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Effects of complex hydrodynamic processes on the horizontal and vertical distribution of Tc-99 in the Irish Sea

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ABSTRACT

The increased discharge of Tc-99 from the Sellafield plant following the commissioning of the Enhance Actinide Removal Plant in 1994 was reflected in higher Tc-99 activity concentrations over much of the Irish Sea. The presence of this radionuclide in the marine environment is of concern not only because of its long half life but also high bio-concentration factor in commercially valuable species, such Norway lobster (*Nephrops norvegicus*) and common lobster (*Homarus gammarus*). Accurate predictions of the transport, and spatial and temporal distributions of Tc-99 in the Irish Sea have important environmental and commercial implications. In this study, transport of the Tc-99 material was simulated in order to develop an increased understanding of long-term horizontal and vertical distributions. In particular, impact of seasonal hydrodynamic features such as the summer stratification on the surface-to-bottom Tc-99 ratio was of interest. Also, material retention mechanisms within the western Irish Sea were explored and flushing rates under various release conditions and meteorological forcing were estimated.

The results show that highest vertical gradients are observed between June and July in the deepest regions of the North Channel and the western Irish Sea where radionuclide-rich saline-poor water overlays radionuclide-poor saline-rich Atlantic water masses. Strong correlation between top-to-bottom ratio of Tc-99 and strength of stratification was found. Flushing studies demonstrate that as the stratification intensifies, residence times within the western Irish Sea increase. In stratified waters of the gyre Tc-99 material is flushed out from the upper layer much quicker than from the bottom zone.

The research also shows that in the gyre the biologically active upper layers above the thermocline are likely to contain higher concentrations than the near-bed region. Long-term horizontal and vertical distributions as determined in this study provide a basis for assessment of a potential biota exposure to Tc-99.

1. Introduction

Technetium-99 (Tc-99) is a long-lived ($t^{1/2} = 2.13 \times 10^5$ years) pure β-emitter (Lederer et al., 1967), which in marine environment forms the soluble pertechnetate ion (TcO_4^-). The controlled release of this radionuclide into the Irish Sea from the nuclear fuel reprocessing plant at BNFL Sellafield, in the north-west of England (see Fig. 1 for location), has been ongoing since the beginning of reprocessing in 1952. Since 1994 the commissioning and operation of the Enhanced Actinide Removal Plant (EARP) treatment has resulted in elevated discharges of Tc-99 to the Irish Sea. Between 1993 and 1995 a thirty-fold increase in discharges of Tc-99 occurred (Long et al., 1998a). This has been reflected in an increase in the activity concentration of this radionuclide at all east coast sampling sites between 1994 and 1999 (Ryan et al., 2000).

Activity concentrations in fish and shellfish from the Irish Sea increased in line with increased discharges from Sellafield (Smith et al., 2009). The accumulation and retention Tc-99 in marine organisms has been well documented (Hunt et al., 1998; Smith et al., 2001; Nowakowski et al., 2004; Copplestone et al., 2004; Oliver et al., 2006). Consequent doses to members of the public from seafood consumption have been also assessed (CEFAS, 2008; Ryan et al., 2000; Colgan et al., 2008).

The potential radiological impact of Tc-99 led to a considerable amount of interest in monitoring the Tc-99 discharges, particularly in the post-EARP period. The monitoring within the Irish Sea has been conducted on a regular basis by both British and Irish authorities. The marine radioactivity monitoring on behalf of the Irish Government is undertaken by the Radiological Protection Institute of Ireland (Pollard et al., 1996; Long et al., 1998b; Ryan et al., 2000; Fegan et al., 2008) and comprises of Tc-99 sampling in seawater, sediment, seaweed, fish and shellfish. On the UK side a series of monitoring reports containing the Tc-99 data (e.g. RIFE, 1998, 2000, 2005) was compiled by the Centre for Environment, Fisheries and Aquaculture Science (CEFAS).

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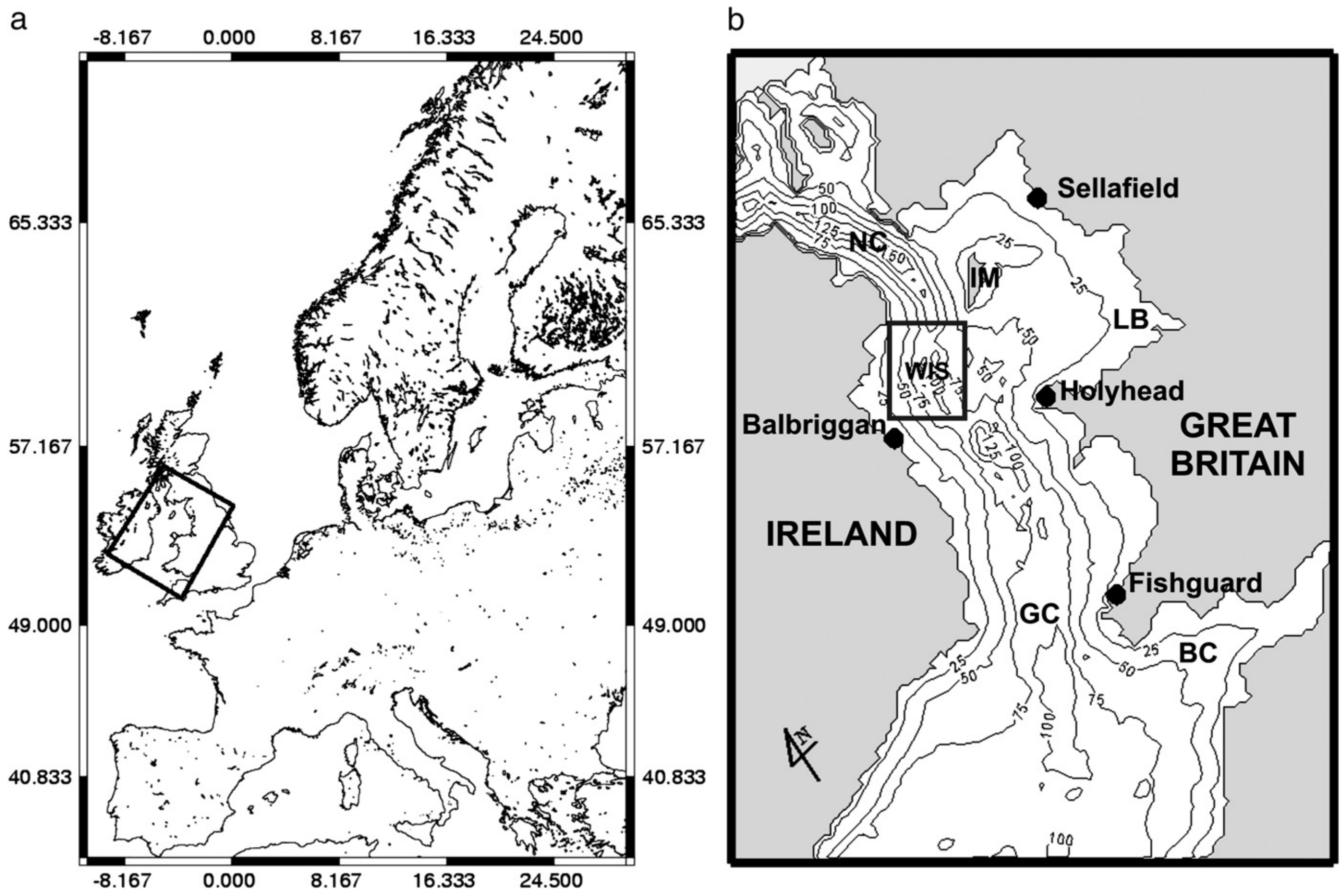

Fig. 1. (a) Irish Sea location and model orientation and (b) bathymetry of the Irish Sea and selected locations. Abbreviations: NC — North Channel, IM — Isle of Man, LB — Liverpool Bay, WIS — Western Irish Sea, GC — St. George's Channel and BC — Bristol Channel.

There is also a substantial number of published papers where Tc-99 seawater data is presented for the period considered in this study (1993–2000). A comparison of pre-EARP and post-EARP Tc-99 surface distributions in the northern Irish Sea was summarized in Leonard et al. (1997) while maps of Tc-99 surfaces distributions over the entire Irish Sea were compiled in McCubbin et al. (2002). Two expeditions aboard the Department of Agriculture for Northern Ireland (DANI) research vessel provided information on surface and bottom seawater samples (Leonard et al., 2004). This dataset allows better understanding of vertical profiles of contamination and has, therefore, important application to this study.

The accumulation of radionuclides in the sub-tidal sediments of the Irish Sea (McCubbin et al., 2006; Leonard et al., 2004; MacKenzie et al., 1998) and the northeast Atlantic continental shelf and slope sediments of Scotland (Mackenzie et al., 2006) have been the focus of much attention . The potential implications for seafood contamination from contaminated sediment have been studied by CEFAS (2008). Time series of radionuclide concentrations in sessile biota in coastal waters have been of great value in determining the extent and rate of spread of contamination by artificial radionuclides (Dahlgaard et al., 1997). However, it is difficult to account for all the observed temporal variability of Tc-99 in biota as a portion of radionuclides may be due to the seasonal effects of uptake (Masson et al., 1995). Historical data from seafood samples provided a comprehensive validation dataset due to the large sampling areas and long periods of collection. On the downside of this approach, the uncertainties were greater when dealing with these data due to mobility of the biota, the definition of the fishing areas, and the uptake and release of activity by marine organisms (Gleizon and McDonald, 2010).

The presence of Tc-99 in the marine environment is of concern not only because of its long half life but its high bio-concentration factor in commercially valuable species. The Irish Sea is a nursery area for many fish species such as cod, haddock, and whiting (Dickey-Collas et al., 1997). Its harvest of shellfish, especially Norway lobster (*N. norvegicus*) and common lobster (*H. gammarus*), is particularly valuable (Briggs, 1995). There is evidence that many of these species are associated during summer months with stratified waters of the western Irish Sea gyre (WISG) which is a retentive system (Hill et al., 1996; Emsley et al., 2005). Some species (e.g. Norway lobster) inhabit the geographically isolated muddy sediment beneath the gyre, while other species (e.g. larva and juvenile fish) occupy the photic zone above the thermocline. For this reason alone, it is important to investigate the vertical distribution of Tc-99 material. The accurate prediction of the hydrodynamics and dispersion of radionuclides in the marine environment of the Irish Sea has therefore important environmental and commercial implications.

A number of mathematical models have been recently used to improve understanding of Tc-99 fate in the marine environment. A fine resolution three-dimensional model of Irish Sea hydrodynamics and solute transport was deployed by Olbert et al. (2010) in order to assess the current monitoring system of Tc-99 by the Radiological Protection Institute of Ireland (RPII). Earlier, a model by Dabrowski and Hartnett (2008) was used to estimate travel and residence times of a conservative tracer released from Sellafield. There was also a substantial amount of work undertaken on Tc-99 dispersion properties in the North-East Atlantic (e.g. Kershaw et al., 1999, 2004) and in the North and Norwegian Sea (e.g. Orre et al., 2007). Karcher et al. (2004) used to hydrodynamic model to study pathways and travel