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The dispersion of ^{99}Tc in the Nordic Seas and the Arctic Ocean: a comparison of model results and observations

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Abstract

The radionuclide ^{99}Tc had been discharged from the nuclear reprocessing facility in Sellafield (UK) into the Irish Sea in increased amounts in the 1990s. We compare the simulated dispersion of ^{99}Tc in surface water as calculated by a hydrodynamic model and an assessment box model with field-observations from 1996 to 1999 to study concentrations, pathways and travel times. The model results are consistent with the observations and show the typical pathway of dissolved radionuclides from the Irish Sea via the North Sea along the Norwegian Coast. Subsequently the contaminated water separates into three branches of which the two Arctic branches bear the potential for future monitoring of the signal in the next decades. The results of the hydrodynamic model indicate a large variability of surface concentrations in the West Spitsbergen Current which has implications for future monitoring strategies. According to the observed and simulated distributions we suggest an improved box model structure to better capture the pattern for concentrations at the surface.

Keywords: Arctic Ocean; Nordic seas; Numerical modelling; Marine dispersion; Technetium

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1. Introduction

^{99}Tc is a highly soluble, beta-emitting man-made radionuclide. The main sources of ^{99}Tc are global fallout from nuclear weapons testing and discharges from reprocessing plants for spent nuclear fuel in North-Western Europe. Global fallout resulted in ^{99}Tc concentrations in the North Atlantic of about 5 mBq/m^3 in surface waters (Dahlgaard et al., 1995b). Radioactive wastes have been discharged from the reprocessing plants at Sellafield (UK) into the Irish Sea and at La Hague (France) into the English Channel since 1952 and 1966, respectively (Kershaw and Baxter, 1995). The discharges from Sellafield increased in the first quarter of 1994 when a new waste treatment plant, the Enhanced Actinide Removal Plant (EARP), began operation. EARP reduces the discharges of plutonium and americium, allowing the processing of stored Medium Active Concentrate (MAC), but does not significantly remove ^{99}Tc. Due to the increased throughput of material for treatment, the released quantities of this radionuclide have increased greatly since the commencement of EARP (Leonard et al., 1998; Kershaw et al., 1999).

^{99}Tc behaves conservatively in seawater and is transported from the Irish Sea into the North Sea via the Scottish Coastal Current, which eventually merges with water flowing through the English Channel, labeled with radionuclides from the French nuclear reprocessing plant at La Hague, in Danish waters near the entrance of the Skagerrak. This water becomes incorporated with the Baltic Sea outflow and forms the Norwegian Coastal Current (NwCC). Atlantic Water (AW) of the North Atlantic Current (NAC)—which has crossed the gaps between Iceland, Faeroes and Scotland from the North Atlantic into the Nordic Seas—continues northward as the Norwegian Atlantic Current (NwAC), and its eastern branch runs parallel to the NwCC. It progressively mixes with the NwCC bearing the radiotracer signal. Near the northernmost end of the Norwegian coast at the western boundary of the Barents Sea (~70°N) the NwAC splits into two currents: the northwards flowing West Spitsbergen Current (WSC) and the eastwards flowing North Cape Current (NCC). At present, ^{99}Tc can be detected along the NwCC and further north, for example in the Barents Sea (Brown et al., 2002) and in the WSC (Gerland et al., 2002a, 2003), a pattern that was observed in earlier years for ^{137}Cs (e.g. Dahlgaard, 1995) following enhanced discharges of this radionuclide in the 1970s. Concentration levels of radioactive material in seawater in the Arctic have been measured in a number of studies both for research and monitoring purposes (e.g. Dahlgaard, 1995; Gerland et al., 2002b; Kershaw and Baxter, 1995; Nies et al., 1998; Strand et al., 1994). Presently, the NRPA conducts monthly seawater sampling at the subarctic and Arctic locations Hillesøy (Northern Norway), Jan Mayen, Bjørnøya, Hopen and Ny-Ålesund (Spitsbergen) for ^{99}Tc level monitoring.

We compare model results from a three-dimensional coupled ice–ocean model which disperses the ^{99}Tc from the source to the Nordic Seas and the Arctic Ocean, with recent measurements of ^{99}Tc levels in seawater. The observations cover the northern part of the Nordic Seas in different years from the late 1990s. Further, we present a comparison of measurements with an assessment box model, which gives

average radionuclide levels involving both large distances (>1000 km) and long timescales, up to the order of 1000 years.

2. Material and methods

2.1. Model descriptions

The dispersion of the recent signal of ^{99}Tc is simulated with the three-dimensional coupled ice–ocean model NAOSIM (North Atlantic–Arctic Ocean Sea Ice Model) of the Alfred Wegener Institute for Polar and Marine Research. The model has been successfully used previously to investigate the circulation of ice and ocean in the Arctic Ocean and the Nordic Seas (e.g. Karcher et al., 2003; Kauker et al., 2003). The same model has also been used in a previous study to calculate the possible spreading of radionuclides from the sunken submarine 'Kursk' (Gerdes et al., 2001). For the present investigation, the model is driven with realistic daily atmospheric forcing data from 1979 to 1999. These atmospheric data are based on data from the European Centre for Medium-Range Weather Forecasts (ECMWF). The model domain covers the Arctic Ocean, the Nordic Seas and the North Atlantic north of 50°N. For a more detailed description of the model and the used forcing, see Karcher et al. (2003).

After a simulation of the period 1979–1989 the release of ^{99}Tc is started in January 1990 for a period of 10 years, using measured monthly discharge data for the period 1994–1999 (Fig. 1) and yearly discharges for the preceding period 1990–1993 (1990: 3.8 PBq, 1991: 3.9 PBq, 1992: 3.2 PBq, 1993: 6.1 PBq). The tracer is released into the uppermost model layer of the ocean (thickness 20 m) at the North Channel of the Irish Sea since the Irish Sea proper is not sufficiently resolved in the model.

For comparison purposes we also run the NRPA box model which is described in detail by Iosjpe et al. (2002). The box model concept is widely used for assessment studies of radioactive doses to biota or man. In contrast to the three-dimensional hydrodynamic model described above, the box model flow is based on a coarse grid with fixed exchange patterns between each of the boxes. On the other hand it includes parameterizations of diffusion of radioactivity through pore water, re-suspension, bioturbation effects and burial of activity in sediments. A novel feature of the NRPA box model, in contrast to previous box model approaches, is its inclusion of dispersion of radionuclides during time in order to provide a more realistic approach. The model box structure is given in Fig. 6. Radioactive decay is included in all model compartments.

2.2. Sample collection and measurements

The ^{99}Tc-measurements were performed by NRPA and CEFAS. For the data obtained by NRPA, 50 or 100 l of sea water were sampled and prefiltered (1 μm) in order to remove suspended matter. Then, ^{99}Tc was separated from other radionuclides by ion-exchange chromatography using AG1-X4 resin (100–200 μm mesh size, BIO-RAD Laboratories, CA, USA) and subsequent separation techniques such as co-precipitation and solvent extraction. The results of seawater have

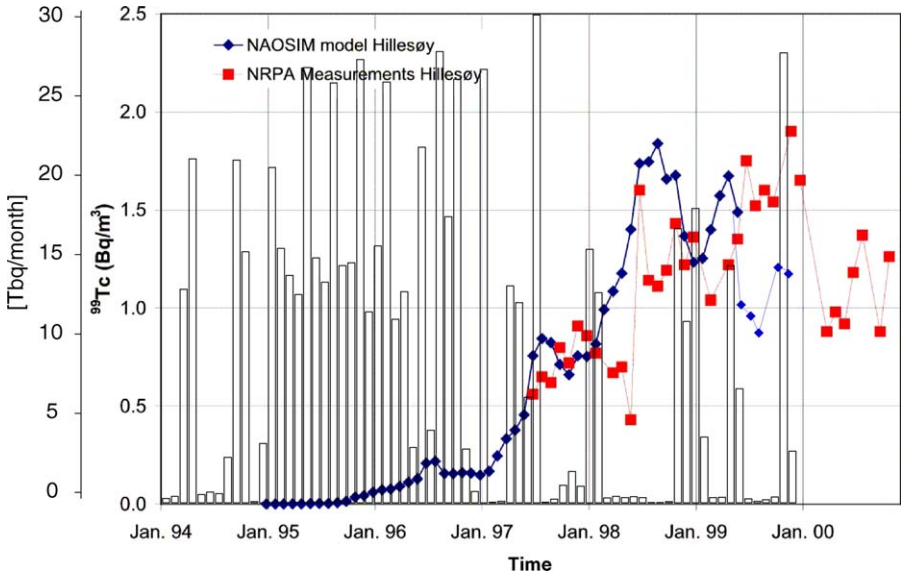

Fig. 1. Time series of the release function at Sellafield (bars), and surface concentrations from observations (squares) and NAOSIM results (rhombs) near Hillesøy at the northern Norwegian coast near 69°N for the period 1994–1999.

an accuracy of ±4% (Kolstad and Rudjord, 2000). The method is described in detail by Kolstad et al. (1999) and Chen et al. (2001). For the data obtained by IMR/CEFAS, discrete near-surface (5 m) water samples (100–150 l) were collected with shipboard pumps. The samples were not filtered prior to analysis, as they were collected in waters with low concentrations of suspended materials. Anion-exchange was used to separate ^{99}Tc prior to beta-counting, using rhenium as the yield monitor (Harvey et al., 1992). The combined analytical error for the measurements, due to chemical yield and counting statistics, was generally below 5%. CEFAS took part in an international intercalibration exercise in an earlier study of ^{99}Tc dispersion from La Hague (Dahlgaard et al., 1995a). This revealed that there was some degree of variability at low concentrations in seawater, for all the participants, but that these were not systematic and did not extend to the two different analytical methods being used. CTD observations were made at all sampling stations and at intermediate points.

3. Results and conclusions

The colour plates presented in Fig. 2 exhibit features of the ^{99}Tc dispersion at the surface during the simulation period, shown as monthly means for September of the years 1996–1999. Despite clear seasonal and interannual variability in the intensity of the surface velocity fields, the basic pattern in the Nordic Seas as described in Section 1 is rather stable over the simulation period. The eastern most

fraction of the NAC which crosses the gap between Iceland and Scotland is contaminated with the radioactive tracer and carries it into the northern North Sea northeast of Scotland (Fig. 2a,b). The contaminated water continues its passage anticlockwise along the British, Dutch, German and Danish coastlines to the Norwegian coast along which it mixes with the main branch of the NwAC on its western flank. A direct link from Scotland to the Norwegian coast exists only for short durations in some years, according to the model, as for example from November 1996 to February 1997 (not shown). At the northern end of Fennoscandia,

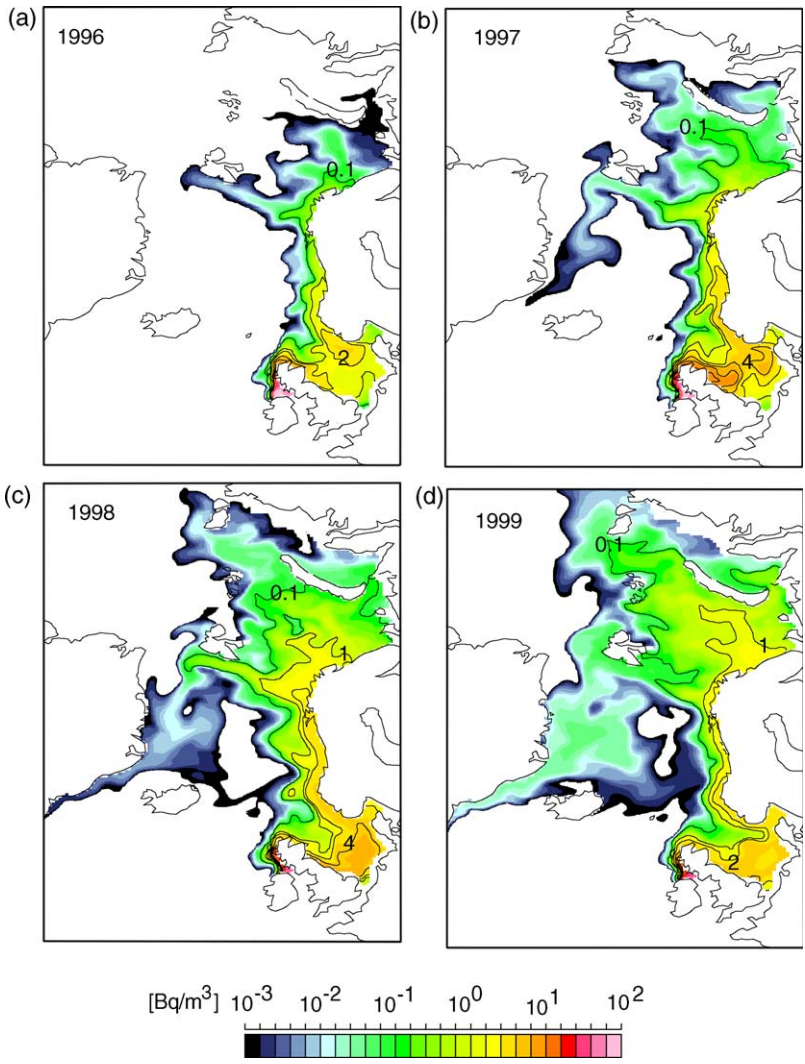

Fig. 2. Distribution of ^{99}Tc (Bq/m^3) in surface water in September (a) 1996, (b) 1997, (c) 1998 and (d) 1999, from model results (lines 0.1, 1.0, 2.0 and every 2.0 Bq/m^3) .

a larger portion of the ^{99}Tc radionuclides is transported eastwards into the Barents Sea and further into the Eurasian Basin of the Arctic Ocean (Fig. 2c,d). A smaller amount moves from the separation point northwards towards Spitsbergen where another separation occurs in the Fram Strait (Fig. 2c). One fraction of the contaminants enters the Arctic Ocean to the north with the WSC (Fig. 2d). Another part of the flow recirculates southward and joins the relatively uncontaminated polar water of the East Greenland Current (EGC) on its eastern flank. Note, the EGC has been contaminated by earlier reprocessing discharges (Aarkrog et al., 1987) and so observed ^{99}Tc concentrations are above the expected background due to global fallout (Kershaw et al., 1999). A fraction which leaves the EGC eastwards with the Jan-Mayen Current contaminates the interior of the Nordic Seas, namely the Greenland Sea Gyre. The EGC leaves the Nordic Sea through Denmark Strait.

A comparison of the modeled surface concentrations (Fig. 2) with observations in the North Sea and along the Norwegian coast (Fig. 3) shows good agreement. The levels of surface concentrations in the North Sea northeast of Scotland in the early phase of the enhanced releases (around 1994) were in the range of 0.5–1 Bq/m^3 in model results (not shown) and observations (Leonard et al., 1997; McCubbin et al., 2002). The input function for the simulation period (Fig. 1) shows the highest monthly input during the period January 1995 to November 1996. This peak release crosses the North Sea in 1996 and 1997. As a consequence, the simulated concentrations northeast of Scotland increase to a maximum of about 10 Bq/m^3 in winter 1996/1997. The available observations from November 1996 in this area reveal maxima of 7–8.5 Bq/m^3 (Fig. 3a), in line with values reported by McCubbin et al. (2002). The highest observed value is found near the Orkney Islands in November 1996, indicating a potentially active eastward shortcut flow across the northern North Sea, as has also been calculated in the model simulation.

The modeled concentrations for late summer 1997 (Fig. 2b) reveal surface values in excess of 2 Bq/m^3 (max 8 Bq/m^3) in a broad band from the British Isles across the North Sea and along the southern Norwegian coast similar to observed data (Fig. 3b). In 1998, levels above 1 Bq/m^3 reached the southwestern Barents Sea moving around the North Cape in the model (Fig. 2c). Observations from the summer of 1998 are not available for the Barents Sea proper but just east of the North Cape maximum concentrations were found to be lower with 0.86 Bq/m^3 at the surface (Fig. 3c). In comparing simulated and observed concentrations one has to keep in mind, however, that the passage of steep gradients may lead to rather large errors caused even by slight inconsistencies in location or time for the comparison. In summer 1998 the steepest gradient of concentrations passes this area. This is evident from a monitoring timeseries for ^{99}Tc taken at Hillesøy, a coastal station at the northwestern coast of Norway near 69°N, occupied since 1997 (Brown et al., 2002) (Fig. 1). A timeseries of modeled surface concentrations from this location reveals a good agreement. At the start of measurements in July 1997 the ^{99}Tc levels reach about 0.6 Bq/m^3 both in the model and in field data. From the modeled timeseries we can deduce that this level is already the result of a first increase of concentrations in 1997 advected with the NwCC. The steep increase in spring/summer 1998 raises the concentration above 1.5 Bq/m^3. The model is approximately

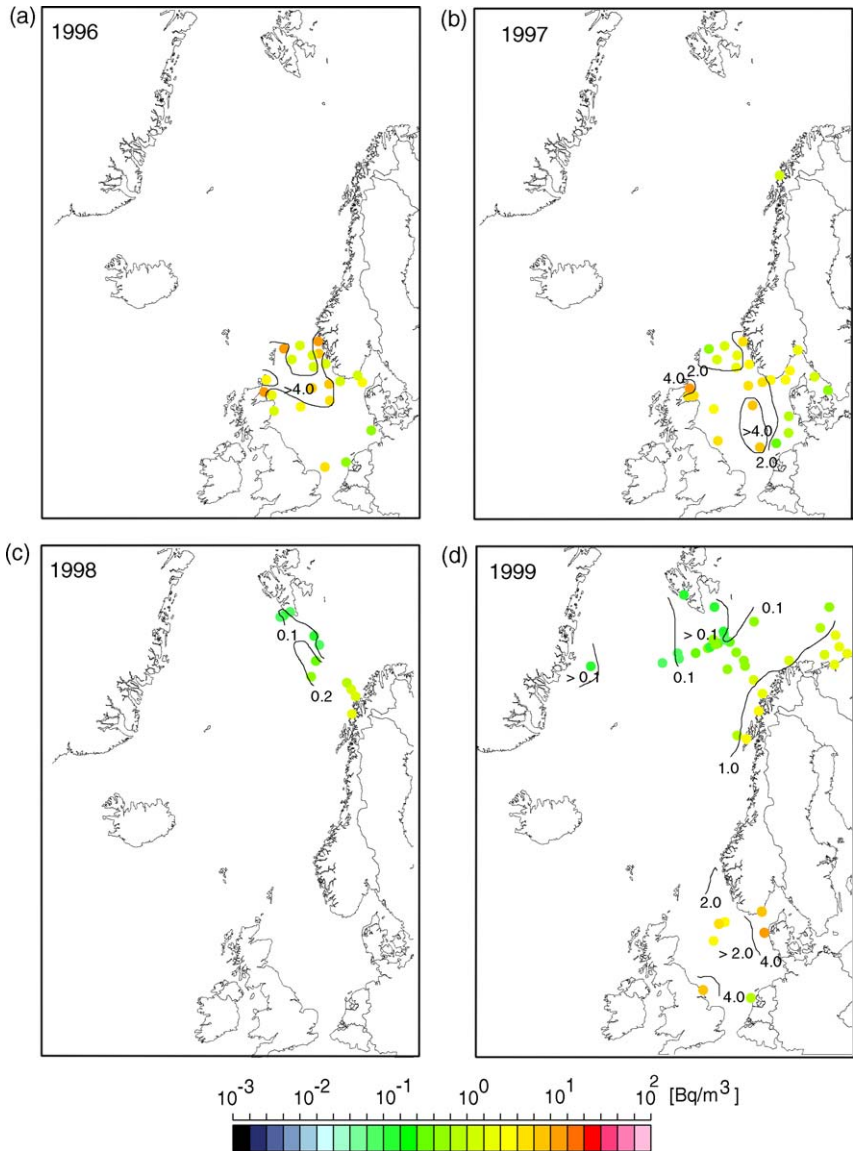

Fig. 3. Distribution of ^{99}Tc (Bq/m^3) in surface water in (a) 1996, (b) 1997, (c) 1998 and (d) 1999, from observations (same colour code as Fig. 2, isolines in Bq/m^3).

3 months ahead in relation to the field results. This can also be deduced from the decreasing concentrations in the simulated timeseries in the second half of 1999, which occur 5 months earlier than in the observed series. This may be due to a shift of the radionuclide release point, in the model, from the original location at Sellafield to the northern outflow channel of the Irish Sea. The

difference of 3–5 months, apparent in the timeseries at Hillesøy, is consistent with the estimated advective timescale of 3 months, in 1994, for a contaminated waterbody from Sellafield to the North Channel (Leonard et al., 1997, 1998). On the other hand local influences in the vicinity of the coastal observing station may lead to some of the differences with the large scale model, the horizontal resolution of which is ~28 km.

Further downstream the northward flowing water of the WSC between the northern Norwegian coast and Spitsbergen has a prominent seasonal to interannual variability of the velocities which is reflected in the modeled surface concentrations of ^{99}Tc. The strong seasonal variability also holds for the modeled vertical structure of the ^{99}Tc signal (Gerland et al., 2003). Until the summer of 1998 the concentrations in the WSC off Bear Island, half way between Norway and Spitsbergen, are below 0.1 Bq/m^3. In the summer of 1998, however, not only does the 'ramp' of ^{99}Tc contamination (i.e. the sudden increase in concentration due to EARP-related ^{99}Tc) enter the WSC, but also the WSC velocity is anomalously strong. This leads to maximum surface concentrations of 1 Bq/m^3 west of Bear Island and 0.5 Bq/m^3 at the southern tip of Spitsbergen in September 1998. This peak in WSC concentrations, however, only lasts for several months. In the summer of 1999 the surface concentrations near Bear Island are back to 0.1 Bq/m^3. The observations gained in June/July of 1998 and 1999 (Fig. 3c,d) both reveal a level of about 0.1–0.2 Bq/m^3 at the southern tip of Spitsbergen and between 0.2 and 0.3 Bq/m^3 in the WSC off Bear Island, potentially missing the peak concentration which might have passed in the previous autumn and winter. While the simulation is consistent with the summer observations in 1998, the simulated concentrations in summer of 1999 are lower than observed.

In the Barents Sea a strong inflow starting in 1998 pushes the ^{99}Tc ramp far northeastward until the summer of 1999 (Fig. 2d). The signal of the contaminant is able to reach the southeastern shores of Spitsbergen with rather large concentrations of 0.1 Bq/m^3 in summer 1999, in line with observations in this area (Fig. 3d). From the Barents Sea contaminated water leaves north- and eastward towards the central Arctic Ocean, and the Kara and Laptev Seas. According to results from previous model investigations we can expect the contaminated surface water to join the Transpolar Drift and finally to leave the Arctic Ocean via the Fram Strait in the next 5–10 years (Nies et al., 1998; Gerdes et al., 2001). Since the eastern Barents Sea is subject to large heat loss to the atmosphere and intense ice formation, dense water is formed here which subsequently leaves the shelf to join the Atlantic water layer at 200–1000 m depth in the Arctic Ocean basins (e.g. Schauer et al., 1997). This water travels on cyclonic loops along the basin margins and also leaves the Arctic via Fram Strait (Rudels et al., 1994), carrying along the contaminant burden from the European reprocessing facilities (Nies et al., 1998). We can expect the water which carries the EARP related ^{99}Tc burden increase to circulate in the Arctic Basins at mid-depth for the next 15–30 years, a timescale known from observation of previous releases and modeling results (e.g. Smith et al., 1999; Karcher and Oberhuber, 2002). A monitoring of the ^{99}Tc signal in the Arctic

Ocean in the following years offers an opportunity to study how Atlantic-derived water circulates in the Arctic Ocean and to validate numerical models.

In the EGC, which is contaminated by the recirculating Atlantic Water south of Fram Strait, peak levels are found in the model run in late 1998 and early 1999 in the offshore part with 0.01–0.1 Bq/m^3. This compares with levels of 0.05–0.22 Bq/m^3 observed in 1994, due to earlier releases (Kershaw et al., 1999). The interior Greenland Sea is affected by ^{99}Tc which circulates cyclonically with the Jan-Mayen Current and the Greenland Sea Gyre. Parts of the EGC contaminants are captured by the East Icelandic Current which moves eastward north of Iceland. Interestingly the interior Norwegian Sea stays rather unaffected by the contaminants due to the strong front on the western flank of the NwAC. Measurements of ^{99}Tc levels for the western Nordic Seas are rare and the only measurement from the late 1990s amounts to 0.13 Bq/m^3 in the recirculating Atlantic Water south of Fram Strait (Fig. 3d) in 1999. The model results indicate a pulsating intensity of the recirculating branch of contaminated AW leaving isolated lenses of increased ^{99}Tc concentration to travel southward with the EGC. The maximum concentration of these lenses in the model amounts to about 0.06 Bq/m^3. Part of this discrepency maybe due to still slightly elevated levels of ^{99}Tc in the EGC exiting the Arctic Ocean in the late1990s. In the mid-1990s these still accounted for 0.1–0.2 Bq/m^3 in the EGC northeast of Iceland (Kershaw et al., 1999).

The propagation speed of the ^{99}Tc 'ramp' from its origin to the entrance of the Barents Sea, according to both the model results and the observations, is between 2.5 and 3.5 years. This is considerably faster than the speed of the ^{137}Cs plume which has passed along the same route in the late 1970s and early 1980s. Kershaw and Baxter (1995) have estimated its travel time to be 4–5 years. A reason for this speed-up of the flow is the intense inflow of Atlantic Water across the Faroe–Scotland gap in 1994/1995 in contrast to 1974/1975. The 'ramp' of increasing discharges started in 1974 for the ^{137}Cs releases and in 1994 for the ^{99}Tc releases, respectively. In addition to the more intense transport into the Nordic Seas for the latter case, a more intense cyclonic circulation in the North Sea is apparent in the model run in 1995 as compared with 1974, and the NwCC was stronger in the 1996/1997 period than in 1975/1976 (not shown). How well do the results of the box model calculation for the dispersion of the ^{99}Tc release in the 1990s compare with the observations and the NAOSIM results? It is evident that due to the coarse resolution of the box model, its predicted concentrations for the oceanic compartment are representative of rather large areas. It is therefore important for the box model that its box structure is chosen according to the shape of the dominant flow regimes along which the dispersion occurs. Otherwise different waterbodies with respect to contamination will be represented by one single box. Fig. 4 shows a comparison of experimental data (Brown et al., 1999; Leonard et al., 1998) with box model results from three boxes: Cumbrian Waters (the box closest to Sellafield), the northern part of the North Sea and the Skagerak. While, in general the predicted timeseries for the Cumbrian Waters shows reasonable agreement, especially the strong increase in concentrations due to the release function in Sellafield (Fig. 1), the detailed correspondence with observations is not satisfying. Since

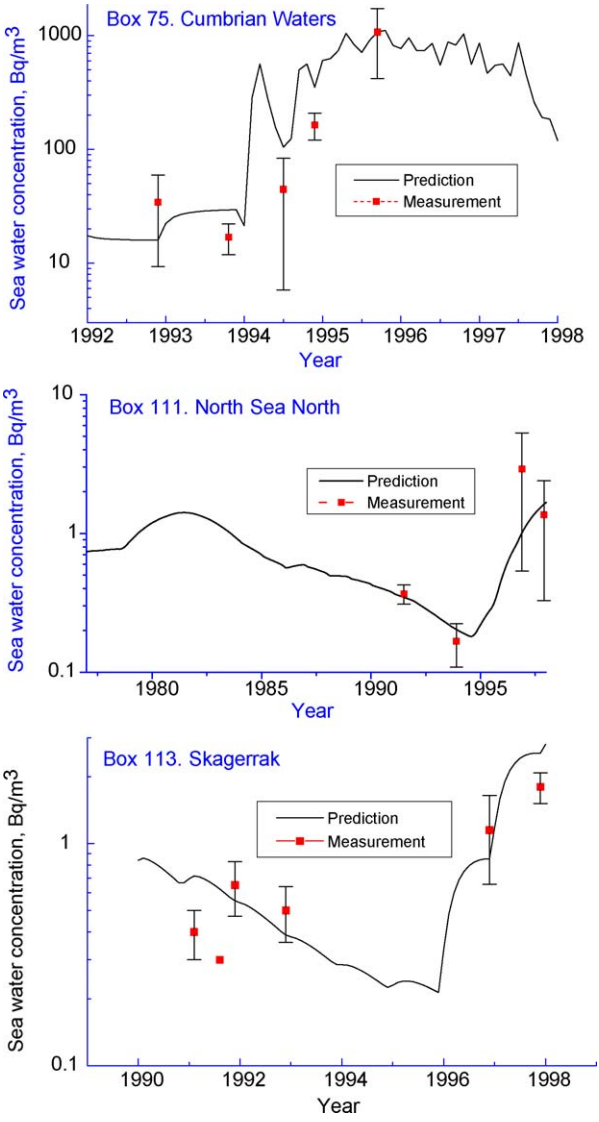

Fig. 4. Comparison of box model concentrations of ^{99}Tc for individual boxes and experimental data. The solid lines correspond to the results of calculations executed by the NRPA box model. Squares show the average values for the marine compartment with one standard deviation for experimental data.

the Northern North Seas box and the Skagerrak box predictions further downstream are in better agreement with observations, the disagreement for the first box can be mainly explained by the uncertainty of the information concerning the discharge of the radionuclide that is important especially for areas closest to the sources of contamination (the Cumbrian Waters). A second reason may also be

Fig. 5. Suggestion for an improved box model structure.

the sparse coverage with observations which leads to errors in the evaluation of spatial average values for comparison with the model. In addition, an inadequate resolution of the pathways of dispersion in this region of the Irish Sea by the box model is a possible flaw.

The comparison between results of the NRPA box model and of the NAOSIM simulation indicates the importance of a box model structure which is well adapted to the main pathways of dispersion. Consequently a reorganised box structure for the NRPA box model has been developed based on the results presented here and suggestions for an improved structure won from hydrodynamical arguments and previous modeling results (Karcher and Harms, 2000) which will be used in forthcoming box modeling efforts (see Fig. 5 in comparison with Fig. 6).

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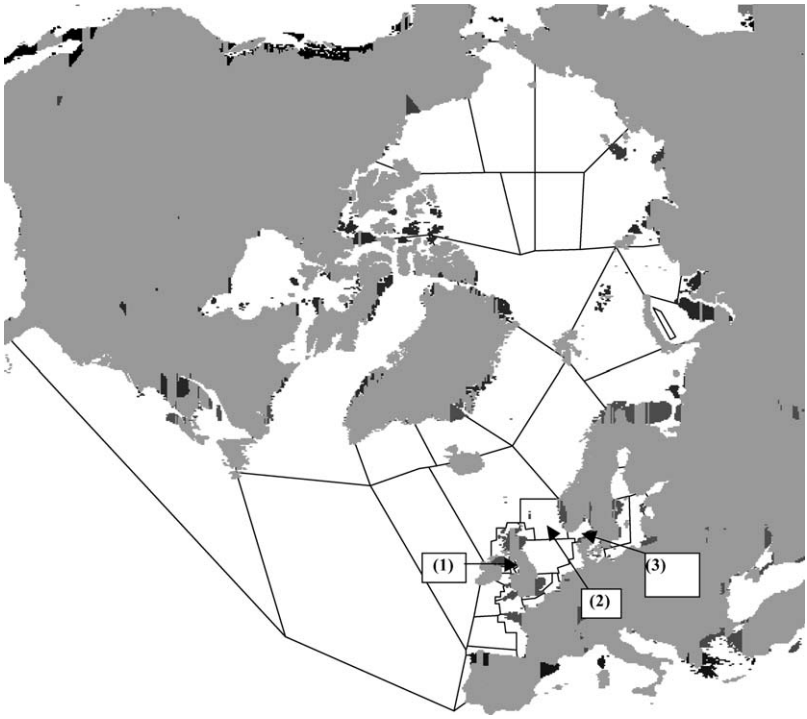

Fig. 6. The structure of the surface water boxes of the NRPA box model with three boxes used for the intercomparison in Fig. 4 (1—Cumbrian Waters; 2—the northern North Sea; 3—Skagerrak).

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