

Deposition of artificial radionuclides in sediments of Loch Etive, Scotland

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ABSTRACT

The nuclear fuel reprocessing plants on the Sellafield site (UK) have released low-level effluents into the Irish Sea under authorisation since 1952. This has led to the labelling of nearby offshore sediments with a range of artificial radionuclides. In turn, these sediments act as a long-term secondary source of both soluble and particle-associated radionuclides to coastal areas. These radionuclides are of interest both in assessing possible environmental impacts and as tracers for marine processes. Here we present results from a study of the geochemistry of natural (²³⁴U, ²³⁸U) and artificial (¹³⁷Cs, ²⁴¹Am, ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, and ²³⁶U) radionuclides and their accumulation in sediments from Loch Etive, Scotland. The data are interpreted in the context of the historical radioactive discharges to the Irish Sea and biogeochemical processes in marine sediments. Loch Etive is divided into two basins; a lower, seaward basin where the sedimentation rate (~0.6 cm/yr) is about twice that of the more isolated upper basin (~0.3 cm/yr). These accumulation rates are consistent with the broad distribution of ¹³⁷Cs in the sediment profiles which can be related to the maximum Sellafield discharges of ¹³⁷Cs in the mid-1970s and suggest that ¹³⁷Cs was mainly transported in solution to Loch Etive during that period. Enrichments of Mn, Fe, and Mo in sediment and porewater from both Loch Etive basins result from contemporary biogeochemical redox processes. Enrichments of ²³⁸U and ²³⁴U in the lower basin may be a result of the cycling of natural U. By contrast, the Sellafield-derived artificial isotope ²³⁶U does not seem to be affected by the redox-driven reactions in the lower basin. The ²³⁸Pu/^{239,240}Pu ratios suggest contributions from both historical Sellafield discharges and global fallout Pu. The uniform sediment distributions of Pu and Am, which do not reflect Sellafield historical discharges, suggest the existence of a homogenous secondary source. This could be the offshore 'mud patch' in the vicinity of Sellafield from which the supply of radionuclides reflects time-integrated Sellafield discharges. This source could also account for the continuing supply of Cs to Loch Etive, even after substantial reductions in discharge from the Sellafield site.

1. Introduction

The behaviour of artificial radionuclides in marine sediments and their interaction with biogeochemical processes is of fundamental importance in developing an understanding of their long-term fate in the environment. Such understanding can be used to underpin both radionuclide tracing of environmental processes and assessment of possible environmental impacts of radionuclide releases to coastal waters, such as occurred in 2011 as a result of the accident at the Fukushima Daiichi Nuclear Power Plant.

The absence of oxygen in sediments drives successive reduction of other terminal electron acceptors (e.g. nitrate, Fe and Mn oxyhydroxides, and sulfate) by anaerobic microorganisms. These processes

could impact the fate of natural and artificial radionuclides in sediments either by direct cycling of redox sensitive radionuclides by anaerobic microorganisms or by interaction of radionuclides with biogeochemical redox cycles of other elements (Brookshaw et al., 2012; Campbell et al., 2015; Kimber et al., 2012; Lovley et al., 1991; Malcolm et al., 1990). Because of the biogeochemical gradients present, sea-lochs provide an ideal natural laboratory to explore these interactions (Sholkovitz, 1983; Williams et al., 1988).

Loch Etive is a sea-loch on the west coast of Scotland with well documented geochemistry and hydrography (Edwards and Edleston, 1977; Overnell, 2002; Ridgway and Price, 1987) which provides perfect conditions (high sedimentation rates, anaerobic conditions, restricted exchange, and reduced erosion) to investigate the geochemical

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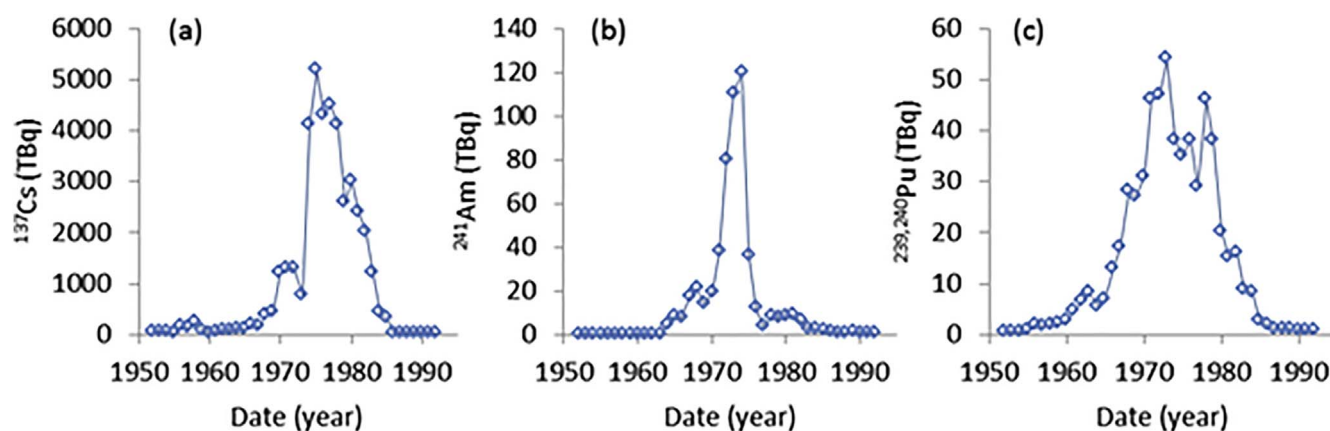


Fig. 1. Total annual quantities of (a) ^{137}Cs , (b) ^{241}Am , and (c) $^{239,240}\text{Pu}$ released from Sellafield between 1952 and 1992 (data from Gray et al., 1995).

behaviour of natural and artificial radionuclides. Sellafield, a nuclear site located on the NW coast of England, is the dominant source of artificial radionuclides in the NE Irish Sea. Authorised radioactive discharges of low level liquid effluents from Sellafield to the Irish Sea started in 1951 with a maximum in the mid 1970's (Fig. 1) (Gray et al., 1995). Other sources, such as weapons testing which had a peak input in 1963 (MacKenzie, 2000), and accidental releases, particularly the Chernobyl accident in 1986 (Camplin et al., 1986; Mitchell et al., 1986) have made smaller contributions to the radioactive inventory of the Irish Sea. Further, accidental release of ^{238}Pu from the burn-up of a US satellite in the atmosphere in 1964 increased the activity ratio of $^{238}\text{Pu}/^{239+240}\text{Pu}$ from global fallout in the northern hemisphere from 0.024 to 0.036 (Hardy et al., 1973).

Due to the seawater circulation in the NE Irish Sea, the discharged radionuclides disperse northwards (Jefferies et al., 1973; Mitchell et al., 1999). Soluble radionuclides such as Cs mostly remain in seawater and are transported northwards along the Scottish coast out of the Irish Sea (Dahlgard, 1995; Jefferies et al., 1973; Kershaw and Baxter, 1995), whereas Pu and Am (particle-reactive species) mainly associate with suspended particles and are then focussed into areas of muddy sediment (the 'mud patch') close to Sellafield. This mud patch then acts as a long-term secondary source of soluble and particle-associated artificial radionuclides, the composition of which are time-integrated due to sediment mixing processes (Brown et al., 1999; Kershaw et al., 1984; MacKenzie et al., 1998). The ingrowth of ^{241}Am from decay of ^{241}Pu ($t_{1/2} = 14.3$ years) has also dominated the direct discharges of ^{241}Am from Sellafield since the late 1970s (Hunt et al., 2013).

Uranium occurs naturally almost entirely as a mixture of three isotopes: ^{238}U , ^{234}U , and ^{235}U but natural ^{236}U also exists in the environment in ultra-trace quantities with a $^{236}\text{U}/^{238}\text{U}$ atom ratio of $\sim 10^{-14}$ (Zhao et al., 1997). Uranium is generally soluble in oxic seawater and present as dissolved U(VI) complexed with carbonate (Cochran et al., 1986), which may also inhibit reduction to U(IV). Diffusion of seawater to the underlying sediments may result in the reduction of U(VI) to insoluble U(IV) at about the depth of Fe(III) reduction, which can lead to accumulation of U at depth in the sediment (Barnes and Cochran, 1990; Cochran et al., 1986). The Sellafield discharges to sea contain irradiated U which would be masked by the natural U in seawater (Zhao et al., 1997) and historical, non-nuclear discharges to the Irish Sea from the Marchon phosphate plant on the Cumbrian coast (Kershaw et al., 1990). However, irradiated U is unique in its enhanced level of ^{236}U ($t_{1/2} = 2.3 \times 10^7$ years) (Zhao et al., 1997) which provides a clear signature for irradiated U and hence, in this setting, Sellafield-derived U, and can be used as a tracer (Al-Qasmi et al., 2016; Marsden et al., 2006). However, ^{236}U has not been widely used as a tracer of environmental processes due to the difficulties of detecting its low mass or activity concentrations with traditional methods. Accelerator mass spectrometry can overcome these difficulties

(Eigl et al., 2013; Zhao et al., 1997).

The inventory of naturally occurring ^{210}Pb ($t_{1/2} = 22.3$ years) in marine sediment has two components: supported and unsupported. The supported Pb is generated *in situ* by decay of ^{226}Ra contained in the sediment minerals themselves. The term 'supported' reflects the fact that this component is in radioactive equilibrium with its parent isotopes and its activity concentration will therefore not change over the timescales of interest. The unsupported Pb originates from decay of ^{222}Rn and this accumulates in surface sediments. Since it is not in radioactive equilibrium with its parent isotopes, the rate at which unsupported ^{210}Pb decays allows estimation of the rate at which the sediment accumulates (Swan et al., 1982). The ^{210}Pb dating method has been widely used to date cores from sea-lochs from the west coast of Scotland (Krom et al., 2009; Teasdale et al., 2011).

The main aim of this paper is to investigate relationships between artificial radionuclides released from Sellafield over a well-defined time period and the biogeochemical cycles which are well documented in Loch Etive, the study site. This will provide deeper understanding of the behaviour of natural and artificial radionuclides in the marine environment, underpinning both assessments of possible environmental impacts and radionuclide tracing of environmental processes.

2. Methods

2.1. Study site and sampling

Loch Etive is a sea-loch on the west coast of Scotland which can essentially be divided into two distinct parts, a lower and upper basin (Fig. 2). The seaward, lower basin has a maximum depth of ~ 70 m and experiences tidally-induced mixing, leading to an oxygenated water column (Overnell et al., 2002). The upper basin is separated from the lower by a submerged sill of 13 m depth and has a maximum depth of 145 m (Howe et al., 2002). In contrast to the lower basin, which is tidally mixed, only the surface water of the upper basin is tidally exchanged. As a result, the bottom water of the upper basin is only exchanged irregularly depending on freshwater input, with a mean repetition time of 16 months (Edwards and Edelsten, 1977) and as a result it is often hypoxic. These environmental conditions are also reflected in the underlying sediments, which generally show oxic conditions in the top 5 cm of surface sediments in the lower basin, and suboxic conditions in top 1 cm in the upper basin (Ridgway and Price, 1987). The upper basin surface sediment is high in Mn beneath the generally hypoxic waters, contrasting with lower surficial Mn in the lower basin beneath water which is usually well mixed (Overnell et al., 2002). Malcolm (1985) studied the geochemistry of Mo in both basins of the loch and established an association of Mo geochemistry with the cycling of organic matter, Mn and sulfur. Previous studies of radioactivity in Loch Etive have used ^{210}Pb and ^{137}Cs as dating tools and showed

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