Circulation features in the northern North Atlantic Ocean inferred from simulated radioactive tracers

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Contents

Preface

An overview part and a collection of papers constitute my thesis presented in partial fulfillment of the requirements for the degree of PhD in physical oceanography at the Geophysical Institute, University of Bergen, Norway.

In the first part, an introduction is given to the main questions addressed in this thesis. The papers are summarized and a synthesis of the conclusions is presented.

The second part consists of three papers published in, or submitted to, international peer review journals:

- **Paper I:** Orre, S., Y. Gao, H. Drange, and J.E.Ø. Nilsen, 2007: A reassessment of the dispersion properties of ^{99}Tc in the North Sea and the Norwegian Sea, *Journal of Marine Systems* **68**, 24-38.

- **Paper II:** Orre, S., Y. Gao, H. Drange, and E. Deleersnijder, 2007: Diagnosing ocean tracer transport by equivalent diffusion and age, submitted to *Advances in Atmospheric Sciences*.

- **Paper III:** Orre, S., J.N. Smith, V. Alfimov, M. Bentsen, and H. Drange, 2008: Simulating transport of ^{129}I and Idealized Tracers in the northern North Atlantic Ocean, to be submitted to *Journal of Geophysical Research*.

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Chapter 1

Overview

1.1 Introduction

1.1.1 On the use of chemical tracers in oceanography

Chemical man-made tracers introduced to the marine environment can be used to infer circulation features and ventilation time-scales in the ocean. Pioneering work in oceanography, for instance on diapycnal mixing and upwelling of the world oceans (Munk 1966) and Wallace Broecker's paradigm "the great ocean conveyor belt" (Broecker 1991), are to a large extent deduced from distributions of tracers. Table 1.1 provide an overview of some of these tracers, their properties, sources and applications. As can be readily seen, the general properties and applications of tracers in the oceans are very different. A short description of some tracers and their applications are given below, as well as some background and motivation for studying radioactive tracers from Sellafield and La Hague, the latter is discussed in Section 1.1.2.

One of the most widely used tracers is radiocarbon (^{14}C). Radiocarbon is a radionuclide of carbon created both naturally and from human activities in the atmosphere, and is dissolved in the surface mixed layer of the ocean in the form of carbon dioxide. Since ^{14}C undergo radioactive decay, the concentration in sea water decrease when being transported to deeper regions in the ocean. When the water masses are transferred beneath the mixed layer, they are shielded from the influence of the atmosphere and then very slowly modify their properties subject to weak mixing in the ocean interior. The longer since a given water mass has been in contact with the surface mixed layer, the more depleted the ^{14}C concentration becomes.

Another set of tracers which is frequently used is chlorofluorocarbons (CFCs) and radioactive tritium (^{3}H). Chlorofluorocarbons are man-made pollutants stemming from a wide variety of industrial applications since the 1930s, and which are extremely inert in sea water and detectable at very low concentrations. Large amounts of tritium were released into the atmosphere by nuclear weapons testing, especially in the 1950s and 1960s. If CFCs or tritium are found in the ocean interior, some fraction of that water mass must have been in contact with the atmosphere since the time at which these chemicals were first introduced to the upper ocean by precipitation, through air-sea ex-

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Table 1.1: An overview of some chemical tracers used for ocean circulation studies. $T_{1/2}$ denotes the time it takes for radioactive tracers to lose half of its radioactivity.

Tracer	Chemical formula	Properties	Main sources	Applications	Time-scales
Radiocarbon	^{14}C	$T_{1/2} = 5730$ yr	Natural isotope, nuclear bomb testing	Global	Centennial
Tritium	^{3}H	$T_{1/2} = 12.5$ yr	Nuclear bomb testing	Global	Decadal
Chlorofluoro-carbons	CCl$_N$F$_M$	stable, inert	Various industry	Global	Decadal
Sulphur Hexafluoride	SF$_6$	stable, inert	Deliberate release	Regional	Monthly, annual
Cesium-137	^{137}Cs	$T_{1/2} = 31$ yr	Reprocessing plants, nuclear bomb testing, Chernobyl	Regional	Annual, decadal
Strontium-90	^{90}Sr	$T_{1/2} = 29$ yr	Reprocessing plants, nuclear bomb testing, Chernobyl	Regional	Annual, decadal
Technetium-99	^{99}Tc	$T_{1/2} = 2.1 \cdot 10^5$ yr	Reprocessing plants	Regional	Monthly, annual, decadal
Iodine-129	^{129}I	$T_{1/2} = 1.6 \cdot 10^7$ yr	Reprocessing plants	Regional	Monthly, annual, decadal

change of the gas, or by contaminated rivers.

Ventilation age is a commonly used quantity in oceanography and represents the time a water parcel has been isolated from the atmosphere. The ventilation age is consequently defined to be zero in the surface mixed layer and then increases with depth as individual water parcels are transported to deeper layers through subduction processes and open-ocean convection. Using the radioactive decay constant for ^{14}C and tritium, and surface and interior concentrations, a "radioactive tracer age" can be calculated. Likewise, using the ratio of CFC-11 and CFC-12 in the surface and in the ocean interior, a "ratio tracer age" can be calculated (Waugh et al. 2003).

Progress has been made in understanding the dynamics of the "ventilated thermocline" in subtropical regions studying observed distribution of tritium and it's decay product [3]He (Jenkins 1998). An important role of isopycnic mixing for ventilating the thermocline in the subtropical gyre of the North Atlantic has been deduced from the distribution of potential vorticity and tritium-[3]He age in the density range of the thermocline (Robbins et al. 2000). More generally, understanding passive tracer transport in the ocean (and in the atmosphere) is crucial for rotating stratified fluid dynamics, because potential vorticity evolves as a dynamically active tracer, and potential vorticity provides the dynamical foundation for many theoretical developments of atmosphere and ocean dynamics (e.g. Gill 1982).

Several attempts of estimating the uptake of anthropogenic CO_2 has been done based on global CFC data sets (e.g. McNeil et al. 2003; Waugh et al. 2006). It is then assumed that the inventory of CFCs into the deep ocean can be seen as an analogue of anthropogenic CO_2. This assumption has been questioned, for instance in the abyssal Weddell Sea, where direct measurements reveal insignificant storage of anthropogenic CO_2, along with a marked increase of CFCs (Hoppema et al. 2001).

Purposefully release of Sulphur Hexafluoride (SF_6) in the ocean is a novel approach to directly study diapycnal mixing, which is difficult to obtain from hydrographic profiles or observed vertical velocity. The strategy has been to release SF_6 onto a single, well defined density surface, and observe its subsequent spreading. It has been shown that

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diapycnal mixing across the pycnocline in the eastern subtropical gyre of the North Atlantic is week compared to expected values from global budgets (Ledwell et al. 1993). However, enhanced mixing in the deep ocean over rough topography has been observed in the Brazil basin (Ledwell et al. 2000). Similar experiments in the central Greenland Sea have revealed important information on mixing and convection processes there (Watson et al. 1999), as well as on the relative composition of the East Greenland Current (Olsson et al. 2005) and overflow waters from the Denmark Strait (Tanhua et al. 2005).

Chemical tracers can be used to assess the simulated circulation and ventilation patterns in Ocean General Circulation Models (OGCMs) (e.g. Heinze et al. 1998; England and Maier-Reimer 2001). Using observed surface values of CFCs as boundary conditions for passive tracers, direct comparison can be made with observed distributions in the ocean interior. CFCs are widely used by climate modelers to evaluate their OGCMs, particularly with respect to their representation of ventilation processes and the resulting deep water pathways in high latitude regions. Coordinated effort with many different OGCMs has been undertaken (Dutay et al. 2002). For instance, it has been shown that ocean models with a good representation of temperature and salinity can have significant errors in simulated distribution of passive tracers such as CFCs. Furthermore, OGCMs with tracers have revealed fatal errors in representing certain water mass formation processes, such as deep water formation in the North Atlantic and in the Southern Ocean. A recent study by Biastoch et al. (2007) compared an eddy-permitting model with a coarser resolution version of the same model with the aim to explore the effect of mesoscale processes on the uptake and spreading of anthropogenic CO_2 and CFCs. While the total inventory of tracers were similar in the two models, local differences were seen, for instance in representing the observed zonal gradients of anthropogenic CO_2 and CFCs in the deep North Atlantic Ocean.

1.1.2 Radioactive tracers from European reprocessing plants

The historically most important sources for radioactive contamination in the northern North Atlantic Oceans has been atmospheric fallout from nuclear weapons testing, discharges from European nuclear re-

processing plants, atmospheric fallout from the Chernobyl accident, dumped radioactive waste in the Kara and Barents Sea, discharge from contaminated Siberian rivers, the sunken nuclear submarine Komsomolets near Bjørnøya, and an airplane crash with nuclear weapons at Thule in northwest Greenland (Nies et al. 1998; AMAP 2004). Of these sources, atmospheric fallout from nuclear weapons testing and discharges from the European nuclear reprocessing plants are by far the major contributors (Nies et al. 1998; AMAP 2004).

Controlled discharges of radioactive waste into the sea have been conducted from Sellafield on the British coast in the Irish Sea since 1952 and from La Hague on the French coast in the English Channel since 1962. These discharges have contributed to elevated concentration of several radioactive species compared to natural background values (AMAP 2004). Discharges from the nuclear reprocessing plant in Dounreay on the northern coast of Scotland have now ceased. Maximum discharges of e.g. ^{137}Cs and ^{90}Sr from Sellafield occurred during the 1970s, with a pronounced reduction in discharges in the recent decades. While discharges of most radioactive species from Sellafield and La Hague have been reduced the last two decades, discharges of ^{99}Tc from Sellafield and ^{129}I from both Sellafield and La Hague greatly increased during the 1990s. Naturally occurring concentration of ^{99}Tc and ^{129}I in the sea is several orders of magnitude less than the concentration in water masses contaminated by discharges from Sellafield and La Hague. Other anthropogenic sources of these contaminants are negligible as well (Kershaw and Baxter 1995; Aldahan et al. 2007).

Radioactive tracers from Sellafield and La Hague differs from traditional tracers such as ^{14}C, CFCs and tritium, because they are released from point sources and not distributed evenly over the surface of the world oceans, and therefore represents European coastal waters (Dahlgaard 1995). Several oceanographic features in the northern North Atlantic Oceans have been investigated using radioactive tracers from these sources.

Along the south-west Norwegian coast (see Fig. 1.1), tracers from Sellafield and La Hague are transported northwards mainly by the Norwegian Coastal Current (NCC), and a lesser extent by the Norwegian Atlantic Current (NwAC) (Gascard et al. 2004). It has been

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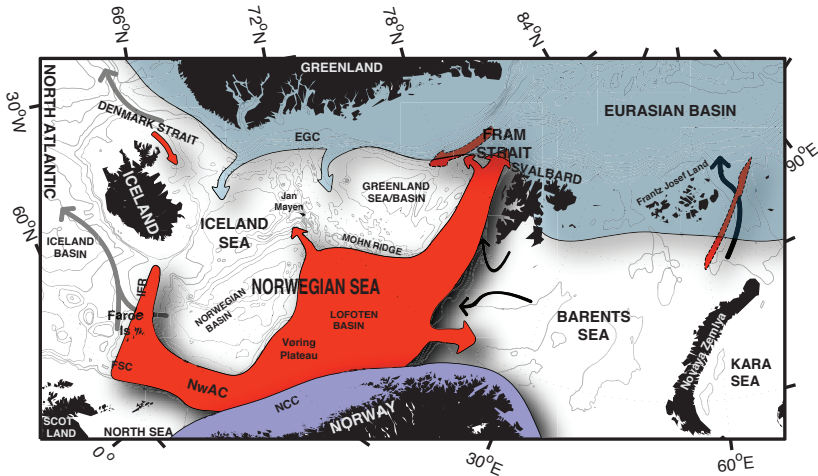
Figure 1.1: Schematic picture of ocean currents and water masses in the Nordic Seas, based on observed hydrography and literature. Courtesy of J. Even Ø. Nilsen.

suggested that the North Atlantic Oscillation (NAO), an atmospheric circulation mode mainly reflecting the strength of winter westerlies in the northern North Atlantic, is strongly influencing the propagation speed of tracers from Sellafield and La Hague in the eastern Nordic Seas (Kershaw et al. 2004). In the Arctic Ocean, the tracer signal is seen in both the Polar Mixed Layer (PML) and in the halocline layer beneath the ice, mainly stemming from Siberian shelf waters originating from the Barents Sea. The tracer signal is also present in the Atlantic Layer (AL), with contribution from both the Barents Sea and inflow of Alantic Waters through the Fram Strait (Smith et al. 1999). For instance, the temporal evolution of ^{129}I in the AL of the Eurasian basin of the Arctic Ocean have suggested that the Amundsen basin is more rapidly ventilated by Atlantic waters than the Nansen and Makarov basins (Alfimov et al. 2004b). Along the eastern coast of Greenland, tracers from Sellafield and La Hague are transport southwards with the East Greenland Current (EGC), and are found both in the return flow of Atlantic Waters, but also in deeper layers from the Arctic Ocean (Alfimov et al.

2004a). In the North Atlantic Subpolar Seas, the concentration of radionuclides from Sellafield and La Hague is highest in the deepest layers, providing a direct signature of overflow waters from the Nordic Seas (Smith et al. 2005). Therefore, radioactive tracers from Sellafield and La Hague can be used as a ventilation tracer for the North Atlantic Deep Waters (NADW). Also the timing of the tracer signal, especially of ^{99}Tc and ^{129}I, is favorable since the releases increased by several hundred percent during the 1990s, and the bulk of these tracers are just now being observed in the overflow regions of the Nordic Seas.

There is another application which is particularly relevant to the more general problem of pollutant spreading in the ocean. The concentration of ^{99}Tc and ^{129}I in the northern North Atlantic Ocean is totally dominated by discharges from Sellafield and La Hague, and the quantity released from these reprocessing plants are well know, especially for the last three decades. A significant fraction of European river pollutants enter the North Sea, so ^{99}Tc and ^{129}I can therefore be used as proxies for other soluble pollutants discharged into the North Sea.

1.2 Objectives

The objectives of this study are:

- To simulate the spatial and temporal evolution of the radioactive tracers ^{99}Tc and ^{129}I in the northern North Atlantic Ocean

- To compare the simulated tracer distribution with observed data

- To investigate the relationship between propagation of tracers to changes in volume transport through specified sections and to changes in the atmospheric wind field

- To simulate idealized tracers and age of tracers to obtain more general knowledge for tracer transport properties in the northern North Atlantic Ocean independent of a particular release rate

- To investigate various measures of mixing which tracers experience in this region, how mixing modify tracer signals, and implications for interpreting tracer transport in general

1.3 Summary of the Papers

A summary of the papers constituting this study is presented in the following section.

1.3.1 Paper I: A reassessment of the dispersion properties of ^{99}Tc in the North Sea and the Norwegian Sea

The dispersion properties of the anthropogenic radionuclide ^{99}Tc in the North Sea and the Norwegian Sea have been studied. The main sources of this radionuclide are Sellafield in the Irish Sea and La Hague in the English Channel. A regional version of the Miami Isopycnic Coordinate Ocean Model (MICOM) was used for simulating the spatial and temporal evolution of ^{99}Tc, as well as an idealized tracer resembling pure water masses from Sellafield and La Hague, for the period 1975 to 2003. Finally, an artificial age-tracer was tagged to the idealized tracers to estimate the mean time elapsed since the tracers left their source regions.

Tracers from both sources are transported into the North Sea, entrained in the Norwegian Coastal Current (NCC) in Skagerak, and transported northwards along the Norwegian coast. Dilution of the radioactice signal occur along the Norwegian coast as the NCC boarders and exchange properties with the Norwegian Atlantic Current (NwAC). Comparison with observational time series from two stations along the Norwegian coast shows that the model fairly accurately captures both the amplitude and the temporal trend of ^{99}Tc from Sellafield and La Hague. The age of the Sellafield-derived tracer at the island Hillesøy on the northern coast of Norway is found to be in the range 5–8 years, while the La Hague-derived tracers are in the range 4–5.5 years at the same place.

From regression analysis of simulated volume transport on sea level pressure fields (NCEP/NCAR-reanalysis data) it is found that the NAO–the leading mode of atmospheric variability over the Atlantic Ocean–is the dominating drive force for ocean volume transport along the Scottish and Norwegian coasts, while the volume transport in the North Channel and the English Channel are mainly influenced by along-channel winds. Thus there is no simple relationship (i.e. with

the NAO) between atmospheric forcing and age of tracers from Sellafield or La Hague entering the Norwegian Sea and beyond.

1.3.2 Paper II: Diagnosing ocean tracer transport by equivalent diffusion and age

Two different expressions of the equivalent diffusion were derived based on a simplified two dimensional advection-diffusion equation for a passive tracer in an infinite domain. To illustrate the use of these expressions, a numerical experiment with idealized passive tracers from the locations of the nuclear fuel reprocessing plants at Sellafield in the Irish Sea and Dounreay on the northern coast of Scotland has been carried out.

Tracers from Dounreay are rapidly carried away from the source region, while the Sellafield-derived tracers are efficiently delayed due to the source being located in the semi-enclosed Irish Sea. The age of the idealized tracers from Dounreay are approximately 2 years younger than the age from Sellafield along the Norwegian coast.

A simulated pulse released from Dounreay in 1989 shows a distinct peak value along the Norwegian coast a few years after the release, and then almost vanish from the entire Nordic Seas by the end of the simulation in 2002. The resulting maximum concentration along the Norwegian coast from the Sellafield-derived pulse is less than half of the maximum concentration stemming from the Dounreay-derived pulse. Hence, differences in tracer transport from Sellafield versus Dounreay cannot be characterized by a simple time lag in the resulting tracer concentration downstream of the source, due to the very different transient evolution of the concentration.

We argue that an order of magnitude higher value of the equivalent diffusion from the Dounreay site explains these qualitative differences. Calculating the single parameter equivalent diffusion has a potential for extracting crucial information from the vast output an OGCM delivers.

1.3.3 Paper III: Simulating transport of ^{129}I and Idealized Tracers in the northern North Atlantic Ocean

Large scale dispersion properties of the anthropogenic radionuclide ^{129}I in the northern North Atlantic Ocean have been studied using a global version of MICOM and observed data. The main source of ^{129}I in the ocean are releases from Sellafield and La Hague, with a pronounced increase during the 1990s.

The OGCM reproduce observed values of ^{129}I in the eastern Nordic Seas quite reasonable. However, the OGCM fails to realistically simulate the inflow of Atlantic Waters into the Arctic through the Fram Strait, which in turns prevents a realistic distribution of tracers there. Vertical profiles of ^{129}I in the Labrador Sea have been sampled since 1997, and the tracer field from the OGCM is close to the observed values, although slightly overestimates the values in surface and in intermediate waters. Interestingly, concentration of ^{129}I is highest in the deepest layers in the Labrador Sea, which is a clear signature of overflow waters from the Nordic Seas. The age of ^{129}I is also youngest in these deep layers, confirming the interpretation of overflow waters as they are exported southwards, while water masses above circulate cyclonically with the Subpolar Gyre. In previous versions of MICOM, the two eastern overflow branches (the Iceland-Faroe Ridge and the Farao-Shetland Channel) have shown a tendency to be trapped on the eastern side of the North Atlantic Ridge. In the present model, these two overflow branches, as well as overflow from the Denmark Strait, are constrained to the western boundary in the North Atlantic, consistent with observations. We argue that utilizing σ_2-coordinates instead of σ_0-coordinates for the vertical discretization is the main reason for this improvement.

We have attempted to estimate the Transit Time Distribution from Sellafield and La Hague from an ensemble average of pulse tracers (Impulse Boundary Propagator, IBP). The first two moments of the IBPs have the physical meaning of the transit time and the width of the transit time. The ratio of these two is a measure of the degree of mixing, and this parameter shows that mixing should be accounted for when describing movements of these tracers.

1.4   Main Conclusions

The main conclusions of this study can be summarized as follows:

- Radioactive tracers from Sellafield and La Hague are transported northwards along the European continental shelf, passing the Norwegian coast and entering the Arctic Ocean through the Barents Sea and the Fram Strait. There is also a large fraction recirculating in the Fram Strait and flowing southwards with the East Greenland Current. In the North Atlantic Subpolar Seas, highest concentration is found in the deepest layers, showing the influence of overflow waters from the Nordic Seas in ventilating the deep North Atlantic Ocean.

- The applied model system is able to reproduce the evolution of radioactive tracers from Sellafield and La Hague in the eastern Nordic Seas region in an encouragingly realistic manner. Due to too strong vertical mixing south-west of Spitsbergen and an unrealistic northward Atlantic Inflow through the Fram Strait in the OGCM, the comparison with observed tracers is less favorable in the Arctic Ocean. From vertical profiles in the Labrador Seas, the OGCM reproduce the general structure of tracers in the water column, with concentrations in the deepest layers exceeding those in the surface.

- The age of tracers from Sellafield is typically 1–3 years in the North Sea, 4–8 years at the entrance to the Barents Sea, 8–15 years in the Eurasion basin of the Arctic Ocean, and 10–25 years in overflow waters in the North Atlantic Subpolar Seas. Tracers from La Hague are typically 1–2 years younger than tracers from Sellafield due to a shorter and more efficient transport route through the English Channel into the North Sea.

- Due to mixing, tracers in the ocean carries a distribution of possible ages, or transit times. From an ensemble average of pulse releases, we estimate a statistical value of the Transit Time Distribution from Sellafield and La Hague. It is demonstrated that the

distribution is continuously broadening away from the source region, thus the relative role of mixing is certainly not negligible for characterizing the tracer transport.

- Radioactive tracers from Sellafield and La Hague offers a unique possibility for studying transport and mixing processes in the Nordic Seas, the Arctic Ocean, and the North Atlantic Subpolar Seas, and to test the performance of OGCMs used for this region.

1.5 Discussion and future work

While a careful examination of radioactive contamination in the ocean is important in its own right for environmental impact assessments, one may also use these tracers to make inferences on the ocean circulation itself.

When simulating the temporal and spatial evolution of chemical tracers in OGCMs, it is recommended to add idealized tracers with constant or pulse time dependencies and age tracers. If an OGCM is able to reconstruct the observed distribution of, for instance, ^{129}I or CFCs, it is reasonable to believe that the simulated idealized tracers and ages are representative of the real oceans behavior as well, and more general results can be obtained.

The future generation of Earth System Models will include the marine carbon cycle simulating a large number of prognostic tracers, it is therefore of uttermost importance to develop efficient and accurate tracer routines in OGCMs. The incremental remapping scheme used in Paper III is a very promising approach, since the computational cost of adding additional tracers is modest, and since incremental remapping ensures monotonicity of the tracers (Dukowicz and Baumgardner 2000).

It has been suggested that increasing fresh water fluxes into the high northern latitudes will take place as a result of global warming, and that this could alter the northernmost extension of the Atlantic Meridional Overturning Circulation (e.g. Curry and Maurtizen 2005). Future melting of the Greenland ice sheet and/or increased fresh water discharge through the Fram Strait from the Arctic Ocean will increase

the southward freshwater transport by the EGC. However, while the EGC carries large amount of fresh water, only a very small fraction is being exported to the interior Nordic Seas (Jonsson 2005). The freshwater transport by the NCC is roughly one order of magnitude less than the freshwater transported by the EGC, but shares the front with the NwAC, and efficient mixing between the two current systems is observed. Lateral sections of ^{129}I along the Norwegian coast have shed light on the transport between the NwAC and the NCC (Gascard et al. 2004), while sections along the eastern Greenland coast illustrates how the EGC is being transformed southwards (Alfimov et al. 2004a). These data-sets could be used to quantify the total amount of freshwater entering the interior Nordic Seas.

Dense water formed in the Nordic Seas and descending across the Greenland-Scotland Ridge is a major source for deep waters in the North Atlantic, and contributes (including entrainment of ambient water downslope of the sill) for roughly two thirds of the Atlantic Meridional Overturning Circulation (Hansen et al. 2004). However, the source regions of overflow waters is still a matter of debate. For instance, Swift et al. (1980) argued that Denmark Strait Overflow Waters (DSOW) origins from the upper few hundred meters in the Greenland and Iceland Seas where winter convection occurs, while Mauritzen (1996) suggest that DSOW is derived mainly from modified Atlantic Waters which has been cooled and freshened during its cyclonic loop through the Nordic Seas and the Arctic Ocean. There has already been attempts to distinguish the different sources of overflow waters from the Nordic Seas based on CFCs and SF_6 (e.g. Tanhua et al. 2005). It is suggested that these tracers along with radioactive tracers from Sellafield and La Hague may further constrain the origin and relative composition of overflow waters from the Nordic Seas.

Wunsch (2002) and Waugh et al. (2003) have shown that age estimates from specific oceanic tracers ("ratio" and "radioactive" tracer ages) are not fundamental properties of the ocean, since tracers having different time dependencies or different radioactive decay would have different ages, and neither of them would be representative for the "true" age. More work is therefore needed to understand the implications and limitations of time-scale diagnostic tools in oceanogra-

phy. There is another type of time-scale which could be useful for the study of radioactive tracers from Sellafield and La Hague, which is not addressed in this thesis: the "residence time" which measures the time spend by a water parcel or a pollutant in a given water body (Bolin and Rodhe 1973; Delhez 2006).

The available studies on the applications of oceanic tracers, including the analysis presented in this thesis, demonstrates the usefulness of chemical tracer for testing and evaluating OGCMs, and for making inferences on the circulation and mixing processes in the ocean.

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