Radionuclides in the Kongsfjorden area, Svalbard

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Abstract. The archipelago of Svalbard is located at the junction of Arctic Ocean, the North Atlantic and the Barents Sea. During a field campaign in May 2000, seawater, sea ice and seaweed, were collected in Kongsfjorden at the western coast of Spitsbergen. Possible sources for artificial radionuclides in the marine Svalbard environment are long-distance transported discharges from nuclear reprocessing plants, dumped radioactive waste, accidents with ships, submarines and power plants containing radioactive material, and global fallout from nuclear test explosions. Samples were taken to measure the radioisotopes technetium (^{99}Tc, sea water and seaweed), plutonium (sea water) and caesium (^{137}Cs, sea water and melted sea ice). Certain processes are unique for the high-arctic environment such as formation of sea ice. The fast ice in Kongsfjorden provides an opportunity to investigate to what degree radionuclides accumulate in sea ice compared with the seawater below. Another important question relates to how radionuclides accumulate in the Arctic in different types of seaweed. First results from laboratory analyses of the samples taken in May 2000 show ^{99}Tc concentrations in seawater similar to levels measured in the western Barents Sea in 1999-2000. Corresponding measurements along the Norwegian coast are one order of magnitude higher. Enhanced values measured at Hillesøy (Northern Norway), in the Barents Sea and off Svalbard and can be explained with increased discharges of ^{99}Tc-contaminated wastewater at the Sellafield reprocessing plant since 1994.

1. INTRODUCTION

Radioactivity in the Arctic is a central topic within environmental pollution issues [1]. Sources, both actual and potential, for radioactive pollution in the Arctic are marine and atmospheric long-range transported radionuclides, originating from nuclear power and reprocessing plants (e.g. Sellafield and La Hague), dumped radioactive waste, accidents with nuclear powered or armed ships and submarines and global fallout from nuclear test explosions in the atmosphere (e.g. [2]). The determination of radionuclide levels, their temporal and spatial variations and investigations concerning Arctic-specific processes provides insight into radionuclide behaviour and long-range transport of contaminants to Svalbard.

1.1 Aims

1.1.1 Uptake and deposition of radionuclides under Arctic conditions

A central aim was to study the uptake and deposition of radionuclides under Arctic conditions. In terms of water temperature, light availability, material in suspension and salinity, the conditions at Kongsfjorden are different compared with other areas. Possibly higher technetium levels in seaweed because of slow growth and smaller sizes (due to low salinity water) are discussed in the literature [3]. Further, influence of enhanced light and water temperature is reported to correlate with increased ^{99}Tc uptake in seaweed [4]. The determination of concentration factors in seaweed was intended to show whether these effects have a measurable input for certain seaweed species.

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1.1.2 Current radionuclide levels for the marine environment at Svalbard

Current levels of radionuclides in the Svalbard area were recorded for providing an inventory for more detailed radioecological studies in the Svalbard area and for possible accident-related surveys. ^{99}Tc concentrations at Svalbard for *Fucus vesiculosus* seaweed were around 10 Bq kg^{-1} d.w. in the early 1980s [5]. In the mid-90s, a dramatic increase in ^{99}Tc discharges from Sellafield began in connection with the EARP (Enhanced Actinide Removal Plant, e.g. [6]), the effect of which was subsequently registered at distant locations in the Norwegian Sea where elevated environmental concentrations of ^{99}Tc could be measured [7]; [8]. It is of interest to speculate as to the spatial extent of contamination arising from the releases from European reprocessing plants. ^{99}Tc levels before the increase from Sellafield could have reached the west coast of Spitsbergen were published and are between 0.04 and 0.06 Bq m^{-3} [9]. The transport of radionuclides from Europe to Arctic locations with ocean currents was studied and results from various groups are published (e.g. [10]-[13]). Transport times from Sellafield to Svalbard are published to be 4-6 years [10]. More recent ^{99}Tc data show levels up to 1.1 Bq m^{-3} in the southern Barents Sea [8].

1.2 Radionuclides

For this study, levels of ^{99}Tc, ^{137}Cs, $^{238, 239+240}$Pu and ^{241}Am were measured in the marine environment of Kongsfjorden and other Arctic locations.

The beta-radiation emitting ^{99}Tc is a long-lived radionuclide formed in fission processes that occur in nuclear reactors and is discharged into the environment at nuclear reprocessing plants (e.g. [7]). It is highly soluble in seawater and has a half life of $2.12 \cdot 10^5$ years. In the marine environment, it accumulates especially in seaweed, which can be therefore used as a bioindicator for ^{99}Tc in seawater. Results of ^{99}Tc in seaweed along the Norwegian coast and in the Baltic Sea were published, for Northern Norway the level in seawater was 1.4 Bq m^{-3} in the second half of 1998 (e.g. [8]). In seaweed (*Fucus vesiculosus*), concentrations of ^{99}Tc were 124 Bq kg^{-1} at Hillesøy (Northern Norway) in early 1998 [7].

The gamma-radiation emitting ^{137}Cs radioisotope (half life: 30 years) in nature originates from nuclear power and reprocessing plants, accidents, test explosion fallout. It is both particle-reactive and soluble. ^{137}Cs levels in the southern and central Barents Sea, measured in 1999, range from 2.8 to 4.0 Bq m^{-3}.[14].$^{238, 239+240}$Pu and ^{241}Am are transuranic radionuclides emitting alpha radiation; the half life of ^{239}Pu is $2.4 \cdot 10^4$ years. Plutonium in the environment may come from nuclear power and reprocessing plants, from nuclear weapon tests, and from accidents with nuclear powered submarines. Plutonium is found both in marine sediments and in seawater. $^{239+240}$Pu levels in seawater in the Barents Sea were 6.6-9.9 Bq m^{-3} [14].

2. SAMPLING AT KONGSFJORDEN

Western Svalbard is characterised by its high Arctic environment with a relatively mild climate, arising from the nearby passage of the West Spitsbergen Current, a branch of the North Atlantic Current. Kongsfjorden is located at 79°N at the western coast of Spitsbergen. This fjord is 25-30 km long and 5-10 km wide. The inner part of Kongsfjorden is covered with seasonal fast ice from about December to June regularly. This ice is typically 60-70 cm thick at the end of the ice-growing season in May [15]. During summer, the fast ice melts and disappears. Then, the water in the fjord is enriched with glacial sediments. At all times of the year, small icebergs and ice pieces from glaciers flowing into Kongsfjorden are abundant in the fjord.

Samples that are used for comparison purposes in this study were taken at Bjørnøya, Hopen, Jan Mayen (Greenland Sea) and Hillesøy ([8], Northern Norway), which are all regular monthly sea water monitoring sites of the NRPA. Among these, Jan Mayen has a unique setting, because it is less influenced by the North Atlantic or West Spitsbergen Current than the other locations. However, [16] states that 85% of the ^{99}Tc in the East Greenland current originates from European coastal discharges.

At Kongsfjorden, fieldwork was performed in May 2000. In the fjord, surface seawater at three locations (^{137}Cs, Pu andAm isotopes, ^{99}Tc), sea ice at one site (^{137}Cs, total γ-analysis), and different types of seaweed at 2 sites (^{99}Tc) were sampled (Tab. 1). Water samples were collected using buckets and 25 litre canisters, whereas sea ice was cored with a 4" Kovacs Mark II corer drill. Seaweed could be collected from rocks (*Fucus distichus*) and by means of scuba diving, near Ny-Ålesund at the southern shore of Kongsfjorden, and from a location near the northern coast of the fjord.

Table 1: Overview on all sample stations.

Site	Setting	Latitude	Longitude	Sampling
1	Outer fjord	N 78° 59'	E 11° 44'	seawater
2	Inner fjord	N 78° 55'	E 12° 13'	seawater, sea ice, seaweed
3	Northern shore	N 78° 59'	E 11° 58'	seaweed
4	Southern shore	N 78° 56'	E 11° 56'	seawater, seaweed

3. MEASUREMENTS TECHNIQUES

For the analyses of the samples, gamma spectroscopy, beta and alpha counting were used.
Concentrations of ^{137}Cs were determined by gamma counting with High-Purity Germanium detectors. Seawater (200 litres) was first filtered through a sorbent filter system. After drying, the sorbents were ashed, and the ash subsequently measured using gamma detectors (Canberra and Ortec HPGe (High-Purity Germanium) detectors). For the gamma spectroscopy on sea ice, ice cores were melted, resulting in 135 liters water. This water was then treated as the other seawater samples.
The method for beta counting is described in detail in [17]. For the ^{99}Tc concentration measurement, gross seaweed samples were dried at +60°C, mechanically powdered and passed through a 500 μm mesh, resulting in a 10 g analytical sample. Seawater samples (50-100 litres) were pumped through a 1 μm polypropylene filter in order to eliminate biotic material. Samples consisted of 100 or 50 litres, depending on existing information on expectable levels. In the pre-measurement sample processing, the ^{99}Tc radionuclides are separated from other nuclides in the sample using radiochemical methods. ^{99m}Tc is used as a tracer in order to quantify the effectiveness of the radionuclide separation. Finally, the ^{99}Tc isotopes are electroplated on a metal disk. The metal disks and ^{99m}Tc standard solution is counted on a NaI detector, before counting for beta radiation on a low-background counter (type GM-25-5, RISØ, Roskilde, Danmark). The results for seaweed and seawater have an error of 5 and 8%, respectively, resulting in an error of about 10% for concentration factors [18].
Before the alpha counting process, water samples (200 litres) are prefiltered and the sample is reduced in volume by chemical precipitation. Later, the radioactive plutonium or americium isotopes are attached to a metal plate in an electrolytic bath (electrode deposition). The plates were then measured in a Canberra alpha counter and analysed with Ortec software.

4. RESULTS

Levels for ^{137}Cs in sea water (Tab. 2) are slightly lower than those measured in surface water of the Southern Barents Sea in 1999 (2.8-4.0 Bq m^{-3}, Brown et al. 2000). The level measured on melted sea ice is in agreement with published ^{137}Cs levels for sea ice (Meese et al. 1997). With an average salinity for Kongsfjorden fast ice of about 6‰ (Gerland et al. 1999), the ratio between salinity in ice and sea water is similar to the ratio of ^{137}Cs levels in both media. This is in agreement with Weeks (1994), who states that, with a few exceptions, substances do not substitute in the ice lattice in measurable amounts.

Site	^{137}Cs (Bq m^{-3})
1	2.32
2	2.19
2 (melted ice)	0.4
4	1.84

Table 2: Levels for ^{137}Cs in sea water and melted sea ice.

Table 3: ^{99}Tc in sea water from Kongsfjorden and other areas.

Site	^{99}Tc (Bq m^{-3})
1	0.26
2	0.21
4	0.25
Bjørnøya (12.00)	0.27
Hopen (12.00)	0.26
Jan Mayen (4.01)	0.13
Hillesøy (8.00)	1.37

Levels for ^{99}Tc in sea water were at all locations measured (1, 2, 4) above 0.2 Bq m^{-3}. This is a similar level as observed in the western Barents Sea (Fig. 1), and about four times as much as [9] published as recorded in 1994 west of Svalbard, giving strong indications that parts of the enhanced discharges from Sellafield reached the Svalbard area latest by May 2000. Recent data from Bjørnøya, Hopen and Jan Mayen indicate also presence of enhanced ^{99}Tc in seawater (Tab. 3, Fig. 1). ^{99}Tc levels in seawater at the Norwegian coast and in the southern Barents Sea are about 4-5 times higher than near Svalbard, due to the setting relative to the Norwegian coastal current ([8]; Fig. 1).

Table 4: Levels of ^{99}Tc in seaweed for Kongsfjorden and Hillesøy (Northern Norway).

Site	Species	^{99}Tc (Bq kg^{-1}) d.w.
3	Alaria esculenta	8.2
3	Laminaria digitata	4.5-13.2
4	Palmaria palmata	0.25
4	Desmerestia aculeata	13.5
4	Laminaria saccharina	2.79
4	Fucus distichus	34.3
Hillesøy (1997-99)	Fucus vesiculosus	179.4

Table 5: Concentration factors for ^{99}Tc in seaweed

Seaweed	CF (. 10^5)	Location/date
Fucus distichus	1.37	Svalbard 2000
Desmerestia acul.	0.54	Svalbard 2000
Laminaria sacch.	0.11	Svalbard 2000
Palmaria palmata	0.01	Svalbard 2000
Fucus vesiculosus	1.45±0.44	Hillesøy, 1997-1999
Fucus vesiculosus	1.19±0.41	Irish Sea [19]
Fucus vesiculosus	0.93±0.19	Utsira, Norway [20]
Fucus serratus	1.21±0.17	Oslofjord, Norway [7]
Fucus spiralis	0.24	Irish Sea [21]

Looking at levels of ^{99}Tc in seaweed, the highest levels were observed in the species *Fucus distichus* (Tab. 4). Among the other species, which were obtained by diving, *Desmerestia aculeata* shows the highest values with about half the level as observed in *Fucus distichus*.

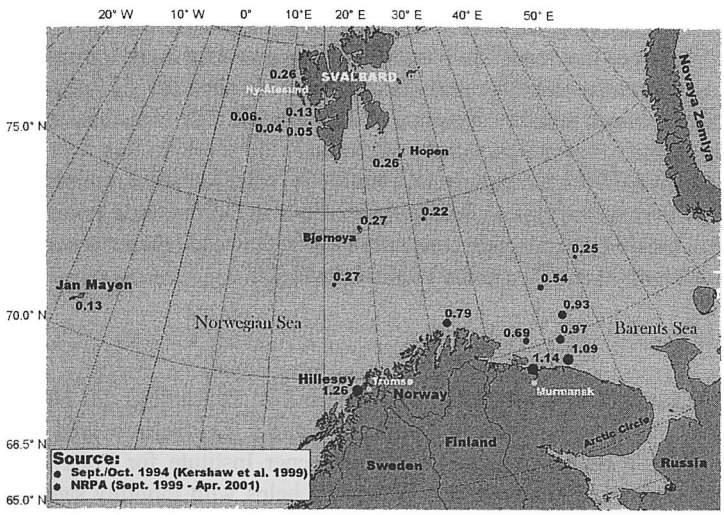

Figure 1: Map of the Norwegian and Barents Seas and adjacent areas with recent results on ^{99}Tc in seawater (in Bq m^{-3}). The data from 1994 are the three measurement sites off the western coast of Svalbard (0.06, 0.04, and 0.05 Bq m^{-3}).

The $^{239+240}Pu$ level for site 1 is 7.2 +/- 1.1 mBqm^{-3} (^{238}Pu concentration was lower than 0.4 mBq m^{-3}, and ^{241}Am was 0.79 +/- 0.67 mBq m^{-3}). The level for $^{239+240}Pu$ is in the same order of magnitude as for the southern Barents Sea (7.5 mBq m^{-3} [22]). The ^{238}Pu /$^{239+240}Pu$ ratio would be 0.056, using the upper limit of the ^{238}Pu level found. However, the concentration of ^{238}Pu was close to the lower limit of detection in seawater, therefore this ratio is not further interpreted.

The concentration factor (CF) for radionuclides in biota is defined as the radionuclides concentration in biota in Bq kg^{-1} (d.w.), divided by the concentration in water from the same location in Bq kg^{-1}. Calculation of ^{99}Tc concentration factors on seaweed from Kongsfjorden showed large variations depending on seaweed types (Tab. 5). Comparison with CF values from other studies (Tab. 5, lower part) shows similar results for *Fucus vesiculosus* as for *Fucus distichus* from Kongsfjorden.

5. DISCUSSION

Radioactivity studies on sea ice often deal with sediment transport and inclusions, because sea ice plays an important role in transporting sediments over long ranges in the Arctic. At a location as Kongsfjorden, it is possible to study the effect of radioactivity in sea ice without impurities and with known location, history and background data on ice formation. Our result from comparison of ^{137}Cs in sea ice and seawater supports the statement that radionuclides accumulate in the brine. Since the porosity of the sea ice changes significantly and with that the coupling between pore space and seawater below the sea ice, bulk radionuclide levels on sea ice can be expected to vary accordingly. Since a variety of ice algae and other biota use the brine channels and sea ice underside as a habitat, it would be of interest to study in the future to what degree radionuclides in the brine may accumulate in the food chain.

It is of interest to speculate as to the spatial extent of contamination arising from the enhanced Sellafield releases. Measurements on seawater west of Svalbard in 1995, undertaken before the increase of ^{99}Tc releases from the mid 90s could have reached the Arctic, show values about four times lower than what we measured on samples from Kongsfjorden 2000 (Kershaw et al., 1999). Transfer times as given in the literature (e.g. Dahlgaard 1995), natural transport from Sellafield to NW Norwegian Current would take 3-4 years, to West Spitsbergen about 5 years, and to the East Greenland current 7-10 years. Time series taken at Hillesøy (Brown et al. in press) confirm the number for Northern Norway, and our results from Svalbard indicate strongly that the enhanced discharge's signal reached the west coast of Spitsbergen. Our result from Jan Mayen gives the impression that the signal arrived also there, although that appears

rather early compared with Dahlgaard's numbers. However, the enhanced level there could be also due to other sources (e.g. discharges that reach Jan Mayen via the Arctic Ocean) or transport with ocean currents from the Norwegian Coastal Current that reach Jan Mayen rather directly.
In May 2001, the NRPA began monthly sampling of seawater for ^{99}Tc measurements. The results of this are intended to help identify seasonal variations of radionuclide levels in this Arctic environment, where the contrast is significant between environmental conditions (solar radiation, temperature, water masses in the fjord, sea ice) in winter and summer. Seasonal variations in radionuclide levels were also observed at other locations. [23] observed radionuclide concentrations in seaweed (*Fucus serratus*) finding a seasonal cycle for iodine, potassium-40 and ^{99}Tc in the English channel.

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