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Toward a radiometric ice clock: uranium ages of the Dome C ice core

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ABSTRACT

Ice sheets and deep ice cores have yielded a wealth of paleoclimate information based on continuous dating methods while independent radiometric ages of ice have remained elusive. Here we demonstrate the application of ($^{234}U/^{238}U$) measurements to dating the EPICA Dome C ice core based on the accumulation of ^{234}U in the ice matrix from recoil during ^{238}U decay out of dust bound within the ice. Measured ($^{234}U/^{238}U$) activity ratios within the ice generally increase with depth while the surface areas of the dust grains are relatively constant. Using a newly designed device for measuring surface area for small samples, we were able to estimate reliably the recoil efficiency of nuclides from dust to ice. The resulting calculated radiometric ages range between 80 ka and 870 ka. Measured samples in the upper 3100 m fall on the previously published age-depth profile. Samples in the 3200–3255 m section show a marked change from 723–870 ka to 85 ka indicating homogenization of the deep ice prior to resetting of the ($^{234}U/^{238}U$) age in the basal layers. The mechanism for homogenization is likely enhanced lateral ice flow due to high basal melting and geothermal heat flux.

1. Introduction

Ice cores drilled from the ice sheets in Greenland and Antarctica have proven to be outstanding archives of atmospheric composition over the past hundreds of thousands of years (Jouzel et al., 1995; Petit et al., 1999; Spahni et al., 2005). They are a unique source of information for documenting the fine-scale changes in the relationship between the concentration of greenhouse gases with other climate indicators such as dust transport and sea ice extent (Delmonte et al., 2004; Wolff et al., 2006). These climate markers found within the ice can also be used as tracers wherein their locations record large scale ice motion, which provides information about the responses of ice sheets to climate change (Clarke et al., 2005). Making full use of these records to investigate global climate and environmental changes requires absolute dating of the ice. Current, precise methods rely on a continuous record (Parrenin et al., 2007) or rare external age markers such as tephra deposits (Narcisi et al., 2006) or enhanced ^{10}Be deposition (Raisbeck et al., 2006). Here, we show that uranium series disequilibria can be used as an independent absolute radiometric dating method for the Dome C ice core based on ^{238}U decay recoil products accumulating from dust trapped within the ice. First we establish that this method is useful and reliable and, second, we apply the dating technique to the deepest section of the ice core where the climate-age record is unreliable (Dreyfus et al., 2007; Jouzel et al., 2007).

2. Uranium series dating of ice

Recoil accumulation as a radiometric dating method for ice was first proposed by Fireman (1986) and more recently revisited by Goldstein et al. (2004), but a combination of analytical limitations, disturbed ice, and incomplete characterization of the materials hindered their ability to produce accurate, precise ages. The principles of the dating scheme are illustrated in Fig. 1, which shows a schematic of the particulate-ice, parent-daughter system for ice. The dust grains bound in the ice contain trace amounts of ^{238}U that alpha decays to ^{234}Th with a half-life of 4.4683 Ga, the energy of which results in the daughter nuclide moving a measurable distance, 20–25 nm (Hashimoto et al., 1985; Fleischer, 2003) in crustal minerals, with a subsequent rapid gamma decay to ^{234}U [Fig. 1, arrows a]. Ice also contains trace amounts of uranium, both ^{238}U and ^{234}U [Fig. 1(b) and (c)]. The concentration of ^{234}U in the ice depends on the initial concentration of uranium in the ice, the

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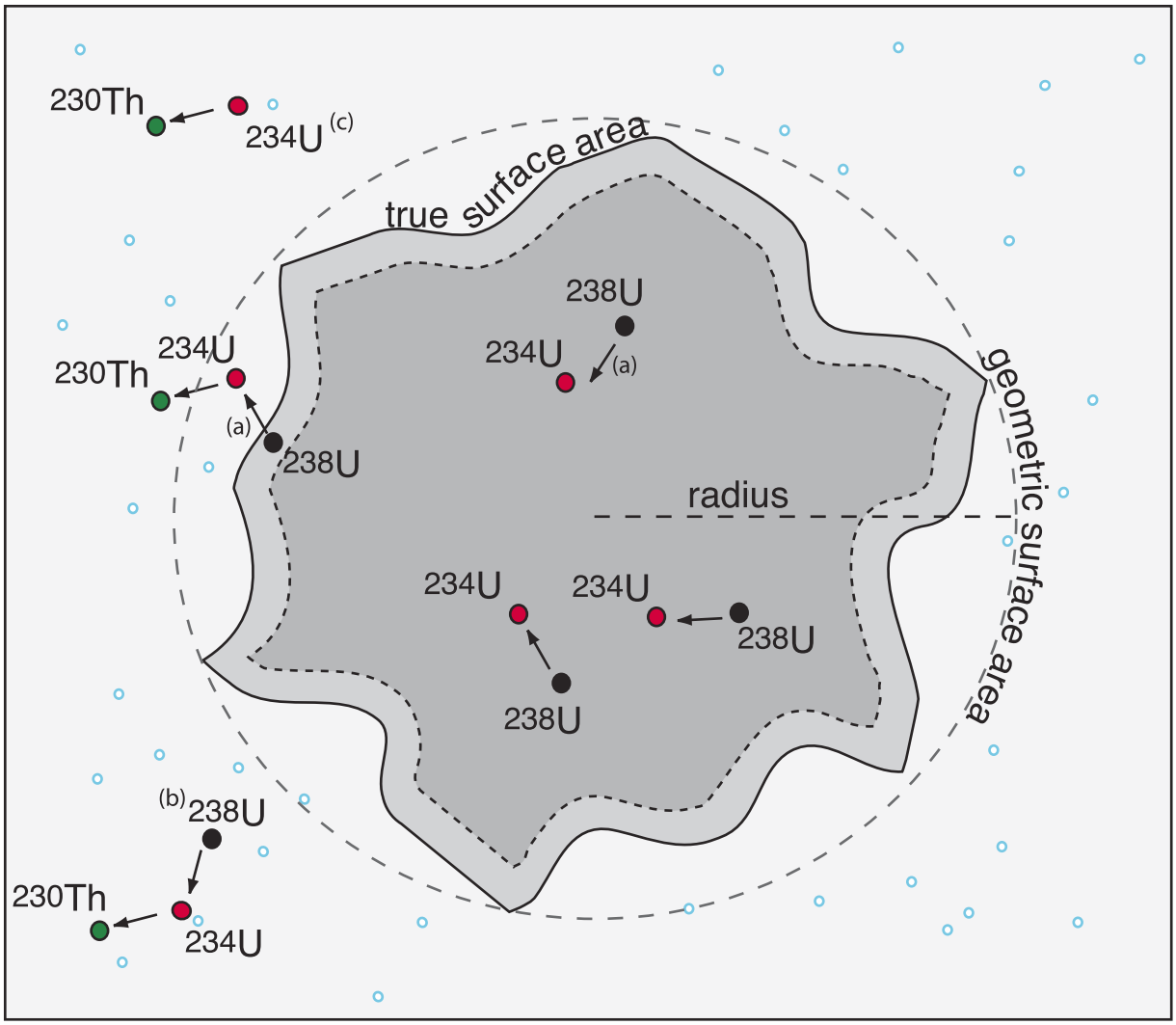

Fig. 1. Schematic of recoil accumulation of ^{234}U daughters in ice from dust.

concentration of ^{238}U in the dust, the fraction of daughters, f, that recoil out of the dust grain into the ice (e.g., in Fig. 1, $f = 0.125$), and the decay of ^{234}U to ^{230}Th. This accumulation-decay defines the timescale over which the radiometric method is applicable, and ages can be calculated using the age equation:

$$t = -\left(\frac{1}{\lambda_{234}}\right)\ln\left(\frac{[U_{234}]_{ice} - f[U_{238}]_{dust} - [U_{238}]_{ice}}{[U_{234}]^{in}_{ice} - f[U_{238}]_{dust} - [U_{238}]_{ice}}\right) \quad (1)$$

where square brackets denote the activity of the respective nuclides (see Appendix for source of equation).

In Antarctica, the two primary sources of trace chemicals within the ice are continental and marine aerosols (Wolff et al., 2010); in the deepest sections, basal sediment incorporation may be a source of chemical species, both as dissolved and particulate material (Jouzel et al., 1999), but in the Dome C ice core, the basal ice appears free of incorporated bedrock sediments (Jouzel et al., 2007). Concentrations of dust within the Dome C ice core range from 10 μg/kg during interglacials to 1500 μg/kg during glacial periods (Fig. 4a). Given average upper crustal uranium concentrations of 2–4 ppm, the concentration of uranium from the dust within the ice core should vary between 0.02 ng/kg and 4 ng/kg.

Constraining the concentration of uranium from marine aerosols, and thus the initial U concentration, is more difficult. Concentrations of sea salt sodium (ssNa), a conservative element that traces marine contributions to ice sheets, vary by a factor of 10 between interglacial and glacial periods, but the ratio of U to Na may not be constant, or at a seawater value. This variability would arise because the source of marine aerosols in ice is not directly from the open ocean. Rather, marine uranium is likely from sea ice where the brine and frost flower environment concentrate elements and fractionate elemental ratios (Rankin and Wolff, 2002; Wolff et al., 2006). While U/Na ratios have not yet been measured in sea ice brines or frost flowers, Voudrias and Means, (1993) found that U is adsorbed to halite in brine environments which would fractionate the U/Na ratio. Thus far, there have been few measurements of uranium in polar ice. Glacial meltwater from the Bartley Glacier in the Dry Valleys of Antarctica shows a uranium concentration of 44 ng/kg (Henderson et al., 2006). Dissolved U in ice samples from the Allan Hills area shows uranium concentrations between 1.5 and 3 ng/kg (Goldstein et al., 2004). Direct measurements of ice cores, Vostok and Dome C, from Antarctica show bulk uranium concentrations varying between 0.01 and 4 ng/kg (Gabrielli et al., 2005; Marteel et al., 2009). Despite this large range in the potential initial uranium concentrations, the concentration can be constrained using the isotopic composition.

The concentration of initial ^{234}U can be calculated from the initial $^{234}U/^{238}U$ in the ice if the source of uranium in precipitation