

Influence of glacial meltwater on global seawater $\delta^{234}\text{U}$ Carli A. Arendt^{a,b,*}, Sarah M. Aciego^{b,c}, Kenneth W.W. Sims^c, Sarah B. Das^d,
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Abstract

We present the first published uranium-series measurements from modern Greenland Ice Sheet (GrIS) runoff and proximal seawater, and investigate the influence of glacial melt on global seawater $\delta^{234}\text{U}$ over glacial-interglacial (g-ig) timescales. Climate reconstructions based on closed-system uranium-thorium (U/Th) dating of fossil corals assume U chemistry of seawater has remained stable over time despite notable fluctuations in major elemental compositions, concentrations, and isotopic compositions of global seawater on g-ig timescales. Deglacial processes increase weathering, significantly increasing U-series concentrations and changing the $\delta^{234}\text{U}$ of glacial meltwater. Analyses of glacial discharge from GrIS outlet glaciers indicate that meltwater runoff has elevated U concentrations and differing ^{222}Rn concentrations and $\delta^{234}\text{U}$ compositions, likely due to variations in subglacial residence time. Locations with high $\delta^{234}\text{U}$ have the potential to increase proximal seawater $\delta^{234}\text{U}$. To better understand the impact of bulk glacial melt on global seawater $\delta^{234}\text{U}$ over time, we use a simple box model to scale these processes to periods of extreme deglaciation. We account for U fluxes from the GrIS, Antarctica, and large Northern Hemisphere Continental Ice Sheets, and assess sensitivity by varying melt volumes, duration and U flux input rates based on modern subglacial water U concentrations and compositions. All scenarios support the hypothesis that global seawater $\delta^{234}\text{U}$ has varied by more than 1‰ through time as a function of predictable perturbations in continental U fluxes during g-ig periods. © 2018 Elsevier Ltd. All rights reserved.

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1. INTRODUCTION

Ice sheet runoff during interglacial (warm) periods contributes substantial meltwater and associated glacially-

derived weathering products to oceans (Blum and Erel, 1995; Anderson, 2005; Vance et al., 2009; Chen et al., 2016). Deglaciation during the onset of interglacial periods is also marked by rapid warming and significant meltwater discharge events that further alter the relationship between elemental ‘inputs’ and ‘outputs’, whereby oceanic inputs are no longer in steady state with oceanic outputs (Henderson, 2002; Vance et al., 2009). If the weathering products contain radiogenic and radioactive nuclides with long half-lives and long ocean residence times, this imbalance

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between input and output causes a noticeable shift in seawater chemistry on glacial-interglacial (*g-ig*) timescales (Vance et al., 2009; Wortmann and Paytan, 2012). In the case of uranium (U), the extent of chemical change in seawater U depends on the chemistry of glacial melt, explicitly U concentrations, [U], and $^{234}\text{U}/^{238}\text{U}$ activity ratios, ($^{234}\text{U}/^{238}\text{U}$). In this study, ($^{234}\text{U}/^{238}\text{U}$) values were converted to the commonly referenced $\delta^{234}\text{U}$ notation using the following equation:

$$\delta^{234}\text{U} = ((U_{234}/U_{238}) - 1) * 1000, \quad (1)$$

where $\delta^{234}\text{U}$ is the deviation of the sample from the ($^{234}\text{U}/^{238}\text{U}$) secular equilibrium value of 0.0 in permil (‰) notation. Hereafter, U concentration is given as [U] and ($^{234}\text{U}/^{238}\text{U}$) is given as $\delta^{234}\text{U}$.

If the $\delta^{234}\text{U}$ composition of glacial meltwater is distinct from seawater and the U flux is high enough, there is the possibility of producing significant changes in the $\delta^{234}\text{U}$ of seawater as a consequence of *g-ig* cycling (Villemant and Fleuillet, 2003; Scholz et al., 2004; Chen et al., 2016).

Globally, past work presumes oceanic U chemistry is stable over *g-ig* timescales (Hamelin et al., 1991; Henderson, 2002; Edwards et al., 2003), as residence times of U in seawater is estimated to be 300–600 thousand years (Henderson, 2002), which far exceeds the mixing time of the world's oceans at approximately 1–2 ka (Broecker, 1982; Chen et al., 1986, 2016; Cheng et al., 2000; Esat and Yokoyama, 2006). However, based on *g-ig* cycles, decay rates, and oceanic mixing timescales, previous studies have speculated that glacial melt is capable of altering seawater $\delta^{234}\text{U}$ on *g-ig* timescales (Robinson et al., 2004; Esat and Yokoyama, 2006). Additionally, numerous fossil coral U-Th records have indicated that seawater $\delta^{234}\text{U}$ may have been as low as 131‰ during the last glacial period (Esat and Yokoyama, 2006), suggesting variability in the seawater $\delta^{234}\text{U}$ record (Cutler et al., 2003; Stirling and Andersen, 2009; Andersen et al., 2010; Thompson et al., 2011; Chen et al., 2016). Esat and Yokoyama (2006) investigated this speculated change in seawater $\delta^{234}\text{U}$ from the perspective of higher $\delta^{234}\text{U}$ inputs from exposed continental shelves during deglaciation events. Other studies have focused on the impact of variations in U fluxes from rivers (Robinson et al., 2004; Andersen et al., 2007) on seawater $\delta^{234}\text{U}$ compositions. In addition, a recent study by Chen et al. (2016) hypothesized ^{234}U enriched waters from subglacial melt reservoirs as an explanation for elevated $\delta^{234}\text{U}$ in corals. These studies establish the framework on which the present study is based: due to prolonged subglacial residence times, subglacial water is enriched in $\delta^{234}\text{U}$, which will influence seawater $\delta^{234}\text{U}$ chemistry on *g-ig* timescales.

To significantly influence the ocean U budget, glacial runoff input must have $\delta^{234}\text{U}$ isotopic compositions distinct from seawater. For glacial meltwater to have elevated [U] and $\delta^{234}\text{U}$, the residence time of subglacial water must be prolonged to allow sufficient weathering to take place, establishing a high baseline of both ^{234}U and ^{238}U concentrations from weathering processes (note that ^{234}U is elevated relative to ^{238}U by weathering processes because of recoil of this nuclide, Kigoshi, 1971). Therefore, glacial melt

contributions should elevate proximal seawater $\delta^{234}\text{U}$ if the melt volumes, weathering rates, residence times and associated $\delta^{234}\text{U}$ are increased, and the corresponding [U] are non-negligible. We postulate that if all these criteria are met, glacial meltwater U contributions to global seawater can alter proximal seawater $\delta^{234}\text{U}$, and potentially global ocean $\delta^{234}\text{U}$ on *g-ig* timescales.

In this study, we test the hypothesis that glacial melt influences seawater U chemistry by collecting samples from a series of sites around GrIS and measuring the U chemistry of GrIS glacial meltwater and proximal seawater. We further use our observations to investigate the influence variations in glacial melt U flux has on seawater $\delta^{234}\text{U}$ on *g-ig* timescales using a simple seawater $\delta^{234}\text{U}$ box model approach (modified from Henderson, 2002).

2. BACKGROUND

2.1. Seawater U chemistry

It is well established that *g-ig* cycles cause changes in major elemental fluxes (e.g. carbon, oxygen) within the ocean geochemical budget. However, changes in seawater trace element and metal isotopic compositions, such as strontium, lead, osmium, iron, boron, sulfur, silicon and calcium, remain the subject of debate (Blum and Erel, 1995; Jones et al., 2002; Vance et al., 2009; Lancan et al., 2012). The ocean U budget is generally thought to have remained relatively stable on *g-ig* timescales, with the global seawater $\delta^{234}\text{U}$ assumed to be uniform within 1‰ (Henderson et al., 1993; Gallup et al., 1994; Stirling et al., 1995; Delanghe et al., 2002; Thompson et al., 2003; Andersen et al., 2010). Notable variations in seawater U chemistry have previously been attributed to local seawater chemistry influences and/or diagenesis (Henderson et al., 1993; Gallup et al., 1994; Esat and Yokoyama, 2006). Other studies, however, have alluded to the possibility of variable U fluxes to the world's oceans (Henderson, 2002; Dunk et al., 2002; Villemant and Fleuillet, 2003; Scholz et al., 2004; Chen et al., 2016). This uncertainty directly impacts climate reconstructions based on U/Th dating (Robinson et al., 2004; Esat and Yokoyama, 2006). Constraints on the impact of [U] and $\delta^{234}\text{U}$ of glacial meltwater on seawater U chemistry and the variables that impact this influence should therefore be further explored in more detail.

2.2. Subglacial U chemistry

2.2.1. U and subglacial weathering

Subglacial physical weathering produces large quantities of small particles with high surface area to volume ratios (Blum and Erel, 1995; Anderson, 2005). These highly reactive particles with freshly exposed surfaces are easily subjected to chemical weathering (Anderson, 2005), and can enhance the U flux from glacial meltwater due to increased sediment surface area from which isotopes, such as ^{234}U and ^{238}U are released (Bonotto and Andrews, 2000). Present day high-latitude rivers have much higher $\delta^{234}\text{U}$ values than present day low-latitude rivers, which is attributed to an increased contribution of U flux from chemical weather-

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