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Assessment of Tc-99 monitoring within the western Irish Sea using a numerical model

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Abstract

Water circulation patterns and associated material transport within a highly dynamic system such as the Irish Sea are complex phenomena. Although Tc-99 monitoring programme undertaken by the Radiological Protection Institute of Ireland provides a good insight to the material distribution on the east coast of Ireland, transport patterns within the Irish Sea have not been fully explored. In this study a validated transport model was used to hindcast transport of Tc-99 discharged from the Sellafield plant to determine extents of Tc-99 migration within the Irish Sea and reassess transit times to east coast of Ireland. Transit times are also estimated within a context of changes in meteorological conditions and fluctuations in discharges. Additionally, seasonal and inter-annual circulation patterns were examined. Relationships between discharge times and timing of far field concentrations are highly variable and are dependant on sea dynamics controlling the accumulation and removal of Tc-99 mass. Transport towards the Irish east coast, and consequently transit times, vary intra- and inter-annually, and depend on the prevailing hydrodynamic conditions resulting from meteorological conditions. The transit times from Sellafield to Balbriggan fall within the wide range of 30-240 days; with summer releases resulting in the shortest transit times. The model also indicated a strong relationship between summer concentration peaks on the east coast of Ireland and the strength of the Western Irish Gyre. Sudden increases of Tc-99 concentrations at Balbriggan coincide with peak of sea surface temperatures when the gyre is strongest and when advection is fastest. The adequacy of the current radionuclide monitoring programme within the western Irish Sea is evaluated, and recommendations are made for the development of a more optimised monitoring programme.

Keywords: Technetium-99, monitoring, dispersion, Irish Sea, numerical model
1. Introduction

Technetium-99 (Tc-99) is a highly soluble radionuclide with a very long half-life \( t^{1/2} = 2.13 \times 10^5 \) years and is predominantly in the stable form of the pertechnetate ion \( (\text{TcO}_4^-) \) in oxygenated seawater (Beasley and Lorz, 1986). Due to its beta-emitting properties it is an important component of high-level radioactive wastes (Wildung et al., 1979). Over many decades Tc-99 has been regularly detected at elevated levels in the Irish Sea. The main sources of this radionuclide in the Irish Sea waters are discharges from the British Nuclear Fuels (BNF) reprocessing plant at Sellafield (Cumbria, UK). Radionuclides derived from Sellafield are now also known to be a major source to the shelf seas of north-western Europe (Orre et al., 2007) and north east Atlantic (Kershaw, 2004). Controlled discharges into the north-eastern Irish Sea began in 1952 and since then have been the subject of national and international controversy. In 1994 an Enhanced Actinide Removal Plant (EARP) was commissioned; the consequence of this has been elevated levels of Tc-99 in Sellafield discharges. The presence of Tc-99 material within the Irish Sea has had a well-recognized impact on a marine ecosystem; the environmental risks to the biota and to the seafood consumer have been widely reported in recent years (Hunt et al., 1998; Smith et al., 2001).

There has been a considerable amount of international interest in monitoring the Tc-99 discharges. Both British and Irish authorities, concerned about radiological impact, established their own monitoring systems within the Irish Sea. The monitoring system of the Irish Sea with a particular interest in Tc-99 distributions on the east coast of Ireland is undertaken by the Radiological Protection Institute of Ireland (RPII) on behalf of the Irish Government. The core of the monitoring programme is an acquisition and processing of standardized monitoring data. The analysis of seawater and biota (seaweeds, fish, crustaceans and molluscs) from the Irish coastal waters yields temporal variations in the concentrations of Tc-99 and geographical distribution. Recently RPII have begun to use models to inform the monitoring system. The increase discharges of Tc-99 from the reprocessing plant at Sellafield since the mid-1990’s and the pulsed release of Tc-99 has provided an opportunity to study transport pathways and
transit times in the Irish Sea and beyond. Tc-99 is not readily absorbed onto particle surfaces, under oxidizing conditions, and is transported considerable distances (Kershaw and Baxter, 1995), therefore, in cases where the seawater is oxygenated, as in the Irish Sea, it is an excellent tracer of water movement (Brown et al., 2002). Tc-99 has been used previously as an oceanographic tracer by measuring its distribution both in the water column (Hermann et al., 1995, Leonard et al., 1997) and in brown seaweeds (Dahlgaard et al., 1997).

A range of mathematical models has been developed since early 1990’s to provide better understanding of Tc-99 fate in the marine environment. Compartmental models (Abril and García-León, 1993; Smith et al., 2000) and grid models (Gleizon and McDonald, 2010) allow observations over long timescales, and calculate the subsequent doses to marine biota and human population. A recently published model of Gleizon and McDonald (2010) is a particularly complex model; it is capable of simulating a long term dispersion and radioactivity transport, sediments absorption, transfer of activity to marine organisms and effective doses to critical groups. There are also a number of short time-scale hydrodynamic and solute transport models that are capable of simulating spatial and temporal transport of radionuclides in the Irish Sea (Prandle, 1984; Periáñez, 1998; Aldridge, 1998). More recently, Dabrowski et al. (2008) used a three-dimensional, fine resolution model to estimate transit and residence times of a conservative tracer released from Sellafield, while Orre et al. (2007) simulated Tc-99 dispersion in the North and Norwegian Sea. In this study, seasonal and inter-annual effects of discharge on the radionuclide spatial and temporal distribution within the Irish Sea were of interest. For these investigations, a spatially high resolution 3D model of short timestep was most appropriate.

This research had a number of objectives. The time-varying Tc-99 distributions in seawater were examined to determine the extent of Tc-99 migration within the complex waterbody of Irish Sea. One of the main objectives of this study was to assess the transit times from Sellafield to east coast of Ireland. The effects of varying hydrographic conditions due to seasonal and inter-annual fluctuations on Tc-99 transit time were investigated. Also, potential impacts of meteorological conditions and variations in discharge rate on far-filed concentrations and material accumulation
were examined. A final objective was to assess the suitability of the radionuclide monitoring programme in the western Irish Sea as devised by RPII.

2. Hydrography of the Irish Sea

The Irish Sea is a part of the Northwest European Continental Shelf and it is located approximately between 51N - 56N and 2.5W - 7W. It is defined as the semi-enclosed body of water connected to the ocean waters through two openings: North Channel and St. George’s Channel (Fig. 1). 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). Its approximate length is 300 km and its width varies from 75-200 km down to about 30 km in the North Channel. The mean water depth averages around 60 m (McKay and Pattenden, 1993); the North Channel, connecting the Irish Sea with the Atlantic Ocean in the north, is the deepest region (exceeding 275 m). The eastern Irish Sea, with average depth of 30m and water depth not exceeding 55m, is the shallowest region. The eastern Irish Sea is defined by the area to the east of the Isle of Man and this region represents 40% of the Irish Sea area and 20% of the total water volume. The complex geometry and bathymetry significantly influence tidal amplitudes and thermal structures, having further implications on other physical features such as the circulation of water within the region and associated transport of dissolved or suspended constituents (Dabrowski et al., 2009).

The hydrodynamics of the Irish Sea is driven mainly by tides with the M2 and S2 constituents having greatest impact. Tides enter the region through both the St. George’s and North Channels, with the two paths meeting along a line running westward from the south of the Isle of Man (McKay and Pattenden, 1993). Tidal currents of the order 1.0-1.5m/s are usually observed in the North and St. George’s Channel (MacDowell, 1997), and they exceed 2.0m/s in the vicinity of headlands (Dabrowski, 2005). The annual residual circulation of the flow through the Irish Sea is northward, however, as a result of wind action and density gradients the movement of water is
Brown and Gmitrowicz (1995) highlighted the importance of wind strength on transport pathways through the North Channel. The strongest response to wind is observed in the shallow eastern Irish Sea (Olbert and Hartnett, In Press), where prevailing wind conditions induce considerable short-term variability in circulation pattern.

An important phenomenon of the Irish Sea is a summer gyre developed in the western Irish Sea region during the heating season. The Western Irish Gyre (WIG) is characterized by summer stratification due to a combination of persistent slack water and water depths exceeding 100 m. A two-layer system develops quickly, over a matter of days (Hill et al., 1997), in late spring (April); the surface layer is 20-40 m thick and up to 7°C warmer than the bottom layer (Horsburgh, 1999). A dome of cold, dense water, composed of water left over from the previous winter, can be found beneath the thermocline. This thermal structure present seasonally in the western Irish Sea is responsible for formation of density-driven thermohaline circulation that reverses the general northward net flow through the Irish Sea. The anticlockwise gyre results in the southward flow along the east coast of Ireland and significantly affects material transport within the Irish Sea. The dome breaks down typically in early autumn, as a result of cooling and strong wind events.

3. Materials and methods

3.1. Data collection

A dataset of Sellafield discharges contains a 55-year long history of annual loadings (Fig. 2a). Detailed chronology of Tc-99 monthly discharges is presented in Fig. 2(b) along with Tc-99 concentration time series recorded at Balbriggan on the east coast of Ireland. From these records an apparent difference in release rates between pre-EARP and post-EARP period emerges. During the pre-EARP period there were relatively small discharges in the 1950’s and 1960’s (8 TBq/year) followed by a significant increase in the 1970’s (40TBq/year) and reduction to c. 5 TBq/year in the 1980’s. In the post-EARP period, the annual discharges increased dramatically
as the new treatment commissioned to reduce concentrations of actinides in effluent prior
discharge was ineffective in removing Tc-99 (Smith et al., 2001). On the annual basis the highest
authorised discharge occurred in 1995 (c. 192 TBq) with levels falling below 100 TBq in 1997,
creating additional large-scale pulse. The mean discharges in the first three years after
commissioning the EARP were 3 orders of magnitude greater than in three years preceding the
plant opening.

The current monitoring programme of Tc-99 within the Irish Sea involves both offshore and
inshore sampling. Shore-based sampling of seawater for radionuclide analysis forms an integral
part of the Irish statutory monitoring programme undertaken by the RPII. The programme, labeled
as the CMP programme, involves sampling at five locations along the east coast of Ireland:
Balbriggan, Bull Island, Carlingford, Cahore and Dunmore East. For the period 1993-2005, in total
139 samples were analyzed, from which 78 were collected at Balbriggan, 31 at Bull Island, 15 at
Carlingford, 11 at Cahore and 4 at Dunmore East.

Additional sources of time-series data were identified to augment the near-shore studies. The
distributions of Tc-99 within the Irish Sea were assessed from offshore sampling. The surface
data were collected during six cruises of the CEFAS research vessel, RV CIROLANA (CIR) and
RV CORYSTES (COR) and are compiled in McCubbin et al. 2002 along with details on analytical
procedures. Two expeditions aboard the DANI research vessel RV Lough Foyle (L.Foyle) collated
surface and bottom seawater samples (Leonard et al., 2004). Inventories from CIR, COR and
L.Foyle cruises were employed to validate the numerical model (horizontal and vertical distribution
of Tc-99) and assess the monitoring system.

3.2. Numerical model description

A hindcast of hydrodynamic conditions within the Irish Sea and transport patterns of Tc-99
originating from Sellafield was successfully performed using a 3D primitive equation numerical
model ECOMSED (HydroQual, 2002; Mellor, 2003). The hydrodynamic module of the model is
essentially the same as widely known Princeton Ocean Model (POM) model (Blumberg and Mellor, 1987; Mellor, 2001). The model equations were solved using a finite difference method and the outputs are time-dependent distributions of: water levels, three components of velocity, temperatures, salinities, tracers, turbulence kinetic energy and turbulence macroscale. Vertical processes are solved on a sigma coordinate system capable of resolving coastal and deep ocean regions.

The model accommodates realistic coastline geometry and bottom topography. Computation was carried out on a 4 km horizontal grid covering a complex area of the Irish Sea and surrounding waters. A bathymetric map with depths referred to the MWL and locations of important sites are presented in Fig. 1.

During this research the model was driven by a variable surface elevation, density distributions and realistic meteorological conditions. At open boundaries a radiation condition related the normal component of the depth mean current to the sea surface elevation accounting for tidal input. Tidal spectrum consisting of 5 constituent (K1, O1, M2, N2 and S2) was extracted from the North-East Atlantic model of Olbert and Hartnett (In Press). Climatological initial conditions for temperature and salinity as well as monthly averages of salinity and temperature along open boundaries were acquired from two sources: (1) LEVITUS94 global dataset (Da Silva et al., 1994), and (2) MPIOM global ocean model of Max Planck Institute, Germany.

A set of meteorological conditions was obtained from the regional reanalysis ERA-40 of European Centre for Medium-Range Weather Forecasts (ECMWF). Hindcast parameters of wind fields, air pressure and temperature, humidity, cloud cover fraction, evaporation, precipitation, shortwave radiation and extinction coefficient were provided as 3-hr instantaneous values.

Wind stresses were parameterized as a quadratic function of the 10 m wind speed. Changes in the roughness of ocean surface were included through varying drag coefficient. The bottom stress term was calculated as a quadratic function of the depth mean flow with a frictional parameter of 0.0025.

The discharge of freshwater rivers was also included in the model. A total of 41 data sets representing monthly average discharges were used to simulate the effect of freshwater input.
With regard to transport model, Sellafield discharges and spatial concentrations for model startup were the primary inputs. The discharges were compiled by the RPII and incorporated into the model in the form of single point source monthly mass loads from the Sellafield plant. Initial distributions of Tc-99 within the Irish Sea were generated from December 1993 survey data (CIR 11/93). At the open boundaries constant inflows concentrations of Tc-99 were assigned on flood tides representing conditions outside the model domain. 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).

4. Results

4.1. Irish Sea Tc-99 data

The Balbriggan station with an average sampling frequency of 0.56 sample/month and sampling history covering the pre- and post-event of 1997 (being of main focus in this study) was selected to investigate the temporal concentrations of Tc-99 on the east coast of Ireland. These data were also used to validate the numerical model. The Tc-99 inventory at other stations is either too rare or does not coincide with a period of model run, and so was not used in the validation process. Recorded concentrations at the Balbriggan station are presented in Fig. 2(b). The baseline levels of 7-10 mBq/l are observed in winter and rise during spring months to reach highest values in summer. The monitoring programme recorded maximum annual concentrations of up to 35, 65, 38, 27 and 23 mBq/l in consecutive years from 1996 to 2000. These changes are associated with an intra- and inter-annual circulation driven by dynamical changing atmospheric conditions as well as pulse-type variable releases from Sellafield.

Spatially throughout the Irish Sea the survey data show that estimates of Tc-99 inventories range from 6 to 166 TBq and are of similar magnitude to the increase in annual Sellafield discharges. The concentrations detected during the December 1993 cruise (CIR11/93) provide a baseline for
the analysis of Tc-99 concentrations in post-EARP period. The average concentration close to Sellafield in December 1993 was c. 30 mBq/l, and this compares to post EARP values of c. 200 in December 1994, c. 1800 in December 1995, c. 500 in December 1996 and c. 40 in September 1998. By December 1996 surface concentrations throughout the Irish Sea were elevated by more than an order of magnitude, compared with pre-EARP levels (Leonard et al., 2004). Leonard et al. (1999) suggested that concentrations have increased from the cumulative effect of continuous discharge. The reduction by an order of magnitude between December 1996 and September 1998 results from greatly reduced discharges for the 6 months proceeding the September 1998. These data provides some illustration of the response of the water body in the eastern Irish Sea to fluctuations in discharges.

4.2. Model validation

The numerical model has been successfully used previously to investigate the circulation and flushing time of conservative tracer within the Irish Sea (Dabrowski, 2005; Dabrowski et al., 2009). 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 2001. The general barotropic and baroclinic circulation within the Irish Sea follows the pattern described in Section 2. Fig. 3 shows a comparison of model predictions of current speeds and surface and bottom seawater temperatures at point T1 in the region of western Irish Sea. The model results correspond closely to field data; comparisons at other locations can be found in Dabrowski (2005). Satisfactory model validation against surface and near-bed temperature is of particular importance in stratified regions of the Irish Sea, where density gradients have been recognised as the significant factor driving the baroclinic, near-surface flow (Hill et al., 1996). Stratification in the western Irish Sea predicted by the numerical model is well reproduced and its spatial extent and net flow fields are in good agreement with those presented in Horsburgh (1999), as shown in Fig.
4. The existence of a gyre within the western Irish Sea and its effect on seasonal transport of Tc-99 will be discussed in following sections.

4.3. Transport model analysis

Performance of the transport model was evaluated by comparison against existing Tc-99 data. The spatial distributions of Tc-99 within the Irish Sea were assessed from offshore samples, while temporal variations were determined from inshore regular sampling. Fig. 5 compares surface distributions of radionuclide in the Irish Sea at 6 times: December 1993 (CIR11/93), December 1994 (CIR12/94), May 1995 (CIR5/95), December 1995 (CIR10/95), December 1996 (CIR11/96) and September 1998 (COR9b/98). In general, modelled and observed concentrations have similar distributions and density gradients match well. The 5, 10, 20, 100, 200 and 500 mBq/l contours are displayed at similar locations as data. Even the COR 9b/98 data are quite well reproduced, in spite of the fact that the Tc-99 distributions and magnitudes are markedly dissimilar to that observed on the preceding surveys.

Although, the Irish Sea is a complex hydrodynamic system with complicated topography, distinct seasonal variations in flow fields and large discharge fluctuations, the model reproduces the transport of Tc-99 surprisingly well, even at remote locations such as Balbriggan. The historic field records, along with their measurement errors, and timeseries hindcast by the model are compared in Fig. 6. Over the 5-year time-slice, the timing of peaks and strength of the model results are in a very good agreement with records. For the years 1996 and 1997 the model results are particularly close to data and the model capabilities to reconstruct a material transport is encouraging.

During June-July 1998 the model predicted a peak of 43 mBq/l, however, this peak was not recorded during monitoring. Although, the reason for this discrepancy is not fully understood, there are a number of possible explanations, such as: (a) overprediction of the strength of the WIG by the numerical model, (b) delay in the computed peak with respect to the measurements...
and (c) sparseness of the monitoring data. This discrepancy will be subject to in-depth analysis in the following sections.

4.4. Seasonal circulation

Varying hydrographic conditions due to seasonal fluctuations and effects of material accumulation are clearly reflected in horizontal distributions of Tc-99. Fig. 7 illustrates model predicted changes in Tc-99 distribution over the year 1996, which is a representative year in terms of meteorological conditions for the 1990’s. In 1996 the east coast of Ireland remains unaffected by Sellafield discharges over the first three months of the year. In this period the strong northward flow transports material along the coast of Scotland and to the NE Atlantic through the North Channel. Southward flow develops in April and gradually becomes stronger; currents advect the plume originating in the North Channel down the Irish coastline. The plume reaches Balbriggan in late May and the peak is observed in June. In the course of the following months the current advects the plume further south to reach Cahore (see Fig. 1 for location map) in October. At this juncture, southward drift gradually reduces as a result of the WIG collapse. In turn, northward flows, gradually increasing in strength, advect the plume northward and concentration in Balbriggan reaches background level by January 1997.

4.5. Transit time estimates

The concentration of Tc-99 along the east coast of Ireland is a function of transport mechanisms and Tc-99 discharge rates from Sellafield. The effect of variations in hydrodynamics on far field concentrations is investigated through idealized experiment. Transit time is defined as the time elapsed after introducing a tracer to the system until the time of occurrence of a peak concentration at a given location. In this study, transit time is calculated on the basis of injections
of an assumed activity of Tc-99 from the Sellafield plant. The material was released at a constant load rate of 0.5 GBq/s over a period of 1 week. Since seasonal variability is likely to have an impact on transit time, the releases were simulated as taking place four times a year: January (winter), April (spring), July (summer) and October (autumn). Additionally, variability in inter-annual circulation was considered by introducing these seasonal releases at four consecutive years 1995-1998. Thus, a total of 16 release scenarios were simulated.

Transit times from Sellafield to Balbriggan, presented in Fig. 8, exhibit high variability depending on both season and year of release. They fall within the wide range of 30-240 days; summer releases resulted in shortest transit times. This is not surprising, since southward net flow accelerates in the course of summer with the amplification of the WIG. The gyre’s retentive character is clearly manifested in Fig. 8 (a) and (b), where material arriving in late spring/early summer has been entrapped there for 4-6 months.

The ultimate collapse of the gyre reverses the net drift from southward to northward, which intensifies over late autumn and winter as a result of prevailing wind conditions. Northern drift prevents material released in January to be transported to Balbriggan during winter and early spring. Consequently, the transit times of January releases are the longest and take between 150-230 days to arrive in Balbriggan.

Timeseries in Fig. 8 clearly reflect the amount of material transported. Spring circulation patterns are effective in the fast transport of highly concentrated material and retaining this material over long timescales. Summer currents, although ensuring short transit times, carry generally much smaller quantities (exception is year 1995) in the first year after release. In turn, the material returns again in the following year and the peak is higher compared to the first year.

From the inter-annual analysis significant differences in transit times between years 1995, 1996, 1997 and 1998 can be seen. The leading edge of material released in January of 1996 and 1998 will reach Balbriggan approximately 60 days ahead of material that was released in January of 1995 or 1997; similar patterns can be found for April releases. The timeseries for autumn releases differ widely for individual years and for this reason are difficult to compare.
In summary, taking into account the seasonal and inter-annual effect, the typical estimates for the leading edge of plume would be within the range of 60-150 days. Conservative estimates range from 40 to 210 days, and extreme estimates range from 30-240 days. Peak concentrations are transported later; the likely transit times of peaks are shown in Table 1.

The above modelling analysis shows that the effects of long-term variability in hydrodynamics on Tc-99 transport are extremely difficult to anticipate from monitoring alone.

4.6. Effect of discharge rate on far field concentrations

It was considered constructive to investigate potential impacts of variations in discharge rate on concentration levels at the east coast of Ireland. Effects of discharge on concentration variability at Balbriggan were quantified by simulating a constant discharge from Sellafield. The load of 13TBq/month represents an average discharge from Sellafield over the year 1996. Fig. 9 demonstrates the extent of fluctuations caused by a constant rate continuous release. Since the constant discharge rate is similar to the actual discharges the timeseries in Fig. 9 for real discharges and constant discharges are very similar. These outcomes indicate that remote from the immediate vicinity of Sellafield, the impact of the fluctuations in the discharge is progressively less marked. Nonetheless, this simple test shows what concentrations at Balbriggan would have been like from year 1998 onward if loads had not been reduced. If loads of 1995-1996 have been maintained over following years, concentrations greatly exceeding the September 1997 would probably have been observed.

4.7. Relationships between discharge and peak concentration

The relationships between discharge time from Sellafield and timing of peak concentrations at Balbriggan are investigated in this section to assess the impact of transport pathways and
material accumulation on peak values and arrival timing. For this purpose, monthly discharges
were grouped into clusters; each cluster is a block of 3-6 monthly discharges as presented in
Table 2.

Effects of discharges on far field concentrations were examined by simulating scenarios, where
single clusters of discharges were excluded (zero discharge during months defined by a cluster)
from a simulation. Fig. 10 shows results of eight runs, each representing exclusion of one cluster
(A-H). The presence of the clusters A, B and C in the simulations has very little effect on activity
in Balbriggan during 1996 and literally no effect on distributions in subsequent years. The
winter/spring release (D) significantly contributes to Tc-99 concentration during summer 1996 and
influences spring concentrations of 1997. The cluster (E) load generates similar effect to spring
release, though overall concentrations are higher as discharges in this cluster are generally
lower. Both summer/autumn (F) and winter (G) releases markedly affect concentrations during
entire 1997 and 1998. The cluster (H) of longest duration but not the largest in terms of discharge
is responsible to a high degree for summer 1997 and 1998 peaks; its effect is evident also in
1999 concentrations.

5. Discussion

5.1. General circulation of Tc-99

The information derived from the Tc-99 transport model is consistent with the monitoring data
provided by the RPII and with existing knowledge pertaining to the surface circulation pattern of
the Irish Sea, particularly with the work of Leonard (1997). Some discrepancies can be attributed
to (1) resolution and accuracy of meteorological conditions, (2) Tc-99 fixed value inflow at the
boundary and (3) accumulation/remobilisation from sediment not reproduced in the model. The
pattern of material transport from Sellafield to the east coast of Ireland predicted by the model is
in good agreement with field observations and with migration maps of Jeffries (1982). Model
results, when complemented by monitoring data, enable to discuss fate of Tc-99 material released from Sellafield.

The annual net flow within the Irish Sea is northward and transports Atlantic water through St. George’s Channel to the west of the Isle of Man. A minor component of the 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). This mechanism is responsible for net advection of Tc-99 off Sellafield in a north-westerly direction along the southern Scottish coastline and later towards the entrance of the North Channel. During winter months the material is further transported northward to the Clyde Sea and Inner Seas on the Scottish coast, from where it takes approximately 3 years to reach Pentland Firth on the east coast of Scotland (Kautsky, 1985). However, as a result of variable meteorological conditions and/or development of seasonal density gradients, the northward annual residual flow may be disturbed inducing a southward net flow over a period of months. The spring and summer southward drift is associated with the development of thermohaline circulation within the WIG. Material becomes entrained into the gyre north-west of the Isle of Man through mixing processes with North Channel waters. It is advected southward along the Irish east coast at rates according to 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). Tc-99 distribution patterns along the east coast of Ireland shows that highest concentrations are found on the northeast coast with levels decreasing southwards down the coast. A small portion of the Sellafield signal is transported out of the Irish Sea to the south via St George Channel, and hence to the English Channel and west of Ireland.

The existence of the WIG is evidently reflected in the historical data recorded at Balbriggan; Fig. 11 presents previously shown field data and model results complemented by local sea surface temperatures to prove strong relationship between Tc-99 peaks and the strength of the WIG. A plume of material originating in the North Channel travels along the Irish coastline for approximately 4-8 weeks until it reaches Balbriggan typically between June and August. This is manifested in a sudden increase of concentrations and is usually preceded by a 2-month period
of relatively constant values. This sudden increase at the initial stage may be substantial; in the course of 2 weeks of August 1997 the concentration raised from background 10 mBq/l to 65 mBq/l, giving a mean daily increase rate to 4 mBq/l and overall increase of 550% in this period. Also, significant increases of 25 mBq/l and 28 mBq/l were observed during August of 1996 and 1998, respectively, giving a daily increase rate of 0.8 and 0.9 mBq/l. This is not surprising, since the rapid rise coincides with a peak of sea surface temperature (July-August) when the WIG is strongest and advection is fastest.

The peak maximum in Balbriggan is typically reached around 4-6 weeks after arrival of the leading edge of plume and is usually followed by a rapid decrease. Consequently, lowest levels are observed during winter months (from December to February concentrations are lowest). The annual cycle exhibiting seasonal oscillations is reasonably well reproduced from one year to another.

Atmospheric conditions affect water circulation patterns in the Irish Sea on the range of timescales and this in turn influences the preferred pathway and rate of transport of the Tc-99 plume. Sensitivity tests (not presented here), carried out to reveal model response to meteorological conditions, showed that wind stresses and heat fluxes are responsible for inter-annual and seasonal variations in Tc-99 transport. The role of air temperature and solar radiation in a development of density-driven baroclinic circulation is well recognized. The effect of wind stresses on longitudinal dispersion and mixing superimposed upon strong southward density-driven advection resulting from exceptionally warm summer were the key factors in the formation of the peak of September 1997. Surprisingly, the effect of variable discharge rates has secondary effects on the September 1997 peak concentration at Balbriggan as discussed below.

As shown in the case study with constant discharge rate, remote from the vicinity of Sellafield, the impact of the fluctuations in the discharge is progressively less marked. The most likely cause is that the residence time in the eastern Irish Sea is long enough to disperse individual Tc-99 pulses over the region before advecting it further. Also complex hydrographic transport pattern and long travel distances are conducive to material mixing and smooth out concentration gradients.
The effect of discharge time on far field concentrations was also investigated. Tc-99 concentration levels within the Irish Sea are highly variable and depend not only on discharge rates but also on discharge time strongly linked to sea dynamics that controls the buildup and removal of Tc-99 mass. The material released from Sellafield travels either southeastward along the coast of Cumbria during calm weather or northwestward during stormy conditions. The total mass of material of the 1995 discharge (highest rate of all annual records) which gradually accumulated over the year was effectively flushed out of the Irish Sea during winter months, and therefore had insignificant impact on concentrations in following years. The fast removal from the Irish Sea basin coincided with a record low winter/spring 1995/1996 sea surface temperatures associated with persistent ‘continental’ winter and a temporary negative phase of the NAO (Loewe, 1996). This result is revealed in a well-defined plume of higher concentrations in the west-central North Sea corresponding to the maximum discharge in 1995 (Kershaw et al., 1999). Tc-99 accumulated during 1996 in the eastern Irish Sea was transported southward during autumn towards Liverpool Bay; that impeded the exit of material through the North Channel in subsequent months. Consequently, a significant portion of material was retained in the Irish Sea system and contributed to exceptionally high levels on the east coast of Ireland during summer of 1997. Smith et al. (2001) maintains that there is a relationship between peak discharges of 1995 and peak values of Tc-99 measured at Balbriggan occurring approximately two years later. The numerical modelling analysis finds, however, that the relationship is more complex due to meteorological forcing.

There is a limited number of studies published in the literature that give estimates of transit times for the Irish Sea; moreover, these estimates are divergent. Smith et al. (2000) suggest a transit time between Sellafield to Balbriggan of around 3 years, whereas Smith et al. (2001) from a monitoring data deduced two-year transit time. Mitchell et al. (1987) found transit times to Dublin of 6-12 months, and proposed an upper limit of 1.6 years. Dabrowski and Hartnett (2008) provide for a seasonal variability in their numerical model and based on a one-year analysis only come up with leading edge transit times to Dublin for 100-140 days. The simple test on transit time conducted here explains why estimates were so different.
5.2. Assessment of Tc-99 monitoring programme

Although the Balbriggan dataset is sparse, it still provides a reasonably good picture of instantaneous values and temporal fluctuations. A comparison between these data and numerical output suggests that a number of short-duration peaks (peak of September 1997, July peaks of the years 1998-2000.) may have been missed by the monitoring programme due to the insufficient frequency of sampling. For this reason it is recommended to increase sampling frequency to observe if such peaks effectively appear. Also, higher sampling frequency is desirable to determine the rate of progress of the Tc-99 signal, particularly to distinguish the arrival of the leading edge of the plume, the peak concentration and the length of the trailing edge. It is suggested that the sampling resolution at Balbriggan in summer months (June-September) is increased to 1 sample per month. During winter months the current frequency should be sufficient as Tc-99 concentrations are rather low in this period.

For other stations on the east coast of Ireland (Bull Island, Carlingford, Cahore and Dunmore East), the same frequency as Balbriggan is suggested. With regard to the locations for monitoring stations they are well selected and give good insight into alongshore transport.

In the period considered, seven surveys (December 1993-September 1998) were performed. While the dataset for the eastern Irish Sea has sufficient horizontal resolution to describe Tc-99 densities and gradients, data for the western part is sparser and hence yields a less precise picture of Tc-99 distribution fields. The southern Irish Sea remains rather under-sampled throughout the survey period; hence greater uncertainty is associated with Tc-99 values in this region. The monitoring of other regions of the Irish Sea is less informative but due to relatively low concentrations is probably not required in a great detail. The current monitoring programme gives a relatively robust picture of spatial distributions at a point in time, though it would be desirable to perform cruise survey with horizontal resolution similar to that of COR 9b/98 (Fig. 5).

Although, offshore sampling provides a good framework for interpretation of spatial distributions and pathways, the temporal scale is too coarse to draw inferences about transport timescales. Four out of six surveys were performed during December when distributions are likely to be
similar over a large region of the Irish Sea. It is recommended that the timing of sampling is adjusted; sampling during January, April, July and October would provide a good insight into the scale of seasonal variability.

6. Summary and conclusions

In this study a validated transport model was used to hindcast the transport of Tc-99 and ultimately to examine the seasonal and inter-annual circulation patterns within the Irish Sea. The modelling work involved numerous tests to establish distribution pattern within the Irish Sea and temporal fluctuations on the east coast of Ireland and effect of meteorological conditions on these fluctuations. Transport pathways and transit times under seasonal conditions, effects of discharge rate and the role of material accumulation within the eastern Irish Sea on Tc-99 levels in remote locations were also investigated. From this research the following conclusions can be drawn:

1) Numerical modelling was found to be necessary to optimize operational monitoring programme of the Irish Sea, which due to its complicated topography (semi-enclose shape, two entrances), tidal input and atmospheric forcing constitutes a complex hydrodynamic system characterized by strong tidal currents and thermohaline circulation.

2) The model indicated a strong relationship between concentration peak levels on the east coast of Ireland and the strength of WIG. Air temperature and solar radiation are responsible for southward drift during summer along the coast of Ireland and highest summer peaks in Balbriggan.

3) Transit times from Sellafield to Balbriggan fall within the wide range of 30-240 days; with summer releases resulting in the shortest transit times.

4) The amount of material transported is highly depended on release times. Spring circulation patterns transport large masses of material and retain it over long timescales. Summer currents carry smaller quantities in the first year from release and higher in the second year.
5) Findings from current study are applicable to any conservative material having properties similar to Tc-99 (long half-life, low accumulation in sediment).

6) This research has contributed to a better understanding of Irish Sea transport processes. Findings from the model were used in an analysis of the RPII's monitoring programme and in an assessment of the adequacy of their sampling regime. Recommendation for optimization of monitoring programme to the RPII was one of the primary goals of this study. The following conclusions were drawn:

1) Sampling at Balbriggan provides a reasonably good picture of instantaneous values and fluctuations despite sparse temporal resolution. It is desirable that inshore monitoring is carried out at all 5 stations on a monthly basis throughout June-September period, and once per 2 months during off-summer periods.

2) Offshore sampling provides a robust picture of spatial distributions, though data densities for the western part are sparse and hence leading edge of the plume or material front can not be clearly determined in this region. Offshore surveys should be conducted at various times of the year in order to obtain a comprehensive image of seasonality.

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Figures

Fig. 1 Bathymetry of the Irish Sea, positions of numerical boundaries and locations of monitoring stations

Fig. 2 (a) Annual discharges to sea of Tc-99 (TBq) from Sellafield (1952-2006). Data up to 1978 are best estimates only (Gray et al., 1995) (b) Comparison between monthly discharges of Tc-99 from Sellafield (bars) and Tc-99 records at Balbriggan (squares)
Fig. 3 Comparison of measured and modelled (a) current speeds at location T1 and (b) water temperatures at location T1. Graphs reproduced from Dabrowski et al. (2009)

Fig. 4 Residual circulation of the surface layer (a) predicted by the ECOMSED model (from Dabrowski, 2005) and (b) similar plot reproduced from Horsburgh (1999)
Fig. 5 Contour plots of Tc-99 surface concentrations (mBq/l) derived from (a) McCubbin et al. (2002) and (b) numerical model.

Fig. 6 Historic and model Tc-99 timeseries at Balbriggan.
Fig. 7 Contour plots of Tc-99 concentrations (mBq/l) predicted by numerical model for January, April, July and October of 1996.
Fig. 8 Tc-99 tracer timeseries at Balbriggan for years 1995-1998. Releases were performed on: (a) winter, (b) spring, (c) summer and (d) autumn.

Fig. 9 Comparison of Tc-99 timeseries modelled using real discharges from Sellafield and constant rate discharges.
Fig. 10. Tc-99 timeseries, control run and run with a cluster of discharges excluded from simulation. These clusters are (a) cluster A, (b) cluster B, (c) cluster C, (d) cluster D, (e) cluster E, (f) cluster F, (g) cluster G, and (h) cluster H. Details of each cluster given in Table 2.
Fig. 11 Historic and model timeseries at Balbriggan along with corresponding SST at the same location