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Radionuclide-decay dating in ice cores

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Age estimates using the radioactive decay of radionuclides in ice cores have the potential to verify and extend existing age scales. The method is currently limited by unexplained variations in radionuclide concentrations and the large masses of ice needed for their measurement.

Radionuclide production

Earth is constantly bombarded by a flux of cosmic rays, which consist mainly of protons and alpha particles, traveling near the speed of light. In our atmosphere, they collide with gases and set off a cascade of nuclear reactions that result in the production of a range of radionuclides, including ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, and ^{81}Kr. Because the atmospheric composition is dominated by nitrogen (^{14}N) and oxygen (^{16}O), most reactions produce radionuclides with a relative isotopic mass below 16; heavier radionuclides are much less abundant (Beer et al. 2012; Poluianov et al. 2016).

Radioactive-decay dating is best known from radiocarbon, which is however, not suitable for most ice-core dating due to its comparably short half-life of 5.73 kyr (Beer et al. 2012). Several other radionuclides have much longer half-lives (Fig. 1a), but face two major challenges. Firstly, the low abundance of most radionuclides translates into low concentrations in ice, necessitating large sample masses for their measurement, as shown in Figure 1b. Secondly, the production of all radionuclides varies over time because the cosmic-ray flux on Earth is not constant. It is subject to modulations from the varying magnetic fields of the Sun and the Earth, since protons and alpha particles are charged particles.

The advantage of radionuclide dating is that it can produce absolute age estimates, independent of other age scales and time markers. Therefore, it can also be applied to ice with a disturbed stratigraphy, making it a valuable tool for the dating of deep ice cores.

^{36}Cl/^{10}Be

With a half-life of $t_{1/2}$ = 301 kyr, ^{36}Cl is well suited for dating ice with ages up to 1.5 Myr, which is what the Beyond EPICA Oldest Ice Core project aims to retrieve. For a sample with an age of 1 Myr, about 1 kg of ice is needed (Fig. 1b). Variations in the production signal affect ^{36}Cl and ^{10}Be similarly, so taking the ^{36}Cl/^{10}Be ratio theoretically removes production-related variations and isolates the decay signal (Delmas et al. 2004). However, measurements from the GRIP (Yiou et al. 1997) and Dome Fuji ice cores reveal significant variations of the ^{36}Cl/^{10}Be ratio unrelated to radioactive decay (Fig. 2a; Kanzawa et al. 2021; ca 7450–7350 yr BP). Most likely, these variations are linked to the different chemical and physical properties of the two radionuclides, which may lead to differences in their atmospheric lifetimes, transport pathways and deposition mechanisms (Beer et al. 2012).

At low accumulation sites, ^{36}Cl can be lost from the ice by reacting with acidic species to form volatile hydrogen chloride (HCl) after deposition, further complicating the interpretation of the measured ^{36}Cl/^{10}Be ratio (Delmas et al. 2004). Ice from glacial periods, however, has the potential to be unaffected by this because the glacial atmosphere contains higher concentrations of alkaline dust, which can neutralize acidic species (Röthlisberger et al. 2003; Wolff et al. 2010). Another uncertainty for age estimates is introduced by the accumulation of ^{10}Be at grain boundaries, where it can be adsorbed on to dust particles in deep ice, creating local concentration spikes and disturbing the stratigraphic signal (Baumgartner et al. 1997; Raisbeck et al. 2006).

Nonetheless, the ^{36}Cl/^{10}Be ratio has been successfully used to date the deepest section of ice from the Dye3 and GRIP cores, where the age estimates agreed with other dating methods (Willerslev et al. 2007).

^{26}Al/^{10}Be

The advantage of using ^{26}Al instead of ^{36}Cl is that it quickly attaches to aerosol particles, similar to ^{10}Be. The transport for both radionuclides should, therefore, be identical, leading to less variations in the ^{26}Al/^{10}Be ratio. Indeed, measurements of atmospheric air around the globe yielded the same ^{26}Al/^{10}Be ratio with deviations of no more than 5% and similar values for measurements in firn from several locations in Antarctica (Auer et al. 2009).

The disadvantage of using ^{26}Al is its very low production rate, which is about 300 times lower than that of ^{10}Be, necessitating at least 7–14 kg of ice for a single measurement of 1-Myr-old ice (Auer et al. 2009; Fig. 1b).

In a pilot study, Auer et al. (2009) measured the ^{26}Al/^{10}Be ratio in the deep, undated section of the EDML ice core (older than 150 kyr) and found that its values varied strongly between samples and were on average 50% higher than in samples from the modern-day atmosphere and Antarctic firn, even though

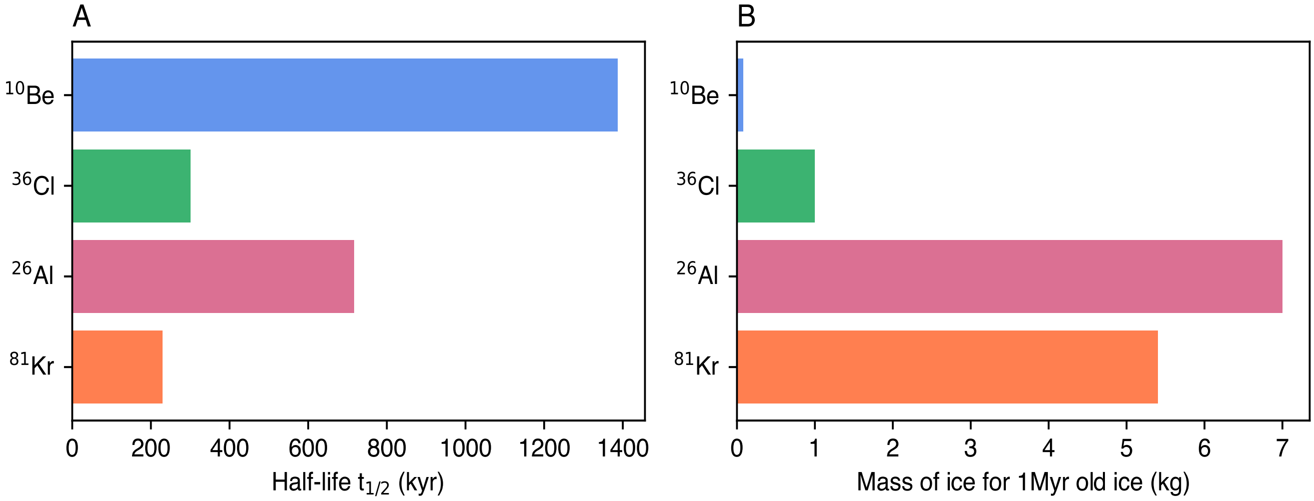

Figure 1: (A) Comparison of the half-lives of different radionuclides and **(B)** the mass of 1-Myr-old ice needed for measuring their respective concentrations.

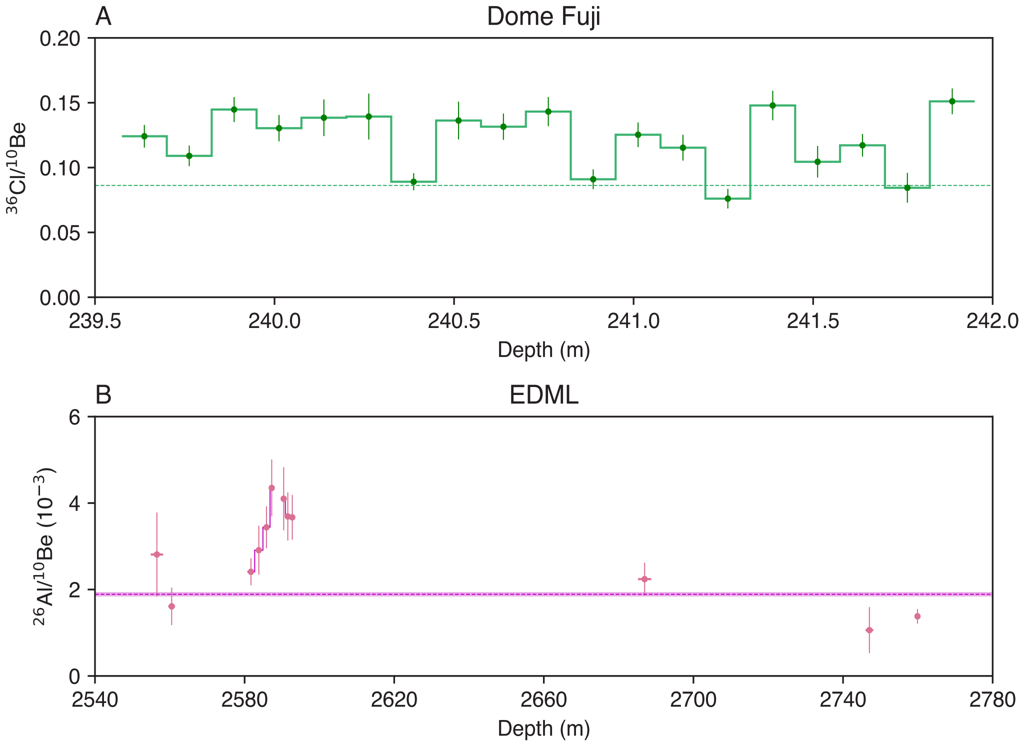

Figure 2: Variations of **(A)** the ^{36}Cl/^{10}Be ratio in the Dome Fuji ice core with the theoretical production rate value of 0.0862 shown as a dashed horizontal green line and **(B)** the ^{26}Al/^{10}Be ratio in the EDML ice core with the modern-day mean atmosphere value of 1.89 ± 0.05 x 10-3 shown as a horizontal magenta line. Data from Kanzawa et al. (2021) and Auer et al. (2009), respectively. Vertical bars indicate analytical errors.

the decay of ^{26}Al ($t_{1/2}$ = 717 kyr) should lead to lower values (Fig. 2b). The authors concluded that recrystallization and high pressure may result in local concentration enhancements at the bottom of the EDML core. However, these alterations and their effects on ^{26}Al and ^{10}Be are not understood, and the ^{26}Al/^{10}Be ratio, therefore, appears to suffer from a similar weakness as the ^{36}Cl/^{10}Be ratio: although changes in the production signal are theoretically removed, the ratio exhibits unexplained variations.

Using only the two deepest measurements, Auer et al. (2009) arrived at an age estimate of 670 kyr with an uncertainty of almost 40% for the deepest EDML sample, which is approximately the minimum possible uncertainty of ^{26}Al/^{10}Be dating with 7 kg of ice, mainly due to the low measurement efficiency.

^{81}Kr

Contrary to the other radionuclides discussed so far, ^{81}Kr is a noble gas, which is largely inert and remains in the atmosphere for most of its lifetime, resulting in a globally well-mixed atmospheric ^{81}Kr/Kr ratio.

The approach for taking production variabilities into account is also different for ^{81}Kr. Instead of using a second radionuclide to correct the signal, a reconstruction of geomagnetic field intensities is used to calculate the theoretical ^{81}Kr/Kr ratio of the last 2 Myr (Zappala et al. 2020). This introduces uncertainties from the past cosmic-ray flux, the absorption cross sections of krypton, and the half-life of ^{81}Kr, t1$_{/2}$ = 229 ± 11 kyr. Nonetheless, the calculation of the theoretical present-day ratio agrees with measurements of modern air (Zappala et al. 2020).

First measurements of ^{81}Kr in three deep samples of the undated Talos Dome ice-core section yielded age estimates with 9–16% uncertainty, and indicated a disturbed stratigraphy, because the deepest sample had a younger ^{81}Kr age than the second deepest (Crotti et al. 2021).

Due to the low abundance of stable krypton (the target element for ^{81}Kr production in the atmosphere) the production rate of ^{81}Kr is even lower than that of ^{26}Al. Measurements with less than 10 kg of ice became feasible only recently (Crotti et al. 2021) and current improvements aim to reduce the required sample mass to just 1 kg of 1-Myr-old ice (Ritterbusch et al. 2022).

Outlook

Several radionuclides have the potential to assist conventional dating of ice cores, especially in the deepest section, where the stratigraphy may be disturbed.

Two main issues complicate the use of radionuclide dating: uncertainty and required sample mass. For the ^{36}Cl/^{10}Be and ^{26}Al/^{10}Be ratios, variations occur over time and are not well understood, while ^{81}Kr suffers from uncertainties connected to the calculation of the historic ^{81}Kr/Kr ratio and its half-life. Because radioactive decay is used for dating, the required sample mass increases exponentially with age. To measure a radionuclide with consistent precision, the required sample mass doubles for each half-life.

Nonetheless, measurement techniques are constantly improving to reduce the required sample size, making radionuclide dating a more viable solution for dating old ice. Simultaneously, research aimed at a better understanding of climatic influences and post-depositional effects on the ^{36}Cl/^{10}Be and ^{26}Al/^{10}Be radionuclide ratios, as well as improved calculations of the historic ^{81}Kr/Kr ratio, are expected to reduce the uncertainties of these three radionuclide dating methods.

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