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Manmade and natural radionuclides in north east Atlantic shelf and slope sediments: Implications for rates of sedimentary processes and for contaminant dispersion

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Abstract

Results are presented for a study of manmade and natural radionuclides in north east Atlantic continental shelf and slope sediments to the west of Scotland. The data are interpreted in the context of sediment mixing and accumulation processes and are used to establish the westward extent of contamination of the sediment system.

Offshore shelf and slope sediments were found to have post-glacial sedimentation rates of the order of 1 cm ky^{-1} but nearshore sediments had much higher accumulation rates of the order of 0.1 cm y^{-1}. Surface mixed layer depths of up to 6 cm were observed and non-local mixing affected most of the slope sediments, resulting in advective transport of surface sediment to depths of up to 10 cm. Biodiffusion coefficients for offshore shelf and slope sediments were dominantly in the range 10^{-8} to 10^{-9} cm^2 s^{-1}.

The study confirmed that seawater contaminated with Sellafield waste radionuclides is dominantly entrained to the east of 7° W and, consistent with this, higher levels of Sellafield derived radionuclides were confined to nearshore sediments, with lower levels to the west of 7° W. $^{238}Pu/^{239,240}Pu$ data indicated that Sellafield contributed 75–91% of the total plutonium in coastal sediment but only about 4–8% of the total in slope sediments. By analogy, it can be concluded that a similar situation will apply to other contaminants in seawater entering the north east Atlantic via the North Channel.

Keywords: North east Atlantic; Shelf and slope sediments; Mixing; Accumulation; Radionuclides; Contaminant dispersion

1. Introduction

Quantitative characterisation of sediment accumulation and mixing processes is of fundamental importance in developing an understanding of marine biogeochemical cycles of both natural and anthropogenic species. The use of natural and manmade radionuclides to establish sediment accumulation rates and mixing characteristics is of central importance in this context and, while such studies have been applied in nearshore coastal regions of the west coast of Scotland (Baxter et al., 1979; Swan et al., 1982; McKay and Baxter, 1985; Williams et al., 1988) and in deep ocean areas of the north east Atlantic (Thomson et al., 1993, 1995a,b, 2000; Brown et al., 1999, 2000; Hughes et al., 2005), little attention has

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been devoted to the extensive area of shelf and slope sediments to the west of Scotland. This has, in part, been due to difficulties in sampling since, with negligible fluvial input, the sediments in this area are generally coarse-grained, carbonate sands, which are inherently unsuitable for coring. Fine sediment, which is suitable for coring, is restricted to limited areas, generally occupying localised depressions in the seabed.

The use of anthropogenic radionuclides as tracers of sedimentary processes in north east Atlantic shelf and slope sediments is complicated by the fact that there has been input from a number of sources, the dominant being discharges from the Sellafield nuclear fuel reprocessing plant in north west England and fallout from atmospheric testing of nuclear weapons. Deposition of weapons testing fallout occurred mainly in the 1950s and early 1960s, with peak input in 1963 (Carter and Moghissi, 1977; UNSCEAR, 1993; MacKenzie, 2000). In the case of plutonium, an additional significant source of atmospheric fallout was from the accidental burn up in the atmosphere in 1964 of a US Satellite containing 1 kg of ^{238}Pu in a SNAP-9A system for auxiliary nuclear power (Hardy et al., 1973). Sellafield has released low level liquid radioactive waste into the Irish Sea since 1952, with peak discharges in the mid 1970s (Gray et al., 1995). Particle reactive radionuclides released from Sellafield were dominantly retained in Irish Sea sediments (Hetherington, 1975; Kershaw et al., 1983, 1984, 1992, 1999a; Hunt, 1985; MacKenzie et al., 1987, 1994, 1998; McCartney et al., 1994; Mitchell et al., 1995, 1999; Charlesworth et al., 2006), whereas more soluble species were transported north along the Scottish coast in solution in seawater (Jefferies et al., 1973; Baxter et al., 1979; McKinley et al., 1981a,b) and eventually to more distant areas (Murray et al., 1978; Livingston et al., 1982; Holm et al., 1983; Kershaw and Baxter, 1995; Kershaw et al., 1995, 1999b; Leonard et al., 1997, 2004; Heldal et al., 2002; Matishov et al., 2002). In the period following the major reduction in Sellafield discharges, redissolution of radiocaesium and, to a lesser extent, plutonium (Hunt and Kershaw, 1990; McCartney et al., 1994; Cook et al., 1997) has resulted in contaminated sediments acting as a secondary source of anthropogenic radionuclides to seawater leaving the Irish Sea.

Transfer of some of the Sellafield-derived radionuclides from solution to the solid phase during transport in the north east Atlantic has resulted in contamination of underlying sediments, but the extent of contamination of slope sediments is undefined and the only reported study of anthropogenic radionuclides in shelf sediments of this area is that of Livingston and Bowen (1977), which involved a single sediment core collected from The Minch (58° 14.6′ N; 05° 49.5′ W) in May 1976. In this study, specific activities of ^{137}Cs were observed to decrease from 27.7 Bq kg^{-1} at the surface to 3.2 Bq kg^{-1} at 12–14 cm, while ^{239,240}Pu showed a corresponding decrease from 1.4 Bq kg^{-1} to 0.01 Bq kg^{-1}. The ^{238}Pu/^{239,240}Pu activity ratio of 0.092 for the surface sediment, was intermediate between the value of about 0.034, which applied to total fallout (weapons testing plus SNAP accident) at this latitude at that time (Hardy et al., 1973), and that of the Sellafield discharge, which had increased from about 0.026 at the start of operations in the early 1950s to about 0.2 by the mid 1970s (Gray et al., 1995). The ^{238}Pu/^{239,240}Pu activity ratio decreased to 0.038 at depth in the core, consistent with the source being either weapons testing fallout or early Sellafield discharges. Inventories of ^{137}Cs and ^{239,240}Pu were 2046 Bq m^{-2} and 115 Bq m^{-2}, respectively. Livingston and Bowen (1977) concluded that the major source of these radionuclides was Sellafield, but noted that there would also be a component from atmospheric fallout and this is supported by the ^{238}Pu/^{239,240}Pu data above, which with the assumption that the plutonium is a binary mixture of fallout and Sellafield-derived components as outlined above, imply a Sellafield contribution of some 37% in the surface sediment of the Minch at that time.

The paucity of analytical data and the complicating factor of input of Sellafield waste radionuclides since 1952 mean that direct evaluation of fallout radionuclide concentrations and inventories in the north east Atlantic shelf and slope sediments is not possible. However, likely ranges for these parameters can be inferred from studies in the north west Atlantic in areas that are relatively remote from nuclear industry sources. For example, Livingston and Bowen (1979) reported data for cores collected in 1975 from an area of fine grained sediments in Wilkinson Basin on the continental shelf of the north west Atlantic, between latitudes 42° 22.0′ N and 43° 0.00.5′ N. Specific activities of ^{137}Cs and ^{239,240}Pu decreased with depth from surface maxima of 2.4 to 5.4 Bq kg^{-1} and 0.37 to 1.7 Bq kg^{-1}, respectively, to values close to, or lower than, the detection limit at depths below 10 cm. The ^{137}Cs/^{239,240}Pu inventory ratio varied in the range 3.70 to 6.67, while the ^{238}Pu/^{239,240}Pu activity ratio ranged from 0.02 to 0.056. Buesseler et al. (1985/86) presented a compilation of available data for north west Atlantic shelf sediments between approximately 31° N and 45° N, which revealed that fine grained shelf sediments generally had ^{239,240}Pu inventories in the range 8.1 to 207 Bq m^{-2}. However, a study by Benninger and Krishnaswami (1981) revealed significantly higher ^{239,240}Pu inventories of up to 932 Bq m^{-2} in fine grained sediments of the New York Bight. In addition, Buesseler et al. (1985/86) noted