techniques are powerful tools to measure atmospheric aerosols on a broad spectral range. In particular, satellite instruments are powerful tools to probe continuously the atmosphere from local to global scale [10–13]. The single channel and viewing angle radiometric observations such as from the Advanced Very High Resolution Radiometer (AVHRR) [11,14] provide estimated values of the Aerosol Optical Depth (AOD). Multi-channel instruments such as MODIS [15] or OMI [16] provide AOD, Angström exponent, and single scattering albedo (SSA) using *a priori* information from other aerosol parameters such as size distribution, or refractive indices. The aerosol information is significantly larger for instruments that perform measurements at multiple viewing angles, such as MISR [17] and AATSR [18], or multi-wavelength polarized radiances such as GOME-II, or PARASOL. Indeed, since the light polarization is very sensitive to the aerosol microphysics [19], many authors [20,21] have shown that these measurements give access to the aerosol loading of fine and coarse modes, the effective radius of at least one mode, and the mean height of the aerosol layer. The high spectral resolution infrared (IR) sounders have the advantage, of being highly sensitive to the aerosol type. Recently, many works have been achieved from IR spectrometers such as AIRS, IASI or TANSO-FTS [22–31]. The aerosol characteristics obtained from latest high spectral resolution observations on wide spectral ranges appear to be very promising to complement the aerosol parameters retrieved from previous observations, mainly recorded on narrow spectral ranges.

Nevertheless, extrapolations of aerosol parameter retrieved from one spectral region to another one may lead to questionable results. This is due to the fact that the majority of aerosol inver-

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sion processes from remote sensing information assume that the complex refractive indices of the airborne particles are well known [32,33]. To compare the aerosol information retrieved from various instruments covering different spectral ranges [34,35], it is essential to have accurate optical properties [22,33] on a broad spectral range. For instance, estimation of the uncertainties in retrieved parameters from IR measurements in the case of uncertainties due to volcanic ash particle type and particle size distribution have been estimated [36]. Uncertainties in the effective radius and the mass loading estimations can reach respectively, 20% for the refractive indices and 50% for the size distribution.

Indeed, accurate knowledge of complex refractive indices is a weak point for aerosol property retrievals (as AOD, SSA, or Angström coefficient) from remote sensing observations [33,37,38]. Common complex refractive indices used are available in open databases such as HITRAN (HIgh-resolution TRANsmission molecular absorption database) [39], GEISA (Gestion et Etude des Informations Spectroscopiques Atmosphériques) [40], ARIA (Aerosol Refractive Index Archive) [41], HJPDOC [42], OPAC (Optical Properties of Aerosols and Clouds) [43] and GADS (Global Aerosol Data Set) [44]. Moreover, it is important to note that these data cover narrow spectral ranges with poor spectral resolution (at best 4 cm^{-1}), which is a problem, particularly in the IR spectral region, where the presence of vibrational bands involves high frequency spectral variations of the indices.

As discussed in the companion paper (hereafter Paper I) [45], it is important that the retrieved optical constants from laboratory measurements correspond to particles in suspension. However, only a few sets of experimental studies have been performed with suspension of aerosol particles in an atmospheric chamber [46–48].

For instance, using AIDA facilities [47], the UV-visible extinction spectra for suspension of airborne Saharan dust samples have been recorded while IR extinction were obtained by using the pellet technique in order to link optical properties and mineralogy of the samples. In 2007, some experiments were performed in a stainless steel chamber for several key components of mineral dust in the IR and the UV-visible spectral regions [48]. The experimental extinction spectra were compared to simulated spectra calculated using Mie theory and published optical constants. Some discrepancies were observed between simulated and experimental extinction spectra mostly in the IR resonance absorption region (resonance peaks shifted by more than 40 cm^{-1}). A reason for such disagreement may be due to the fact that complex refractive indices from the literature used are not suitable for aerosol particles. As an example, for a kaolinite sample, authors had calculated new optical constants to obtain good agreement between experimental and simulated extinction spectra. Similar experiments were performed later where only IR spectra were recorded with the size distribution of the particles. Here also comparisons between experimental and simulated extinction spectra were achieved using the Mie theory and the Rayleigh model. The latter gives better results for peak position and band shape of the IR characteristic features.

Another difficulty arises when retrieving the real and imaginary parts of the complex refractive index from an experimental measurement (extinction spectra for example). In that case, the real and imaginary parts are linked by the Kramers-Kronig relations [49]. The mathematical expression of these equations shows that the experimental spectra should be recorded in the broadest possible spectral range. The later point is not always checked for some studies where data from other published work or data obtained from the IR spectral region only are used for the inversion process [50,51]. In this work, a complete methodology has been proposed for measuring optical properties of airborne particles in order to overcome the limitations mentioned above.

In this paper, we describe how this methodology has been applied for pure and calibrated silica (amorphous SiO_2) spherical particles. Silica exists in crystalline and noncrystalline form, the latter being also referred to as amorphous silica [52]. Silica has specific features in the IR spectral region ($8-12\,\mu m$) [6] which are the result of lattice vibrations [53]. For methodology validation, calibrated silica spherical particles are available from commercial sources. Optical properties of silica have often been studied throughout the literature [53] and have mainly been derived from experiments using pellets [50,53], and references therein. Lastly, silica is the main component of mineral dust and volcanic ash [54].

In this study, IR and UV-visible extinction spectra and size distributions have been recorded simultaneously for a suspension of calibrated silica spherical particles in order to retrieve complex refractive index. The structure of the paper is as follows: the experimental set-up and experimental protocol are described in Section 2. Section 3 presents an application of the complete methodology to amorphous silica spherical particles. Extinction spectra recorded in the IR and the UV-visible spectral regions are exposed for particles with diameter of 0.5 and 1.0 μm. A unique set of the complex refractive index for silica is retrieved in this whole spectral range ($650-32{,}500\ cm^{-1}$). Lastly, using the derived complex refractive index and the Mie theory, the extinction spectra are calculated and are compared to the experimental one for each size distribution. Finally, Section 4 summarizes our results and presents perspectives for future applications.

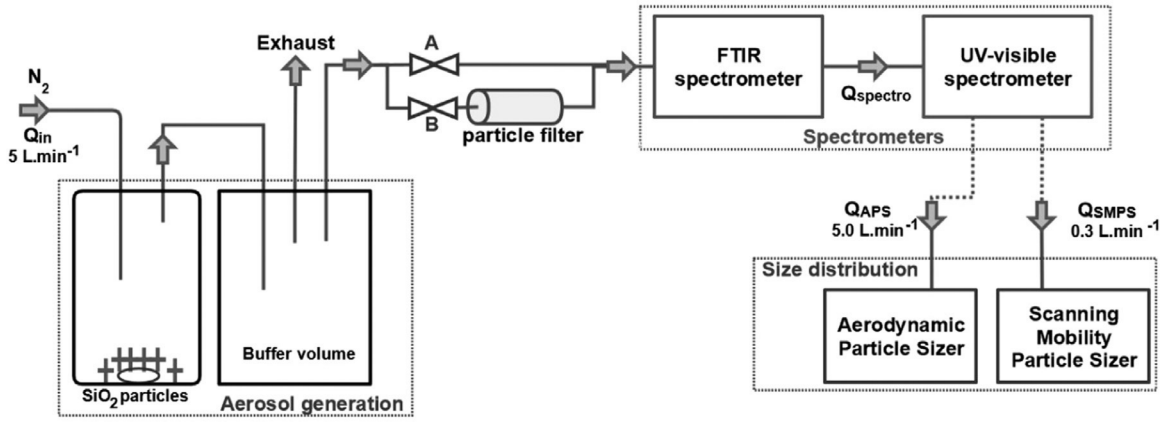

Fig. 1. Experimental set-up composed by an aerosol generation system, a FTIR spectrometer, an UV-visible spectrometer and, depending on the particle size, either a Scanning Mobility Particle Sizer or an Aerodynamic Particle Sizer spectrometer.