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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.
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.

Keywords: Technetium-99, Irish Sea, western Irish Sea gyre, numerical model, dispersion, stratification, flushing, bioaccumulation

1. Introduction

Technetium-99 (Tc-99) is a long-lived ($t^{1/2}=2.13\text{x}10^5$ 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 Figure 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 pubic 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) 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 (*Nephrops norvegicus*) and common lobster (*Homarus 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 times of Tc-99 from Sellafield to the Nordic Seas and the Arctic Ocean. Apart from the short time-scale advection-diffusion models discussed above there is also a range of compartmental models (Cefas, 1998; Abril and García-León, 1993; Smith et al., 2000) and grid models (Gleizon and McDonald, 2010) that allow observations over long timescales and calculation of dose to marine biota and human population. Good review of hydrodynamic models and compartment models and their application to radionuclide transport in marine environment can be found in Harms et al. (2003). In this research, seasonal and inter-annual effects of discharge on radionuclide spatial and temporal distributions within the Irish Sea were of interest. For such type of simulations, a fine resolution 3D model with a short timestep was considered to be appropriate.

The main objective of the present research was to undertake high resolution 3D transport modelling of the Tc-99 material released from the Sellafield plant into the Irish Sea. In particular, the vertical distribution and impact of seasonal hydrodynamic features, such as summer stratification, on the surface-to-bottom Tc-99 ratio were of interest. Individual contributions of salinity and temperature effects to this ratio were quantified. Also, the effects of the presence of the WISG on material retention mechanism within the system were explored and flushing rates under various release conditions and meteorological forcing assessed. Finally, long-term horizontal distributions of Tc-99 were analyzed in a view to likely biota exposure to Tc-99 activity.

2. Hydrography and circulation of Tc-99 within the Irish Sea

The Irish Sea is a part of the Northwest European Continental Shelf; our region of interest is designated by coordinates: 51N - 56N and 2.5W - 7W as shown in Fig. 1a. The geographical domain extends northwards from St. David's head and Carnsore Pt on the Welsh and Irish coasts to the North Channel between Larne and the Mull of Galloway (Gaffney, 2001). The Irish Sea is a semi-enclosed body of water connected to the ocean waters through two openings: the North Channel and St. George's Channel. Further hydrographical details of the Irish Sea can be found in Olbert et al. (2010). The Irish Sea is characterized by complex geometry and bathymetry (Fig. 1b) that markedly influence tidal amplitudes and thermal structures, having further implications on the circulation of water within the region and associated transport of dissolved or suspended constituents (Dabrowski et al., 2010). An important phenomenon of the Irish Sea is the WISG (Fig. 1b) developed annually during the summer season in the western Irish Sea (WIS). The mechanism of gyre formation is well recognized now (Hill et al., 1997; Horsburgh et al., 2000) but its retention properties on material transport and marine biota have not been fully analysed.

The annual net flow through the Irish Sea is northward; Atlantic water enters the Irish Sea through St. George's Channel and is transported to the west of the Isle of Man. A small component of flow enters the eastern Irish Sea to the north of Anglesey and moves anticlockwise round the Isle of Man before rejoining the main flow to exit through the North Channel (Howarth, 1984). Along with a seasonal formation of the density-driven WISG, the general northward net flow through the Irish Sea is disturbed from spring till late summer. The cyclonic gyre results in the formation of a southward flow along the east coast of Ireland; flow rates depend on the strength of the gyre and wind fields. This current is additionally amplified by the inflow from the western side of the North Channel (Brown and Gmitrowicz, 1995).

The presence of Tc-99 material in the Irish Sea is primarily due to discharges from the Sellafield nuclear reprocessing plant to the eastern Irish Sea. The net advection of Tc-99 away from Sellafield is in a north-westerly direction along the southern Scottish coastline and later towards the entrance of the North Channel. Tc-99 becomes entrained into the gyre northwest of the Isle of Man through mixing processes with the North Channel waters. The highest concentrations along the east coast of Ireland are found on the northeast coast, with levels decreasing southwards down the coast. A small portion of the Sellafield discharge is transported out of the Irish Sea to the south via St George Channel, and hence to the English Channel and the west of Ireland (Olbert et al., 2010).

Strong relationships between Tc-99 peaks and the strength of the WISG are reflected in the historical Tc-99 data recorded by the RPII along the east coast of Ireland. Olbert et al. (2010) demonstrated that the rapid rise of Tc-99 concentrations on the east coast of Ireland (July-August) coincides with the peak of sea surface temperature when the WISG is strongest and advection is fastest. Collapse of the gyre in late summer/early autumn reverses the southward net flow. Consequently, the lowest levels of Tc-99 along the east coast of Ireland are observed during winter months. The annual cycle, exhibiting seasonal oscillations, is reasonably well reproduced from one year to another.

3. Methods and materials

3.1. Tc-99 data

A dataset of Sellafield discharges containing a 55-year long history of annual loadings is presented in Fig. 2a. Differences in discharge rates between pre-EARP and post-EARP period are apparent; annual load increased from 6 TBq in 1993 to 192 TBq in 1995. From year 1997 onward continuous reductions in discharges are observed and by 2005 discharges reached the pre-EARP levels. Tc-99 monthly discharge time series presented in Fig. 2b were the main inputs of Tc-99 fluxes to the transport model.

Tc-99 surface samples inventories collected during 6 cruises of the CEFAS research vessel (RV CIROLANA and RV CORYSTES) between 1993-1998 and compiled in McCubbin et al. (2002) were used for the numerical model validation. The transport model was additionally validated against surface and bottom water samples compiled in Leonard et al. (2004). This dataset consists of CTD and Tc-99 profiles at offshore sampling sites located within the western and eastern Irish Sea. The samples were collated during three expeditions to sea aboard the DANI research vessel RV Lough Foyle (L.Foyle 6/98 and 6/99) and the CEFAS research vessel RV CORYSTES (COR 9b/98).

3.2. Model description

A numerical model simulation of hydrodynamic conditions and transport of Tc-99 released from Sellafield was performed using the three-dimensional numerical model ECOMSED. Full description of the model can be found in HydroQual (2002) and Mellor (2003), while details of the model setup for the Irish Sea domain are reported in Olbert et al. (2010) and augmented with additional information in Dabrowski (2005). The model equations calculate time-dependent distributions of: water levels, three components of velocity, temperatures, salinities and tracers. Computation was carried out on a 4 km horizontal grid covering a complex area of the Irish Sea and surrounding waters. Vertical diffusion determined from transport of the vertical turbulence kinetic energy and the turbulence macroscale is defined by the advection-diffusion equations of Mellor-Yamada (Mellor and Yamada, 1982). The central upwind scheme is selected for interpolation of solutes.

With regard to the transport model, the Sellafield discharges and spatial concentrations for model initial conditions were the primary inputs. Monthly discharges were specified to the model according to rates presented in Fig. 2b. The transport model was initialised from the Tc-99 distributions recorded within the Irish Sea during CIR 11/93 survey (McCubbin et al., 2002). At the open boundaries constant Tc-99 inflows concentrations of 8mBq/l and 0.4mBq/l were assumed along the western and southern boundary, respectively, in line with data published in McCubbin et al. (2002).

3.3. Calculations of residence time

Effects of complex hydrodynamic processes on the distribution of Tc-99 in the Irish Sea are discussed through flushing properties of the region. Average residence time is defined as the expected time during which material resides in the region under consideration. The concept was developed by Takeoka (1984) and utilized by Murakami (1991). The average residence time (τ) is calculated by fitting a remnant function representing an exponential distribution to the actual Tc-99 decay curve using the least square method and integration over a time range from 0 to infinity.

4. Results

4.1. Validation of the hydrodynamic model

The hydrodynamic model of the Irish Sea described above was used in previous studies; validation results for 3-D currents, temperature and salinity profiles are presented in Dabrowski (2005), Dabrowski and Hartnett (2008) and Dabrowski et al. (2010) and for brevity are not repeated here. The model results correspond closely to field data and the general barotropic and baroclinic circulation within the Irish Sea follows the pattern described in Section 2.

In this study, considering the availability of discharge data (monthly values 1990-2006), field records for initial conditions and validation data, the numerical simulation was performed for a period of December 1993 to December 2000. Additional validation was performed for the period considered; CTD temperature (L.Foyle 6/98 and 6/99, COR 9b/98) of surface and bottom waters within the WIS were compared to numerical predictions. The agreement between model and data is satisfactory; the temperature profiles and the dome of cold water are well reproduced by the model (Fig. 3). Bottom temperatures of 9.5-10.0°C and 8.5-9.0°C simulated by the model for June 1998 and July 1999 sampling, respectively, correspond well to bottom temperature records.

Similarly, modelled surface temperatures match the data well. This demonstrates that the model is capable of simulating stratification in the WIS, which is of great importance for this study.

4.2. Validation of transport model

The evaluation of the transport model accuracy was conducted in Olbert et al. (2010), where Tc-99 surface values predicted by the numerical model were compared against existing sea surface Tc-99 data. The comprehensive validation was performed on both spatial and temporal scales and an extensive dataset was involved. Modelled and observed surface concentrations have similar distributions, and gradients match well; Fig. 4 compares modelled Tc-99 concentrations on the east coast of Ireland and historic measurements provided by the RPII (Pollard et al., 1996; Long et al., 1998; Ryan et al., 2000).

Further validation involved a comparison of concentrations in both surface and bottom layers. Fig. 5ai-aiii illustrates recorded surface and near-bed Tc-99 distributions at several sites within the WIS, while Fig. 5bi-biii presents numerical outputs as a cross-section through the region. Both modelled surface and bottom Tc-99 concentrations are in general comparable with data for all three periods (June 1998, September 1998 and July 1999). In particular, bottom concentration gradients along the A-B transect are in good agreement with bottom records at transect-nearby sampling points. The surface distributions predicted by the model also correspond generally well to the field surface concentrations.

The only distinctive discrepancy between the computed concentrations and field records were found in the proximity to the west coast of the Isle of Man, where the model simulated higher concentrations. These higher values result from overpredicting the strength of anticlockwise jet-like circulation around the Island that carries high concentrations from the eastern Irish Sea. However, other validation results confirm that the flow patterns in the vicinity of the Isle of Man, as inaccurately simulated by the model, seem to have only local effect on Tc-99 predictions. Therefore, the model’s ability to reconstruct the material transport is overall encouraging.

4.3. Horizontal distribution of Tc-99 in seawater

In this section long-term horizontal distributions of Tc-99 within the Irish Sea are discussed based on the model predictions. Fluctuations in monthly Tc-99 discharges combined with variations in hydrodynamic conditions on seasonal and inter-annual timescale influence the horizontal distribution of this nuclide. Annual averages can vary considerably from one year to another; the seawater concentrations of Tc-99 in years immediately after EARP commissioning are much higher than those in subsequent years. The total discharge in years 1995-1997 was a factor of 2.5 higher than that in the period of 1998-2000. Two three-year-periods: (1) 1995-1997 and (2) 1998-2000 are selected for arithmetic averaging of concentrations; the averaged concentrations for these periods are shown in Fig. 6a and b, respectively. The first period represents the highest discharges on record, the second represents typical Tc-99 releases from late 90's to mid 2000's. The general pattern of modelled spatial distribution of Tc-99 over the Irish Sea shows highest concentrations in the vicinity of Sellafield, with concentrations progressively decreasing with distance from the source. The Tc-99 concentrations change over the study area by at least one order of magnitude from the source point to Holyhead (Welsh coast) and Balbriggan (Irish coast). The most southerly parts of the Irish Sea experience approximately two orders of magnitude lower values. The rate of decrease is not uniform over the Irish Sea; from Sellafield to Holyhead the decrease is about four times greater than between Holyhead and Fishguard.

With regard to discharge levels and corresponding seawater concentrations, a considerable difference was observed between the Tc-99 spatial distribution in the first and second periods, and unsurprisingly the differences were greatest close to the Sellafield plant. In the eastern Irish Sea to the Isle of Man, the average Tc-99 concentration was 340 mBq/l and 200 mBq/l, respectively for both periods. Although, Tc-99 discharges in the second period were much reduced, the concentrations in the eastern Irish Sea remained considerably high, particularly at the entrance to Liverpool Bay. High contamination in the region is likely to be a combined effect of high discharges in the first period and the relatively poor flushing properties of that region, as Dabrowski and Hartnett (2008) demonstrated. In the WIS similar concentrations were observed for both periods. This was expected as the concentrations away from the source are progressively diluted and, therefore, the impact of discharges on concentration decreases with distance from the discharge source. The average Tc-99 concentrations in the WIS were reduced by approximately 10-fold compared with those in the eastern Irish Sea. Interestingly, in the southern Irish Sea the plume stretches along Irish and British coastline more to the south during years 1998-2000 than during 1995-1997. Dabrowski et al. (2010) claims that this stronger southward flow is a baroclinic feature resulting from stratification, which coincides with a lack of strong wind events during summer. This flow becomes a part of geostrophic flow along the southern coast as presented in Brown et al. (2003) and Young et al. (2004). The shape of contours in the central part of southern Irish Sea suggests strong northward net inflow of uncontaminated Atlantic water.

4.4. Vertical distribution of Tc-99

The vertical distribution of Tc-99 was analysed by introducing a relative concentration expressed as a ratio of surface (Tc_s) to bottom (Tc_b) Tc-99 concentration ($R_{sb} = Tc_s / Tc_b$). Fig. 7a shows a map of modelled relative concentrations defined as

$$R_m = \frac{\sum_{i=1}^{n} R_{mi}}{n} \qquad (1)$$

where R_{mi} is a monthly average of R_{sb} for year i and n is the total number of years equal to 7 in this case. For instance, the R_m value for January is the arithmetic mean of 7 monthly-averaged values (R_{mi}) for January obtained for the years 1994-2000.

As can be seen from Fig. 7a, generally higher Tc-99 concentrations are observed in surface waters. The ratio R_{sb} is highly spatially varied throughout the Irish Sea. Higher R_{sb} are found predominantly offshore; the ratio R_m ranges from 1.0 along the coastline up to 2.6 in the deepest region of North Channel.

With regards to timescale of R_m peaks, in the WIS the highest R_m is observed in July and is followed by a slightly lower value in August, as demonstrated in Fig. 8a. The shape of the temporal distribution in most of the Irish Sea area is broadly similar to that in the WIS; though at sites adjacent to the coastline the timings may be different. Close to the Irish coast highest peaks are reached in June, as in the North Channel shown in Fig. 8b. Along the Cumbrian coast peaks are generally exhibited in May-June (Fig. 8c) whereas peaks on the Welsh coast occur in August.

Spatial distributions of R_{sb} agree well with distribution patterns of potential energy anomaly (PEA) shown in Fig. 7b and expressed as

$$PEA_m = \frac{\sum_{i=1}^{n} PEA_{mi}}{n} \qquad (2)$$

where PEA_{mi} is a monthly average of PEA for year i. This relationship explains higher R_{sb} in stratified waters and ratios close to 1 in vertically well-mixed shallow waters. In a stratified system high ratios are due to mixing of waters of two origins; surface salinity-poor technetium-rich waters are attributed to inflow from the eastern Irish Sea, whereas bottom heavy waters are due to incursions of saline-rich radionuclide-poor Atlantic waters. Atlantic waters enter the North Channel through the north opening (Knight and Howarth, 1999), whilst in the WIS both close proximity to the North Channel and northward winter net flow are responsible for the presence of oceanic water (Bowden, 1980).

4.5. Effect of stratification on Tc-99 vertical profile

Since PEA quantifies the strength of stratification, R_{sb} depends on the parameters that induce stratification. Fig. 9a shows a map of correlation coefficients between R_{sb} and salinity ratio S_{sb} (salinity at the surface/salinity at the bottom). In total 720 samples uniformly distributed in time were used in the determination of Pearson correlation coefficients at each numerical grid point. In general, there is a strong correlation between the two variables and the relationship is statistically significant (correlation coefficients are given at the 95% significance level). On average coefficients in the range -0.6 and -0.8 are observed throughout the Irish Sea. The highest magnitudes (<-0.8) occur in proximity to the Isle of Man, while the lowest (close to zero in a positive and negative range) are determined at the entry to and within the Bristol Channel. The strong negative correlation within the Irish Sea provides indication of mixing between salinity-low radionuclide-rich coastal waters and salinity-high radionuclide-poor seawater flowing into the Irish Sea from St. George's Channel and North Channel. The inverse relationship apparent in offshore waters was not evident so clearly inshore. The positive correlation observed at some of the coastal sites is likely to be associated with freshwater influx and is found only in the immediate proximity to coast characterized by marked river discharges. Freshwater fluxes result in the formation of stratification that is ultimately perturbed with increasing distance from the coast due to strong tidal mixing.

Relationships between temperature profile T_{sb} (temperature at the surface/ temperature at the bottom) and R_{sb} were also examined (Fig. 9b); correlation coefficients ranged between 0.4-0.8 for the central Irish Sea and along Irish coastline. This compares to the weak negative correlation along British coast. The strength of the relationship between R_{sb} and the corresponding S_{sb} and T_{sb} can be visually assessed from Fig. 9a and b. Salinity has a stronger effect on R_{sb} than temperature throughout most of the Irish Sea domain. The vertical distribution of temperature appears to have a stronger impact on the Tc-99 vertical distribution along the east coast of Ireland.

4.6. Effects of wind stress and stratification on flushing properties

Flushing rates within the WIS are expected to vary vertically during periods of stratification; however, neither the flushing-stratification relationship nor the scale of vertical variations in flushing has been investigated to date. Since seasonal variability is likely to have an impact on the residence time, the instant releases were simulated as taking place three times a year: spring (March), summer (June) and autumn (September). Additionally, variability in the inter-annual circulation and stratification pattern was considered by introducing these releases for each year during the period 1994-2000. In total 21 residence times were obtained giving a representation of intra- and inter-annual variations within the WIS. Also, effects of wind forcing on flushing rates were quantified by running a model with a zero wind stress. The same analytical procedures were applied as in the first case. A region of instant releases of uniform concentrations over the water column is marked as a box in Fig. 1b.

Values of residence time (τ), averaged over the 7-year period for each seasonal release, are summarized in Table 1 along with accompanying statistical properties: standard deviation (σ_τ) and 7-year minimum ($\tau_{\min}$) and maximum ($\tau_{\max}$). The results from the runs with complete meteorological conditions indicate comparable residence times for spring and summer releases, and approximately 25% longer residence time for the autumn release.

Relative to the non-zero wind conditions, outcomes are considerably different when wind forcing is excluded. During spring and autumn periods, under zero-wind stress conditions the average residence time decreased by 30% (15 days) and over 80% (33 days) for both periods, respectively. This analysis shows that the high wind intensity and frequency between autumn and spring increase the residence time, and this is possibly associated with the effect that wind stress has on northward net flow. However, the mechanism of alteration of water circulation and associated transport under various wind conditions (wind speed and direction) is not explored in this study.

As expected, small discrepancies in residence times when wind stress was included and excluded were found during summer. Winds enhance flushing through increased turbulent mixing so the overall residence time becomes shorter. The analysis also reveals considerable effects of stratification on the residence time. Under zero-wind stress increasing stratification within the WIS causes residence times to increase. Due to the properties of the gyre higher concentrations are retained in the dome of stagnant water beneath the thermocline. Fig. 10 a-c shows a strong positive correlation between residence time and potential energy anomaly within the WIS. Determination coefficients of 0.43, 0.73 and 0.66 were obtained for spring, summer and autumn release, respectively. Correlation between residence time and stratification strength is much weaker (and negative for summer release) when wind stress is included in the simulation. This outcome highlights the important effect of wind action on the circulation and dispersion processes. As far as the inter-annual variability is concerned, from one year to another, considerable discrepancies in residence times were found. The computed minimum residence time is almost three times shorter than the 7-year average; this is observed for the March simulation of year 2000 (τ =24.6 days). In contrast, the highest maximum-to-average residence time ratio is c. 1.5 and it was observed for each release period. This apparent inter-annual variability within the WISG is caused mainly by two factors. Firstly, different tidal conditions prevailed at the start of each simulation. It is well know that both the spring-neap cycle and high-low tide phase have great impact on flushing rates. Secondly, meteorological conditions affect circulation pattern to a great extent. Variability in wind conditions strongly influences surface transport, while summer heating is responsible for density-driven circulation. Consequently, for the zero-wind stress case, the spring and autumn residence times as well as standard deviations exhibit relatively small variations. This may be explained by more uniform inter-annual hydrodynamic conditions under no wind conditions. Interestingly, summer release under wind forcing is characterised by shorter residence times, lower standard deviation and lower minimum and maximum values than when wind stress is excluded.

4.7. Vertical patterns of flushing properties within the WIS

The analysis of vertical patterns of flushing within the western Irish Sea was considered by running the model where an instant release was performed either in the upper zone (3 top layers) or in the near-bed region (3 bottom layers) of the WIS (region of release shown in Fig. 1b). Consequently, the residence times were calculated for upper and lower zone separately; this approach better accounts for effects of vertical mixing. As expected, vertical discrepancies in flushing properties are remarkable (Table 2). For summer release, Tc-99 near surface flushed out markedly faster than at the bottom. The surface-to-bottom ratio of residence time varied in the range of 0.4 – 1.1, with the mean value of 0.7 reflecting a sharp thermocline and suppressed vertical turbulent mixing over summer heating. For March and September release the average ratios of 1.0 and 0.9 were observed, respectively.

5. Discussion

5.1. Role of stratification in Tc-99 vertical distribution and flushing

The analysis undertaken in this research shows that vertical profiles of Tc-99 concentrations vary both seasonally and regionally. Inhomogeneity throughout the water column is primarily driven by the stratification as strong correlations between R_{sb} and strength of stratification, PEA, illustrates. Spatial maps of R_{mi} are generally in good agreement with those of PEA_{mi}. Minor divergences are due to fact that PEA is a vertically averaged quantity, and, therefore, does not necessarily coincide with top-to-bottom density difference. Timing of the R_m peaks also correlates with timing of highest PEA_m levels.

The highest surface-to-bottom Tc-99 ratios occur in the North Channel and western Irish Sea which are characterised by strong stratification. In the WIS the stratification is controlled by temperature gradients that increase as the heating season progresses. Along with the strengthening of the thermocline, by late June the water column becomes a two-layer system with clearly separated fresher water from a dome of relict saline water. In such a system the surface layer becomes a warm, low-salinity water body with the potential to contain high concentrations of radionuclide material discharged from Sellafield. In contrast, the bottom stagnant waters of the gyre are composed of dense, cold and saline water advected into the Irish Sea from the Atlantic, and hence tends to be radionuclide-poor.

Relationships of top-to-bottom ratios of Tc-99 concentration with salinity and temperature as stratification drivers were examined. Comparison of R_{sb} against T_{sb} or S_{sb} indicates a particularly strong correlation between R_{sb} and S_{sb}. Although, stratification is controlled by the temperature in regions of highest R_{sb}, salinity, as an indicator of water mass origin, correlates more strongly to R_{sb}.

The nonuniform vertical salinity profile is also responsible for Tc-99 vertical gradients in the eastern Irish Sea observed around April and May (Fig. 8c), but unlike the WIS, this shallow region stratifies due to fresh water discharge. As riverine discharges into the Irish Sea are highest in November-February and lowest in June-September, salinity is a key stratification driver in spring. The role of salinity diminishes as the summer season progresses and by the end of June temperature dominates stratification.

Despite the similarity of R_{sb} contours with PEA, R_{sb} depends also on two other mechanisms: tidal residual currents and wind-driven flows. The effects of these factors on flow fields, and therefore Tc-99 distribution and R_{sb}, are discussed indirectly through flushing properties of the region. A tracer well-mixed over the water column within the region of WIS is used in flushing studies to examine retention times of Tc-99. From the comparison of residence times under three seasonal regimes the important role of wind action on flushing emerges. Along with an increase in wind strength and frequency over autumn-spring period, substantial increase in residence times is observed. In the period of strong winds, the generally northward net flow is disturbed leading to a delay of material removal from the region. With regards to flushing during summer, wind action increases levels of turbulent mixing within the WIS and this results in faster flushing. Wind variations were also shown to have influence at an inter-annual scale and this is reflected in considerable discrepancies in flushing rates.

Stratification has apparent effect on gyre's flushing properties and is responsible for different rates of material removal at different vertical levels. Material removal from the WIS proceeds faster in the upper layer, while near-bed regions, characterized by generally lower flows, exhibit longer retention times. Additionally, weak exchange between the static dome and the gyre is conducive to slower tracer removal. In regions that do not thermally stratify (e.g. eastern Irish Sea) this phenomenon is not observed (Dabrowski and Hartnett, 2008).

The above assessment of temporal and vertical flushing characteristics explains aspects of the fate of Tc-99 within the WISG. Although Tc-99 concentrations in the upper layer during summer can become relatively high, the residence time suggests that the material is removed relatively fast. The near-bed layer due to weak flows and low mixing may retain material on much longer timescales; however, as the model and field data indicate this region remains rather unaffected by the Tc-99 material.

5.2. Effect of horizontal and vertical Tc-99 distribution on biota

In recent years contemporaneous assessments of residual flow, stratification and marine organism distribution in the Irish Sea were undertaken by several researchers in order to assess the level of interactions between water physics, chemistry and biota. The WISG is now known to be a retention system for Norway lobster (Hill et al., 1996). The importance of gyre on juvenile fish distribution was highlighted by Dickey-Collas et al. (1997). Emsley et al. (2005) investigated horizontal and vertical movement of copepods, larval stages of fish and *Nephrops* in stratified waters. Routes, migration patterns and Tc-99 accumulation in crabs were explored by Copplestone et al. (2004).

Limited data on the Tc-99 variations in seaweed, mussel, winkle and seawater concentration indicate that biota respond to changes in seawater concentration over longer time periods ranging from a few tens of days to 12 months or more depending upon the species (Copplestone et al., 2004). In the case of seaweeds the activity concentrations are lower in the summer and higher in the winter, concurrent with a growth-dilution hypothesis (Nowakowski et al., 2004). In contrast, there is no short-term or seasonal pattern between the Tc-99 release and uptake by crab and lobster resulting from physiological factors but there are inter-species differences in the degree of Tc-99 bioaccumulation.

The rate of radionuclide accumulation from seawater by biota is expressed by the concentration factor (CF) defined as the ratio of radionuclide concentration in biological tissue to that in the surrounding seawater environment. CF may vary with uptake and retention rates, the duration of pulse discharge and with the inter-pulse hiatus (Copplestone et al., 2004). In many cases however, the use of annual average seawater concentration data in establishing CF can be justified as organisms generally respond slowly to change in ambient water concentration (Olsen and Vives i Batlle, 2003).

In this research, monthly average seawater concentrations within the Irish Sea were calculated in order to establish a potential biota exposure to Tc-99. The analysis was based on the numerical simulation of Tc-99 transport for the period 1993-2000. Fig. 11 shows maps of duration of Tc-99 occurrence in the seawater at a particular concentration. Contours refer to average number of months in a year, when Tc-99 is present in concentrations equal or greater than 10 mBq/l, 50 mBq/l and 100 mBq/l. Model outputs show that marine biota within the Irish Sea in the period considered is likely to be exposed to detectable Tc-99 activities throughout the whole year. Concentrations of at least 10 mBq/l (Fig. 11a) are predicted to prevail across the eastern and western Irish Sea and the North Channel for 12 months per year. Concentrations of at least 50 mBq/l (Fig. 11b) are expected to be observed all year round in the eastern Irish Sea and for approximately half a year in the North Channel, while concentrations of 100 mBq/l (Fig. 11c) or more are likely to occur in the inner part of the eastern Irish Sea and, in particular, close to the Cumbrian coastline.

The analysis undertaken in this research also provides information on vertical distribution of Tc-99 material, based on which the risk of Tc-99 to marine organisms can be more fully assessed. In particular, knowledge of the distribution over the stratified western Irish Sea is important as vertical gradients in this region can be considerable. Reports mentioned above suggest that the retentive mechanism of the gyre is conducive to high abundance of fauna during the stratification period. Surveys of pelagic juvenile fish in 1994, 1995 and 1996 showed that fish locations were coincident with the centre of the gyre, rather than with spawning areas of shallow vertically mixed waters off the Irish coast and the Isle of Man. The abundance of whiting, cod, haddock, dab and witch were positively correlated with stratification (Dickey-Collas et al., 1997) and their entrainment into the gyre could be mediated by both the transport and retention mechanisms. Within the WISG various groups of organisms occupy various depths in the stratified water column. The larva of many commercial fish species found in this region show peak abundance between depths of 10 and 15 m, whereas smaller copepods remain mostly within layers above the thermocline (20 m). The numerical model analysis herein shows that these species are more exposed to higher Tc-99 concentrations than those in deep zones. There is also a group of organisms that perform vertical migration. The majority of krill occupy zone between 60 and 80m during daytime and ascended to depth 10 - 30m during the night. Emsley et al. (2005) found that organisms performing midnight sinking achieve a greater level of retention within the gyre, causing animals to cluster in the deep stratified region where production is relatively high. As the midnight sinking increases the amount of time organisms spend in the deeper layers, the exposure to Tc-99 is likely to be reduced.

WISG is a retention system for the commercially important crustacean Norway lobster (*Nephrops norvegicus*). The planktonic larvae are released in the water column from late March onwards and spend approximately 50 days in the plankton. During this time they must reside over a muddy substrate if they are to survive. By this time juvenile settle on the bed and colonize the muddy substrate found below the centre of the gyre. As the lower layers consist of Atlantic technetium-free water and as strong summer vertical and horizontal fronts prevent from mixing, this region remain relatively unaffected and consequently, marine biota settling in deeper layers of WISG are likely to be less exposed to contamination than these occupying upper zone or vertically mixed regions.

6. Conclusions

This research extends previous work by Olbert et al. (2010) and focuses on long-term horizontal distributions and seasonal effects of hydrodynamic features on vertical distributions of Tc-99 within the Irish Sea. In particular, behaviour in the western Irish Sea and effects of the WISG on Tc-99 vertical distributions are analysed. Features of the gyre were further explored and retentive properties of the system investigated. Findings from the numerical model were used in the assessment of potential risk to marine organisms as a result from exposure to Tc-99 activity. The following are the main conclusions:

- Vertical gradients of Tc-99 vary considerably both temporarily and regionally. Highest vertical gradients are observed in deep regions of the Irish Sea where radionuclide-rich saline-poor seawater overlay radionuclide-poor saline-rich Atlantic water masses. Maximum gradient is usually observed between June and July.
- WISG acts as a mechanism to concentrate Tc-99 in surface layers. The stagnant dome of winter water, about which the gyre circulates, contains lower levels of Tc-99 due to weaker vertical mixing.
- High correlation between top-to-bottom ratio of Tc-99 and the degree of stratification specified as PEA is found. Salinity, as a stratification driver, exhibits strong negative correlation with R_{sb} throughout most of the Irish Sea. Temperature correlates with R_{sb} better than salinity in regions of high fresh water input.

- An increase of wind intensity and frequency between autumn and spring is conducive to considerably longer residence times within the WIS. In contrast, during summer, wind stress increases turbulent mixing within the gyre and leads to faster flushing.
- Summer stratification has evident effect on vertical pattern of flushing.Tc-99 material is flushed from the upper layer much quicker than from the bottom part due to weaker currents in lower layers and little turbulent mixing in the vertical. In the absence of stratification, the material is removed at similar rates throughout the water column.
- Marine fauna and flora in shallower vertically well-mixed regions are exposed to Tc-99 concentrations to the same extent regardless of the inhabiting depth. In the case of stratified regions, like WIS, the biologically active photic zone above the thermocline may contain higher amounts of material then the near-bed region, and inhabitants of this zone are consequently more exposed to contamination.
- Although conclusions from the study are based on the analysis of Tc-99, these observations can be also applied to other dissolved materials with properties similar to Tc-99, including other Sellafield radionuclides and nutrients.
- Outcome from this research can be used for improvement of current Tc-99 monitoring. Processes described in this paper were not understood during the planning stages of previous discharges into the Irish Sea. However, it is imperative to consider such processes in future discharges.

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Figures

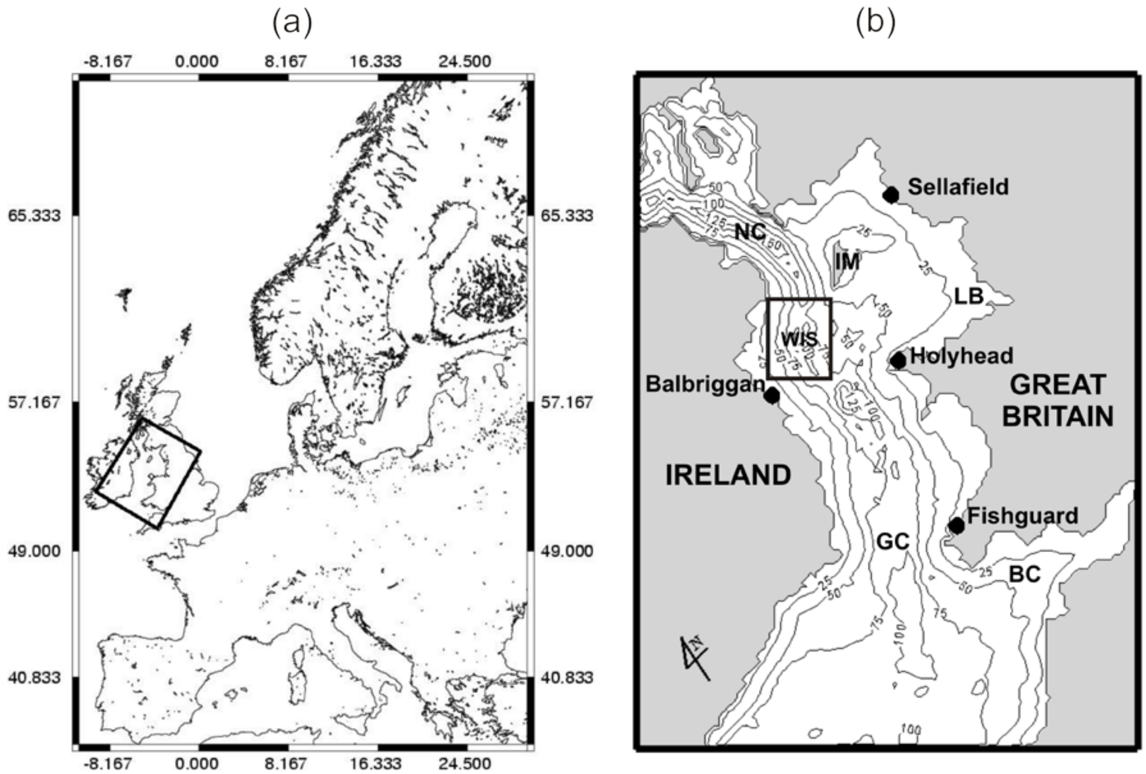

Fig. 1. (a) Irish Sea location and model orientation, (b) bathymetry of the Irish Sea and selected locations. Abbreviations: NC – North Channel, IM – Isle of Man, LB – Liverpool Bay, WIS – Western Irish Sea, GC – St. George's Channel and BC – Bristol Channel.

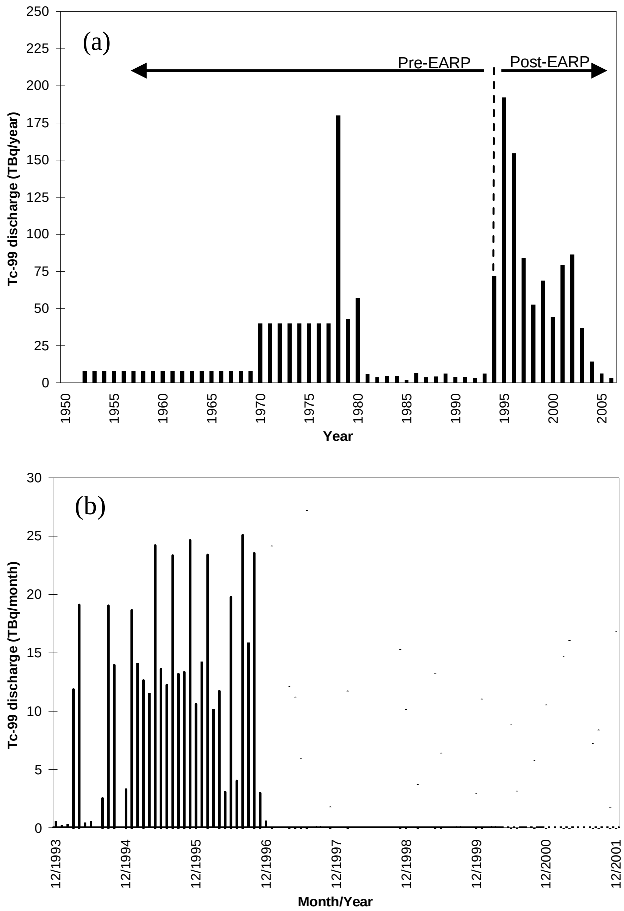

Fig. 2. (a) Annual and (b) monthly discharges of Tc-99 (TBq) from Sellafield. Data up to 1978 are best estimates only (Gray et al., 1995)

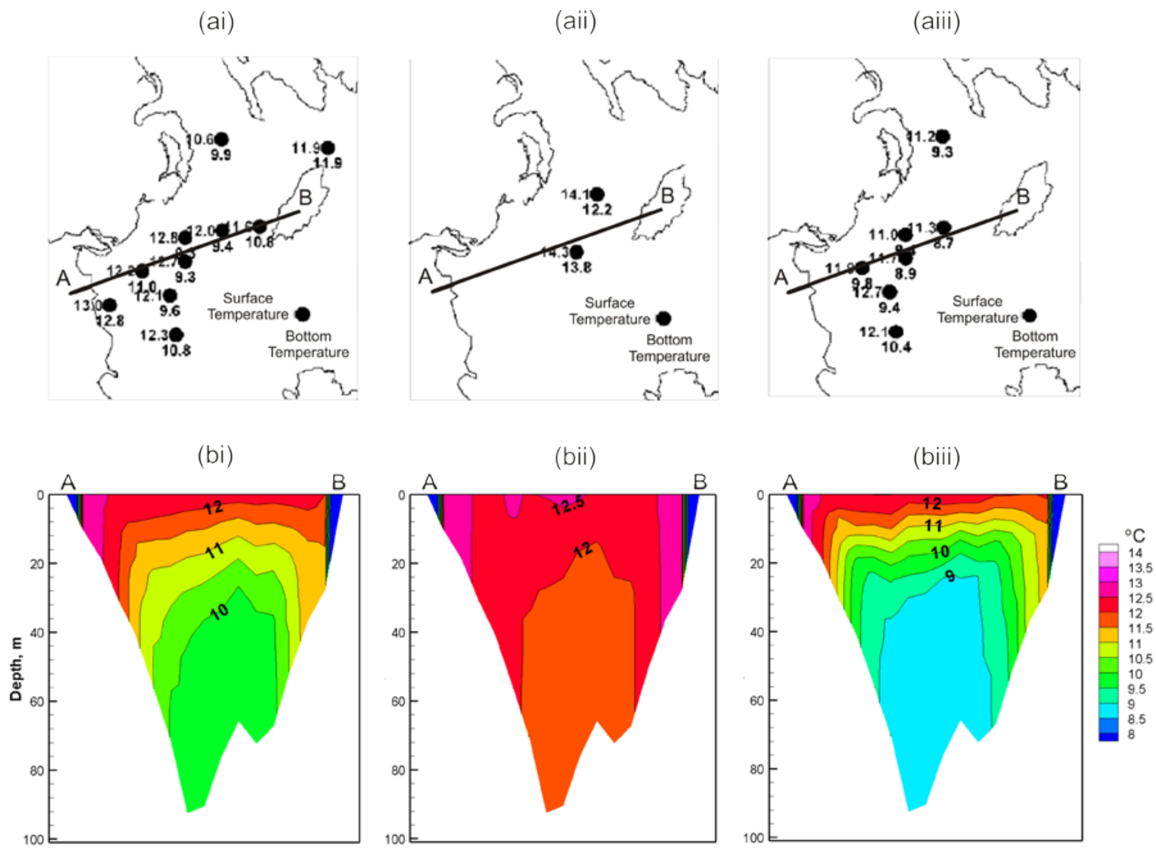

Fig. 3. (a) Temperature (˚C) of surface and bottom waters in the Irish Sea (from Leonard et al., 2004), (b) temperature vertical profiles predicted by the model along transect A-B. (i) June 1998, (ii) September 1998 and (iii) July 1999

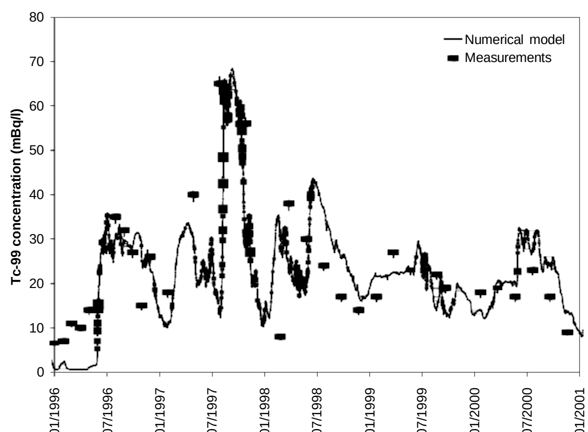

Fig. 4. Historic and model Tc-99 timeseries at Balbriggan

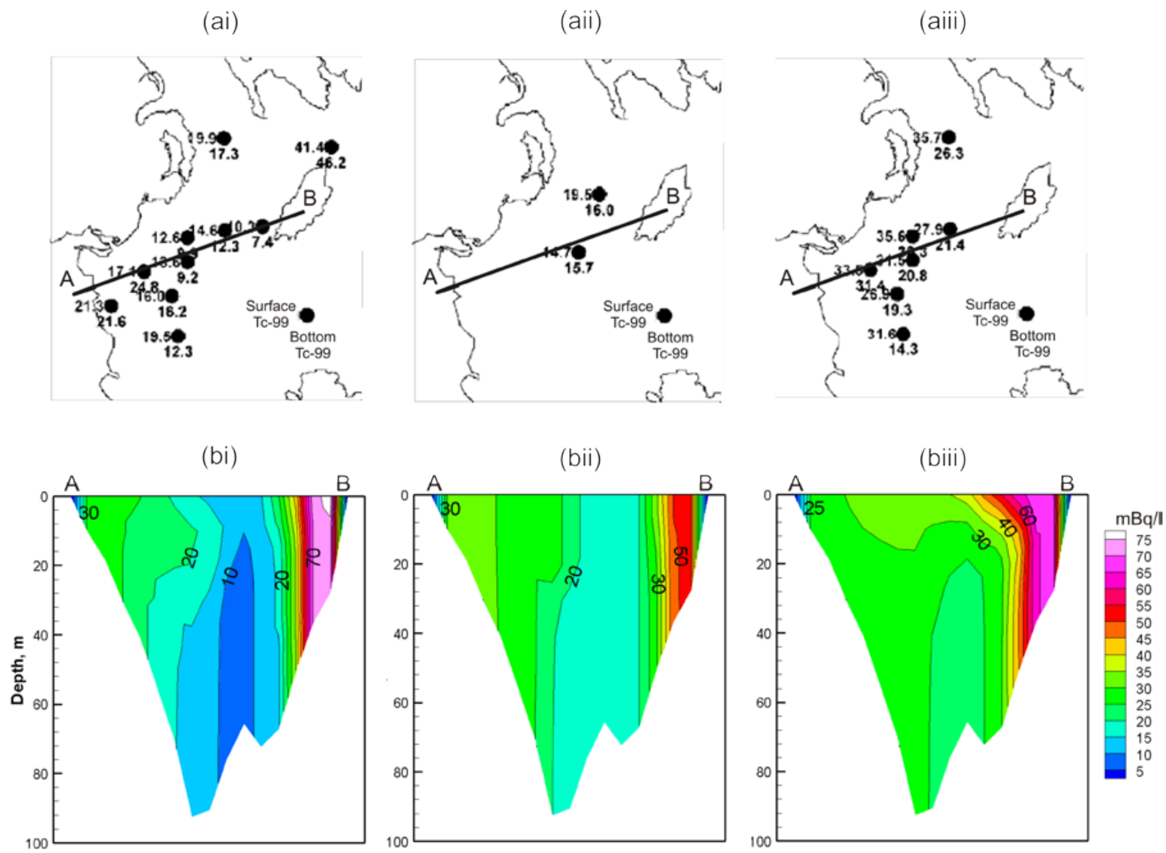

Fig. 5. (a) Tc-99 concentrations (mBq/l) of surface and bottom waters in the Irish Sea (from Leonard et al., 2004), (b) Tc-99 vertical profiles predicted by the model along transect A-B. (i) June 1998, (ii) September 1998 and (iii) July 1999

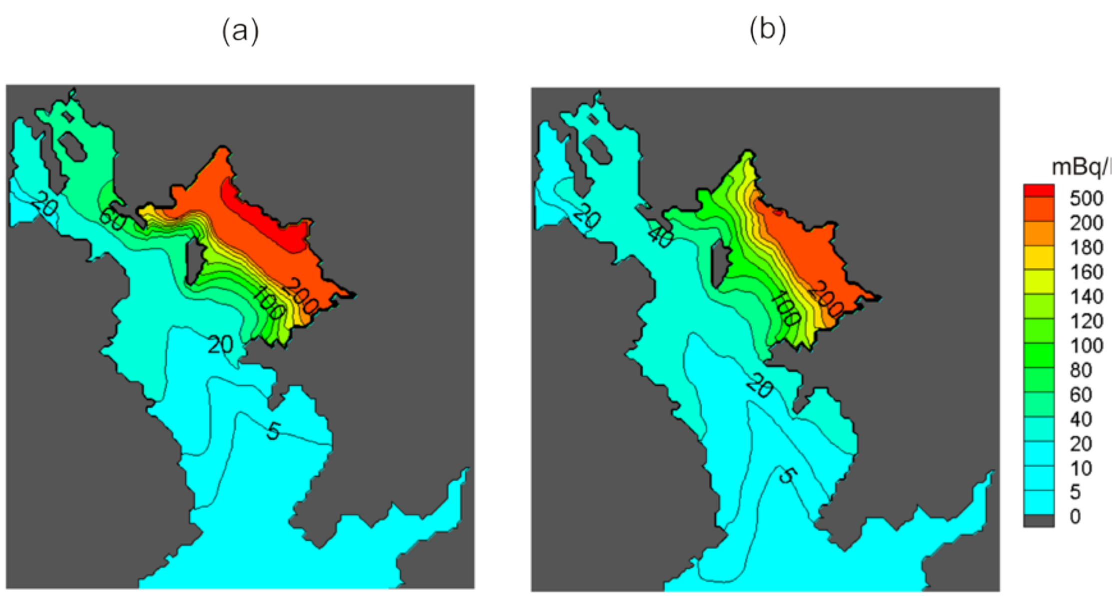

Fig. 6. Depth-averaged Tc-99 concentration in period (a) 1995-1997 and (b) 1998-2000

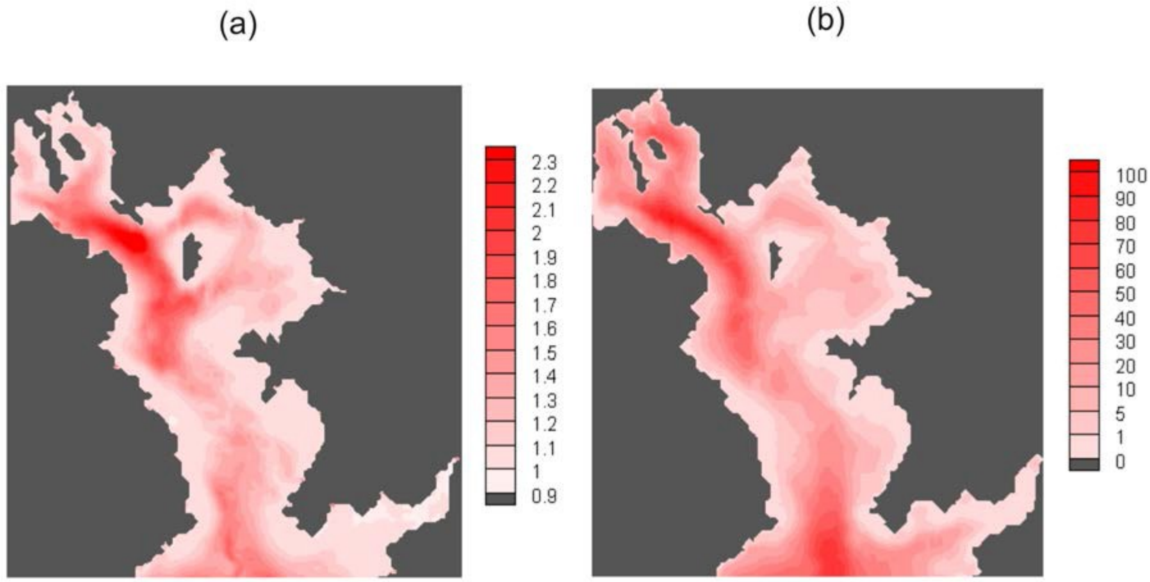

Fig. 7. Contour maps of (a) R_m and (b) PEA_m (J/m^3)

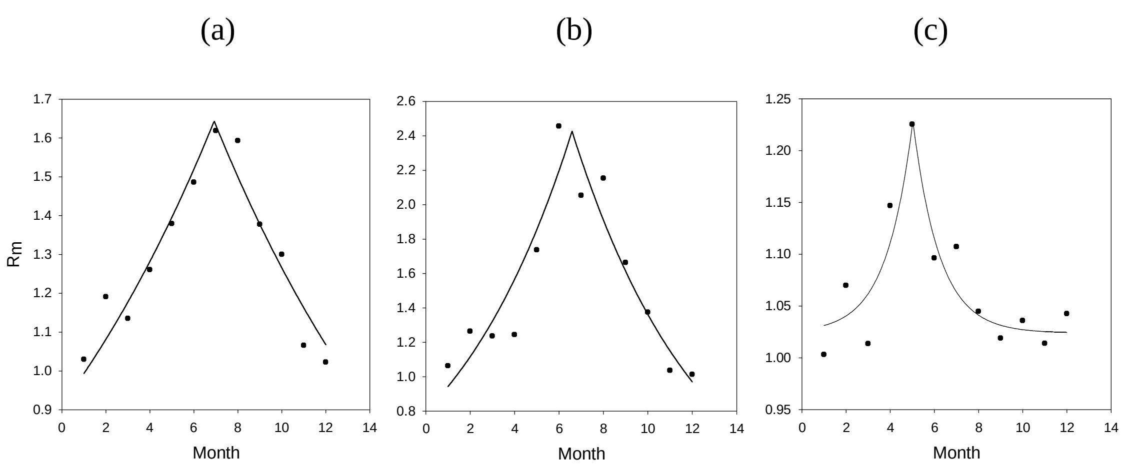

Fig. 8. R_m values over a year in (a) western Irish Sea and (b) North Channel and (c) eastern Irish Sea

(a) (b)

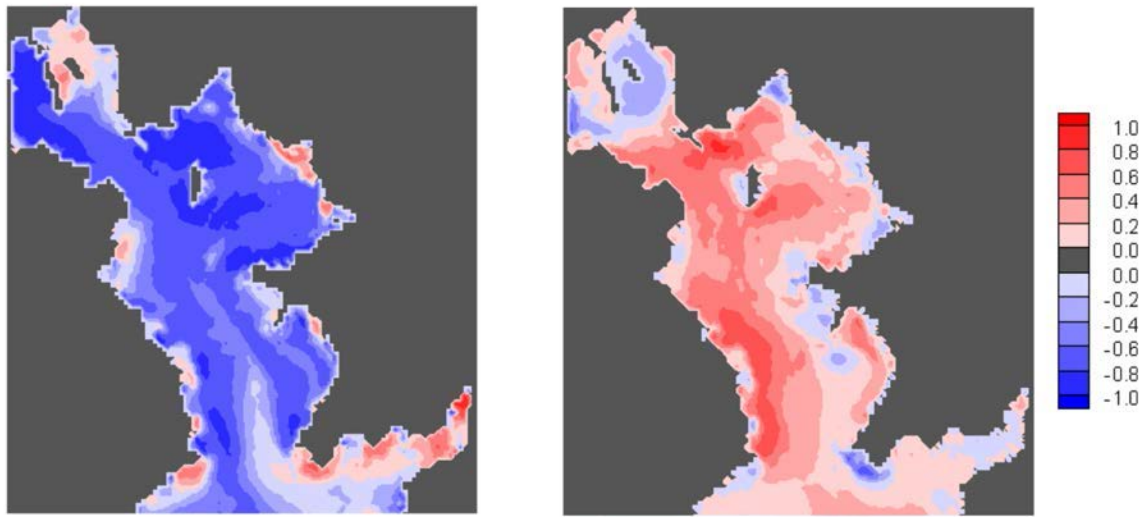

Fig. 9. Correlation coefficient between (a) S_{sb} and R_{sb}, (b) T_{sb} and R_{sb}

(a) (b) (c)

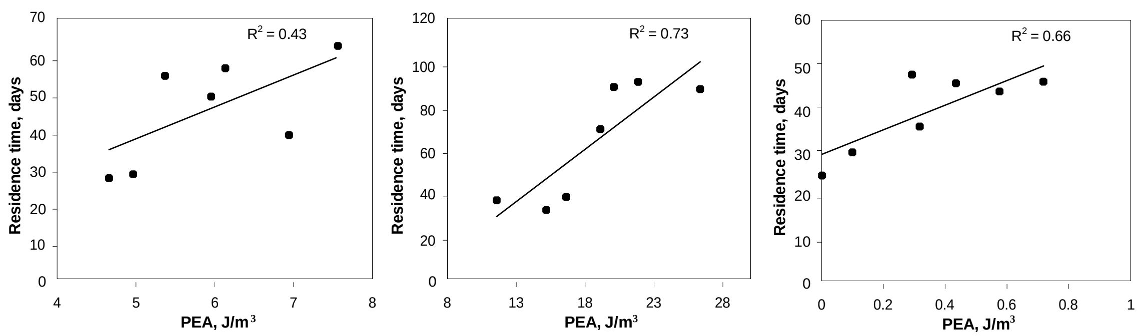

Fig. 10. Correlation between residence time and potential energy anomaly for (a) March, (b) June and (c) September instant release. Wind stress excluded from simulation

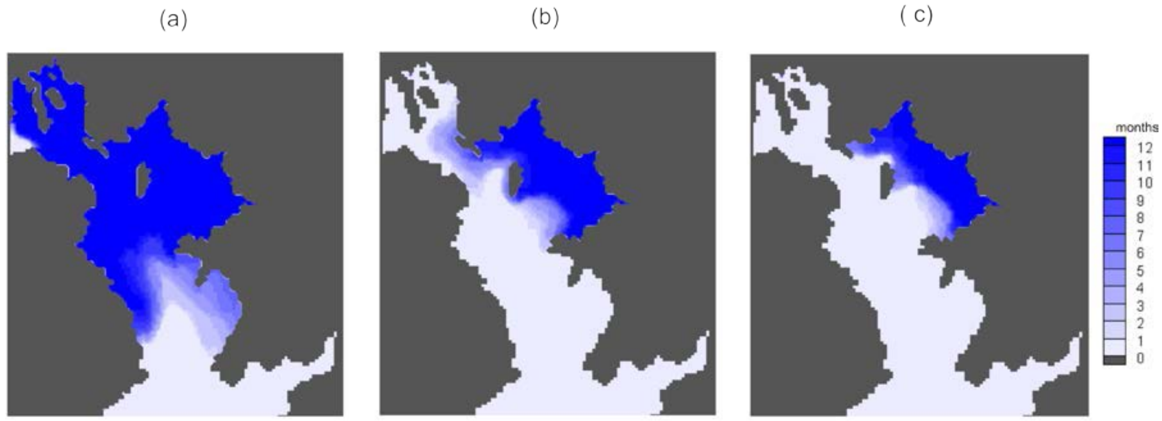

Fig. 11. Average number of months in a year of occurrence of Tc-99 material within the Irish Sea. Minimum depth-averaged concentration of (a) 10 mBq/l, (b) 50 mBq/l and (c) 100 mBq/l