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^{99}Tc in seawater in the West Spitsbergen Current and adjacent areas

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Abstract

^{99}Tc levels were measured in seawater samples collected between 2000 and 2002 in the West Spitsbergen Current (WSC) and along the western coast of Svalbard or Spitzbergen and compared with available oceanographic 3-D modelling results for the late 1990s. Additional data from related regions are also presented in order to support the data interpretation. The seawater in the Arctic fjord Kongsfjorden on the western coast of Svalbard is influenced by the WSC, as shown by the ^{99}Tc levels in surface water. By means of the WSC, ^{99}Tc reaches the Eastern Fram Strait, where one branch of the WSC turns west into the East Greenland Current (EGC), and another branch continues northwards into the Arctic Ocean. Surface seawater collected in the central part of the WSC during a cruise on board the R/V "Polarstern" in the summer of 2000, showed higher levels of ^{99}Tc than samples measured in Kongsfjorden in the spring of 2000. However, all levels measured in surface water are of the same order of magnitude. Data from sampling of deeper water in the WSC and EGC provide information pertaining to the lateral distribution of ^{99}Tc. In all vertical profiling surveys (conducted in spring and summer), the highest levels of ^{99}Tc were found in surface water. Comparison with oceanographic 3-D modelling indicates both significant seasonal variations in the lateral stratification of the WSC and variations with depth over shorter vertical distances. This information can be applied in sampling strategies, environmental monitoring, long-range transport of pollutants and physical oceanography.

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1. Introduction

Ocean currents are able to transport artificial and natural radionuclides over long distances. This is important for environmental pollution studies and monitoring (e.g. AMAP, 1998) and oceanographic information resulting from the investigation of ocean currents is useful for improving the understanding of ocean dynamics. The anthropogenic, highly soluble and soft β-emitter, ^{99}Tc (half-life: 213 000 y) is discharged into the seas of central Europe by nuclear reprocessing plants (Sellafield, UK, and La Hague, France). Known pathways to the Arctic are the Norwegian Coastal Current (NCC), and further north the West Spitsbergen Current (WSC) (e.g. Haugan, 1999). Levels in the eastern North Atlantic after enhanced discharges in the mid 1990s have been documented in a number of publications (e.g. Brown et al., 1999, 2002; Gerland et al., 2002a,b). Contributions to the global background levels of ^{99}Tc in the environment are earlier releases from nuclear installations and from fallout in connection with atmospheric nuclear weapon tests (Smith et al., 1997). After Dahlgaard (1994), the ^{99}Tc levels observed over the period 1988–1990 in seawater in the EGC can be divided into one part of about 85% originating from European coastal discharges and another part of 15% from nuclear weapons test fallout. The transport of radionuclide discharges from European nuclear reprocessing plants was studied in detail in the 1970s and 1980s, when enhanced amounts of ^{137}Cs were discharged (e.g. Dahlgaard, 1995; Smith et al., 1990). These discharges could also be detected in Fram Strait and in the Arctic Ocean. The corresponding transfer time was reported to be four years from the reprocessing plant at Sellafield on the Irish Sea, to Fram Strait (Dahlgaard, 1995).

The Norwegian Radiation Protection Authority (NRPA) commenced investigations into long-range transported radionuclides off the western coast of Spitsbergen between the spring of 2000 and the summer of 2002 (Gerland et al., 2002a,b), with sampling programs in Kongsfjorden (Hop et al., 2002; Svendsen et al., 2002), Isfjorden (only in 2001), and further radioecological fieldwork in Fram Strait (RV "Polarstern" cruise in 2000).

Although the amount of new samples in this study is rather limited we find that the results give some valuable indications which are of use in further radioecological and oceanographic studies, especially in combination with oceanographic modelling. However, this study is meant to be the first phase of a longer period of long-term observation and monitoring, combined with further improved models.

2. Samples, measurements and the oceanographic model

Seawater samples of 50–100 litres each were obtained in Kongsfjorden from May 2000 to July 2002 (Gerland et al., 2002a,b), in Fram Strait in August 2000 (R/V

"Polarstern" cruise, Gerland and Grøttheim, 2001) and in Isfjorden on the R/V "Lance" cruise in 2001. At three stations (PS 197, PS 173 and PS 264), samples were also taken below the surface layer using information from vertical salinity and temperature profiling (CTD measurements) prior to sampling. In the western part of Fram Strait, where the EGC runs southwards parallel to the edge of the East Greenland shelf, seawater could be sampled at 100, 350 and 1000 m depths (station PS 197-1). On the eastern side of the Strait, off the coast of Spitsbergen, the water sample PS 264-2 was taken at 100 m below the surface (see Table 1), and the corresponding CTD profile (Fig. 1, personal communication from U. Schauer, 2001; Krause and Schauer, 2001), measured before sampling, indicated by its salinity maximum the presence of the WSC water masses. The sampling depth for PS 173-3 (152 m) was also chosen according to CTD information. Deep water samples were obtained at 1000 m (PS173-1, PS197-1) and at 465 m (PS264-2). In Kongsfjorden, only surface water was sampled. In Isfjorden, a sample was obtained at 55 m depth, since CTD information indicated Atlantic water influence at this level, exhibiting a 20–30 m thick layer with enhanced temperature and salinity (personal communication from V. Tverberg, 2003).

The method for measuring ^{99}Tc in seawater is described in detail by Kolstad et al. (1999) and Chen et al. (2001). Seawater samples were prefiltered (1 μm) for removal of suspended matter. ^{99m}Tc was used as a tracer in order to quantify the effectiveness of radionuclide separation. In the pre-measurement sample processing,

Table 1
Sample locations, location coordinates for seawater sampling, station ID, sampling depth and date, and technetium-99 results with associated errors (sorted from east to west)

Location	Latitude	Longitude	Station ID	Sampling depth (m)	Sampling date	^{99}Tc in seawater (Bq m^{-3})
Isfjorden	N 78° 17.7	E 15° 18.7	SV01-1c	55	18-05-01	0.34±0.12
Inner Kongsfj.	N 78° 58.1	E 12° 15.1	SV01-11b	surface	28-05-01	0.21±0.04
Inner Kongsfj.	N 78° 55.0	E 12° 13.4	SV00-2a	surface	17-05-00	0.21±0.03
Central Kongsfj.	N 78° 55.7	E 11° 56.3	SV00-4b	surface	19-05-00	0.25±0.03
Central Kongsfj.	N 78° 55.7	E 11° 56.3	SV01-9b	surface	24-05-01	0.20±0.03
Outer Kongsfj.	N 78° 59.1	E 11° 43.5	SV00-1b	surface	16-05-00	0.26±0.03
Outer Kongsfj.	N 78° 59.2	E 11° 55.3	SV02-MS1	surface	10-07-02	0.18±0.07
WSC	N 78° 55.2	E 8° 28.1	PS 264-3	surface	18-08-00	0.39±0.05
WSC	N 78° 55.2	E 8° 28.1	PS 264-2	100	18-08-00	0.27±0.03
WSC	N 78° 55.2	E 8° 28.1	PS 264-2	465	18-08-00	0.17±0.03
West WSC	N 78° 50.1	E 4° 2.4	PS 173-3a	surface	04-08-00	0.22±0.03
West WSC	N 78° 50.1	E 4° 2.4	PS 173-3	152	04-08-00	0.14±0.03
West WSC	N 78° 50.1	E 4° 2.4	PS 173-1	1000	04-08-00	0.04±0.02
Western EGC	N 78° 59.9	W 5° 0.0	PS 197-1	100	08-08-00	0.18±0.03
Western EGC	N 78° 59.9	W 5° 0.0	PS 197-1	350	08-08-00	0.16±0.04
Western EGC	N 78° 59.9	W 5° 0.0	PS 197-1	1000	08-08-00	0.05±0.02

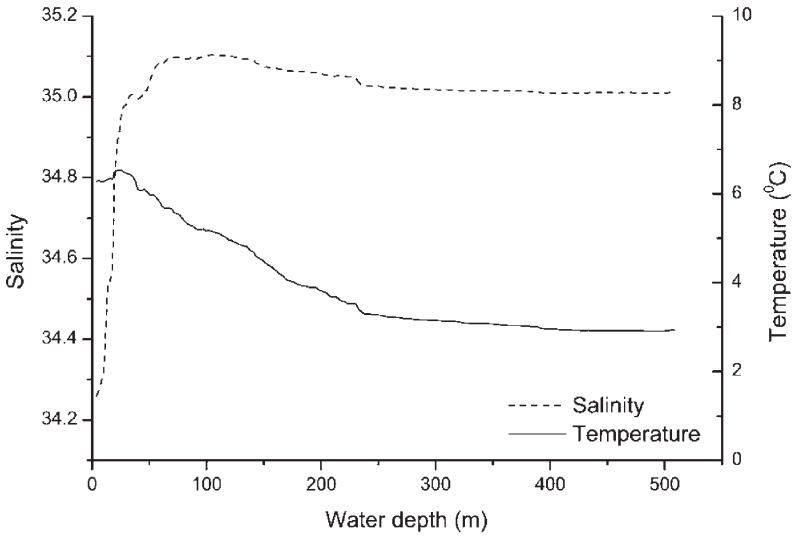

Fig. 1. CTD (Conductivity–Temperature–Depth) profile from station PS264. (Personal communication from U. Schauer) see also Krause and Schauer (2001) and Gerland et al. (2002b). In addition to surface sampling, the water sampling depths were chosen at 100 m and 465 m, because the maximum in salinity indicates Atlantic Water from the West Spitsbergen Current (WSC), and near the bottom deep water can be expected.

^{99}Tc is separated from other nuclides by ion-exchange chromatography using AG1-X4 resin (100–200 μm mesh size, Bio-Rad Laboratories, Hercules, CA, USA) and subsequent separation techniques. Then, the technetium isotopes were electroplated onto steel disks. The yield of the ^{99m}Tc tracer (typically 70–80%) on the disks was counted on a NaI well detector (Canberra, CT, USA). Later, the ^{99}Tc activity on the disks was measured with a low-background anti-coincidence β-counter (type GM-25-5, Risø, Roskilde, Denmark).

In order to be able to evaluate ^{99}Tc levels with better spatial and temporal resolution than could be achieved by sampling, the dispersion of the recent signal of ^{99}Tc was simulated with the 3-dimensional coupled ice-ocean model NAOSIM (North Atlantic–Arctic Ocean Sea Ice Model) of the Alfred Wegener Institute for Polar and Marine Research, Bremerhaven, Germany. The same model had also previously been used successfully, e.g. to calculate the possible spreading of radionuclides from the sunken submarine “Kursk” (Gerdes et al., 2001). Here, the model was driven with realistic daily atmospheric forcing data from 1979 to 1999, based on data from the European Centre for Medium-Range Weather Forecast (ECMWF). The model domain covers the Arctic Ocean, the Nordic Seas and the northern North Atlantic Ocean north of 50° N. For a more detailed description of the model and the forcing used see Karcher et al. (2003a,b). Monthly discharge data for the period 1994–1999 from the Sellafield reprocessing plant and yearly discharges for the preceding period of 1990–1993 were used as a tracer released into the uppermost model layer of the ocean (layer thickness 20 m) at the North Channel of the Irish Sea since the Irish

Sea proper is not sufficiently resolved in the model. The model output as used in this study covers a cross-section from Greenland to Svalbard at 76° N.

3. Results

The sample locations in Fram Strait form a profile from the shelf edge off the East Greenland Coast (station PS 197 in the Western EGC, water depth: 1320 m), to the deep sea west of the WSC (station PS 173, water depth: 2298 m), via the shelf edge (station PS 264, water depth: 509 m) to the outer (station SV00-1b), central (stations SV00-4b/01-9b, water depth <10 m) and inner Kongsfjorden (stations SV00-2a/01-11b). For Kongsfjorden stations, exact water depths are not available. On this profile, the levels of ^{99}Tc (see Table 1) in surface water were highest in the centre of the WSC with 0.39 Bq m^{-3}. The measurements west (PS 173) and east of the WSC show that the WSC is fairly narrow (Gerland et al., 2002b), in agreement with conclusions from oceanographers (Haugan, 1999; Krause and Schauer, 2001). However, levels east of the WSC in Kongsfjorden remain higher than west of the WSC (PS 173) indicating substantial influence by Atlantic Water from the WSC into the fjord. Additional results from sampling in Kongsfjorden in 2001 and 2002 are in reasonable agreement with the observations from 2000 (see Table 1). One sample from Isfjorden (SV01-1c), taken in May 2001 at 55 m depth in obviously Atlantic Water, showed also a high level with 0.34 Bq m^{-3}. The numbers of stations and depths levels at which samples could be taken, are not large enough to permit an estimation of the quantity of ^{99}Tc that reaches the EGC by recirculation in Fram Strait from the WSC. Nevertheless, the similarity of concentrations at 100 and 350 m depths at station PS 197 may indicate that the water at 100 m is more affected by weakly contaminated polar water masses, whereas at 350 m the influence of ^{99}Tc from the WSC is plausible. Results from below-surface water and deep water sampling from stations PS 173, PS 197 and PS264 show a decrease in ^{99}Tc levels with depth (Fig. 2, Table 1). An explanation for this depth distribution could be that the water with the highest levels of the conservatively behaving ^{99}Tc was found near the surface, because the discharges were also released to surface water (no or only little mixing; personal communication from U. Schauer, 2002). Variable, depth-dependent velocities of the WSC could be another process generating the observed distribution, by superimposing water with discharges from different discharge events.

The output from the NAOSIM model also shows decreasing ^{99}Tc levels with depth for November 1998 in Fram Strait (Fig. 3, top). Furthermore, strong lateral gradients in surface water are visible, agreeing well with observations in the WSC from our samples taken in spring and summer, 2000. However, for June 1999 (Fig. 3, bottom), the model gives a significantly different picture with higher ^{99}Tc levels at depths in the deeper part of the model range (300–500 m).

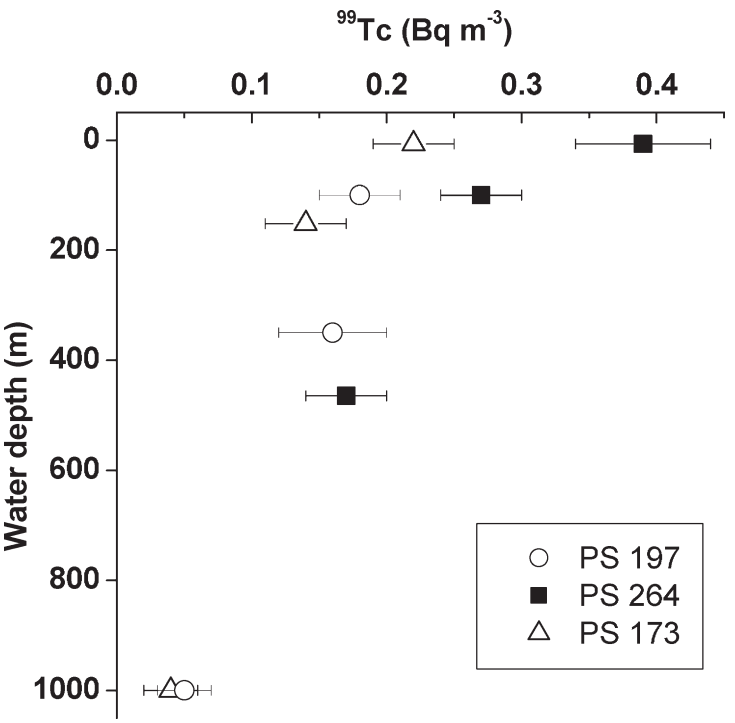

Fig. 2. ^{99}Tc data from seawater sampling at the locations in the areas of the East Greenland Current (EGC; PS 197) and the West Spitsbergen Current (WSC, PS 173 and PS 264) versus sampling depth.

4. Discussion

The ^{99}Tc levels observed west of Svalbard in 2000 are between six to seven times higher than those of surface water sampled in the same area in the mid-1990s (Kershaw et al., 1999), before the enhanced ^{99}Tc discharges from Sellafield could have reached Svalbard. This difference in levels and results from oceanographic 3-D modelling (Karcher et al., 2003a) give reasons to link the enhanced discharges from the nuclear reprocessing plant at Sellafield to the ^{99}Tc level changes observed in the Arctic. However, levels measured at Hillesøy on the coast of northern Norway, also in 2000 (Kolstad and Lind, 2002), are again five times higher than in Kongsfjorden at the same time.

Although the amount of ^{99}Tc data presented here is small, it gives clear indications of the coupling of the WSC and the fjords to the east of the Current, Isfjorden and Kongsfjorden. This supports observations made in the same area by oceanographers and sea ice researchers (Ingvaldsen et al., 2001; Gerland and Winther, 2000; Saloranta and Svendsen, 2001; Svendsen et al., 2002; see also Gerland et al., 2002b).

The result from the NAOSIM model (cross-sections in Fig. 3) indicate large changes in the vertical distribution of ^{99}Tc in the ocean west of Svalbard over a relatively short time. Reasons may be a strong seasonal variability in the upper part

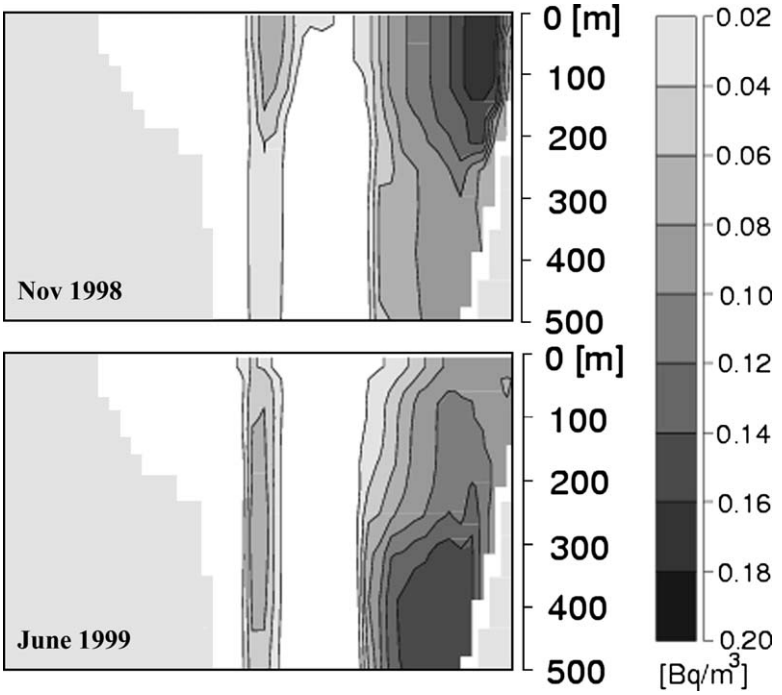

Fig. 3. 3-D modelling results for ^{99}Tc levels in seawater for a section across the Fram Strait from Greenland in the west (left) to Svalbard in the east (right) at ca. 76° N for November 1998 (upper diagram) and June 1999 (lower diagram), showing substantial changes in the spatial ^{99}Tc distribution over a short period.

of the WSC intensity or a strong month-to-month variability of surface concentrations as observed and simulated in the NCC further upstream (Karcher et al., 2003a). Unfortunately, corresponding NAOSIM model simulations later than 1999 are not yet available and a direct comparison of the observed levels and the model is not possible at this stage. Such a comparison will be necessary to identify whether it was accidental that no profile with a deep ^{99}Tc maximum was observed, or whether the model simulation exhibits inadequencies. For northern Norway, data from surface water observations at Hillesøy and NAOSIM results show good agreement. The same holds for the surface concentrations further upstream in the NCC and in the North Sea between 1996 and 1999 (Karcher et al., 2003a).

In order to understand the large variability of ^{99}Tc concentrations in time and space, surveys with significantly higher sampling density would be necessary, along with information on other properties of sea water. The fact that ^{99}Tc measurements are very expensive and time consuming will limit the possibilities of obtaining very large sample quantities as long as more practicable measurement techniques are not available. However, since 2000/2001, the NRPA has commenced monthly monitoring programs for ^{99}Tc in seawater at Ny-Ålesund, and the islands of Bjørnøya, Hopen, and Jan Mayen, as part of the Norwegian marine surveillance program. Along

with monitoring at Hillesøy, northern Norway (running since 1997), this data is intended to resolve, along with improved numerical oceanographic models, the complex transport processes of this radionuclide to the Arctic. Furthermore, as a result of enhanced international collaboration, data obtained from institutions from different nations is becoming more commonly available and integrated due to improved communication and database possibilities (e.g. AMAP, 2002, Karcher et al., 2003a).

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