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Extension of $^{239+240}$Pu sediment geochronology to coarse-grained marine sediments

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ABSTRACT

Sediment geochronology of coastal sedimentary environments dominated by sand has been extremely limited because concentrations of natural and bomb-fallout radionuclides are often below the limit of measurement using standard techniques. ICP-MS analyses of $^{239+240}$Pu from two sites representative of traditionally challenging (i.e., low concentration) environments provide a "proof of concept" and demonstrate a new application for bomb-fallout radiotracers in the study of sandy shelf-seabed dynamics. A kasten core from the New Zealand shelf in the Southern Hemisphere (low fallout), and a vibracore from the sandy nearshore of North Carolina (low particle surface area) both reveal measurable $^{239+240}$Pu activities at depth. In the case of the New Zealand site, independently verified steady-state sedimentation results in a $^{239+240}$Pu profile that mimics the expected atmospheric fallout. The depth profile of $^{239+240}$Pu in the North Carolina core is more uniform, indicating significant sediment resuspension, which would be expected in this energetic nearshore environment. This study, for the first time, demonstrates the utility of $^{239+240}$Pu in the study of sandy environments, significantly extending the application of bomb-fallout isotopes to coarse-grained sediments, which compose the majority of nearshore regions.

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1. Introduction and background

During the past few decades, hundreds of studies have employed naturally occurring and bomb-fallout isotopes (e.g., ^{210}Pb, ^{7}Be, ^{234}Th, ^{137}Cs, $^{239+240}$Pu) to quantify sediment processes occurring in terrestrial and marine environments. Such studies have enjoyed great successes in measuring important environmental parameters, such as: sediment deposition and accumulation rates (e.g., Robbins and Edgington, 1975; Nittrouer et al., 1979); biological mixing coefficients (e.g., Aller and Cochran, 1976; McKee et al., 1983); biological mixing depths (e.g., Peng et al., 1979; Nittrouer and Sternberg 1981); and physical resuspension depths (e.g., Kuehl et al., 1996; Dellapenna et al., 1998). However, an inherent bias in most of these studies is that they have typically been limited to fine-grained sediments (muds and/or organic-rich sediments), in large part because traditional decay counting techniques are not sensitive to extremely low activities expected in sandy sediments as a function of their relatively low surface area. Such studies of environments characterized both by modern sands and muds (e.g., the majority of coastal and nearshore marine settings) have been severely biased toward the fine-grained regions. This study presents results from inductively coupled plasma mass spectrometry (ICP-MS) analysis of bomb fallout $^{239+240}$Pu in two environments where traditional decay counting proved inadequate, and demonstrates the applicability of this approach to regions with characteristically low activities or sandy sediments.

Fallout isotopes from nuclear-weapons testing, primarily ^{137}Cs, have been widely used in sedimentation studies because of their known depositional history and inventory, particle-reactive chemical properties, and global distribution (e.g., Ritchie and McHenry, 1990). Significant ^{137}Cs deposition first occurred worldwide in 1954, and monthly deposition peaked in mid-1963, just prior to the August 1963 signing of the Limited Test Ban Treaty. ^{137}Cs has proven extremely valuable as a specific chronostratigraphic marker, and as an indicator of physical and biological sediment disturbance from particle resuspension and mixing.

^{137}Cs has been used extensively due to its high-fission yield, but also because its measurement by gamma spectrometry is relatively straightforward. Unfortunately, ^{137}Cs has a half-life of only 30 years; in many settings, it has already decayed below levels readily detectable with reasonable sample sizes and counting times. The detection of ^{137}Cs in the Southern Hemisphere is especially problematic, because fallout levels were approximately

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four-fold lower than in the Northern Hemisphere (Beck and Bennett, 2002). In addition, Cs has been shown to be mobile in reducing environments (particularly marine), complicating its interpretation. In saline environments with coarse-grained sediments, two different factors tend to preclude the binding of ^{137}Cs to sediment particles: (A) there is a paucity of fine-grained smectite/illite clay particles into which Cs can intercalate, leaving few actual sites for Cs to bind to sediments, and (B) the high ionic-strength environment has a very large supply of cationic competitors for the few mineral sites where Cs could bind.

Plutonium isotopes, on the other hand, are expected to be more successful than Cs at binding to sediments in coarse-grained marine environments. Pu is highly particle reactive in marine environments with a variety of sediment types (Higgo and Rees, 1986); Pu tends to bind irreversibly to sediments in these environments with little chance of chemical re-mobilization once incorporated into sediment particles (Cf. Gouzy et al., 2005). For these reasons, it would be anticipated that Pu would be widely used in preference to Cs in marine-sediment studies. In coarse-grained marine sediments, one would expect that Pu is mainly associated with Fe- and Mn-oxide particle coatings, organic matter, and apatite phases; this association with several different solid phases is significantly more complex than the simple associations between Cs and clays.

Nevertheless, Pu has not found widespread application in sediment studies. This is apparently because the traditional analytical approach for Pu determinations, alpha spectrometry (Livingston et al., 1975; Vajda et al., 2009), involves a time-consuming sequence of radiochemical separations, source preparation, and lengthy counting times (i.e., several days for low-activity samples). Since a large-scale sediment study can entail collection of many sediment cores and the subsequent analyses of several hundred individual core intervals, alpha spectrometry is rendered impractical for generating Pu-activity data for these high-throughput analytical needs.

Over the past 10–15 years, ICP-MS has experienced significant advances in the sensitivity and reliability of commercially available instruments. The ICP-MS is now widely used to measure the concentrations and isotopic ratios of a number of long-lived radionuclides (Becker, 2005), and is well suited for routine determination of all of the Pu isotopes except ^{238}Pu. Both sector-field ICP-MS systems and quadrupole ICP-MS systems have been used successfully for routine Pu determination (Ketterer and Szechenyi, 2008), and ICP-MS is quite competitive or superior to alpha spectrometry in terms of detection limits for ^{239}Pu and ^{240}Pu. Some studies have demonstrated detection limits as low as 0.5 fg g^{-1} (Kenna, 2002). In comparison, a rough equivalent in terms of radioactivity is a single disintegration per day, which is not practically measurable using decay counting. The improvements in sensitivity for ICP-MS vs. alpha spectrometry and, particularly, improvements in sample throughput vs. alpha spectrometry (minutes vs. days per sample), generate many new opportunities for extending the use of $^{239+240}$Pu as a powerful tracer of recent processes in the Earth′s surface environment.

In this study, we investigate the application of $^{239+240}$Pu chronostratigraphy in two environments: (A) a low-fallout, Southern Hemisphere shelf site in New Zealand dominated by riverine inflow, and (B) a sandy environment of the inner continental shelf of North Carolina, USA. The major goal herein is to conduct a "proof-of-principle" investigation of ICP-MS-based $^{239+240}$Pu-activity data for applications in chronostratigraphy of sandy sediments.

2. Study sites

Core samples were collected in 2005 from two environments where previous attempts to measure ^{137}Cs by gamma spectrometry were unsuccessful: (1) a muddy-shelf environment off the Waipaoa River, New Zealand (Fig. 1A), and (2) a sandy nearshore environment off Kill Devil Hills, North Carolina (Fig. 1B).

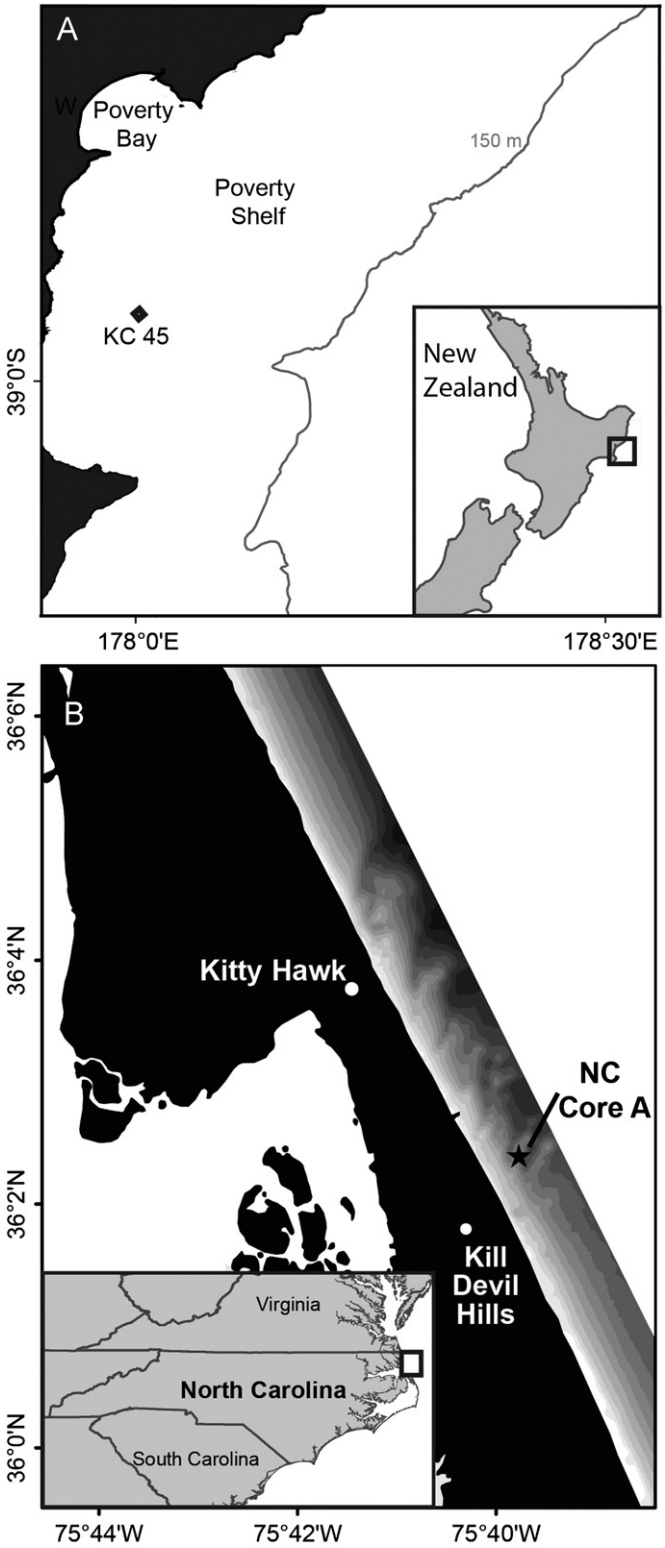

Fig. 1. (A) Study area off the Waipaoa River (entering Poverty Bay), New Zealand. The kasten core (KC45) location is shown by a solid triangle, in 50 m of water depth. (B) Study area off North Carolina, USA. The location of core A described herein is indicated by the black star, in 8.5 m of water depth.

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