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Uranium comminution ages: Sediment transport and deposition time scales

Âges de comminution de l'uranium : échelles de temps de transport et de dépôt de sédiment

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ABSTRACT

The uranium isotope comminution age is determined from the $^{234}U/^{238}U$ ratio and reflects the timescale associated with the transformation of bedrock to sediment. The comminution age is applicable to Late Pleistocene sediments and measures the amount of time elapsed since sediment generation by mechanical weathering and erosion. The age significance of the $^{234}U/^{238}U$ ratios is based on physical disruption of the ^{238}U-decay series by recoil loss of ^{234}Th that occurs in mineral grains smaller than 50 μm. Results from study of fine-grained deep sea sediments in the North Atlantic Ocean, alluvial sediments in California and Australia, and modern glacial outwash are encouraging, but critical aspects of the method require further investigation. Particular issues are the effects of laboratory chemical leaching treatment on sediment samples and estimation of ^{234}U loss rates as a function of grain size. In the North Atlantic marine environment the U isotope variations are inferred to reflect differences in the transport time of the sediment–the time elapsed between the generation of the small sediment particles by glacial action in Iceland and Fennoscandian source areas, and the time of deposition on the seafloor in the North Atlantic Ocean at a drift site south of Iceland. Calculated transport times vary from less than 10 kyr to about 400 kyr, and correlate with provenance and glacial cycles. Application to alluvial sediments in California and Australia suggests that where sediments are glacially-derived and transported short distances, the U comminution age may approximate the sedimentation age, but in larger basins that are not glaciated the sediments retain information about residence/transport times that can extend to ca. 400 kyr. To verify that initial $^{234}U/^{238}U$ ratios for glacial sediments are close to the secular equilibrium ratio, outwash from several major glaciers around the world was measured and found to be within ± 1% of the accepted equilibrium $^{234}U/^{238}U$ value.

Mots clés :
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RÉSUMÉ

L'âge de comminution de l'isotope de l'uranium est déterminé à partir du rapport $^{234}U/^{238}U$ et reflète l'échelle de temps associée à la transformation de la roche mûre en sédiment. L'âge de comminution est applicable aux sédiments du Pléistocène supérieur et mesure la quantité de temps qui s'est écoulée entre la production de sédiment par altération et érosion. La signification des rapports $^{234}U/^{238}U$ est fondée sur la rupture

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physique de la série de désintégration de ^{238}U par perte recul de ^{234}Th qui se produit dans des grains minéraux inférieurs à 50 μm. Les résultats de l'étude de sédiments océaniques fins de profondeur dans l'Atlantique Nord, de sédiments alluviaux en Californie et en Australie et de dépôts glaciaires actuels sont encourageants, mais les aspects critiques de la méthode requièrent des investigations plus poussées. Celles-ci pourraient être les effets du lessivage chimique en laboratoire sur des échantillons de sédiment et l'estimation des taux de perte de ^{234}U en fonction de la taille des grains. Dans l'environnement océanique de l'Atlantique Nord, les variations isotopiques de U sont supposées refléter les différences dans le temps de transport du sédiment – le temps écoulé entre la production de particules de petite taille par action glaciaire à partir de zones sources en Islande et en Scandinavie et le temps de dépôt sur le plancher océanique de l'Atlantique Nord sur un site de drift au sud de l'Islande. Les temps de transport calculés varient entre moins de 10 ans et environ 400 000 ans, selon la provenance et les cycles glaciaires. Une application aux sédiments alluviaux de Californie et d'Australie suggère que là où les sédiments ont une origine glaciaire et sont transportés sur de courtes distances, l'âge de comminution de l'U correspond approximativement à l'âge de sédimentation, mais que dans de plus grands bassins qui ne sont pas englacés, l'information que retiennent les sédiments à propos du rapport temps de transport/résidence peut atteindre ca. 400 kans. Pour vérifier que les rapports initiaux ^{234}U/^{238}U sont proches du rapport d'équilibre séculaire, des dépôts glaciaires en provenance de plusieurs importants glaciers à travers le monde ont été analysés et se situent à ± 1 % de la valeur d'équilibre ^{234}U/^{238}U admise.

1. Introduction

There are now a large number of approaches to isotopic geochronology – the use of radioactive decay or other natural nuclear processes to measure the ages of geologic rock formations and features. When applied to sediments and sedimentary rocks, however, isotopic geochronology is typically ineffective, or at least challenging (e.g., Dickin, 1995, Rasmussen, 2005). The more successful approaches to dating sedimentary formations depend on the presence of intercalated volcanic ash layers that are more amenable to common geochronologic methods, or correlation between fossil assemblages that have been dated using volcanic materials. Sedimentary formations that contain neither fossils nor volcanic interbeds are usually not dateable. One of the difficulties with dating clastic sediments is that the rock and mineral grains that comprise them had a prior existence, potentially for a long time, as parts of other rock formations before they were broken down by erosion, transported, and deposited as a sedimentary formation. The challenge for dating is in finding a physical or chemical change that happens at or near the time of sediment deposition, and that expresses itself as a disruption to a radioactive decay system.

Recent attempts at dating clastic sedimentary rocks involve the uranium and thorium radioactive decay series or cosmogenic nuclides. In both cases, there are difficulties in establishing when the isotopic clocks are set, and this difficulty, as well as analytical limitations, translates into considerable uncertainty in the determined age. For the use of U-Th isotopic disequilibrium (e.g. Chabaux and Riotte, 2003; Dosseto et al., 2008; Granet et al., 2007, 2010; Vigier et al., 2001), the assumption is that U and Th are chemically separated (fractionated) during weathering in soils where the sediment originates. For cosmogenic nuclides, which are sensitive to exposure of rock material to the cosmic ray-produced neutrons at the Earth's surface, the exposure age measured in sediments is a combination of the time since deposition and time spent close to the Earth's surface in a soil or regolith before or during transport (Phillips et al., 1997; Bierman and Nichols, 2004).

This article summarizes recent research using the U radioactive decay series, but with a specific approach referred to as the "U comminution age". The comminution age method (DePaolo et al., 2006) is designed to measure the time that elapses subsequent to bedrock being reduced by *physical* weathering to small grains–small being defined functionally as less than about 50 μm in diameter. The U radioactive decay series is sensitive to the size of the particles in which the U is contained, as described below. The term comminution refers to any process that accomplishes the reduction of rock material to silty-sand, silt, or clay-sized mineral grains. A key conceptual problem is to determine (or define) when sand and silt-size particles are produced from bedrock. Glacial erosion represents one geologic situation where this time may be well defined.

2. Comminution age model

The decay of ^{238}U to ^{206}Pb occurs in several steps (cf. Dickin, 1995; Ku, 2000). The first few steps in the sequence produce ^{234}U via an intermediate daughter isotope ^{234}Th. In most rocks and minerals that are older than a million years, and even in many that are younger, the ^{234}U/^{238}U ratio is almost exactly equal to the inverse ratio of the decay constants (λ) for these two U isotopes. This condition is referred to as radioactive equilibrium, and implies that the rate of production of ^{234}U by the decay of ^{238}U is equal to the rate of decay of ^{234}U, so that the ratio of ^{234}U/^{238}U is constant and equal to $\lambda_{238}/\lambda_{234}$. The equilibrium condition can also be described in terms of the activity ratio $(^{234}U/^{238}U)_{AR}$ being equal to unity.

In a small sediment grain with diameter of order 10 μm or less, it is impossible for the ^{234}U/^{238}U ratio to be maintained at the equilibrium ratio. The failure to maintain equilibrium is due to the fact that in the process