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ScienceDirect

Geochimica et Cosmochimica Acta 152 (2015) 143–165

Geochimica et Cosmochimica Acta

www.elsevier.com/locate/gca

Uranium isotopes and dissolved organic carbon in loess permafrost: Modeling the age of ancient ice

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Received 24 June 2012; accepted in revised form 13 November 2014; available online 24 November 2014

Abstract

The residence time of ice in permafrost is an indicator of past climate history, and of the resilience and vulnerability of high-latitude ecosystems to global change. Development of geochemical indicators of ground-ice residence times in permafrost will advance understanding of the circumstances and evidence of permafrost formation, preservation, and thaw in response to climate warming and other disturbance. We used uranium isotopes to evaluate the residence time of segregated ground ice from ice-rich loess permafrost cores in central Alaska. Activity ratios of ^{234}U vs. ^{238}U ($^{234}U/^{238}U$) in water from thawed core sections ranged between 1.163 and 1.904 due to contact of ice and associated liquid water with mineral surfaces over time. Measured ($^{234}U/^{238}U$) values in ground ice showed an overall increase with depth in a series of five neighboring cores up to 21 m deep. This is consistent with increasing residence time of ice with depth as a result of accumulation of loess over time, as well as characteristic ice morphologies, high segregated ice content, and wedge ice, all of which support an interpretation of syngenetic permafrost formation associated with loess deposition. At the same time, stratigraphic evidence indicates some past sediment redistribution and possibly shallow thaw among cores, with local mixing of aged thaw waters. Using measures of surface area and a leaching experiment to determine U distribution, a geometric model of ($^{234}U/^{238}U$) evolution suggests mean ages of up to ~200 ky BP in the deepest core, with estimated uncertainties of up to an order of magnitude. Evidence of secondary coatings on loess grains with elevated ($^{234}U/^{238}U$) values and U concentrations suggests that refinement of the geometric model to account for weathering processes is needed to reduce uncertainty. We suggest that in this area of deep ice-rich loess permafrost, ice bodies have been preserved from the last glacial period (10–100 ky BP), despite subsequent fluctuations in climate, fire disturbance and vegetation. Radiocarbon (^{14}C) analysis of dissolved organic carbon (DOC) in thaw waters supports ages greater than ~40 ky BP below 10 m. DOC concentrations in thaw waters increased with depth to maxima of >1000 ppm, despite little change in ice content or cryostructures. These relations suggest time-dependent produc-

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http://dx.doi.org/10.1016/j.gca.2014.11.008

tion of old DOC that will be released upon permafrost thaw at a rate that is mediated by sediment transport, among other factors.

1. INTRODUCTION

Over half the belowground terrestrial organic carbon (C) pool resides in permafrost that is warming and thawing at high latitudes in response to recent warming (Hugelius et al., 2014; Lachenbruch and Marshall, 1986; Osterkamp et al., 2009; Romanovsky et al., 2010; Tarnocai et al., 2009). The release and decomposition of the large amounts of organic carbon stored in permafrost soils may constitute a substantial positive feedback to the climate system (Zimov et al., 2006, 2009; Ping et al., 2008; Schuur et al., 2008; Koven et al., 2011), subject to the complex interaction between permafrost extent and factors such as plant community changes and decreasing sea ice extent (Euskirchen et al., 2006; Lawrence et al., 2008). It has been argued that much of the carbon in permafrost soils was sequestered during the Late Pleistocene, followed by extensive thaw of permafrost and release of carbon during the Pleistocene–Holocene transition (Zimov et al., 2006, 2009). Understanding the relationship between known temperature fluctuation and the vulnerability or resilience of permafrost over multiple glacial cycles is critical for understanding the global carbon balance and predicting its fate in the future (Jorgenson et al., 2010; Grosse et al., 2011). Development of multiple chronometers is therefore essential for understanding the development and evolution of permafrost systems that may have existed over tens of thousands to millions of years.

Development of tools for dating permafrost has been limited by two factors. The first is the temporal constraints of commonly used chronological tools: ^{14}C has a half life of 5730 y and can be used to date materials up to about 50 ky BP (Reimer et al., 2009). Tritium can be used to directly date ice, but its 12.32 y half life restricts its use to residence times from one year to a few hundred years (Morgenstern et al., 2010). Both ^{14}C and tritium are complicated – though not intractably – by residually elevated concentrations due to atmospheric testing of nuclear weapons in the mid-twentieth century. The second limiting factor is the potential for ice bodies to be younger than the surrounding sediment, due to thaw and drainage followed by re-accumulation and freezing of meteoric water. This possibility necessitates secondary geochronological evidence in support of dates obtained through ^{14}C analysis of solids, luminescence analysis of loess, and tephrachronology (Schirrmeister et al., 2002; Froese et al., 2008a; Jensen et al., 2008; Blinov et al., 2009). Given evidence for preservation of Pleistocene ice over timescales of 10^5 y (Froese et al., 2008; Reyes et al., 2010a; Vaks et al., 2013), a direct measure of the age of ice in permafrost at timescales of 10^4–10^6 y is needed.

Permafrost formed in conjunction with loess deposition occurs throughout the Arctic and Subarctic, and represents a specific mode of "syngenetic" permafrost formation, also termed yedoma (French and Shur, 2010; Kanevskiy et al., 2011). Gradual addition of eolian and organic material to frozen land surfaces – particularly in the context of productive Pleistocene steppe-tundra ecosystems – results in syngenetic loess permafrost that stores large quantities of carbon (Zimov et al., 2006), slows subsequent thaw due to high ice content (Romanovsky et al., 2010; O'Donnell et al., 2011a), preserves distinct ice morphologies (French and Shur, 2010), and by definition must increase in age with depth. Moreover, the restricted particle-size distribution in eolian-derived loess and the likelihood of equiaxial grains minimizes the effect of variable soil materials over time (Fleischer, 1983; Muhs et al., 2008). Hence this type of permafrost offers a useful medium for testing indicators of permafrost chronology based on rock–water contact, and associated carbon dynamics.

In this study, we evaluate radioactive disequilibrium among uranium isotopes in thaw water (specifically, $^{234}U/^{238}U$ activity ratios, here denoted as ($^{234}U/^{238}U$) values) as a direct measure of the age of ice in syngenetic loess permafrost. U-series disequilibrium occurs over timescales of up to 10^6 y, and results from several natural processes, either as a consequence of the distinct chemical behavior of different elements in the decay chain, or as a result of alpha-recoil effects from radioactive decay (Fig. 1) (Bourdon et al., 2003). Recoil-driven disequilibrium occurs as a function of rock–water contact at low temperature and has been extensively observed in groundwaters (Osmond et al., 1983; Paces et al., 2002; Porcelli and Swarzenski, 2003), as well as marine and riverine environments as a function of weathering (Vigier et al., 2001, 2006; Chabaux et al., 2003a,b, 2011; Robinson et al., 2004; Dosseto et al., 2006a,b, 2008, 2010; Pogge von Strandmann et al., 2011). Recoil effects have also been documented from mineral-ice contact (Fireman, 1986; Goldstein et al., 2004; Aciego et al., 2011) and in weathering rinds and soils (Pelt et al., 2008, 2013; Suresh et al., 2013). In general, recoil disequilibrium effects occur as a function of time, U concentrations in minerals and water or ice, and mineral grain size and surface area. Ice-rich loess permafrost that formed syngenetically with eolian loess deposition over time is an ideal substrate for testing a U-series age model for permafrost ice for four reasons. This variety of permafrost (1) should increase in age with depth (French and Shur, 2010; O'Donnell et al., 2011a), (2) should produce measureable signal due to small grain sizes and abundant mineral surfaces (Fireman, 1986; DePaolo et al., 2006, 2012; Maher et al., 2006; Bourdon et al., 2009), (3) should