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A review of optical measurements at the aerosol and cloud chamber AIDA

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ABSTRACT

This paper provides a survey of recent studies on the optical properties of aerosol and cloud particles that have been conducted at the AIDA facility of Forschungszentrum Karlsruhe (Aerosol Interactions and Dynamics in the Atmosphere). Reflecting the broad accessible temperature range of the AIDA chamber which extends from ambient temperature down to 183 K, the investigations feature a broad diversity of research topics, such as the wavelength-dependence of the specific absorption cross sections of soot and mineral dust aerosols at room temperature, depolarization and infrared extinction measurements of ice crystal clouds generated at temperatures below 235 K, and the optical properties of polar stratospheric cloud constituents whose formation was studied in chamber experiments at temperatures well below 200 K. After reviewing the AIDA research activity of the past decade and introducing the optical instrumentation of the AIDA facility, this paper presents illustrative examples of ongoing and already published work on optical measurements of soot aerosols, mineral dust particles, and ice crystal clouds.

1. Introduction

The AIDA aerosol and cloud chamber of Forschungszentrum Karlsruhe (Aerosol Interactions and Dynamics in the Atmosphere) has been established during the past decade as a unique experimental facility to study the optical properties of complex aerosol particles and to investigate the formation of cloud particles under realistic atmospheric conditions [1]. As the AIDA aerosol vessel can be operated over a broad temperature range from ambient down to 183 K, the research activity includes the formation and optical characterization of supercooled liquid water clouds, ice crystal clouds (cirrus), and polar stratospheric clouds (PSCs). Apart from its excellent temperature control, another unique feature of the AIDA chamber is its huge size of 84.3 m^3, thereby enabling experiments on typical tropospheric aerosol life times of several days.

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As a prerequisite for the cloud formation experiments, the AIDA chamber can be evacuated with two vacuum pumps down to a final pressure of about 0.01 hPa. The mechanical pumps can be operated at variable pumping speeds, allowing for controlled expansion cooling experiments which mimic the adiabatic expansion cooling of rising air parcels in the atmosphere. The formation of supercooled liquid water and/or ice clouds is triggered when the relative humidity inside the vessel has exceeded a threshold value whose magnitude depends on the type of the pre-added seed aerosol particles and the temperature.

In the present contribution, we want to review the AIDA research activities on optical measurements of aerosol and cloud particles. In Section 2, we will present a brief summary of recent journal publications using AIDA and draw attention to the benefits of performing these experiments in the AIDA chamber. Section 3 will describe the optical measurement techniques and numerical models which are currently employed in the AIDA investigations, followed by a detailed discussion of selected optical measurements in Section 4.

2. AIDA research on optical properties of aerosol and cloud particles—an overview

2.1. Aerosol optics

The first investigations with AIDA focused on the optical properties of airborne soot particles [2,3], combining broadband extinction measurements in the 230–1000 nm wavelength range [4] and scattering measurements with a three-color integrating nephelometer to derive accurate extinction, scattering, and absorption coefficients which are needed to assess the influence of soot aerosol on the atmospheric radiation budget. The experimental data were compared to model calculations with the aerosol behavior code COSIMA (*Co*mputer *Sim*ulation of *A*erosols) which simulates the dynamics and optical properties of agglomerate aerosols, employing Rayleigh–Debye–Gans (RDG) theory to model light absorption and scattering [5]. Due to the large volume of the AIDA chamber, the residence time of the soot aerosols extended up to several days. Therefore, not only the optical properties of freshly emitted soot aerosols, but also their change due to aging processes like coagulation, could be investigated. The experiments were conducted for soot particles of different morphology and chemical composition. In many laboratory studies on soot aerosols, an artificial soot sample from a commercial spark discharge generator (GfG1000, Palas) was used as surrogate for combustion soot. In the AIDA chamber, also the optical properties of soot particles emitted from a VW Diesel engine, being available as part of the infrastructure of Forschungszentrum Karlsruhe, have been investigated. This study revealed that the mass specific extinction cross section of Diesel soot is by a factor of two higher than that of spark discharge soot at visible wavelengths [3]. More recently, a new flame soot generator (Combustion Aerosol Standard, CAST, Jing-CAST Technologies) was introduced which allows for generating carbonaceous aerosols with different contents of organic carbon (OC). It could be shown that an increasing amount of OC material, presumably consisting of condensed polyaromatic molecules, leads to a strong decrease in the absorption cross sections in combination with a strong increase in the absorption Ångström exponent [6].

After having determined the absorption and scattering coefficients of pure soot particles, further AIDA studies addressed the influence of non-absorbing coating layers of organic and inorganic materials on the optical properties of soot aerosols [7,8]. Such coating layers might grow on the soot particles during their atmospheric life time through aging processes like heterogeneous coagulation and condensation. Several modeling calculations have predicted that the heating potential of atmospheric black carbon would strongly increase in an internal mixture with non-absorbing material [9,10]. In these studies, the enhancement factors for the increase in the specific absorption cross sections between uncoated and coated soot particles were found to be in the range from 2.5 to 4, depending on the layer thickness and the diameter of the soot core. For the first time, this predicted absorption amplification of internally mixed soot particles has been evidenced experimentally in AIDA chamber studies where pure soot aerosols were coated *in situ* with non-absorbing organic material from the ozonolysis of α-pinene [8]. As a result of the long aerosol residence time in the chamber, this coating step could be performed repeatedly, thereby successively increasing the thickness of the coating layer. A Mie model for concentrically coated spheres was employed to reproduce the measured amplification factors. Details of this analysis, as well as selected results from the optical characterization of pure soot aerosols, are presented in Section 4.1.

Another important class of aerosols with a substantial contribution to the overall atmospheric aerosol load and which may alter the earth's radiative budget by absorption and scattering of light at visible and infrared wavelengths is mineral dust. In recent AIDA investigations, the wavelength-dependence of the specific absorption cross sections of a variety of Saharan mineral dust samples has been measured [11]. Instead of deriving the absorption cross sections indirectly by the difference method from simultaneous measurements of the aerosol extinction and total scattering coefficients, the absorption coefficients were directly measured with a novel multi-wavelength photo-acoustic absorption spectrometer at wavelengths of 1064, 532, 355, and 266 nm. The optical measurements were supplemented by chemical and mineralogical analyses of the dust samples, with particular emphasis on quantifying the amount of the iron oxides hematite and goethite. The study revealed that just the contribution of these phases significantly increases the absorption cross sections of the mineral dust aerosols at visible and near UV wavelengths. Therefore, ongoing AIDA investigations also focus on the optical characterization of these pure iron oxide minerals like hematite. In contrast to the high diversity of the dust grains in terms of size, shape, and surface structure which is observed for the natural dust probes, hematite samples can be synthesized as a monodispersed ensemble of particles with uniform and well-defined particle shapes like cubes or ellipsoids. This allows