Journal of Environmental Radioactivity 151 (2016) 94–104

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Spatial trends on an ungrazed West Cumbrian saltmarsh of surface contamination by selected radionuclides over a 25 year period

Jane A. Caborn [a, b, *], Brenda J. Howard [a], Paul Blowers [c], Simon M. Wright [a]

[a] Centre for Ecology & Hydrology – Lancaster, Lancaster Environment Centre, Library Avenue, Bailrigg, Lancaster, LA1 4AP, UK
[b] NNL, Preston Laboratory, A709, Springfields, Salwick, Preston, PR4 0XJ, UK
[c] Cefas Lowestoft Laboratory, Pakefield Road, Lowestoft, Suffolk, NR33 0HT, UK

ARTICLE INFO

Article history:
Received 12 December 2014
Received in revised form
7 September 2015
Accepted 9 September 2015
Available online 3 October 2015

Keywords:
Saltmarsh
Radionuclides
Spatial variation
Temporal variation
Sedimentation
Monitoring

ABSTRACT

Long term spatial and temporal variations in radionuclide activity have been measured in a contaminated ungrazed saltmarsh near Ravenglass, Cumbria. Over a twenty–five year period there has been a decrease in activity concentration with ^{106}Ru and ^{137}Cs showing the highest rate of change followed by Pu alpha and ^{241}Am. A number of factors contribute to the reduction with time; including radiological half lives, discharge and remobilisation. For ^{241}Am the lower reduction rate is partially due to ingrowth from ^{241}Pu and partially as a result of transport of sediment from the offshore Irish Sea mud patch. Considerable spatial variation for the different radionuclides was observed, which with time became less defined. The highest activity concentrations of long-lived radionuclides were in low energy areas, typically where higher rates of sedimentation and vegetation occurred. The trend was reversed for the shorter lived radionuclide, ^{106}Ru, with higher activity concentrations observed in high energy areas where there was frequent tidal inundation. Surface scrape samples provide a pragmatic, practical method of measuring sediment contamination over large areas and is a sampling approach adopted by most routine environmental monitoring programs, but it does not allow for interpretation of the effect of variation in sedimentation rates. This paper proposes a method for calculating indicative sedimentation rates across the saltmarsh using surface scrape data, which produces results consistent with values experimentally obtained.

1. Introduction

Between 1952 and 1992 the reprocessing facilities on Sellafield Limited nuclear site in NW England have, under authorization, released liquid effluents containing low levels of activity into the Irish Sea. The liquid effluents contained actinide and fission elements which are discharged via pipeline and are comprised of the purge water from waste storage ponds and process liquors from spent fuel reprocessing. Discharge histories showed the activity concentrations for radionuclides including ^{137}Cs, Pu-α and ^{241}Am reached a maximum in the 1970s and then substantially declined (Gray et al., 1995).

Once discharged, the radionuclides become attached to varying degrees dependent on particle reactivity to sedimentary particles with some becoming incorporated by sedimentary deposition and suspension processes into intertidal and estuarine environments resulting in the saltmarshes being contaminated with a wide range of radionuclides (Howard et al., 1986). Parallel to the coastline is an area of mud and muddy sediments (approximately 15 km long × 3 km wide) commonly known as the 'mud patch' (Kershaw et al., 1992; Mackenzie et al., 1994; Pentreath et al., 1984). Fine grained particles with associated radionuclides accumulate on the patch as a result of tidal movement and currents in the shallow Western Irish Sea basin (depth approximately 30 m) (Hetherington, 1978a, b; MacKenzie et al., 1994). Particulates are redistributed and deposited onto the saltmarsh as a result of tidal processes and storm events.

The Ravenglass estuary (NW, England) is one of the most radioactively contaminated saltmarshes within the Irish Sea and provides a unique resource for understanding the behaviour of radionuclides in the environment. This study focuses on the spatial

Abbreviations: dw, dry weight; CEH, Centre of Ecology and Hydrology; GAU, Geosciences Advisory Unit; NPL, National Physical Laboratory; IAEA, International Atomic Energy Authoirty; PIPS, passivated implanted planar silicon.

* Corresponding author. NNL, Preston Laboratory, A709, Springfields, Salwick, Preston, PR4 0XJ, UK.
E-mail address: jane.a.caborn@nnl.co.uk (J.A. Caborn).

http://dx.doi.org/10.1016/j.jenvrad.2015.09.011
0265-931X/

and temporal changes in the activity concentrations of deposited radionuclides in surface sediment over a period of 25 years, determines sedimentation rates across the ungrazed saltmarsh and relates the data to discharge history.

2. Experimental site and sampling details

A survey in 1980 described the spatial distribution of activity concentrations for various radionuclides in the surface silts of an ungrazed saltmarsh in the River Esk estuary, Cumbria (Horrill, 1983). Further surface samples were taken in 1992 and 2005 by the Centre for Ecology and Hydrology (CEH), based on the sampling grid established in 1980, to enable assessment of the long term environmental processes for the 25 year period. Additional reported data from a study conducted in 1997 by Oh et al. (2009) have also been included.

Ravenglass saltmarsh is situated approximately 10 km south of the Sellafield site in the River Esk Estuary in Cumbria, UK. The site is on the northern shore of the River Esk (National Grid reference SD089948), inland of a railway viaduct (Fig. 1). The sampling area was 300 × 250 m.

2.1. Site sampling

The site was originally selected in 1980 on the basis that the total gamma radiation levels, using a field ratemeter, were relatively high compared with other saltmarshes in the estuary. In 1980 a permanent 25 m survey grid was established on the marsh (Fig. 1) with a total of 100 sampling points. In August 1992 and March 1997 a survey of all the sample points was carried out similar to that of the original study of July 1980 (Horrill, 1983). A subsequent, less intensive survey was carried out in July 2005 comprising of 26 sampling points (alternate rows and columns).

For each of the surveys, a 250 × 250 mm quadrant was placed at the grid point. The vegetation within the quadrant clipped and collected taking care to avoid the inclusion of surface silt. The surface silt was then removed over the quadrant area by scraping to a depth of approximately 20 mm. In many cases the sample silt included a considerable portion of root mat.

2.2. Site characteristics

A diagram of the saltmarsh in 1980 and the associated sampling grid is given in Fig. 1. Horrill (1983) reported a linear trend in vegetation distribution across the saltmarsh. The size and shape of the saltmarsh has changed over time due to continuous accretion and erosion. By 1992, an erosion bank had developed at the lower end of the marsh and three sampling sites were lost (B10, X8 and D8). The vegetation cover on the saltmarsh was similar to that recorded during the original survey (Horrill, 1983).

By 2005 it was observed that the degree of erosion had increased significantly, markedly reducing the number of available sampling points near the river. A new triangular area of saltmarsh covered in vegetation was present at the lower end of the sampling transect (Fig. 2). Over the sampling period, the extent of vegetation biomass on the remaining saltmarsh has markedly increased except at sites B5, C5 and C7 where there was no vegetation coverage.

3. Radiochemical analysis

Following collection, the samples were dried at 105 °C, ground and analysed for radionuclide content. CEH Lancaster (formerly ITE Merlewood) performed the analysis for the 1980, 1992 and 2005 surveys. The 1997 survey was conducted by the Geosciences Advisory Unit (GAU), Southampton (Oh et al., 2009).

The measurement of gamma-emitting radionuclides (^{106}Ru

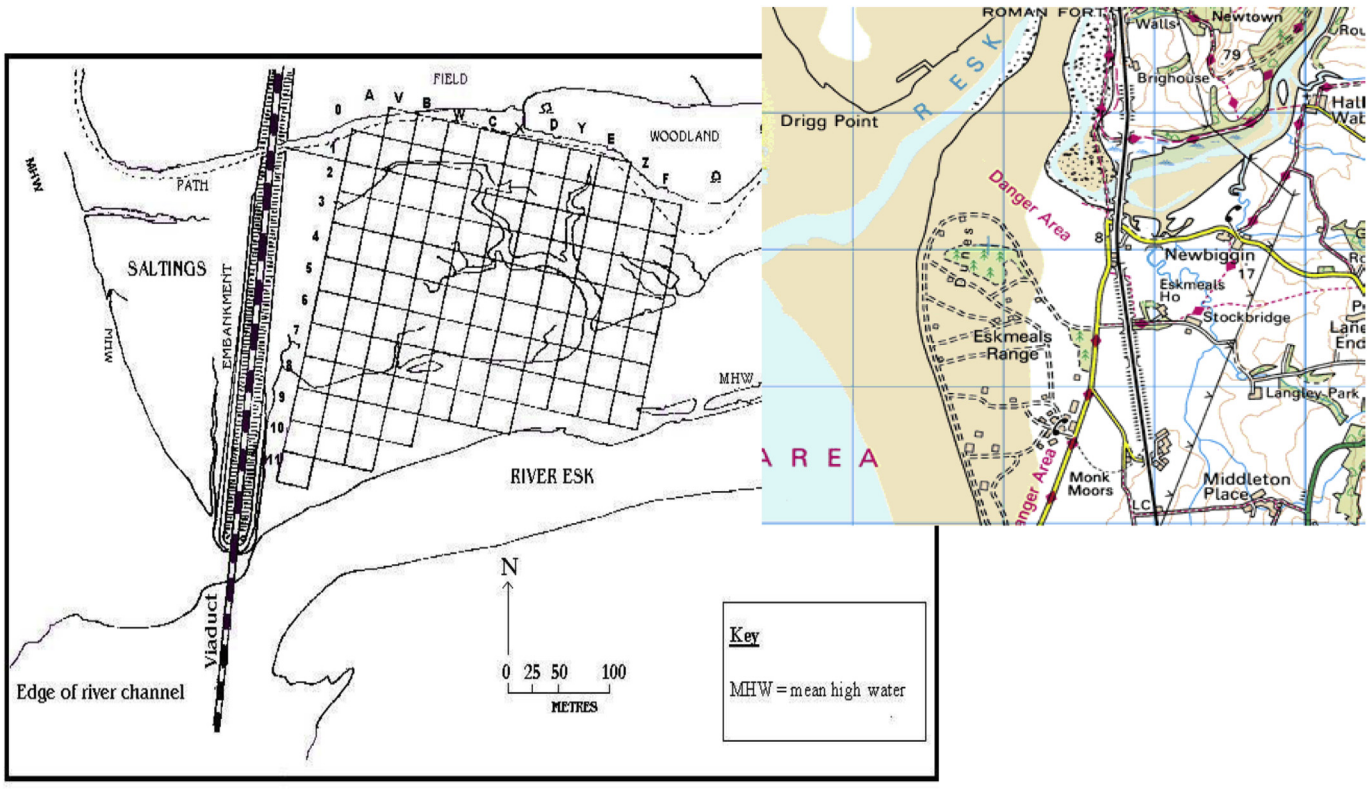

Fig. 1. Location of saltmarsh and sampling grid (Ordnance Survey, 2006; Horrill, 1983).