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Light scattering properties of sea-salt aerosol particles inferred from modeling studies and ground-based measurements

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Abstract

Direct climate radiative forcing depends on the aerosol optical depth τ, the single scattering albedo ϖ, and the up-scatter fraction β; these quantities are functions of the refractive index of the particles, their size relative to the incident wavelength, and their shape. Sea-salt aerosols crystallize into cubic shapes or in agglomerates of cubic particles under low relative humidity conditions. The present study investigates the effects of the shape of dried sea-salt particles on the detection of light scattering from the particles. Ground-based measurements of scattering and backscattering coefficients have been performed with an integrating nephelometer instrument for a wavelength $\lambda = 0.55\,\mu m$. The measurements are compared to two models: the Mie theory assuming a spherical shape for the particles and the Discrete Dipole Approximation (DDA) model for the hypothesis of cubic shape of the sea-salt aerosols. The comparison is made accurately by taking into account the actual range of the scattering angles measured by the nephelometer in both models that is from 7° to 170° for the scattering coefficient and from 90° to 170° for the backscattering coefficient. Modeled scattering and backscattering coefficients increase for nonspherical particles compared to spherical shape of particles with diameter larger than about $1\,\mu m$. However, the comparison of the modeling results with the measurements gives best agreement for particles diameter less than about $1\,\mu m$. The size distribution of the particles is measured with two instruments with different size bins: an electrical low-pressure impactor (ELPI) and an aerodynamic particle sizer (APS). It is found that the size of the bins of the instruments to determine the number concentration of the particles in accordance with their diameter is critical in the comparison of measurements with modeling.

Keywords: Scattering and backscattering coefficients; Ground-based measurements; Modeled cubic sea-salts

1. Introduction

Atmospheric aerosols present the greatest uncertainties in estimation of the global radiative forcing (Intergovernmental Panel on Climate Change, IPCC report, 2001). Natural and anthropogenic aerosols affect the Earth's climate directly through absorbing and scattering the radiation of the solar spectrum and indirectly through modification of cloud microphysical properties [1]. In situ measurements of the chemical composition of aerosols and the spatial variation of their size distribution are needed, with the knowledge of the shape of the particles to infer accurately the optical properties of aerosols particles [1,2]. The optical quantities directly

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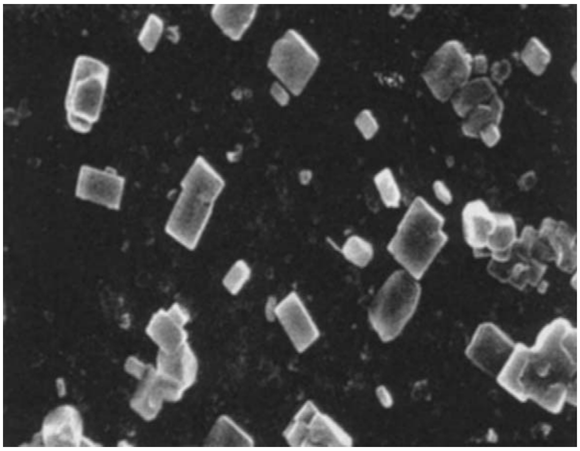

Fig. 1. Scanning electron photographs of dried sea-salt particles for marine air conditions collected on filters at Mace Head on the west coast of Ireland [9]. The width of the picture represents 68 μm.

involved in the radiative forcing equation are the aerosol optical depth τ, the single scattering albedo ω_0 and the up-scatter fraction β. The optical depth is related only to the scattering coefficient σ_{sp} for non-absorbing particles, and the up-scatter fraction is equal to the backscattering coefficient σ_{bsp} for solar zenith angles of 0° [1].

Sea-salt aerosol particles in the maritime environment are usually modeled with a spherical shape [3–5] due to their hygroscopic nature [6–9]. However, in dry conditions as encountered during aerosol sampling, sea-salt aerosols can exhibit a cubic shape or can be found in agglomerates of cubic particles as shown in Fig. 1. Models using nonspherical particles are usually applied only to dust-like aerosols while only few studies investigate nonspherical sea-salt aerosols [10–12]. Moreover, ellipsoidal-shaped particles are usually considered for these nonspherical particles and *T*-matrix calculations are applied [4,13,14]. Ground based measurements of the scattering and backscattering coefficients with size distribution measurements at the Global Atmosphere Watch (GAW) atmospheric research station in Mace Head, located near Carna, at the west coast of Ireland (53°19′N, 9°54′W) and at the eastern edge of the North Atlantic Ocean give insights into the physical and optical properties of marine aerosols. The experimental setup of all the measurements is indoors allowing the consideration of dried particles inside the nephelometer whose relative humidity never exceeds 50% due to internal heating of the instruments. This occurrence offers a possibility of studying optical properties of dried sea-salt aerosols whose ground-based detection is investigated in this work. Section 2.1 is a description of the experimental set-up during a measurement campaign at Mace Head from the 23rd of April to the 28th of April 2002. Meteorological parameters of wind speed and wind direction were also measured at Mace Head. In Section 2.2, the modeling approaches are briefly described as already detailed in [15]. Section 3 contains a detailed study of the effect of the width of the size bins of the sizing instruments in the modeling for a wavelength $\lambda = 0.55\,\mu m$. Comparison of the scattering and backscattering measurements using an integrating nephelometer with predicted values based on calculation of the scattering parameters using Mie [16] and Discrete Dipole Approximation (DDA) formulation [17], for the spherical and cubic assumptions of the shape of the particles, respectively, and applied to the measured aerosol size distribution is presented in Section 4 for a wavelength $\lambda = 0.55\,\mu m$.

2. Methods

2.1. Experimental

2.1.1. Aerosol scattering and backscattering coefficient

The measurements reported here cover the period April 23–28, 2002. The total aerosol scattering coefficient (σ_{sp}) as well as the hemispheric backscattering coefficient (σ_{bsp}) is measured with an integrating nephelometer,