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Journal of Marine Systems 68 (2007) 24–38
JOURNAL OF MARINE SYSTEMS
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A reassessment of the dispersion properties of ^{99}Tc in the North Sea and the Norwegian Sea

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Received 16 June 2006; received in revised form 29 August 2006; accepted 27 October 2006
Available online 13 December 2006

Abstract

The spatial and temporal evolution of the beta-emitting, anthropogenic radionuclide Technetium-99 (^{99}Tc) in the North and Norwegian Seas have been simulated with a regional isopycnic Ocean General Circulation Model (OGCM), forced with realistic, daily averaged atmospheric forcing fields for the period 1975 to 2003. The main source of this soluble radionuclide is the reprocessing plants at Sellafield in the Irish Sea and La Hague on the French coast in the English Channel. The radioactive contaminants follow the general ocean circulation in the area, and are eventually transported northwards along the Norwegian coast heading towards the Arctic Ocean. Comparison with observational time series from two stations along the coast of Norway shows that the model fairly realistically captures both the amplitude and the temporal trend of ^{99}Tc from Sellafield and La Hague. To isolate the effect of changes in the ocean circulation and mixing on the released tracers, idealized tracers with a clock attribution are included for both sources. These idealized tracers measure the age along the pathways of the tracers from the two sources. The age from the Sellafield-derived tracer at the island Hillesøy on the northern coast of Norway is found to be in the range 5–8 years, which is slightly older than previous estimates of the related concept of transit time. A complex pattern of variability is identified, with local and regional atmospheric forcing influencing both the northward tracer flux and the time elapsed since the tracer left the source.

Keywords: Technetium-99; Tracer dispersion; Atmospheric forcing; Age diagnostic

1. Introduction

Technetium-99 is a highly soluble, long-lived anthropogenic radionuclide with concentration (or activity) level in the North Atlantic surface waters of about 0.005 Bq m^{-3}. This background concentration is mainly a result of global fallout from nuclear weapons testing in the 1950s and 1960s (Dahlgaard et al., 1995). Controlled discharges of radionuclides into the sea have been conducted from Sellafield in the Irish Sea (Fig. 1) since 1952 and from La Hague on the French coast in the English Channel since 1962, contributing to elevated concentrations of ^{99}Tc as well as other man-generated radionuclides.

In April 1994, a new radioactive waste treatment plant, the Enhanced Actinide Removal Plant (EARP), began

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doi:10.1016/j.jmarsys.2006.10.009

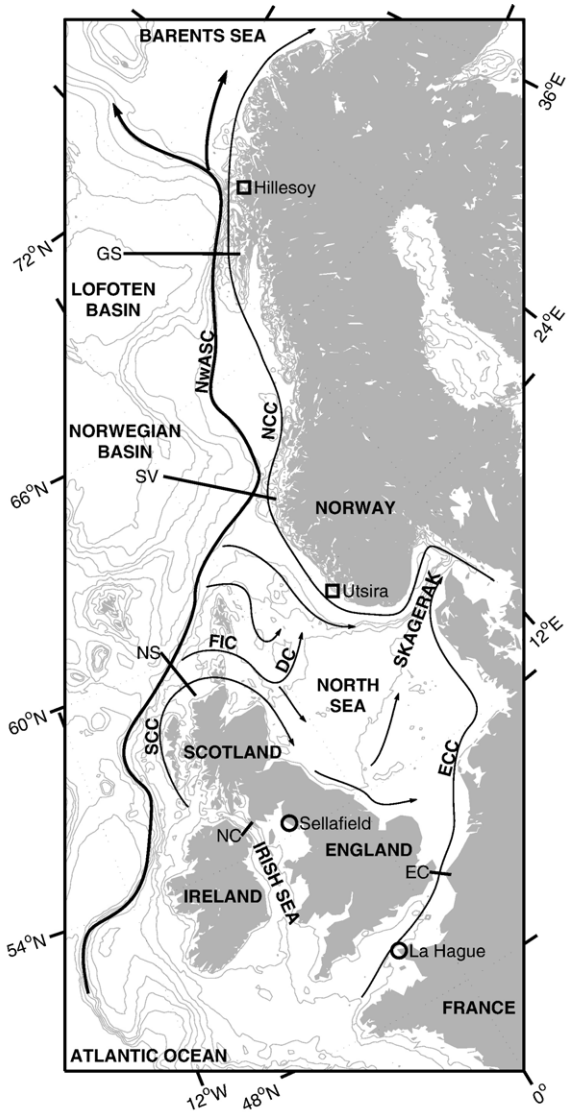

Fig. 1. Schematic figure of the most relevant ocean currents in the North Sea and the Norwegian Sea (Lofoten and Norwegian basins). The Norwegian Atlantic Slope Current (NwASC) is the eastern branch of the broader Norwegian Atlantic Current (NwAC). Other abbreviations are explained in the text. Isobaths are drawn for every 50 m to 200 m and every 500 m from 500 m. Marked straight lines indicate the sections used for transport estimates: Gimsøy (GS), Svinøy (SV), North Scotland (NS), North Channel (NC), and English Channel (EC).

operation at Sellafield (AMAP, 2004). The main goal of EARP was to reduce the discharges of Plutonium and Americium, but as a side effect resulted in increased discharges of ^{99}Tc (Fig. 2). The high release rates of ^{99}Tc from the mid-1990s were observed along the western coast of Norway and in the Barents Sea region several years later (Kershaw et al., 1999). The release of this long-lived radionuclide has, however, decreased considerably since the late 1990s (AMAP, 2004).

It has been shown that the current generation OGCMs are capable of simulating the dispersion of radionuclides like Strontium-90, ^{99}Tc and Caesium-137 in a realistic manner (e.g. Karcher et al., 2004; Gao et al., 2004, 2005). However, the variability of the ocean transport due to variable atmospheric forcing, and how ocean variability influences the transport of passive oceanic contaminants, are not straightforward. Based on observations, Kershaw et al. (2004) report a faster transport of the EARP-releases in the 1990s than the propagation of ^{137}Cs in the late 1970s, and argues that this is mainly due to a strengthened North Atlantic Oscillation (NAO), an atmospheric circulation mode mainly reflecting the strength of winter westerlies in the northern North Atlantic (Hurrel, 1995), in the latter period.

In this paper, the dispersion of ^{99}Tc from Sellafield and La Hague is simulated from 1975 to 2003. The available time series from Sellafield and La Hague, annual release rates of ^{99}Tc from 1975 to the end of 1989 (Fig. 2a) and monthly release rates from 1990 to 2003 (Fig. 2b), are used. However, it is important to note that the first years of release from Sellafield (1975 to 1980) are estimates based on observed concentration of ^{99}Tc in seawater just off the reprocessing plant (Dahlgaard et al., 1997). This will possibly add an extra bias to the simulated concentration field in the late 1970s and the early 1980s. Due to the fact that most of the observations of ^{99}Tc from the North Sea and along the Norwegian coast are from the late 1980s and onwards, we will focus on this part of the simulated period. The evolution of an idealized tracer resembling pure water from the two source regions is simulated over the same time period to distinguish the role of release rate from the influence of underlying ocean dynamics. Recent advancement of numerical tools for calculating the *age* of tracers (Deleersnijder et al., 2001) are implemented and used to rigorously estimate the mean time elapsed since the tagged water masses left their source regions. It should be realized that a tracer signal in the ocean carries a continuous distribution of *ages* from one region to another. Due to the presence of mixing, no single age can completely nor uniquely summarize the transport. Furthermore, tracers with different release rates also produce different age estimates.

In Section 2 the OGCM and the tracer module used in this study are presented. Section 3 gives an overview of the possible contaminant pathways of ^{99}Tc in the area of interest, along with a careful analysis of how the atmospheric forcing influence the ocean currents and mixing in the different regions. The simulated spatial and temporal

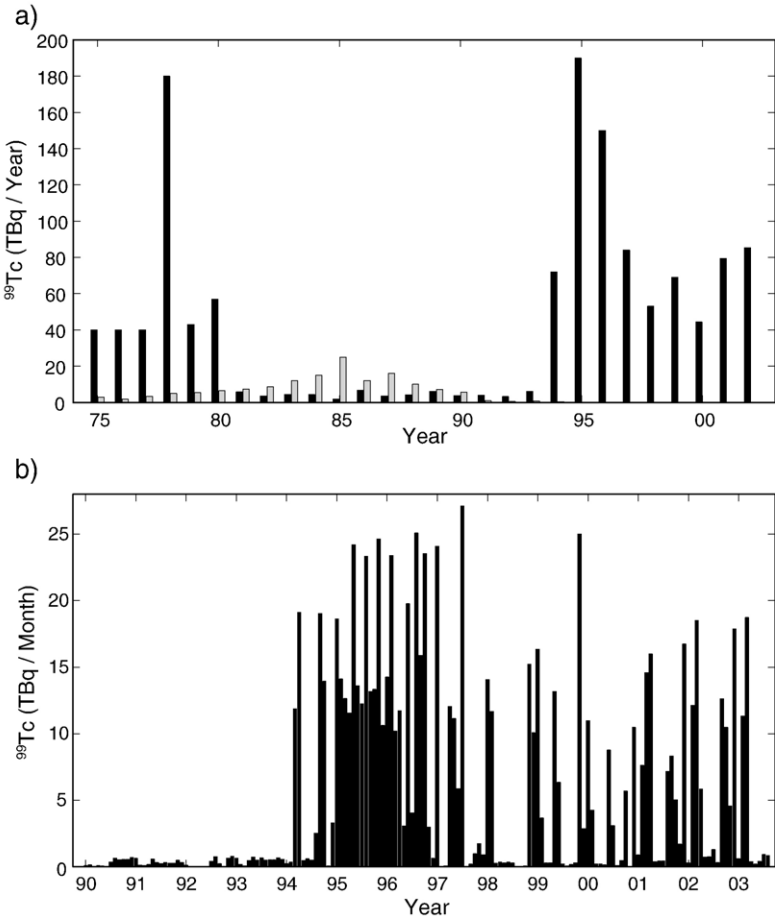

Fig. 2. (a) Annual releases of ^{99}Tc (10^{12} Bq) from the reprocessing plant at Sellafield (black bars) and La Hague (grey bars). (b) Monthly releases of ^{99}Tc (10^{12} Bq) from the reprocessing plant at Sellafield during the 1990s.

distribution of ^{99}Tc and comparison with available observations are presented in Section 4. Finally, discussion and concluding remarks are given in Section 5.

2. Method

2.1. Ocean general circulation model

The model system applied in this study is a nested, medium-resolution regional version of Miami Isopycnic Coordinate Ocean Model (MICOM; Bleck et al., 1992), fully coupled to a sea ice module consisting of the Hibler (1979) rheology in the implementation of Harder (1996), and the thermodynamics of Drange and Simonsen (1996). The regional model has demonstrated good skills in reproducing observed anomalies in the climate systems of the northern North Atlantic (Hatun et al., 2005a,b; Mauritzen et al., 2006). In the vertical, the model has 25 layers with fixed potential densities, and an uppermost prognostic mixed layer (Gaspar, 1988) with freely varying temperature and salinity, and thus density. The horizontal grid resolution in the region of interest is about 20 km. The model domain covers the northern North Atlantic Ocean and the Nordic Seas (from about 30°N to about 80°N). At the lateral boundaries, weekly-resolved velocity, temperature and salinity fields from a global 40 km version of the same model (e.g. Nilsen et al., 2003; Drange et al., 2005) are forced onto the regional model. The OGCM is driven by daily atmospheric forcing fields provided by NCEP/NCAR for the period 1948–2003 (Kalnay, 1996). Although 6-hourly forcing fields are available, the main features of the atmospheric forcing are transferred to the OGCM by using daily forcing fields.

2.2. Eulerian tracer module

The spatial and temporal evolution of the concentration distribution from a conservative radioactive

tracer like ^{99}Tc is governed by the Eulerian transport equation:

$$\frac{\partial C}{\partial t} + \nabla \cdot (\mathbf{v}C - \mathbf{K} \cdot \nabla C) = q - \lambda C.$$

Here C ($\mathbf{x}$, t) is the concentration (Bq m^{-3}), $\mathbf{v}(\mathbf{x}, t)$ is the velocity field, $\mathbf{K}$ is the diffusivity tensor, $q(\mathbf{x}, t)$ is the source function for C (specified as flux of tracers into the surface grid cell at the source divided by the volume of that grid cell), and λ is the radioactive decay term ($\sim$213 000 years; implying that ^{99}Tc can be treated as an inert component on the time scales considered here). This transport equation is linear since the tracer concentration has no influence on the flow.

In order to investigate the dispersion properties without taking into account the complicating factor of a variable release rate and the radioactive decay, we also simulate a passive tracer without varying release rates from Sellafield and La Hague. To make these tracers resemble pure seawater from a point source, we prescribe a constant value for the surface grid cell at the source and set $\lambda = 0$. We emphasize that the boundary condition for the idealized tracer is qualitatively different than the one used for the ^{99}Tc tracer; there is no mass flux into the model domain at the source, non-vanishing concentrations of the tagged tracer are therefore governed by dispersion of tracers that has been in contact with the surface grid cell at the source. In the following we denote these tagged water masses C^* and refer to the tracers as *idealized* tracers. Finally, we add an Eulerian age to C^* following Deleersnijder et al. (2001):

$$\frac{\partial \alpha}{\partial t} + \nabla \cdot (\mathbf{v}\alpha - \mathbf{K} \cdot \nabla \alpha) = C^*,$$

where the age concentration is given by $\alpha(\mathbf{x}, t) = a(\mathbf{x}, t) \cdot C^*(\mathbf{x}, t)$, hence the age $a = \alpha / C^*$. Initially, the age from both sources is zero in the model domain. During the integration, the age is prescribed to zero for the source water C^* at the time of its production. Due to the presence of mixing, a region downstream of the source consist of constituents taking different routes after leaving the source, hence they have different ages. The computational procedure outlined above delivers the mass-weighted average of the age distribution (for simplicity, we will refer to this quantity as the age). The tracer equations are discretizised on a C-grid, and are run *online* with the OGCM.

3. Ocean circulation

3.1. Contaminant pathways on the north-west European shelf

Before studying the model results we will here give a brief overview of the known circulation patterns and contaminant pathways in the prime area of interest. Fig. 1 schematically shows the main features of the current systems involved in the study area.

From Sellafield in the Irish Sea the dominant pathway of radioactive contaminants is northward through the North Channel, although a small proportion of the Sellafield signal is transported south, via the St. Georges Channel to the English Channel (EC), and then to the southern North Sea. A northward tidal residual is present in the Irish Sea, but the flushing is largely controlled by the speed and the direction of the wind, with along-channel winds yielding the strongest volume transport, with a complex transverse structure (Davies and Hall, 2000). Contaminants escaping through the North Channel are entrained into the Scottish Coastal Current (SCC), which enters the North Sea shoreward of the Atlantic Water influenced Fair Isle Current (FIC). All the way from the Scottish Shelf and along the British coast there is mixing between the two currents, and thus radionuclides are transported towards the south as well as into the interior of the North Sea (Kershaw et al., 1999).

The dominant circulation pattern in the North Sea is widely acknowledged to be anti-clockwise, while the circulation in the central North Sea is unsystematic and mainly wind driven (Dooley, 1974; Winther and Johannessen, submitted for publication). There are some observed currents across the North Sea, the most well known being the Dooley Current (DC), a continuation of the FIC, leaving the Scottish east coast heading towards Norway, following the edge at the 100 m isobath (Dooley, 1974; Svendsen et al., 1991). The other Atlantic inflows to the North Sea, the diffuse Shetland Shelf inflow and the inflow to the Norwegian Trench from its opening, may also serve as contaminant pathways toward the Norwegian side of the North Sea. For the contaminants southbound along the British coast there are also possible pathways across the North Sea before reaching the English Channel area. Baroclinic features may occur during summer when the deep northern North Sea is thermally stratified (Otto et al., 1990) and a seasonal eastward jet-like current have been observed across the central North Sea (Brown et al., 1999).

Radioactive contaminants from La Hague are transported through the English Channel by the European

Coastal Current (ECC), following the north-west European coastline, and are eventually mixed with contaminants from Sellafield in the southeastern North Sea and in the Skagerak (Dahlgaard, 1995).

The water in Skagerak, as described here and with addition from the Baltic Sea and the Norwegian Coastal Water, constitute the main outflow from the North Sea; the Norwegian Coastal Current (NCC). This relatively fresh and persistent current propagates along the Norwegian coast all the way to the Barents Sea and into Russian waters. Along its path the NCC borders the Norwegian Atlantic Current (NwAC), and areas of strong mixing between the two are the northern parts of the Norwegian trench, the Svinøy area and outside the Lofoten Islands (68° N, see Fig. 1 for the geographic locations) (Jakobsen et al., 2003). This mixing dilutes the radioactive signal from the North Sea. For instance Gascard et al. (2004) found that about half of the radionuclide Iodine-129 (^{129}I), mainly originating from La Hague, was transferred from the NCC to the NwAC before reaching the Lofoten Islands. Dilution from Norwegian runoff (0.011 Sv; Mosby, 1962) is negligible compared to the volume transport of the NCC (>0.7 Sv; Björk et al., 2001).

3.2. Atmospheric forcing on volume transport

Analyses have shown that the volume transport in the narrow North Channel and the English Channel are controlled by the amount of along-channel winds (Davies and Hall, 2000), while on the northern tip of Scotland and in the Svinøy section the volume transports are largely controlled by geostrophic adjustment, with strong westerlies generating high positive volume transport anomalies (Nilsen et al., 2003).

In order to investigate how atmospheric forcing influences the transport of radioactive contaminants, volume transports from the ocean model in four sections relevant for the Sellafield and La Hague releases are examined. These are the North Channel (NC), the English Channel (EC), the North Scotland Section (NS), and the Svinøy section (SV) (Fig. 1). Volume transports were calculated as net fluxes (inflow minus outflow), and defined as positive in the direction of the mean flow perpendicular to the chosen section.

Monthly mean volume transports for the winter months (December–March) as well as winter mean values using the same months were regressed on corresponding mean Sea Level Pressure (SLP) data fields from NCEP/NCAR-reanalysis. Fig. 3 shows the results of the monthly regressions. On the variability of the simulated NC and EC transports, wind variability in a cyclonic pattern centered in the North Atlantic just off the British Isles has the strongest influence (Fig. 3a,b). Both patterns contain variability aligned with the respective channels, especially their outlets (for the EC this is through the North Sea).

The corresponding regression patterns for transports through the NS and SV Sections (Fig. 3c,d) are practically identical. They are also similar to the typical NAO-pattern since the mid-1970s (as was also reported for the Faroe Shetland Channel Atlantic Inflow by Nilsen et al., 2003). The interannual variability (winter means) show similar results (not shown), but for EC and NC there was a slightly less direct alignment with the local geography and more influence from the dominant variability in the large scale atmospheric circulation (i.e. NAO).

4. Dispersion of ^{99}Tc from Sellafield and La Hague

4.1. Simulated concentration field

The simulated surface distribution of ^{99}Tc over the period March 1992 to March 2002 is presented in Fig. 4. The surface is in this text defined as the upper 200 m of the water column. For MICOM, with a prognostic bulk mixed layer, 200 m can be taken as a typical annual mean depth value of the mixed layer thickness. In the early 1990s (Fig. 4a) the concentration is 2–5 Bq m^{-3} in the English Channel and the central North Sea. In 1994 and onwards, EARP-related tracers escape the Irish Sea in a plume-like structure through the North Channel (Fig. 4b), entering the North Sea with the SCC (Fig. 4c, d). Elevated concentrations of ^{99}Tc are gradually dispersed farther north along the Norwegian coast with the NCC. In March 2000 (Fig. 4e), values of 2–5 Bq m^{-3} were seen before reaching northward to the Lofoten Islands. Eventually, after discharges of ^{99}Tc from Sellafield were reduced in the second half of the 1990s, lower concentrations are found along the Norwegian coast in 2002 (Fig. 4f) while the concentration in the North Sea is still relatively high, 2–10 Bq m^{-3}. The concentration of ^{99}Tc does not drop down to the pre-EARP values. A relevant question here is to what extent and for how long time will an instantaneous pulse-release from Sellafield or La Hague influence the North Sea or the Norwegian Sea. Some of these aspects will be discussed later in this paper.

Increased concentrations of ^{99}Tc has also been observed in the core of the Atlantic inflow water north of Scotland, hence indicating the possibility for Sellafield-derived contaminants taking a shortcut northwards following the NwAC (Kershaw et al., 1999). From

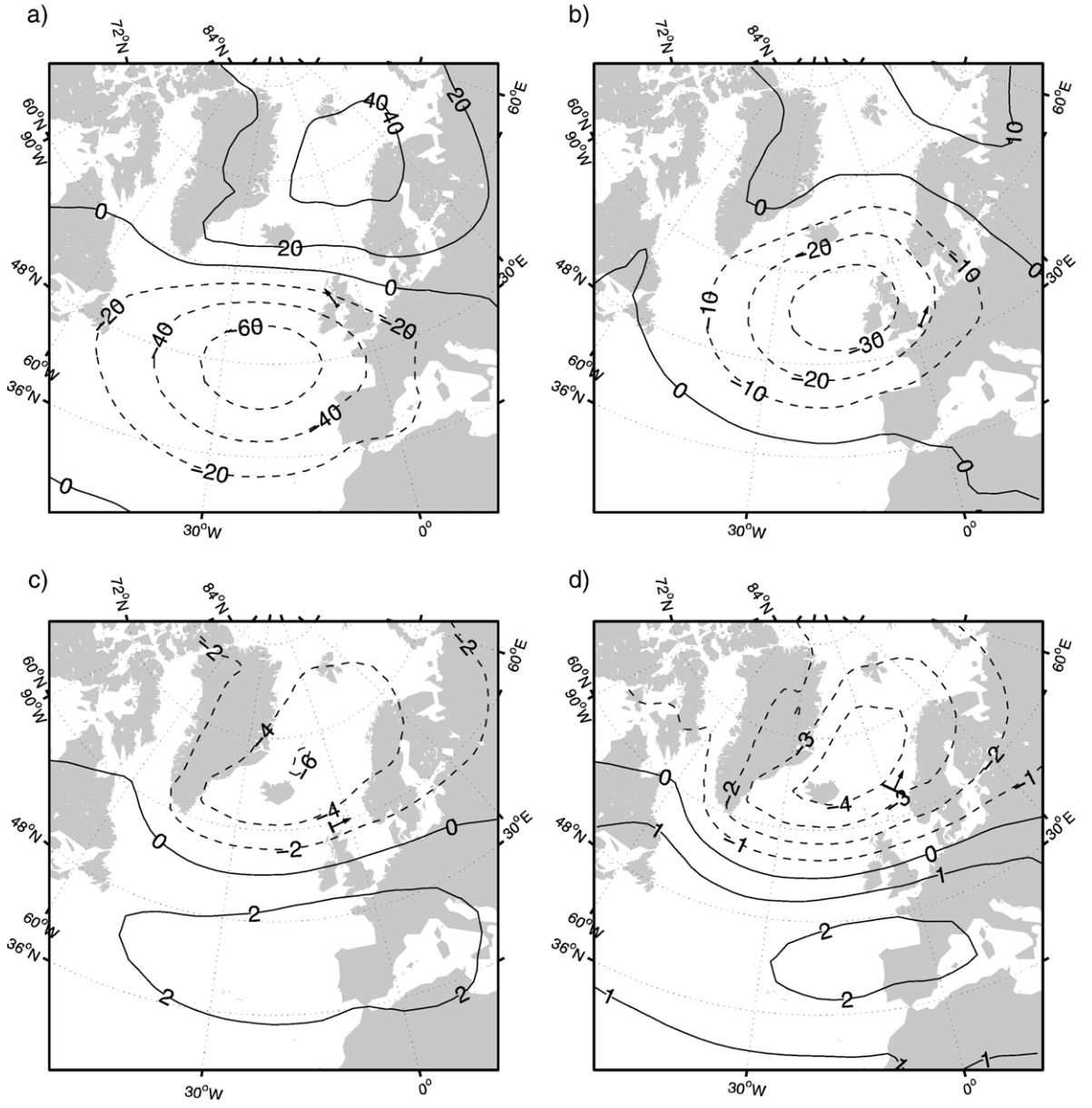

Fig. 3. Regression coefficient maps for NCEP/NCAR monthly mean SLP (Dec–Mar) regressed on simulated volume transports through the (a) North Channel, (b) the English Channel, (c) the North Scotland section, and (d) the Svinøy section. Straight lines and arrows indicate location and positive direction of the respective volume transports. Correlation coefficients in negative centers of action (center closest to the transport locations) reach 0.6 (a,b) and 0.8 (c,d), and all non-zero contours are significantly different from zero at the 95% confidence level.

vertical sections of the tracer transport north of Scotland and on the south-west coast of Norway derived from our model (not shown), about 20% of the Sellafield-derived contaminants escapes the North Sea transport route.

The radionuclides released from La Hague are initially entrained in the NCC, but mixing with the NwAC along the Norwegian coast causes a net transport of radionuclides from the NCC to the NwAC, also seen from observations (Gascard et al., 2004). It is beyond the scope of this paper to quantify the exchange rate between these two current systems, although an attempt to do this with another ocean model with higher horizontal resolution will be made in the near future based on passive tracers initially carried by the NCC in the Skagerak region.

Fig. 5 shows the simulated ratio of the ^{99}Tc concentration originated from Sellafield to the total concentration from Sellafield and La Hague. In 1994 (Fig. 5a), the

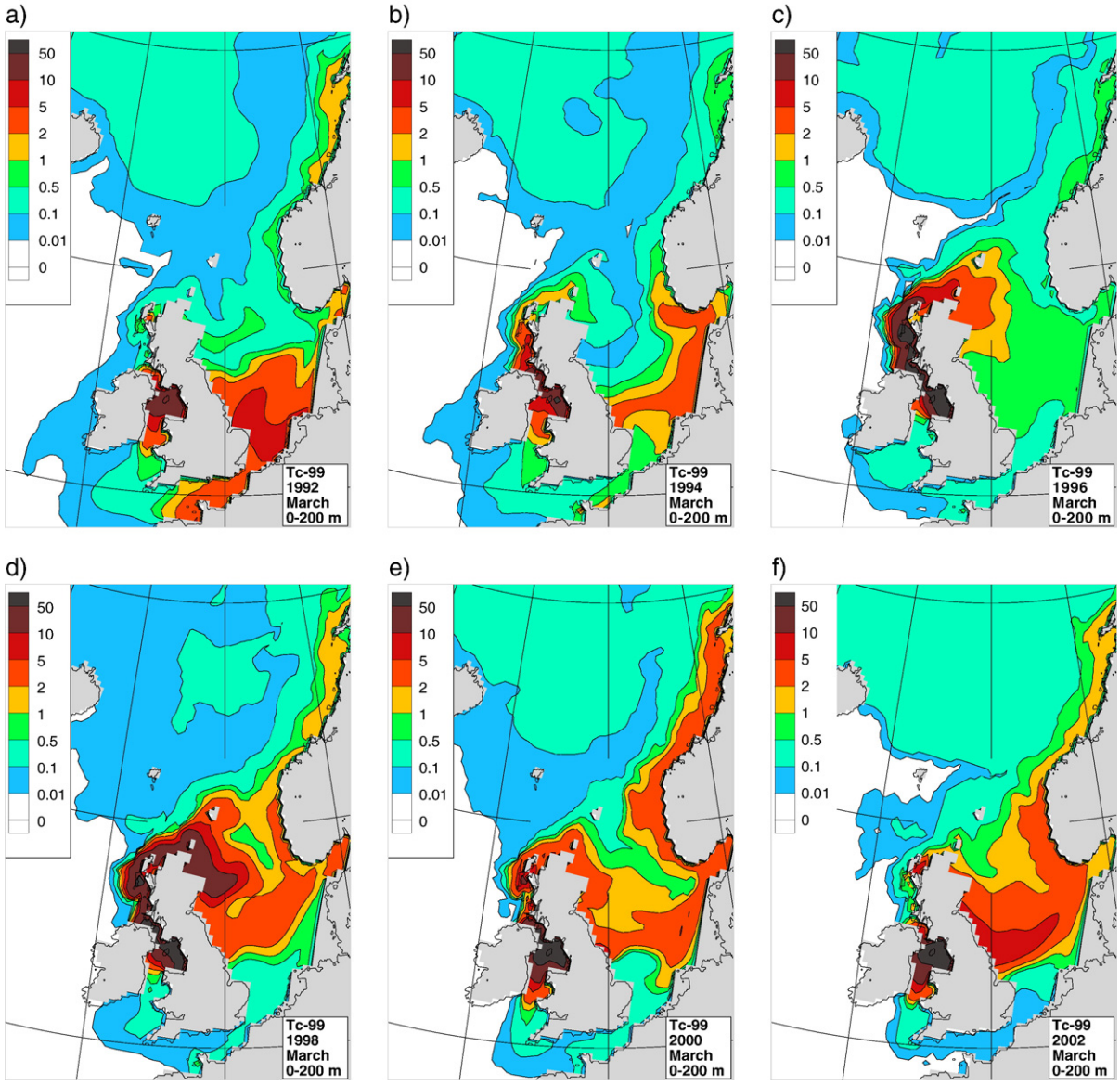

Fig. 4. Simulated concentration of ^{99}Tc (Bq m^{-3}) in the surface from Sellafield and La Hague, (a) March 1992, (b) March 1994, (c) March 1996, (d) March 1998, (e) March 2000, and (f) March 2002.

contribution from La Hague is still significant, in fact, in the southern North Sea and along the Norwegian coast, the Sellafield-derived contamination only accounts for 20–30% of the total concentration. In 1998 and 2002 (Fig. 5b,c), the EARP-related contribution from Sellafield rapidly overtakes La Hague as the dominant source in the whole region. As mentioned earlier, a small portion of the Sellafield signal is transported south via the St. Georges Channel into the English Channel. Therefore, due to the much lower input from La Hague in this period (roughly two orders of magnitude difference), the Sellafield signal dominates in the English Channel, even though the bulk of tracers from Sellafield is transported through the North Channel.

4.2. Data — model comparison

To evaluate the performance of the OGCM, two time series of observed ^{99}Tc concentration in seawater and one time series of observed concentration in seaweed from the coast of Norway are used. The first time series is observed seawater concentration from the island Utsira located off the west coast of Norway (Fig. 1). These observations are provided by the *Institute for*

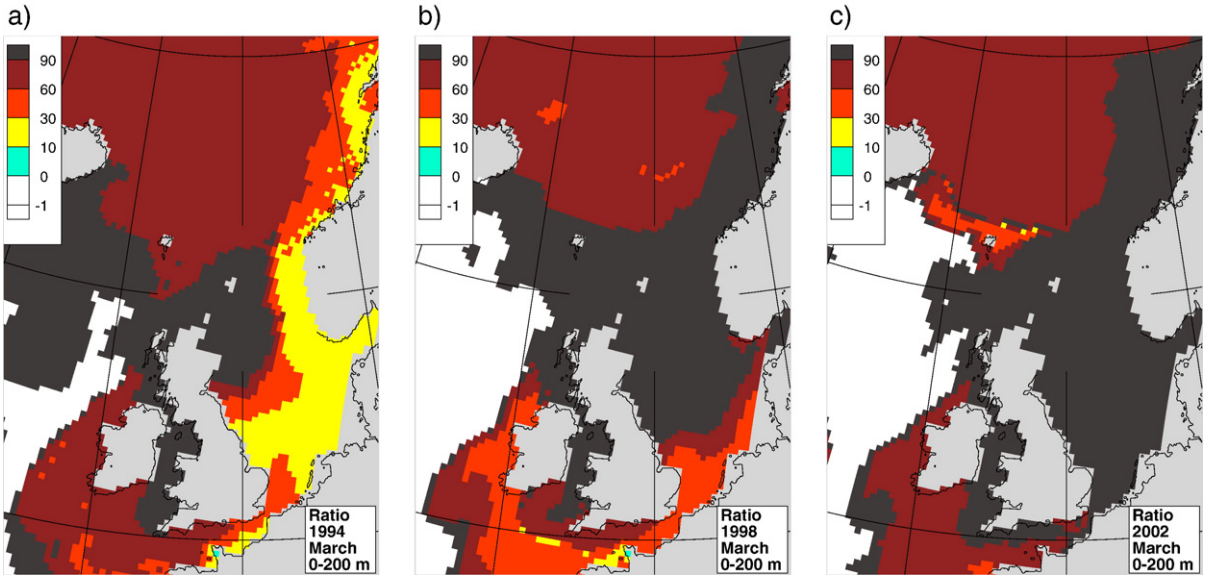

Fig. 5. Ratio (in percent) of simulated concentration of ^{99}Tc from Sellafield to the total concentration (Sellafield + La Hague) in the surface, (a) March 1994, (b) March 1998, and (c) March 2002. White areas denote low total concentration, less than 0.001 Bq m^{-3} (−1 on the legend scale).

Energy Technology (IFE) (Gordon Christensen, pers. comm., 2006) and covers the period March 1986 to April 1993. The model captures the interannual trend and amplitude fairly well, although the seasonal variations are in general not represented in a realistic manner (Fig. 6). The simulated concentration from Sellafield corresponds roughly to half of the total concentration, indicating that contamination from Sellafield and La Hague are equally important at this location and for the displayed period.

It is difficult to precisely determine when the ^{99}Tc-enriched water originating from the EARP-releases reaches Utsira (or elsewhere on the Norwegian coast). The simulated concentration time series suggest that it occurs during the fall of 1996, which is indirectly confirmed from observation of radioactivity in sea-weed located at Utsira (Fig. 7; data obtained from IFE). Maximum concentration is observed in May 2000, while the simulated seawater shows maximum concentration in December 1999. It should be noted that seaweed accumulates ^{99}Tc almost immediately, but its concentration show a time lag when seawater concentration decreases (Dahlgaard et al., 1997). As a consequence, concentration in seaweed do not decrease at the same speed as seawater. Another ”problem” with seaweed is the seasonality in uptake of radioactivity. If exposed to constant concentration of ^{99}Tc in seawater, the concentration in seaweed would be higher in the winter than in the summer, while for ^{137}Cs this mechanism is opposite (Dahlgaard et al., 1997). For these reasons, a one-to-one comparison between radioactive concentration in seawater and in seaweed should be made with care.

The third time series is from observed concentration in seawater at the island Hillesøy on the north coast of Norway (Fig. 8). These data are obtained from the *Norwegian Radiation Protection Authority* (NRPA) (Morten Sickel, pers. comm., 2005) and covers the period July 1997 to December 2002. Here we see an increase in the concentration some years after the EARP-releases, starting around the fall of 1997, with gradually decreasing levels starting in mid-2001 (Fig. 8). The observations show two distinct peaks, one in the winter 1999/2000 and the other in the following winter. These peaks are clearly seen and are comparable in time and amplitude in the OGCM as well. Sellafield is by far the dominant source at this location for this period.

4.3. The influence of ocean dynamics on tracer dispersion

The concentration of idealized tracers from Sellafield and La Hague, described in Section 2, is only affected by the underlying ocean dynamics. In other words, enhanced ocean transport gives higher concentration downstream of the source, reduced ocean transport gives less concentration. In contrast, the ^{99}Tc tracers are dependent on both the ocean dynamics and the release rate. Comparing these two tracers will therefore show

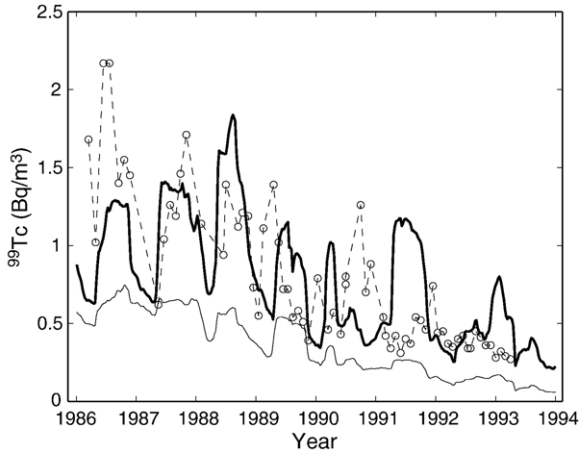

Fig. 6. ^{99}Tc concentration (Bq m^{-3}) in seawater at Utsira (59.18° N, 4.53° E) from observation (open circles) and the surface of the OGCM (bold solid line) for the period 1986 to 1994. The simulated contribution from Sellafield only is indicated with the thin solid line.

the indirect effect of the release rate (Fig. 2) on the subsequent concentration evolution at downstream locations.

Normalized concentration at Hillesøy of ^{99}Tc and idealized tracers from both Sellafield and La Hague are shown for the simulated time period in Fig. 9. Apparently, variations in the ^{99}Tc tracers from Sellafield follows the idealized tracers from Sellafield. However, some features can only be explained by the release rate. For instance, the two peak values at Hillesøy for the ^{99}Tc tracers already discussed are not as dominating for the idealized tracers, hence only the release rate can explain them. The variability of the idealized tracers from Sellafield is much larger than that of ^{99}Tc in the late 1980s and early 1990s, this is most likely an effect of the relatively low input from Sellafield in this period. There was a steady decrease in the ideal tracer concentration from Sellafield at Hillesøy from 1988, followed by a rapid increase from mid-1995 and onwards. The flux of idealized tracers out of the Irish Sea is certainly an important factor to explain these changes. The simulated tracer flux through the North Channel reached a record low value in 1992, accompanied by a record high tracer flux through the St. Georges Channel in the south. An anomalously high tracer flux through the North Channel was seen in the following three years: 1993, 1994, and 1995 (not shown). Also, the idealized tracers from La Hague exhibits substantial variations with a dominant seasonal cycle. The ^{99}Tc tracers from La Hague are largely constrained to these variations, although the amplitude differs as a result of the varying release rate.

In Fig. 10 the extent of lateral spreading is illustrated by the simulated distribution of ^{99}Tc from both Sellafield and La Hague in two horizontal sections along the Norwegian coast, the Svinøy and Gimsøy sections (see Fig. 1). Contaminants carried by the NCC are mixed on the way northwards with outerlaying Atlantic Waters, therefore the lateral spread is more significant at the Gimsøy Section (Fig. 10b) than the Svinøy Section further south (Fig. 10a). This is particularly clear from the lateral extent given by the concentration interval 0.1–0.5 Bq m^{-3}. The maximum concentration is also less at Gimsøy due to continuous vertical mixing. As for the one-point concentrations at Hillesøy discussed above, the ideal tracer signal and its variability are also partly resembled by the ^{99}Tc tracer signal seen in these Hovmo¨ller plots (not shown).

It is apparent from these figures that an interplay between the strong seasonal dependence of the ocean currents and mixing in the area (Nilsen and Falck, 2006) forces the tracer signal to show a strong seasonal signal, without having such variability in the release rate (remember that the idealized tracers do not even have a release rate, but a constant value prescribed at the source point). The ideal tracers exhibit much of the same seasonal behavior as the ^{99}Tc tracers, showing that the ocean dynamics significantly modulates the initial tracer signal. For this reason, observations of radioactivity along the Norwegian coast done only once or a few times a year will in general lead to an erroneous representation of the actual concentration throughout the year. Based on geostatistical techniques applied on the

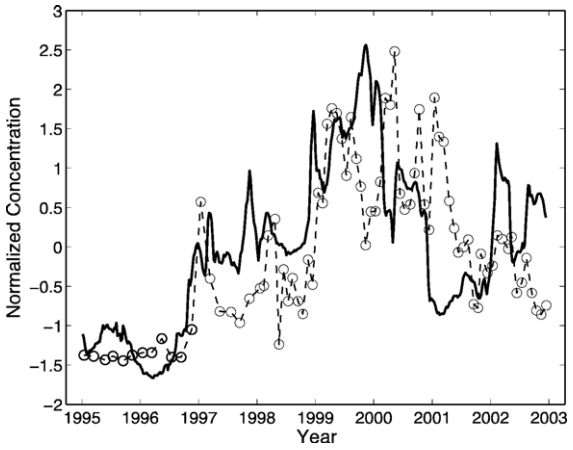

Fig. 7. Normalized ^{99}Tc concentration (Bq kg^{-1}) in seaweed, *Fucus vesiculosus*, from observations (open circles) and normalized simulated concentration in the surface (Bq m^{-3}) of ^{99}Tc in seawater (solid line) at Utsira, in the period 1995 to 2003. Mean values and standard deviation for seaweed is 241 and 153 Bq kg^{-1}, respectively, and 2.21 and 1.04 Bq m^{-3} for simulated seawater concentration.

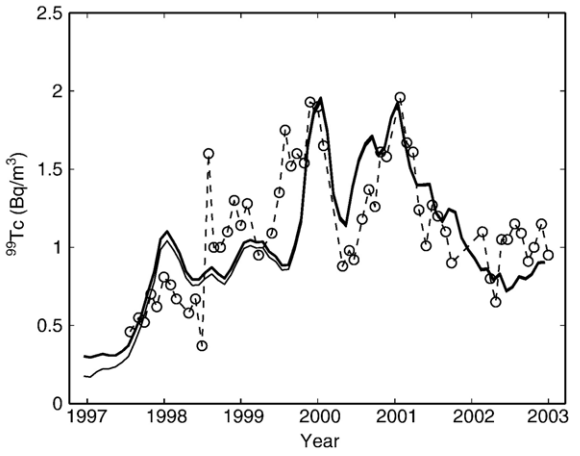

Fig. 8. ^{99}Tc concentration (Bq m^{-3}) in seawater from observation (open circles) and the surface of the OGCM (bold solid line) at Hillesøy (69.65° N, 17.95° E) for the period 1997 to 2003. The simulated contribution from Sellafield only is indicated with the thin solid line.

observed monthly time series of ^{99}Tc at Hillesøy, Dowdall et al. (2005) found that reducing a sampling frequency to less than approximately 50 days will deteriorate the information provided by the time series.

4.4. Simulated age field

Three snapshots of the spatial distribution of the age of idealized tracers originating from Sellafield are shown in Fig. 11. By construction, differences in the age are depending on both variations in the ocean dynamics and the release rate. However, since these tracers represent surface water masses from the source, differences are merely a result of variable ocean dynamics. It follows from the figure that for this period the age along the Norwegian coast is in the range 4–7 years, while accumulation of up to 8 years old water masses are seen in the English Channel and in the southern North Sea due to the slow southward transport in the Irish Sea. It should be noted that the age field seen here does not necessarily reflect the particular circulation pattern, in contrast to the concentration field (Delhez and Deleersnijder, 2002). For instance, the relatively old water masses in the English Channel and the southern North Sea do not have a significant impact on the age along the Norwegian coast since the relative concentration is low compared to the waters in the northern North Sea (roughly an order of magnitude) which is contaminated by tracers following the route north of Scotland.

The simulated time history of the age of idealized tracers at Utsira and Hillesøy from Sellafield and La Hague are shown in Fig. 12. These results suggest the age at Utsira to be in the range 4–7 years from Sellafield and 3–4.5 years from La Hague. At Hillesøy further north, the age are 5–8 years from Sellafield and 4–5.5 years from La Hague. From 1995 and onwards, a tendency is seen for younger idealized tracers from Sellafield at Hillesøy, which is also evident from Fig. 11.

Since the age at these remote sites provides an integrated picture of the total effect ocean currents and mixing have on transporting tracers from the source, it is unlikely that a single index (such as the local wind forcing or the volume transport in a specified section) can explain the variations of the age. However, the sharp increase of the idealized tracer concentration seen in the same period as the age decreases at Hillesøy (see Fig. 9; discussed in the previous section) is believed to be a key

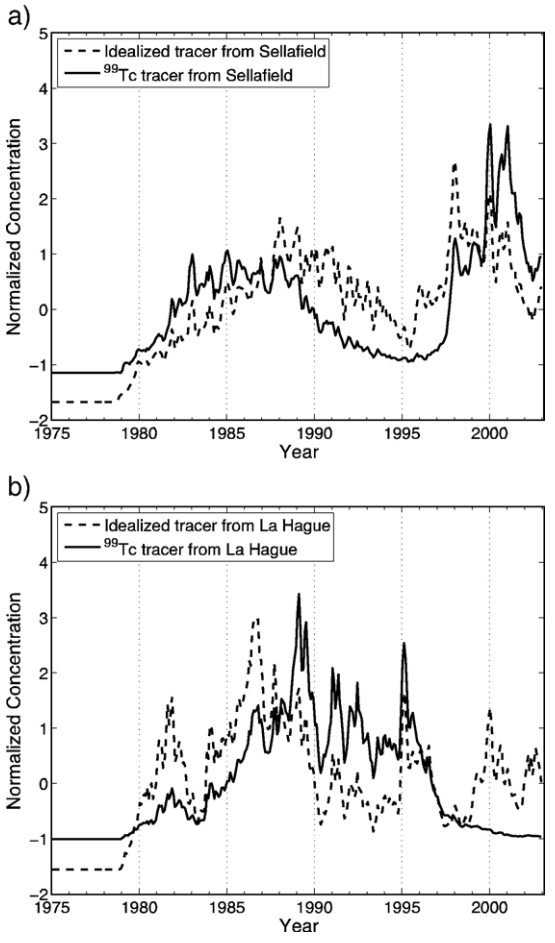

Fig. 9. Simulated normalized concentration in the surface at Hillesøy of ^{99}Tc (solid line) and idealized tracers (dashed line) from (a) Sellafield and (b) La Hague. Mean value and standard deviation of ^{99}Tc from Sellafield is 0.49 and 0.43 Bq m^{-3}, respectively, and 0.16 and 0.16 Bq m^{-3} from La Hague. Normalized concentration of tracer C at station $\mathbf{x}_s$ is calculated as $(C(\mathbf{x}_s,t)-\text{mean}(C(\mathbf{x}_s,t)))/\text{std}(C(\mathbf{x}_s,t))$.

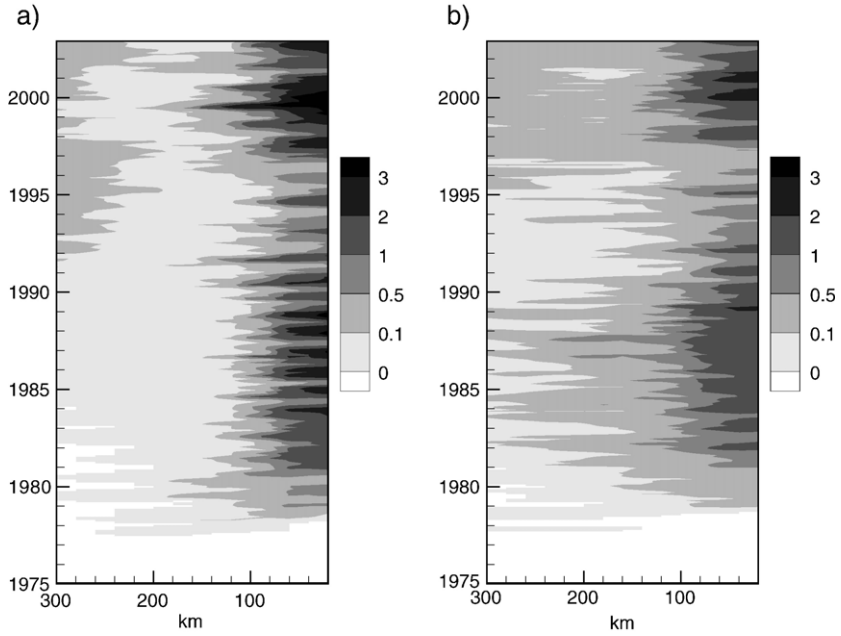

Fig. 10. Hovmöller plot of simulated ^{99}Tc concentration (Bq m^{-3}) in the surface from both Sellafield and La Hague at (a) the Svinøy section and (b) the Gimsøy section.

factor. We therefore suggest that increasing tracer concentration leads to decreasing age at a given location (this has also been demonstrated by Delhez and Deleersnijder, 2002).

It follows from Fig. 12 that the age is zero before the signal arrives at a specific location downstream the source, then it rapidly jumps up to an age value representative for the leading edge of the tracers, followed by a slow growth period. The transient response time for the age along the Norwegian coast, defined loosely here as the time taken when the age reaches a quasi steady-state behavior, is very long: Around 5 years for La Hague and at least 10 years for Sellafield.

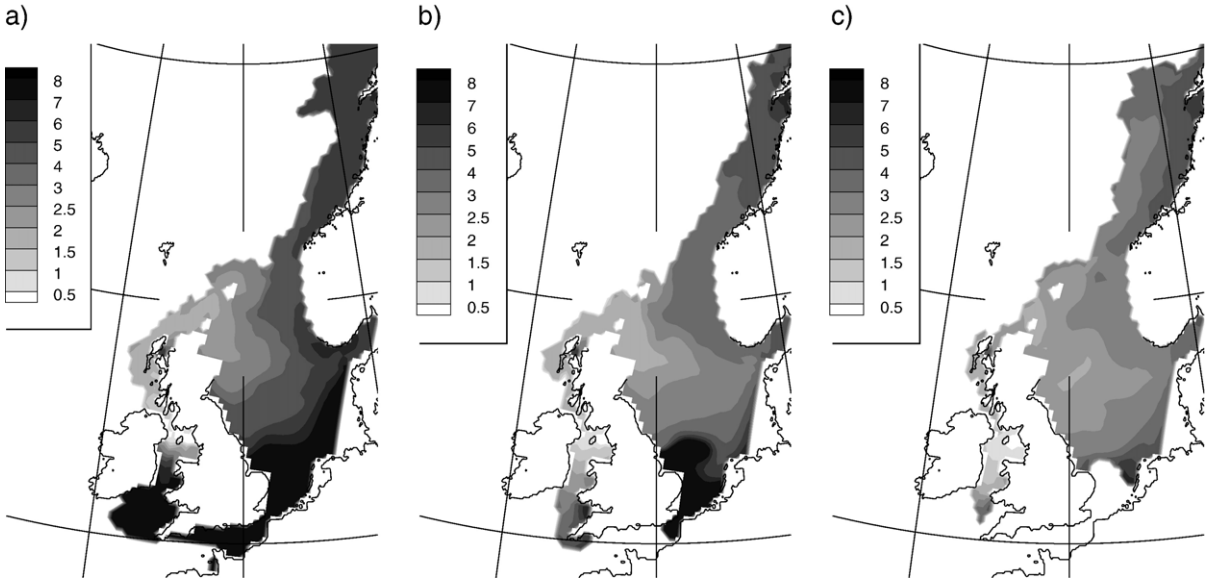

Fig. 11. Simulated age field in the surface of idealized tracers from Sellafield, (a) March 1998, (b) March 2000, and (c) March 2002.

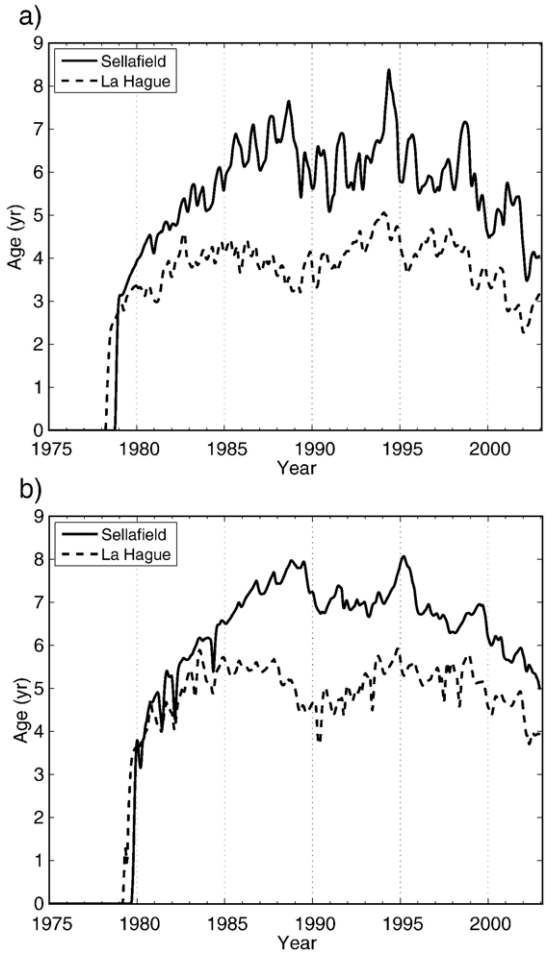

Fig. 12. Simulated age in the surface of idealized tracers from Sellafield and La Hague at (a) Utsira and (b) Hillesøy during the time period considered.

It has been suggested by Delhez and Deleersnijder (2002) that this response time should be twice the mean age, depending on the "width" of the age distribution. If mixing is negligible, the shape of the initial tracer signal is maintained when transported away with the ocean currents, and the width of the age distribution is negligible. On the other hand, if mixing is significant, one would expect a large fraction of the tracers to arrive both faster and slower than the mean age at an observational point. Therefore, the spreading of a tracer signal in different age classes is an integrated measure of mixing occurring from the source region to a specific location.

An idealized experiment with the same model setup has been performed to further assess transport and timescale properties associated with passive tracers from Sellafield (Orre et al., submitted for publication). In the latter study it was found that a pulse released in January 1989 passed Hillesøy for a period of more than 10 years, and that the standard deviation of the concentration from the pulse, which is representative for the width of the age distribution, was approximately 6 years. Although these results could differ if the pulse was released in another period of the year or another year, we believe that the width of the age of tracers from Sellfield at Hillesøy is comparable to the age itself. Consequently, passive tracers released into the Irish Sea experience a considerable amount of mixing before reaching northern Norway and the entrance to the Barents Sea. Since the transient response time for the age of tracers from La Hague is much shorter than that from Sellafield, we speculate that the width of the age from La Hague at Hillesøy is less than the age itself.

5. Discussion and conclusion

The dispersion properties associated with the soluble radionuclide ^{99}Tc released from Sellafield in the Irish Sea and La Hague in the English Channel have been reassessed. We emphasize the complexity of the combined transport pathways these tracers experience as they are transported from their source regions towards the Arctic Ocean. Simulated data fields from the regional Nansen Center version of MICOM are used, along with three observed time series from the Norwegian coast. To gain more accurate insight into the timescales involved, idealized age tracers are implemented. Variability of the age is an integrated effect of all transport and mixing processes that has occurred since the tracer was released at the source. It is therefore difficult to use just one index, being the ocean volume transport from a section or the intensity of the atmospheric circulation, to explain changes of the simulated age.

The applied model system reproduces fairly accurately both the spatial and temporal distribution of ^{99}Tc at the coastal stations Utsira and Hillesøy located on the western and northern coast of Norway (see Fig. 1). Interestingly, the amplitude and interannual trends for both of these time series in seawater are realistically captured by the model, whereas the seasonal variability is only reproduced for the Hillesøy time series, see Section 4. One reason for the erroneous seasonal cycle at Utsira could be that only annual mean release rates from Sellafield and La Hague are available until 1990, and from Fig. 2 we see that the monthly releases from Sellafield in the 1990s vary quite substantially. Another reason might be the uncertainties in the release rate from Sellafield in the 1970s, which would influence the concentration along the Norwegian coast also during the

1980s. It should also be mentioned that observations done at coastal stations like Utsira and Hillesøy are not necessarily representative for the concentration in the core of the flow, the NCC in this case. When analyzing the model results, one cannot a *priori* rule out the possibility that unresolved physical processes from the applied 20 km OGCM, as well as model deficiencies and uncertainties in the applied atmospheric forcing, may lead to unrealistic dispersion of the radionuclides. However, the good comparison at Hillesøy (Fig. 8) are from the period when we have accurate monthly release rates. This, along with realistically simulated ocean transport and temperature variations of the poleward flowing Atlantic water (Nilsen et al., 2003; Drange et al., 2005) give us some confidence to state that the applied model system are able to reproduce several of the key features of the dispersion properties of radioactive tracers in the region on the timescales considered here (months to decades). A very attractive opportunity when simulating tracers with state-of-the-art OGCMs, is to use the model system as a numerical laboratory to assess and to quantify the relative contribution from different sources of contamination, the Sellafield and La Hague locations in this case (Fig. 5).

Detailed understanding of the underlying ocean circulation and mixing processes on seasonal to interannual timescales is still quite difficult. The presented results provide insight into the variability in the ocean state and how it influences the pathways and age of the tracers considered. Obviously, further work needs to be done to identify and to quantify physical mechanisms behind the variability in the ocean circulation and mixing, and the subsequent implications for the evolution of the tracers. However, we make the following remark regarding the transport of tracers from Sellafield and La Hague to the Nordic Seas: While the NAO – the leading mode of atmospheric variability over the North Atlantic Ocean – is the dominating driving force for ocean transport along the Scottish and Norwegian coasts (Fig. 3c,d), the transports in the North Channel and the English Channel are mainly influenced by along-channel winds (Fig. 3a,b). Thus the suggested hypothesis that enhanced NAO results in faster transport of radioactive contaminants from Sellafield to the Norwegian coast (Kershaw et al., 2004), might be too simplistic. Focussing on the oceanic transports directly, Gao et al. (2005) found that the strength of the Atlantic Inflow to the Nordic Seas is also a far too simple index for characterizing the temporal evolution of tracers from Sellafield to the Nordic Seas. The North Channel being the direct transfer route for tracers from the Sellafield location to the downstream current systems, its out-of-phase relationship with the NAO (Fig. 3a,c) is likely what prevents a simple relation between the NAO and the age of tracers transported into the Nordic Seas.

From hydrological sections across the Lofoten Basin in May 2000, Gascard et al. (2004) estimate that the mass flux of ^{129}I only accounts for one third of the annual discharge from La Hague and Sellafield. From the idealized tracers presented in Figs. 9 and 10, it is apparent that seasonal variations in tracer concentration are significant, and just one snapshot of the mass flux in the highly variable NCC/NwAC system is in general not representative for the annual mean. In addition, our model results do not support any important transfer pathways of radioactive contaminants from Sellafield and La Hague outside the NCC/NwAC system, suggested as giving one possible explanation for the relatively low mass flux of ^{129}I seen at this time by Gascard et al. (2004).

It is interesting to see that the age presented here is considerably older than what is called the *transit time* previously seen in the literature for various radioactive tracers from the same sources (e.g. Dahlgaard, 1995; Karcher et al., 2004; Kershaw et al., 2004). For instance Kershaw et al. (2004) report that the transit time for EARP-related tracers at Hillesøy is about 3.5 years, while in this study the age of water masses from Sellafield is in the range 5–6 years for the same time period. These discrepancies could very well be due to the definition of different tracer-based time scale concepts. As an example, consider the special case of a radioactive tracer signal dumped into the Irish Sea to reach Hillesøy. Are we then interested in

(1) the time it takes for the first tracer signal to arrive at Hillesøy,
(2) the time lag between a peak release and a subsequent peak concentration observed at Hillesøy, or
(3) the average time the tracer signal spends on its way from the source to Hillesøy?

While it is obvious that the first definition provides a shorter estimate of the age than the two others due to mixing, it is not obvious that the latter two definitions provide different time scales.

In a modeling study of radionuclides discharged from La Hague in the English Channel and the southern North Sea, Salomon et al. (1995) define the transit time as "the time lag maximizing the correlation between the concentration near the source and the concentration at a given location downstream of the source". If this definition is meant to provide an estimate of the age, one has to assume that the *shape* of the release rate is

somehow conserved when transported away with the ocean currents. Now, neither the Hovmöller-plots presented in Fig. 10 nor the concentration at Hillesøy shown in Fig. 9 reveal the shape of the release rate as displayed in Fig. 2. In fact, a great deal of the variations at Hillesøy can only be attributed to the ocean dynamics, since the idealized tracers resembling pure water masses contain much of the same variability as the realistic ^{99}Tc tracer. Assuming these modelled concentrations are realistically simulated, Salomons method would lead to a spurious estimate of the age, at least on the time scales considered here. Following Brown et al. (2002), the transit time is defined as "the time taken for the maximal effect from a discharge to be observed at a remote site". While this is without doubt a useful measure for environmental assessments, it is only representative for the age if diffusion is negligible, i.e. for pure bulk advection (e.g. Holzer and Hall, 2000; Delhez and Deleersnijder, 2002). As a consequence, a tracer signal in a fluid flow where diffusive processes cannot be neglected does not propagate with the mean velocity of the fluid. Numerous studies of geophysical transport problems, based on both observations and models (see Waugh et al., 2003, for a general overview), have shown that the width of the age can be of the same size as the age itself. Consequently, the transit time defined as above could potentially be very different from the age. To summarize, if one is interested in estimating the average time passive tracers spend on their way from the source to an observational point, the method suggested by Salomon et al. (1995) and Brown et al. (2002) are biased towards younger values in the presence of diffusive processes. On the other hand, the method for age-tracers employed in this study is not a useful diagnostic tool if one is interested in the timing of a peak concentration value downstream of the tracer source.

The concern about ^{99}Tc and other long-lived radionuclides from European reprocessing plants is not as high today as it was 10 or 20 years ago. In general, the current level of radioactive contamination in the North Sea and the Nordic Seas is considered to be low (AMAP, 2004). However, the presence of radioactive contaminants in the marine environment still makes it highly relevant to simulate accidental and continuous releases of anthropogenic radionuclides for the present day climate and possible states of the climate in the 21st century. Despite the controversy of releasing liquid radioactive waste in the marine environment, discharges of soluble radionuclides from reprocessing plants (in this case ^{99}Tc from Sellafield and La Hague) do offer a unique opportunity to trace water masses and mixing processes in the ocean, processes that are of crucial importance for assessing the reliability of OGCMs in, for instance, climate research.

Acknowledgments

The authors would like to thank Gordon Christensen from IFE, Norway, for the valuable comments on the paper and for providing the time series of observed concentration of ^{99}Tc in seawater and seaweed at Utsira. Thanks to Morten Sickel from the NRPA, Norway, for providing the time series of observed concentration of ^{99}Tc in seawater at Hillesøy. We would also like to thank Peter J. Kershaw at CEFAS, UK, and Pascal Bailly du Bois at IRSN, France, for providing the ^{99}Tc release rates from Sellafield and La Hague, respectively. Dag J. Steinskog at NERSC is thanked for the statistical help and advise. This study is supported by the project Arctic Radioactive Contamination (ARC) and the Programme of Supercomputing, both under the Research Council of Norway. Support from the G. C. Rieber Foundations is acknowledged. This is publication Nr A 154 from the Bjerknes Centre for Climate Research.

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