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Inspecting the radiative properties of insoluble impurities stored in ice cores

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In addition to air bubbles and ions, ice cores contain insoluble particles, mainly mineral dust. These particles provide a temporal record of the atmospheric aerosol content of the past, which is key to understanding Earth's energy balance.

In an era of rapid, global, and significant climatic changes, the scientific community is devoting great efforts to the study of the current and past major drivers of these changes. Fully understanding these changes also means studying their trends, determining their periodicity, and possibly assessing the extent to which they can be dealt with or mitigated. In this context, atmospheric aerosols have attracted much attention in the literature, as they have a significant impact on the climate system (Kok et al. 2023). Cloud cover depends primarily on the ability of water molecules to condense or crystallize around condensation nuclei, i.e. aerosols. Cloud cover is a determining factor in planetary albedo and is highly dependent on the aerosol species that populate the atmosphere. In addition, aerosols themselves contribute to the Earth's energy balance by scattering and absorbing solar and terrestrial radiation, thus cooling or warming the climate (Forster et al. 2021). While direct measurements in the atmosphere and remote sensing can be used for information about the present time, measurements of impurities in ice cores can be relied upon to obtain valuable information about the past.

Overview

In the framework of studying the Earth's energy balance and climate, the optical properties of impurities in ice cores give us insight into the radiative properties of aerosol particles, and provide data that can be used for radiative transfer models. Light scattering, absorption, and albedo depend on many characteristics of the particles, including their size, shape, and composition, and are challenging to retrieve without direct measurements. Nonetheless, radiative transfer models still use the spherical shape approximation, which contributes to the uncertainties in the estimate of the impact of aerosols on the Earth's energy balance. This requires some further steps toward the integrated measurement of as many parameters as possible, on an experimental basis. A continuous flow analysis (CFA) system is currently being developed in the EuroCold laboratory in Milan, oriented toward the light-scattering characterization of particles in polar and Alpine cores using single-particle extinction and scattering, an optical particle sizer, and digital holography microscopy. The aim is to characterize dust records and contribute to the monitoring of the fast evolution of climate, which is having a detrimental impact on glaciers, among other consequences.

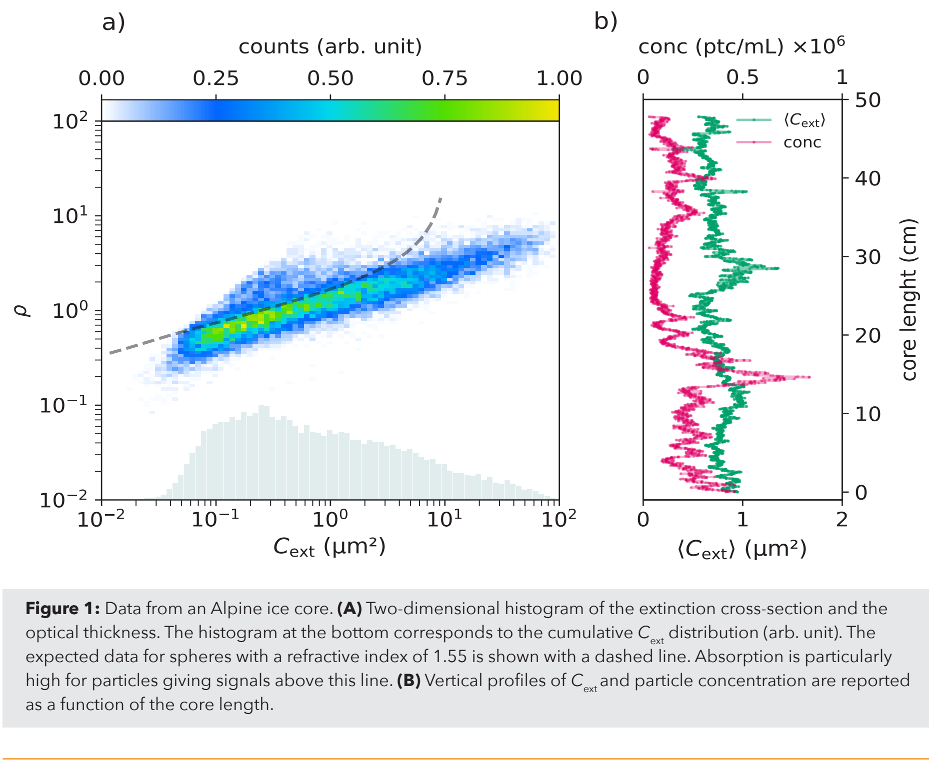

Figure 1: Data from an Alpine ice core. **(A)** Two-dimensional histogram of the extinction cross-section and the optical thickness. The histogram at the bottom corresponds to the cumulative C_{ext} distribution (arb. unit). The expected data for spheres with a refractive index of 1.55 is shown with a dashed line. Absorption is particularly high for particles giving signals above this line. **(B)** Vertical profiles of C_{ext} and particle concentration are reported as a function of the core length.

What to look for with optical-based instruments, in a nutshell

A powerful system for studying ice cores is the CFA of different chemical and microphysical compounds. Significant temporal resolution can be achieved by slowly melting cores from one end to pump meltwater into in-line instruments (Bigler et al. 2011; Erhardt et al. 2019). Here we provide a snapshot of what can be observed with optical-based instruments, i.e. light scattering within the optical spectrum. The radiative properties of a particle can be predominantly traced back to its extinction cross-section (C_{ext}), which has the units of a surface area. This is the area that effectively interacts with solar light, or any other incoming radiation. Similar definitions exist for the fraction of light that is scattered and absorbed and give the scattering and the absorption cross-sections, respectively. These optical cross-sections are determined by the conservation of radiative energy and may have little to do with the geometric cross-section of the particle, which is another parameter that may be of interest in its own right. From the scattering and the extinction cross-sections, we can assess the single-scattering albedo of an aerosol particle and make an educated guess about its refractive index. Other radiative parameters of single particles include the optical thickness and effective polarizability, related to their refractive index, shape, and size (Cremonesi 2020).

Light extinction and forward scattering

In a recent CFA campaign on an alpine ice core, we used the single particle extinction and scattering (SPES) method. With this in-line instrument, in addition to particle concentration, two optical parameters can be measured without calibration on a particle-by-particle basis (Potenza et al. 2015): the extinction cross-section and the optical thickness (C_{ext}, ρ). These parameters tell us how much power the particle removes from the incident light and the phase lag of the wave scattered by the particle (Potenza et al. 2016). As a general rule, a larger extinction cross-section corresponds to larger particles and vice versa; similarly, optically dense particles exhibit correspondingly high optical thickness. Some spikes in the particle

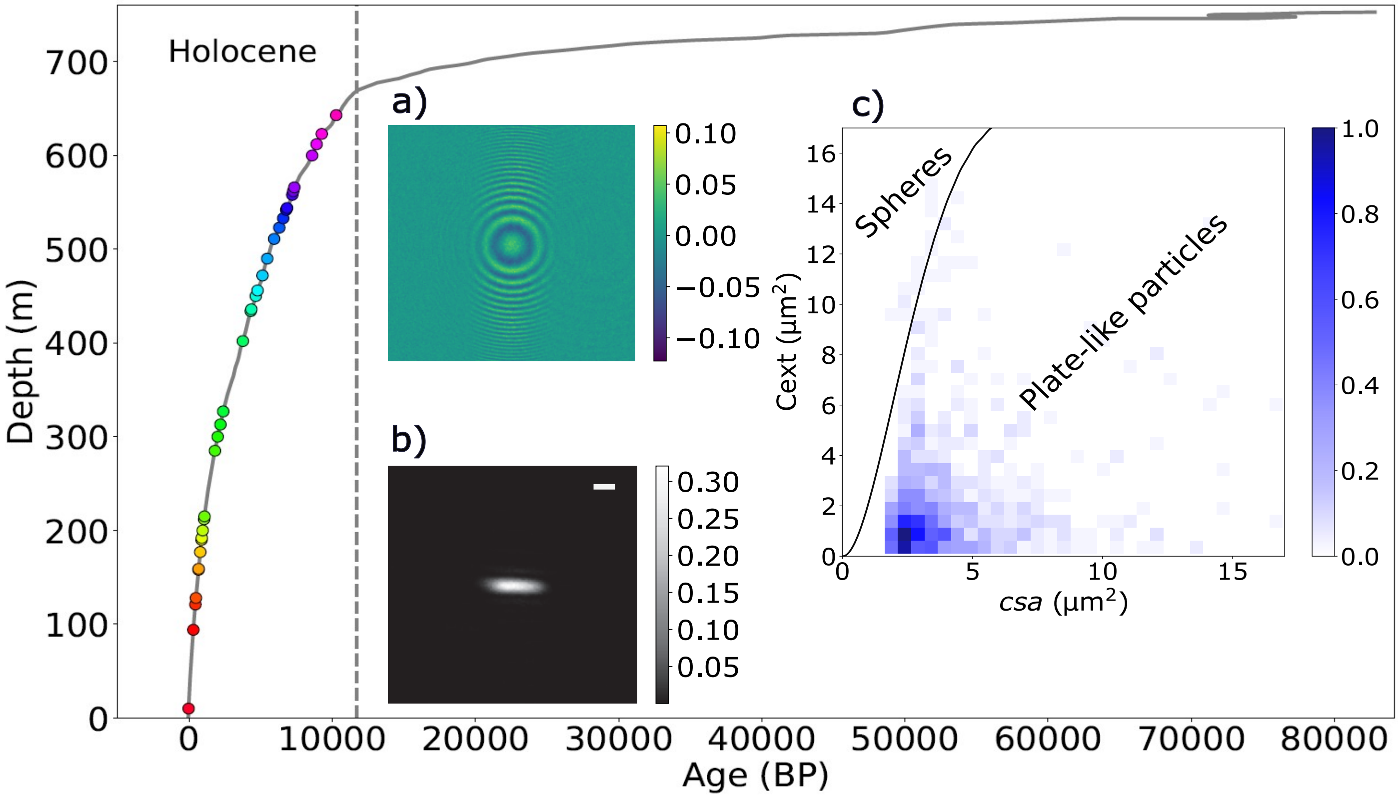

Figure 2: Age-depth relationship at Roosevelt Island ice core (Lee et al. 2020; Winstrup et al. 2019). The samples of Holocene age analyzed with the holographic technique are shown as dots. **(A)** Hologram fringes of a mineral dust particle and **(B)** corresponding reconstructed image. The white bar on the top-right corner is set to 2 μm. **(C)** (*csa*, C_{ext}) plane reported as a two-dimensional histogram and normalized on its maximum (dark blue), from a 313 m-deep sample.

concentration, related to advection events, show a peculiar trend of the combination of C_{ext}, and ρ. Figure 1 shows a snapshot of the optical parameters of a ~15 m deep Alpine ice core from the Adamello glacier (Eastern Alps, Italy). This location is affected by local natural and anthropogenic sources, in addition to the long-range transport of aerosols. Figure 1a shows the cumulative two-dimensional histogram of the extinction cross-section and the optical thickness for all the particles in the ice core. A variety of particle shapes, compositions, and sizes gives a widespread distribution along the two axes (Simonsen et al. 2018). The dashed line corresponds to the expected data for spherical particles with a refractive index of 1.55 (ranging from 0.3 to 2 μm in diameter), which is a threshold for identifying highly absorbing particles. The vertical profiles of C_{ext} and particle concentration are reported as a function of the core length in Figure 2b. Both parameteres depend on the characteristics of the particles and the transport pathways, therefore, C_{ext} and particle concentration do not always covariate.

Characterization by digital holography

Another technique that we integrated into our CFA system is digital holography microscopy, by which we investigate the optical and geometric properties of larger dust particles in the micrometer size range (Berg et al. 2022). As an example, we report an ice-core record from the eastern Ross Sea, as part of the Roosevelt Island Climate Evolution (RICE project, see Bertler et al. 2018; Lee et al. 2020; and Winstrup et al. 2019). In Figure 2, we show the age-depth relationship (gray continuous line), while samples of Holocene age are identified on the timeline by different dots.

With the holographic technique we acquire the so-called hologram patterns (Fig. 2a), i.e. interferometric images where information about the size and optical properties of the particles are encoded. The holographic pattern is then processed numerically in real-time or post-measurement, without the need to check when particles are in the field of view. Moreover, multiple objects can be imaged at different focal planes simultaneously. The result of the reconstructed algorithm is an image of the silhouette of the particle, as shown in Figure 2b. We characterized insoluble particles suspended in meltwater as described in Ravasio et al. (2021; 2022). We obtained the value of C_{ext}, the cross-sectional area (*csa*), and the thickness over diameter ratio (*tdr*) of each particle, as well as the particle count.

The importance of performing both optical and size characterization is shown in Figure 2c. Here, we show the (*csa*, C_{ext}) data from one of the samples (313 m of depth), represented as a two-dimensional histogram and normalized on its maximum, and selecting only isometric-shape particles. We show with a black solid line the expected result for spherical particles from Lorenz–Mie theory (1.4–2.8 μm in diameter, refractive index of 1.55), as reference. We note that most of the data falls below this line and spans a considerable range of both *csa* and C_{ext}. Indeed, many particles are plate-like with *tdr* values between 0.1 and 0.25, which lowers the actual C_{ext} of the particles compared to spheres with the same geometrical cross-section.

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