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A decade of in situ cosmogenic ¹⁴C in Antarctica

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Abstract

Cosmogenic nuclide measurements in glacial deposits extend our knowledge of glacier chronologies beyond the observational record. The short half-life of in situ cosmogenic ¹⁴C makes it particularly useful for studying glacier chronologies, as resulting exposure ages are less sensitive to nuclide inheritance when compared with more commonly measured, long-lived nuclides. An increasing number of laboratories using an automated process to extract carbon from quartz has led to in situ ¹⁴C measurements in Antarctic samples at an accelerating rate over the past decade, shedding light on deglaciation in Antarctica. In situ ¹⁴C has had the greatest impact in the Weddell Sea Embayment, where inferences on the thickness of ice and timing of deglaciation were limited by inheritance in other cosmogenic nuclide systems. Future subglacial measurements of the nuclide hold much potential as they can provide direct evidence of proposed Holocene thinning and subsequent re-thickening of parts of the Antarctic ice sheets.

Introduction

Cosmogenic nuclides are rare nuclides made in near-surface rocks and minerals by cosmic rays. The concentration of a cosmogenic nuclide in a surface is directly proportional to the time the surface was most recently uncovered by receding ice. As such, measuring cosmogenic nuclide concentrations is a common way of studying glacier chronologies (Schaefer and others, 2022). By measuring cosmogenic nuclides at different elevations above glaciers, we can constrain both the past thickness and timing and pattern of thinning (Ackert and others, 1999; Stone and others, 2003), typically following the Last Glacial Maximum (LGM). These geologic constraints are used to validate the results of numerical ice sheet models investigating deglaciation (e.g., Whitehouse and others, 2012; Pittard and others, 2022), informing model parameter selection and ultimately reducing uncertainties when these same models are used to simulate the future response of ice sheets to a changing climate.

Concentrations of cosmogenic nuclides are converted to exposure ages using production rates and, for radioactive nuclides, their half-lives. Most exposure dating studies use ¹⁰Be or combine it with ²⁶Al (with half-lives of 1.4 and 0.7 Myr, respectively) (Balco, 2011). Exposure dating studies rely on the assumption that concentrations accumulated in a single phase of exposure. Cosmogenic nuclides are predominantly produced in the upper few metres of rock, and we rely on erosion during glaciations to 'reset' surfaces. Preserved beneath coldbased (nonerosive) ice, long-lived nuclides like ¹⁰Be can persist for multiple glacial-interglacial cycles, breaking the assumption of one period of exposure. Another cosmogenic nuclide, in situ ¹⁴C, has a much shorter half-life (5700 ± 30 yr), making concentrations and resulting exposure ages less sensitive to this nuclide 'inheritance'. The half-life is so short that ¹⁴C accumulated prior to the LGM will have decayed away, regardless of how much erosion took place. In situ ¹⁴C exposure ages are therefore essentially free of inheritance, providing unambiguous evidence for the timing of glacier thinning or retreat.

Another useful aspect of in situ ¹⁴C is the potential to constrain the maximum extent of LGM ice. A balance between production and decay is reached after about 5.5 times the half-life of a radioactive cosmogenic nuclide, at which point a surface is 'saturated'. This means a surface is saturated with in situ ¹⁴C after \approx 30 ka of exposure, assuming minimal erosion. When we measure a concentration equivalent to saturation, we know that the sample has been exposed for at least 30 ka, and thus was not covered during the LGM. Hence, surfaces saturated with in situ ¹⁴C provide unambiguous evidence for the extent of ice during the LGM. In summary, in situ ¹⁴C is useful for studying deglaciation because (i) concentrations are essentially uninfluenced by previous periods of exposure, providing exposure ages that are more likely to reflect the true age of deglaciation when compared with those from long-lived nuclides and (ii) measurements can provide upper constraints on the extent of ice at the LGM, a type of constraint that cannot be provided by measuring long-lived nuclides. Both of these aspects of in situ ¹⁴C mean measuring it is particularly useful for benchmarking the results of numerical ice sheet models.

A growing number of laboratories capable of extracting in situ ¹⁴C and automation of the extraction process have led to the nuclide being measured at an enhanced rate over the last decade (Fig. 1a). These measurements are advancing our knowledge of the most recent deglaciation in Antarctica, especially where inferences from long-lived nuclides are limited. How in situ ¹⁴C is measured, where in Antarctica it has been measured and what these measurements have shown us about deglaciation, are described below. Potential research questions

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(a) 140 Weddell 0 Number of in situ ¹⁴C measurements Sea OMI 120 SR Prvdz 100 Bay 80 EAIS 1 - 5 WM (o) 6 - 10 60 ٩M 15 40 16 - 20 Ross 21 - 25 20 Sea 0 26 - 30 2010 2012 2014 2016 2018 2020 Year No. of in situ 14C measurements

Fig. 1. (a) Cumulative (black) and yearly (grey bars) total in situ ¹⁴C measurements from Antarctica (excluding CRONUS A). (b) Sampling locations of all published subaerial in situ ¹⁴C measurements from Antarctica, excluding those of CRONUS-A (purple star). WAIS and EAIS are the West and East Antarctic Ice Sheets, respectively. Measurements sourced from the following studies: Antarctic Peninsula (AP), Jeong and others (2018), Lassiter Coast (LC), Pensacola Mountains (PM) and Shackleton Range (SR), Nichols and others (2019), Ellsworth Mountains (EM), Fogwill and others (2014) and Spector and others (2019), Whitmore Mountains (WM), Spector and others (2019), Amundsen Sea Embayment (ASE), Johnson and others (2017, 2020), Transantarctic Mountains (TAM), Hillebrand and others (2021), northern Victoria Land (NVL), Goehring and others (2019b) and Balco and others (2019), Prydz Bay, Berg and others (2016) and White and others (2011), Queen Maud Land (QML), Akçar and others (2020). Map made with Quantarctica (Matsuoka and others, 2018).

that can be addressed using this nuclide are also outlined, including estimating glacial erosion rates by combining measurements of in situ ¹⁴C and ¹⁰Be, assessing the outputs of numerical ice sheet models with single measurements of in situ ¹⁴C, and investigating and quantifying a proposed Holocene thinning (beneath present) episode with subglacial measurements of in situ ¹⁴C. Much of our knowledge of the past of the Antarctic ice sheets is based on periods when the ice sheets were larger than today, such as during the LGM, because evidence for past ice extent is preserved in rock and sediments above, adjacent to and offshore of present ice margins. Knowledge of contracted configurations of the Antarctic ice sheets, gained through subglacial measurements of in situ ¹⁴C, is key to understanding the future of the ice sheets given that they are predicted to continue losing mass (DeConto and others, 2021).

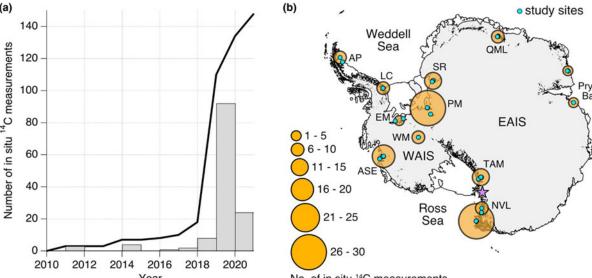
How is in situ ¹⁴C measured?

The utility of in situ ¹⁴C exposure dating has long been known (e.g., Lal, 1987, 1988; Lal and Jull, 2001) but it was not until the 2010s that improved reproducibility, increased reliability in extraction systems and an accompanying reduction in blank levels, helped make measuring it more routine. Building on methods for measuring in situ ¹⁴C in extraterrestrial samples (Goel and Kohman, 1962; Suess and Wänke, 1962), Lifton and others (2001) developed the methods for extracting carbon from quartz used in laboratories today. While methods differ with laboratory, the key steps are similar: carbon is liberated through the heating of quartz under vacuum, oxidised to form CO₂, then purified using liquid nitrogen. Some extraction lines use a tube furnace and fuse quartz in a lithium metaborate flux (Lifton and others, 2015; Lamp and others, 2019; Goehring and others, 2019a), while others use an electron bombardment or resistance furnace to release in situ ¹⁴C by diffusion through the crystal lattice (Fülöp and others, 2015, 2019; Lupker and others, 2019). Samples are sent for AMS measurement as CO2 (Hippe and others, 2013; Lupker and others, 2019) or after dilution and graphitisation (e.g., Lifton and others, 2015). Isotope ratios are used to determine in situ ¹⁴C concentrations following Hippe and Lifton (2014). In situ ¹⁴C concentrations, combined with sample density, thickness, elevation, latitude and longitude and topographic shielding, are then used to calculate exposure ages, usually using an online exposure age calculator such as the online calculators formerly known as the CRONUS-Earth online calculators (Balco and others, 2008).

The rise in the number of studies applying in situ ¹⁴C (Fig. 1a) is fuelled by a number of factors, among which most notable are a growing number of extraction lines, automation of extraction, decreasing blank levels and the widespread adoption of data reduction and production rate calibration methods. Most importantly, an increasing number of laboratories are capable of extracting carbon from quartz. Automation of the extraction process has increased sample throughput, particularly at Tulane University (Goehring and others, 2019a). A gradual reduction in ¹⁴C in process blanks has improved the detection limit. Repeat measurements of the in situ ¹⁴C concentration of the interlaboratory comparison material CRONUS-A (Jull and others, 2015) have been used to characterise the reproducibility of in situ ¹⁴C measurements (approximately 6%; Nichols and others, 2019) and calibrate the production rate used by the online exposure age calculators (Balco and others, 2008) and the Informal Cosmogenic-Nuclide Exposure-age Database (ICE-D, ice-d.org, Balco, 2020). Standardisation of data reduction (Hippe and Lifton, 2014) and the identification of a source of contamination from a commonly used method of quartz isolation (Nichols and Goehring, 2019) have also contributed to the now relatively routine measurement and application of in situ ¹⁴C.

Advances based on in situ ¹⁴C

Measurements of in situ ¹⁴C are reported from all sectors of Antarctica but are focused in the Ross, Weddell and Amundsen sea embayments, with a dearth of measurements in East Antarctica and few on the Antarctic Peninsula (Fig. 1b). Post-LGM exposure ages constrain deglaciation at most sites, and saturated measurements constrain the limit of LGM ice in



the Shackleton Range (Nichols and others, 2019), close to the West Antarctic Ice Sheet (WAIS) Divide (Spector and others, 2019) and adjacent to Prydz Bay (Berg and others, 2016). Samples saturated with in situ ¹⁴C are also observed on blue ice moraines in Queen Maud Land (Akçar and others, 2020). CRONUS-A, a sand-stone sample sourced from 1679 m asl in Arena Valley in the Dry Valleys, Antarctica (Jull and others, 2015; Fig. 1b), is saturated with ¹⁴C and has been measured at least 75 times.

The most obvious places to measure in situ ¹⁴C for exposure dating studies are those yielding solely or primarily pre-LGM exposure ages from long-lived nuclides, and thus inferences on the extent of LGM ice are limited. This is the case at the Lassiter Coast in the Weddell Sea Embayment (Fig. 1b), where the majority of ¹⁰Be exposure ages of deposits, presumably from the LGM or most recent deglaciation, exceed 100 ka (Fig. 2a). Taken at face value, one could infer that ice has not been thicker here for hundreds of thousands of years, certainly not during the LGM. However, in situ ¹⁴C measurements made at the same site, with many of the same samples, yield Holocene exposure ages (Fig. 2a), showing that (i) ice was at least 380 m thicker than present at the LGM, (ii) deglaciation occurred relatively rapidly and (iii) this region was covered by cold-based ice that preserved ¹⁰Be that accumulated during previous periods of exposure.

A similar pattern of pre-LGM 10Be exposure ages and post-LGM in situ ¹⁴C exposure ages is observed at other sites in the Weddell Sea Embayment. Limited LGM thickening inferred from predominantly pre-LGM ¹⁰Be exposure ages in the Shackleton Range (Hein and others, 2011) and Pensacola Mountains (Balco and others, 2016; Bentley and others, 2017) was used to benchmark ice sheet models for some time (e.g., Whitehouse and others, 2017; Kingslake and others, 2018). These interpretations led to relatively little post-LGM ice volume change in the Weddell Sea Embayment (when compared with previous reconstructions, see Bentley and Anderson (1998)) becoming the predominant reconstruction among the palaeo community (Hillenbrand and others, 2014). Subsequent measurements of in situ ¹⁴C yielded post-LGM exposure ages at both locations, showing that, rather than limited thickening, ice was at least 310 and 800 m thicker than present at the LGM (Nichols and others, 2019). Other locations with multiple samples yielding pre-LGM exposure ages from long-lived nuclides and post-LGM in situ ¹⁴C exposure ages are the Flower Hills and Meyer Hills in the Ellsworth Mountains (Fogwill and others, 2014) and the Darwin-Hatherton Glacier System in the Ross Sea Embayment (Hillebrand and others, 2021).

In situ ¹⁴C can also be useful at sites yielding post-LGM ¹⁰Be exposure ages. For example, in the Amundsen Sea Embayment, Johnson and others (2020) use measurements of in situ ¹⁴C to identify a smaller degree of inheritance in their exposure ages. Here, ¹⁰Be exposure ages (n = 9) indicate deglaciation happened about 17 ka, while in situ ¹⁴C exposure ages (n = 8) show it occurred about 6 ka, a difference of 11 ka, which is significant for establishing an accurate deglacial chronology of the region. Samples with post-LGM ¹⁰Be exposure ages and younger in situ ¹⁴C exposure ages are also observed at sites in northern Victoria Land (Balco and others, 2019; Goehring and others, 2019b), with an additional sample in the Flower Hills (Fogwill and others, 2014). Evidently, even when ¹⁰Be exposure ages at a site postdate the LGM and thus we know the degree of ice thickness change, there could still be a detectable amount of inheritance skewing our understanding of the timing of deglaciation.

Glacier chronologies are constrained solely with in situ ¹⁴C measurements (without accompanying long-lived nuclides) at some locations, such as the Whitmore Mountains close to WAIS Divide (Spector and others, 2019) and some sites in northern Victoria Land (Goehring and others, 2019b). Additionally, concordant in situ ¹⁴C and ¹⁰Be exposure ages are observed at many sites in Antarctica (White and others, 2011; Balco and others, 2019; Goehring and others, 2019b; Hillebrand and others, 2021).

Future research priorities

Measuring in situ ¹⁴C

While we have learnt much about deglaciation in Antarctica from in situ ¹⁴C in recent years, we have also learnt much about measuring the nuclide itself, and some questions remain unanswered. Some studies observe in situ ¹⁴C concentrations in excess of theoretical limits (Balco and others, 2016; Akçar and others, 2020), while another observes measurement reproducibility lower than that expected from measurement uncertainties alone (Nichols and others, 2019). When sample contamination can be ruled out, mass movement and supraglacial transport could explain elevated concentrations (Balco and others, 2016), while unrecognised measurement error could explain the limited reproducibility. Further work dedicated to method development is needed to isolate what is (i) limiting measurement reproducibility and (ii) contributing toward concentrations exceeding theoretical limits. Most studies measure in situ ¹⁴C in quartz, but the nuclide is produced in other materials such as calcium carbonate

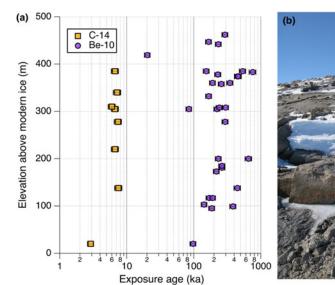


Fig. 2. (a) Exposure ages from the Lassiter Coast (Johnson and others, 2019; Nichols and others, 2019) sourced from ICE-D using the LSDn scaling method. Error bars show external uncertainties but are often smaller than symbols. (b) Collection site of a bedrock sample (P11-11-4) on the Bowman Peninsula, Lassiter Coast (Johnson and others, 2019). This bedrock sample has a ¹⁰Be exposure age of 410 ± 30 ka and an in situ ¹⁴C exposure age of 7.4 ± 0.6 ka. Photo credit: Joanne Johnson (British Antarctic Survey).

(Handwerger and others, 1999) and olivine (Pigati and others, 2010). Establishing methods for the extraction of carbon from these materials would expand the number of locations we can study with in situ ¹⁴C beyond only those rich in quartz.

Applying in situ ¹⁴C

Further exposure dating studies using in situ ¹⁴C would be useful in areas unstudied with cosmogenic nuclides or those yielding solely or primarily pre-LGM exposure ages from long-lived nuclides (e.g., Hodgson and others, 2012). By filling spatial gaps in our knowledge of deglaciation in Antarctica, these measurements of in situ ¹⁴C would provide more geologic constraints for the benchmarking of numerical ice sheet models. Additionally, there are a few applications beyond traditional exposure dating yet to be used (or to their full potential) that could improve our knowledge of the history and glaciology of the Antarctic ice sheets.

How much did glaciers erode into bedrock during the Holocene? This question can be answered by combining measurements of in situ ¹⁴C and ¹⁰Be in recently exposed proglacial bedrock. Making direct measurements of subglacial erosion is complicated by the difficulty of accessing the beds of glaciers. Using cosmogenic nuclides to estimate past erosion rates provides knowledge of glacial processes over a longer period than contemporary point measurements, extending our knowledge of glacial erosion rates beyond the observational record. The relationship between the in situ ¹⁴C and ¹⁰Be concentration of a proglacial bedrock sample is related to the depth to which a glacier eroded into bedrock in the Holocene, allowing the estimation of Holocene glacial erosion rates (Rand and Goehring, 2019). Because this method requires proglacial bedrock, it may be limited to smaller glaciers such as those on the Antarctic Peninsula or at high elevations on the continent.

How closely do numerical ice sheet model outputs reflect the timing of both the advance and retreat phases of deglaciation contained in geologic archives? Measurements of in situ ¹⁴C, rather than long-lived nuclides, can answer this question. Many numerical ice sheet models are benchmarked against exposure age datasets recording only deglaciation. By assuming samples were saturated prior to LGM burial, individual measurements of in situ ¹⁴C can be used to assess the timing of both advance and retreat phases of model outputs (Spector and others, 2019), reducing uncertainties when these same models are used to simulate the future of ice sheets. More generally, targeting exposed surfaces high above modern ice elevations could help provide more upper constraints on LGM ice thicknesses to help validate numerical ice sheet model outputs.

To what extent, and where, did parts of the Antarctic ice sheets readvance in the Holocene? This is perhaps the most important question that in situ ¹⁴C can answer. A number of studies, both through geologic observations (Siegert and others, 2013; Wolstencroft and others, 2015; Greenwood and others, 2018; King and others, 2022) and modelling (Kingslake and others, 2018), infer that some parts of the Antarctic ice sheets were smaller than present in the Holocene and subsequently grew to their present configuration. Through measuring carbon in subglacial sediments, two studies (Venturelli and others, 2020 and Neuhaus and others, 2021) report the first direct evidence of a Holocene grounding line readvance in the Ross sector. Further direct evidence for a Holocene readvance can be obtained through in situ ¹⁴C measurements in subglacial bedrock, because significant concentrations in subglacial bedrock unambiguously requires Holocene exposure, either complete or through relatively thin ice (Johnson and others, 2022). Constraining the scale of this readvance, both in ice thickness change and geographic extent,

could shed light on the processes causing the mass loss and subsequent gain (e.g., ocean forcings, glacioisostatic adjustment), information that can then be used with numerical models to replicate this ice sheet behaviour. Given that current Antarctic ice sheet mass loss is predicted to continue (DeConto and others, 2021), knowing the processes that helped recover ice mass loss in a climate relatively similar to that of today is key to understanding the reversibility of current and future Antarctic ice mass loss. While previous studies have investigated long term changes in the Greenland Ice Sheet by measuring long-lived nuclides in subglacial material (Schaefer and others, 2016; Christ and others, 2021), there are no published subglacial measurements of in situ ¹⁴C from beneath any ice sheet. If above background in situ ¹⁴C indicative of a Holocene readvance is measured in samples collected from beneath the Antarctic ice sheets, multiple studies will be required to confirm if this ice sheet behaviour is widespread or localised.

Conclusions

To summarise, the cosmogenic nuclide in situ ¹⁴C has been measured at an enhanced rate over the last decade, fuelled by the automation of the extraction process and an increasing number of laboratories now capable of extracting it. Measurements of in situ ¹⁴C have been used in exposure dating studies to shed light on deglaciation in all sectors of Antarctica, but especially in the Weddell Sea Embayment. Some studies observe in situ ¹⁴C concentrations exceeding theoretical limits and also measurement reproducibility lower than expected, which can hopefully be addressed with dedicated work on understanding the extraction process and geomorphic scatter. While there are many locations in Antarctica where traditional in situ ¹⁴C exposure dating studies would be useful, there are also a number of other applications of the nuclide that hold much potential, including using subglacial measurements to constrain episodes of thinning and rethickening in the Holocene.

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