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Where does the mineral dust in Greenland ice come from?

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The Asian deserts were identified as the primary source of mineral dust in Greenland ice. However, secondary sources may be overlooked when quantifying contributions of different sources in large bulk samples. Single particle studies can help overcome these limitations.

How mineral dust is archived in Greenland ice

Various types of aerosols are produced in different environments on Earth. For example, volcanoes emit aerosols and precursor gases during their eruptions, oceans generate sea-salt aerosols, and deserts produce mineral dust aerosols (Fig. 1). Among these aerosols, mineral dust plays a crucial role in aerosol composition.

The emission of soil-derived dust into the atmosphere occurs when the wind velocity is high enough to disperse the dust. The threshold wind velocity depends on the particle size, mass, and soil moisture content in the source region. In quantitative terms, the flux of dust generated by the wind is nonlinearly dependent on wind speed (Marticorena and Bergametti 1995). Reconstructing the strength of dust emission from measured ice-core concentrations can help us understand climate change in the source regions.

Many of the major dust sources are located in arid regions of the extratropics due to their dry and windy conditions. This restricted location of dust sources implies that dust must be transported over long distances by atmospheric circulation to reach Greenland. The atmospheric lifetime of dust depends on its size, mass, and the degree of washout by precipitation en route. Thus, climatic changes, such as alterations in atmospheric circulation patterns and precipitation, impact the transport efficiency of dust to Greenland. Long-range transported dust is deposited onto the surface of the Greenland Ice Sheet through both wet and dry processes. Size-dependent dry deposition of aerosols always occurs, whereas wet deposition can only occur if a precipitation event happens on the ice sheet. In summary, for efficient dust export to the Greenland Ice Sheet, dry and windy conditions at the source region and little precipitation en route are necessary (which typically implies transport at high altitudes). Ideally, precipitation over the ice sheet should occur at the time the dust plume reaches Greenland.

Different approaches to determine where the mineral dust came from

Various methods have been used to investigate the sources of dust in Greenland ice. Satellite imagery has been used to observe the transport of dust from its sources to Greenland directly. For example, Prospero et al. (2002) used satellite pictures to identify the desert belt extending from North Africa to Eastern Asia as the primary source of dust in the Northern Hemisphere. Since satellite imagery can only be obtained from cloud-free scanned regions and only for the last few decades, it falls short of providing a comprehensive understanding of the influence of past climate on the sources of dust in Greenland ice.

Atmospheric circulation modeling also enables the exploration of dust transport also for past and future conditions. Kahl et al. (1997) modeled back trajectories of air masses from Summit Station, Greenland. They found that approximately 60% of all winter trajectories to Summit, Greenland, were connected to a cluster that allows for aerosol transport from the East Asian desert dust source region, whereas a cluster connected to the North American dust source region was dominant (46%) in summer. However, this approach has limitations in accurately determining the source apportionment when atmospheric conditions are poorly defined in the model. This is especially true for past climate conditions that cannot be validated against meteorological observations, compromising the accuracy of the model's results in such cases.

Direct analysis of the dust deposited onto the Greenland Ice Sheet is another effective way to determine its origin. Mineralogy, elemental composition, and isotopic composition are widely used (often in combination) for source identification. Mineralogy can help identify the type of soil using crystal information. For example,

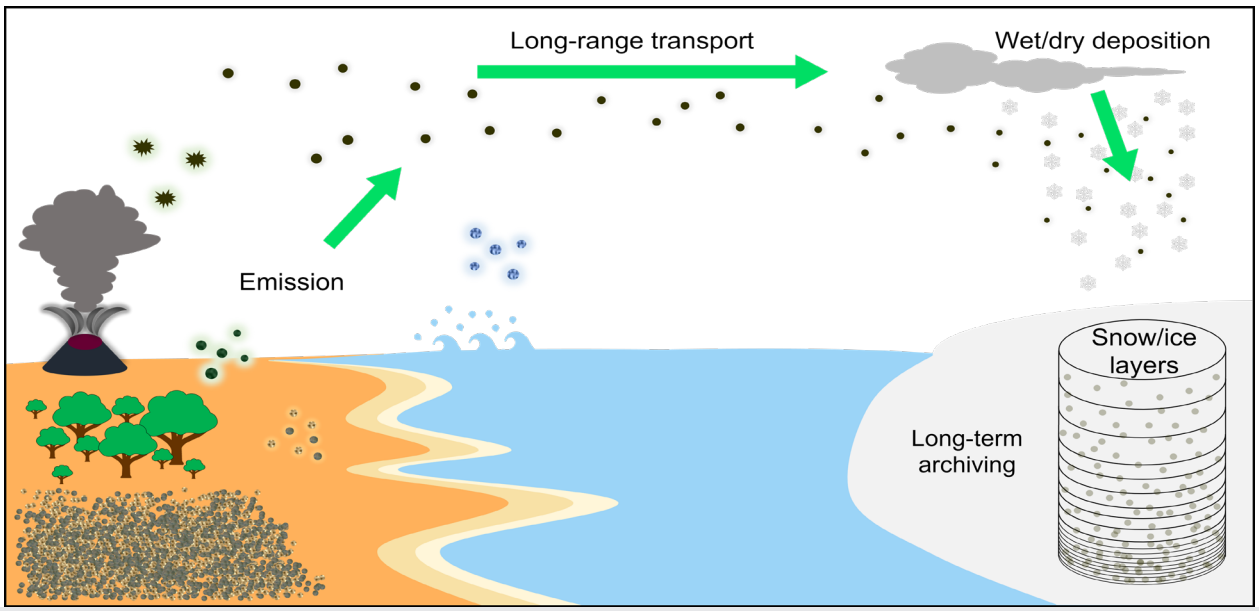

Figure 1: Life cycle of aerosol components archived in ice cores. Aerosol species are emitted from various sources, such as deserts, vegetation, volcanic activities, and oceans, and then transported over long distances by atmospheric circulation. Aerosols (such as dust) are wet- and dry-deposited en route and onto the ice sheet, and preserved in ice layers.

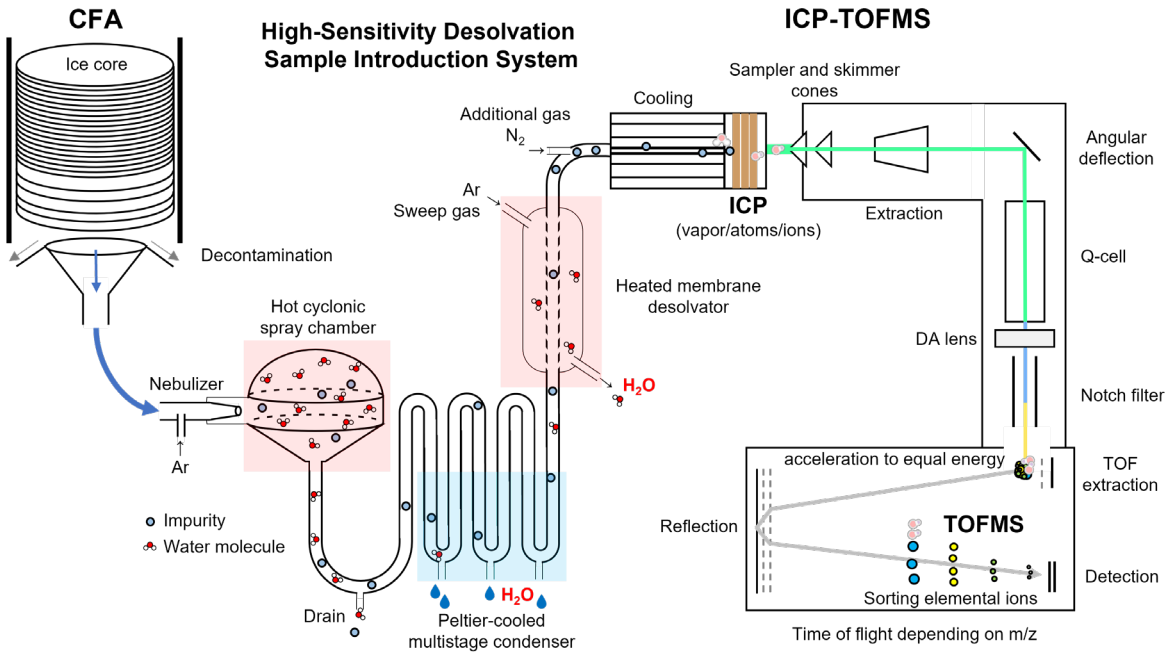

Figure 2: Schematic diagram of CFA-sp-ICP-TOFMS with a high-sensitivity desolvation sample introduction system. The meltwater sample from the CFA flows into the desolvation unit and undergoes a drying process through heating, condensation, and membrane diffusion during sample introduction. The ions reaching the TOFMS can be analyzed over the 23 to 254 mass range, and due to the high time-resolution of sp-ICP-TOFMS, individual dust particles can be detected, in addition to the dissolved background.

Maggi (1997) characterized the mineralogical composition of dust from glacial and interglacial periods in the Greenland Ice Core Project ice core and compared it with dust samples from low- and high-latitude source regions. The results suggest that warm periods are characterized by a higher contribution from chemical weathering in low-latitude source regions compared to more mechanical weathering in mid-latitudes during cold periods.

The elemental and isotopic composition of dust can serve as unique fingerprints for identifying dust sources, and are typically measured using mass spectrometry. Bory et al. (2003) used the mineralogy and isotopic signatures of the dust in snow-surface samples to identify present-day dust sources for Greenland. The mineralogical signatures found in all the dust samples from Greenland snow pits, characterized by low kaolinite to chlorite ratios, provide clear evidence that the sources are dominated by East Asian desert areas. This finding rules out North America and Northern Africa as major sources (Bory et al. 2003). The isotopic signatures of the samples not only agreed with the East Asian origin determined by their mineralogy, but also showed pronounced seasonal variation in sources within Asian inland regions. The Taklamakan Desert contributes substantially to the dust transported to northern Greenland during the maximum dust concentration in spring, while the main source of dust for autumn is shifted to the Tengger and Mu Us deserts (Bory et al. 2003).

However, some recent studies have suggested that the sources of dust may be more complex than previously thought, and may vary in response to abrupt climatic changes, such as Dansgaard-Oeschger events (Han et al. 2018; Újvári et al. 2022).

Single particle analysis: A new approach to dust characterization

Most of the aforementioned chemical methods which describe bulk parameters of large ice samples after melting and pretreatment, like acid digestion, have inherent limitations. This implies that (i) chemical characterization by these methods cannot distinguish between particulate and dissolved dust tracers and (ii) bulk samples average a mixture of many individual dust particles which may originate from different source regions.
To overcome these limitations, we follow a new approach that takes advantage of the millisecond time resolution of single-particle inductively coupled plasma time-of-flight mass spectrometry (sp-ICP-TOFMS) (Fig. 2). This novel approach enables the detection and elemental characterization of single dust particles in addition to the dissolved impurities in a meltwater stream provided by the continuous flow analysis (CFA) developed at the University of Bern (Kaufmann et al. 2008). First results on the elemental composition of single dust particles from Greenland meltwater samples support Asian deserts as the main origin of the long-range transported particles to Greenland (Erhardt et al. 2019).

Conclusions and outlook

The origin of dust in Greenland has been determined through various approaches, such as remote sensing, atmospheric circulation modeling, and direct measurements of dust in ice cores. Many previous studies, using diverse approaches, agree that Asian deserts are the primary sources of dust transported to central Greenland. However, we stress the importance of high-resolution single dust particle analysis (Erhardt et al. 2019), in addition to bulk analysis. The sp-ICP-TOFMS enables the investigation of elemental composition of individual dust particles in samples. This allows the precise characterization of the fingerprints of individual soil-derived aerosols from both source samples and Greenland ice, including the internal variation within each source region and source mixing.

Within the DEEPICE project, we are furthering these studies by improving our analysis system with a desolvation sample introduction, which dries the sample stream before it enters the plasma (Fig. 2). Drying the sample helps to reduce loss of analytes during the introduction, and minimizes spectral interferences caused by the input of water into the system. Furthermore, we are developing automated data analysis routines to characterize different clusters of dust origin. However, diverse methods should be used complementarily to improve our understanding of the origin of dust in Greenland ice under different climatic conditions.

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