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Origin of artificial radionuclides in soil and sediment from North Wales

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ABSTRACT

During the operations at the Sellafield nuclear fuel reprocessing complex, artificial radionuclides are discharged to the Irish Sea under authorisation, where they are dispersed. In this study, the southern distribution and transport of Sellafield derived radionuclides have been investigated. Both natural and artificial radionuclides have been studied in a soil core from the riverbank of the Afon Goch in Anglesey, North Wales. Particulate input is dominant for all artificial radionuclides (including the more soluble ^{137}Cs and ^{236}U) with an estimated lag time of about a decade. The preferential northward seawater movement in the NE Irish Sea limits solution input of ^{137}Cs and ^{236}U to the areas south of Sellafield. The relatively long lag time reflects both the water circulation pattern and distance between the study site in north Wales and the source point in Cumbria. Two redox active zones are observed in the top and the bottom of this core, although there is no evidence for any redistribution of Pu and natural uranium by these redox processes. However, ^{236}U, derived from irradiated uranium, showed variable distribution in the core. This could be a potential response to the geochemical conditions, showing that ^{236}U may be a promising tracer for the environmental processes and a signature of the Sellafield historical discharges of irradiated uranium.

1. Introduction

The Sellafield nuclear fuel reprocessing plant in northwest England is the dominant source of anthropogenic radioactivity to the Irish Sea. Accidental releases such as Chernobyl (1986) and global fallout from weapons testing have also contributed to the Irish Sea's inventory of artificial radionuclides. Permitted discharges of a complex mixture of radionuclides from Sellafield to the Irish Sea in low level effluents have occurred since 1951 (Gray et al., 1995; Kershaw et al., 1990). These discharges have changed considerably from 1951 until present, with a maximum in the mid-1970s (Fig. 1) (Gray et al., 1995; MacKenzie et al., 1994). Thereafter, the discharges declined significantly because of improvements in effluent treatment (Sellafield Ltd, 2013). However, a proportion of the historical discharges has been retained in offshore sediments which act as a long-term secondary source of soluble and particle associated artificial radionuclides.

Particle-reactive radionuclides such as Pu and Am predominantly associate with suspended particles and sediments close to the discharge point and then a fraction of this material transports to intertidal areas (Kershaw et al., 1995; MacKenzie et al., 1987). By contrast, more soluble radionuclides such as Cs are relatively conservative in seawater and are mostly dispersed northwards along the Scottish coast into the North Sea and beyond (Dahlgaard, 1995; Jefferies et al., 1973; Kershaw and Baxter, 1995). Although solution transport is more important for Cs, a fraction of Cs (~10%) is retained in the sediment and transports with particulates (Jefferies et al., 1973). Natural uranium in general is soluble in seawater and is present as U(VI) (Cochran et al., 1986) and any uranium released from Sellafield to the environment would be diluted with the natural uranium background (Zhao et al., 1997) and historical, non-nuclear discharges from a phosphate plant (Marchon) at Whitehaven (Kershaw et al., 1990). However, ^{236}U ($t_{1/2}$ 2.3 × 10^7 years) is a unique marker for irradiated, and hence Sellafield-derived, uranium and could be used as a tracer to study its transport and accumulation. So far, very few studies have used ^{236}U as a tracer of environmental processes because traditional methods are not sensitive enough to measure the low mass or activity concentrations of ^{236}U and accelerator mass spectrometry

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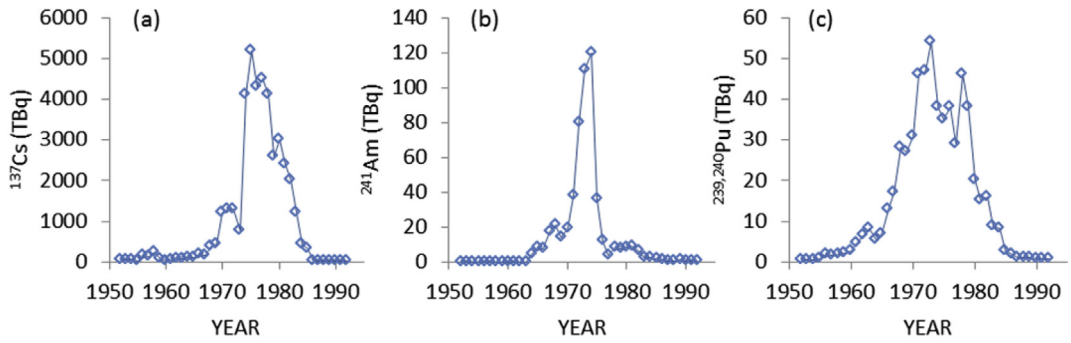

Fig. 1. Temporal variations in the annual quantities of (a) ^{137}Cs, (b) ^{241}Am and (c) ^{239,240}Pu discharged from the Sellafield nuclear fuel reprocessing plant (data from Gray et al., 1995).

needs to be used for this purpose (Eigl et al., 2013; Zhao et al., 1997). The dispersion of Sellafield-derived, anthropogenic radionuclides has been extensively studied in the area around and to the north of Sellafield because of the preferential northward seawater movement in the Irish Sea (Jefferies et al., 1973; Mitchell et al., 1999). However, relatively little attention has been devoted to Wales, south of Sellafield (Fig. 2). For example, Garland et al. (1989) investigated the presence and transfer of radionuclides from sea to land on the Welsh coast in 1986. They detected measurable activities of Pu isotopes, ^{241}Am and ^{137}Cs along the north coast and noticed sea to land transfer of Pu and Am. After 15 years of this study, Bryan et al. (2008) provided an update on these data and found a consistent results with no increase in the supply of these Sellafield-derived radionuclides to the north Wales coast. Their results also suggested that sea to land transfer of Pu was occurred by either direct inundation or deposition of marine aerosol. Parys Mountain is an abandoned copper mining site in Anglesey, North Wales. The mine has led to contamination of the Afon Goch and the

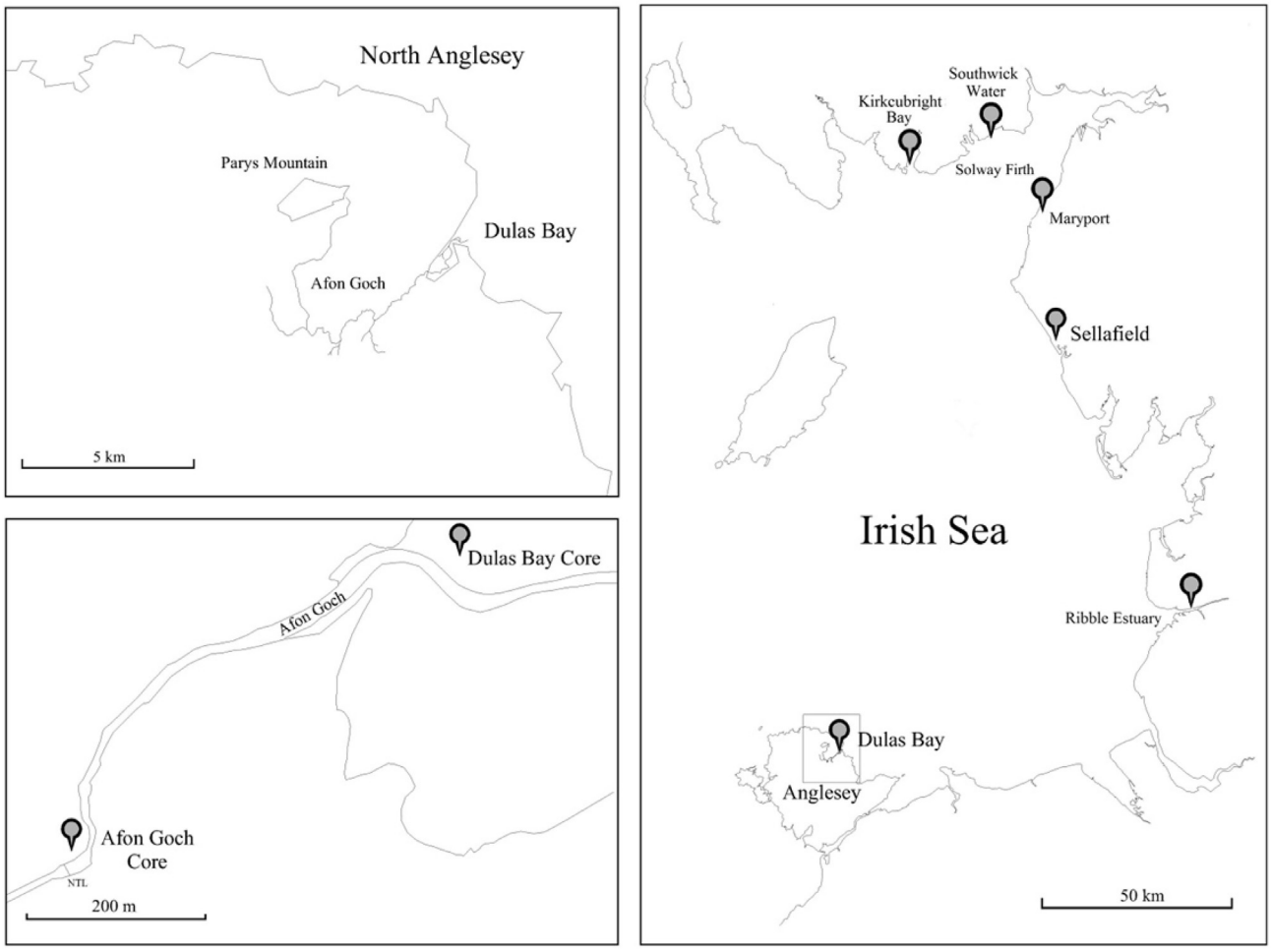

Fig. 2. Right: A map showing the study site in Anglesey and the Sellafield site; Top left: North Anglesey showing Parys Mountain, the Afon Goch and Dulas Bay; Bottom left: Locations of the Afon Goch core (described here) and the Dulas Bay core (results are described in Al-Qasmi et al., 2015).