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## Distribution of natural and anthropogenic radionuclides in sediments from the Vefsnfjord, Norway



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#### ABSTRACT

Areas in central Norway were heavily contaminated with fallout from the Chernobyl accident in 1986. In this study, we assess <sup>137</sup>Cs in surface sediments and sediment cores collected in the Vefsnfjord in Nordland county. Concentrations of <sup>137</sup>Cs in surface sediments ranged from 159 to 191 Bq kg<sup>-1</sup> dry weight (d.w.). Sub-surface peaks of <sup>137</sup>Cs were observed in all cores, with a maximum concentration of 432 Bq kg<sup>-1</sup> d.w. Given that little is known about the distribution of naturally occurring radionuclides in Norwegian fjords and coastal areas, a better understanding of the total burden of radioactivity is important for the Norwegian fishing and aquaculture industries. Therefore, analyses of the natural radionuclides <sup>40</sup>K, <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>210</sup>Pb were included in the study. Analyses of total sulphur (TS), total carbon (TC), total organic carbon (TOC) and grain size distribution have been performed to provide a sedimentologic context for interpreting the radionuclide results.

## 1. Introduction

Knowledge about the distribution of radioactive contamination in the environment after a nuclear accident is vital to both food resource management and nuclear preparedness (e.g. Minoura et al., 2014; Vives i Batlle et al., 2018). For example, predictive scenarios of contaminant dispersal in marine areas and biological uptake are important to the aquaculture and fishing industries.

Cesium-137 (<sup>137</sup>Cs) (half-life  $30.05 \pm 0.08$  years (Bé et al., 2013)) is one of the most common anthropogenic radionuclides, for which main sources in the terrestrial environment in Norway are fallout from the atmospheric nuclear weapons tests in the 1950s and early 1960s and fallout from the Chernobyl accident in April 1986. Norway was among the countries receiving most Chernobyl fallout (Backe et al., 1986). Due to rainfall during the first few days after the accident, some areas in central Norway (Oppland, Hedmark, Trøndelag and Nordland counties, Fig. 1) were particularly contaminated. Levels in Norwegian soils were mapped country-wide shortly after the accident and showed an average <sup>137</sup>Cs activity concentration of 7.1 kBq m<sup>-2</sup> (Backe et al., 1986, 1987). In 1986, the municipalities Vefsn, Hattfjelldal and Grane, surrounding Vefsnfjord, had mean values of <sup>137</sup>Cs deposition between 19 and 24 kBq m<sup>-2</sup> (Backe et al., 1986). The levels in these municipalities (different localities) were 7–16 kBq m<sup>-2</sup> in 1995 and 3–16 kBq m<sup>-2</sup> in 2005 (Gjelsvik and Steinnes, 2013), involving local spatial variations (pers. comm. R. Gjelsvik).

Results from Norway's national monitoring programme Radioactivity in the Marine Environment (RAME; www.dsa.no) show that Vefsnfjord is one of the Norwegian fjords with highest levels of <sup>137</sup>Cs in surface sediments with activity concentrations up to 348 Bq kg<sup>-1</sup> dry weight (d.w.) in the period 2002–2018 (NRPA, 2004, 2011, 2012; Skjerdal et al., 2015, 2017, 2020; IMR/DSA unpublished data). These levels are two orders of magnitude higher than in open Norwegian sea area sediments (e.g. Skjerdal et al., 2020), and confirm that Vefsnfjord is heavily affected by <sup>137</sup>Cs contamination, even more than 30 years after the Chernobyl accident.

Little is known about the distribution of naturally occurring radionuclides in Norwegian fjords. A better understanding of the total burden of radioactivity in Norwegian coastal areas is important for the fishing and aquaculture industries. Natural radionuclides (e.g. potassium-40 (<sup>40</sup>K), half-life  $1.2504 \pm 0.0030 \cdot 10^9$  years (Bé et al., 2013) are much more abundant in the marine environment than anthropogenic radionuclides, and some of them are of much higher radiological significance than <sup>137</sup>Cs (e.g. lead-210 (<sup>210</sup>Pb), half-life 22.30 ± 0.22 years (Schötzig and Schrader, 1993)). Important sources for natural radionuclides to

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marine sediments are weathering and erosion of terrestrial rocks containing  $^{40}$ K and radionuclides of the uranium and thorium radioactive series (e.g. radium-226 ( $^{226}$ Ra), half-life 1600  $\pm$  7 years (Bé et al., 2013), radium-228 ( $^{228}$ Ra), half-life 5.75  $\pm$  0.04 years (Bé et al., 2013) and  $^{210}$ Pb), and atmospheric deposition due to rain, snow or dry fallout.

The Vefsnfjord is about 50 km long and reaches maximum depths of approximately 490 m in the inner part and 270 m in the outer part (www

.norgeskart.no) (Fig. 1). The bathymetry is complex with several sills and shallow areas. The hydrography and currents in the fjord are influenced by freshwater discharges, tides and winds. Riverine runoff mixes with fjord water to form a brackish surface layer flowing out the fjord. In periods of large freshwater discharge from the rivers (mainly Vefsna, see Fig. 1), the surface layer has a thickness of 2–5 m and salinity as low as 5 psu in the inner part of the fjord (Haugen et al., 1981). The



**Fig. 1.** Map of Norway showing the counties Oppland, Hedmark, Trøndelag and Nordland and the municipalities Vefsn, Hattfjelldal and Grane (in italics). The geographical location of Vefsnfjord is shown. Inset: Bathymetry of Vefsnfjord and positions of four sampling sites and their echo depths (m). The main rivers draining into Vefsnfjord include Vefsna, Fusta, Drevja and Hundåla. Vefsna (163 km long and total elevation of 877 m) is the largest with a catchment area of 4229 km<sup>2</sup> (Molvær, 2010). Mean and maximum water discharge rates of the Vefsna river in the period 1978–2008 were 174 and 1800 m<sup>3</sup> s<sup>-1</sup>, respectively (Molvær, 2010). According to NVE (2004), the maximum discharges take place during the snow melting season in May–June, but flooding events caused by heavy rainfall is frequent in the autumn. Sills in Vefsnfjord are found between the inner and outer part (~170 m), in the sound Sundet (~40 m) and at the fjord mouth between Tjøtta and Mindlandet (~80 m).

velocity of the surface layer flowing out of the fjord is 0.1–0.3 m s<sup>-1</sup> (Molvær, 2010). A layer of compensating inflowing water is found below the outflowing surface layer. The sills limit water exchange between the deeper parts of the fjord and the Norwegian Coastal Current, but according to Haugen et al. (1981), it is likely that the bottom water is replaced in winter every year or every other year. The reduced water exchange and complex bathymetry of the fjord is affecting the sedimentation regime in the fjord.

Bedrock geology in the western part of the Vefsna drainage area comprises of Cambrosilurian strongly metamorphosed sedimentary and volcanic rocks. Gneissic and calcareous rocks with varying metamorphic degrees occur further to the south. Granitic rocks are present in the northern part of the Vefsna drainage area, while the eastern part of the drainage area consist of carbonates, calcareous shales, phyllites, sandstones and conglomerates (Gustavson, 1981). Quaternary deposits are present mainly in the major valleys where sediment accumulation takes place whereas the mountainous areas have very little Quaternary deposits present due to ice-erosion during the last glaciation. The quaternary geology has been mapped and published by Follestad (1990).

In this study, surface sediments and sediment cores from four locations in a gradient along the Vefsnfjord (Fig. 1) were analysed for the content of <sup>137</sup>Cs, <sup>228</sup>Ra, <sup>226</sup>Ra, <sup>210</sup>Pb and <sup>40</sup>K. We assess the spatial and stratigraphic distribution of radionuclides in the fjord sediments and evaluate the impact of the <sup>137</sup>Cs contamination. Further, to elucidate the radionuclide distribution in the fjord, the sediments were also analysed for grain size (GS) and the content of total sulphur (TS), total carbon (TC) and total organic carbon (TOC). Sediment ages and sediment supply rates were also determined using <sup>210</sup>Pb- and <sup>137</sup>Cs-profiles. These results are reported in a separate paper (Heldal et al., 2021).

#### 2. Materials and methods

#### 2.1. Sample collection and preparation

Sediment samples were collected onboard R/V "Kristine Bonnevie" 28. October 2018 using a "Smøgen" box corer (area of 30 cm  $\times$  30 cm and depth of 40 cm) at four sites in the Vefsnfjord (Fig. 1, Table 1). One surface sample (0–2 cm) and three parallel sediment cores were collected from a single box corer at each site. One core was collected to study the vertical distribution of GS, TS, TC and TOC whereas two cores were collected to study the vertical distribution of a range of radionuclides (the natural radionuclides <sup>40</sup>K, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>210</sup>Pb and the anthropogenic radionuclide <sup>137</sup>Cs). The results from analyses of GS, TS, TC, TOC and radionuclides are reported in the present paper. The <sup>137</sup>Cs and <sup>210</sup>Pb/<sup>226</sup>Ra data were used to determine the geochronology at each site, which is reported in Heldal et al. (2021).

The sediment cores were collected by pushing PVC tubes of 40 cm length and 10 cm inner diameter into the box core. The bottom ends of the PVC tubes were sharpened to minimize disturbance and the effect of sediment compaction. The cores were cut into 1 cm slices (0–10 cm) and 2 cm slices from 10 cm to the bottom of the core onboard the ship. The samples were transferred to pre-weighed aluminium containers and their wet weighs determined. The samples were stored frozen at  $\div$ 20 °C until further preparation took place at the Institute of Marine Research (IMR) in Bergen. At the laboratory, the frozen samples were freeze-dried using a CHRIST ALPHA 1–4 freeze dryer until constant dry weight was achieved. Dry weight of all samples was determined. The samples were homogenized using a Retsch Planetary Ball Mill PM 100.

## 2.2. Analyses of grain size (GS), total sulphur (TS), total carbon (TC), total organic carbon (TOC) and calculation of carbonate content

Grain size (GS) measurements were carried out at the Geological Survey of Norway (NGU) in Trondheim using laser diffraction instrument Coulter LS 2000 (for details, see Xu, 2000, and references therein) that measures the fractions from 0.4  $\mu$ m to 2 mm. The GS distribution is

determined with respect to percent volume, with the assumption of uniform density of the sample.

Contents of total sulphur (TS), total carbon (TC) and total organic carbon (TOC) were determined using a LECO SC-632 instrument. For determination of TC and TS, the sediment sample is combusted in an oven with high temperature (normally 1350 °C) with continual flow of oxygen to produce  $CO_2$  and  $SO_2$ . For determining the TOC, the samples were first treated with hydrochloric acid (HCl) to remove inorganic carbonate carbon. The difference between TC and TOC is assumed to be carbonate C primarily originating from biogenic shell material in the sediments. Other possible sources for carbonate in the sediments might be carbonate in eroded bedrocks draining to Vefsnfjord. The carbonate content is calculated according to the following formula:

Carbonate, weight% =  $(TC \div TOC) \times 8.33$ 

where the factor 8.33 is derived from the molecular weight  $\mbox{CaCO}_3/\mbox{C}$  ratio.

## 2.3. Analyses of <sup>40</sup>K, <sup>137</sup>Cs, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra by gamma spectrometry

All samples were analysed by gamma spectrometry for  $^{40}$ K,  $^{137}$ Cs,  $^{210}$ Pb,  $^{226}$ Ra and  $^{228}$ Ra at IMR in Bergen. Homogenized samples were transferred to 60 or 200 ml polypropylene (PP) counting geometries. The sample sizes varied from 43.6 to 185.6 g d.w. The samples were vacuum sealed in an aluminium-lined bag using a Turbovac T20 Table Top Vacuum Packing Machine to prevent loss of radon-222 ( $^{222}$ Ra). The samples were thereafter stored for at least four weeks prior to gamma analysis to establish a secular equilibrium between  $^{226}$ Ra and its progeny (Gäfvert and Mauring, 2013).

The analytical method for measuring <sup>137</sup>Cs is accredited in accordance with the standard ISO 17025. The calibration and validation sources are traceable to national standards (NPL B180222, VNIIM 252/ 2000). The methods for determining <sup>40</sup>K, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra are not accredited, but are calibrated and verified by analysing reference sources traceable to national standards (PTB SRM numbers RARB15075 and RBRB15076) and other reference materials (KCl-salt merck for analysis), IAEA-RGU-1 and IAEA RGTh-1). The calibration and validation sources had the same geometry and similar density as the samples. The methods are regularly verified by participation in national and international intercomparison exercises.

The <sup>40</sup>K, <sup>137</sup>Cs, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra content was determined using two low-background ORTEC High Purity Germanium (HPGe) detector systems: one N-type coaxial HPGe-detector (model no. GMX-M5970P–S) with preamplifier (model no. 257N) equipped with X-Cooler electric cryostat cooling system and DSPEC multichannel analyser; and one Ptype coaxial HPGe-detector (model no. GEM-S8530P4-RB) with preamplifier (model no. A257P) equipped with X-Cooler III electric cryostat cooling system and DSPEC-50 multichannel analyser. Relative efficiencies of the detectors at 1.33 MeV were 47% and 52%, respectively. Analytical uncertainties are due to uncertainty in sample preparation, calibration standards, calibration methods, counting statistics and background correction, and are in the results given as  $\pm 2\sigma$ . Counting times varied from approximately 65,000 to 270,000 s.

The <sup>40</sup>K content was determined using the 1460.8 keV gamma peak. Potassium-40 isotopic abundance of 0.0117% was assumed (www.nuc leide.org). Background peak were accounted for by Peak Background Correction (PBC) in the Gamma Vision® software.

The  $^{137}\mathrm{Cs}$  content was determined using 661.7 keV gamma peak. No  $^{137}\mathrm{Cs}$  in background was detected.

The <sup>210</sup>Pb content was determined according to the method described by Sværen (2010). The method includes corrections for selfabsorption of the 46.5 keV gamma peak of <sup>210</sup>Pb. Corrections were carried out using a 255 kBq <sup>210</sup>Pb point source (QSA Global GmbH).

The <sup>226</sup>Ra content was determined using gamma peaks of the decay

Table 1

Sediment samples collected in the Vefsnfjord and type of analyses.

Sample ID	Sample type	Latitu	ıde		Longi	tude		Echo depth (m)	Length of core (cm)	Number of samples	Type of analyses
1121-S	Surface	65	57.39	Ν	12	44.90	Е	226	_	1	Gamma spectrometry
1121-1	Core	65	57.39	Ν	12	44.90	Е	226	20	15	Gamma spectrometry
1121-2	Core	65	57.39	Ν	12	44.90	Е	226	19	15	Gamma spectrometry
1121-3	Core	65	57.39	Ν	12	44.90	Е	226	20	15	GS, TS, TC, TOC
1122-S	Surface	65	57.72	Ν	12	54.77	Е	487	-	1	Gamma spectrometry
1122-1	Core	65	57.72	Ν	12	54.77	Е	487	14	12	Gamma spectrometry
1122-2	Core	65	57.72	Ν	12	54.77	Е	487	14	12	Gamma spectrometry
1122-3	Core	65	57.72	Ν	12	54.77	Е	487	15	13	GS, TS, TC, TOC
1123-S	Surface	65	56.46	Ν	12	58.31	Е	486	-	1	Gamma spectrometry
1123-1	Core	65	56.46	Ν	12	58.31	Е	486	18	14	Gamma spectrometry
1123-2	Core	65	56.46	Ν	12	58.31	Е	486	16	13	Gamma spectrometry
1123-3	Core	65	56.46	Ν	12	58.31	Е	486	18	14	GS, TS, TC, TOC
1124-S	Surface	65	54.79	Ν	13	6.15	Е	448	-	1	Gamma spectrometry
1124-1	Core	65	54.79	Ν	13	6.15	Е	448	12	11	Gamma spectrometry
1124-2	Core	65	54.79	Ν	13	6.15	Е	448	13	12	Gamma spectrometry
1124-3	Core	65	54.79	Ν	13	6.15	Е	448	12	11	GS, TS, TC, TOC

products <sup>214</sup>Pb (295.2 keV and 351.9 keV) and <sup>214</sup>Bi (609.3 keV) and the <sup>228</sup>Ra content was determined using gamma peaks of <sup>228</sup>Ac (338.3 keV, 911.2 keV and 969.0 keV) (see e.g. Kahn et al. (1990) and Köhler et al. (2002)). Variation in the radon and thoron background levels was controlled by routine background measurements. Background peaks were accounted for by Peak Background Correction (PBC) in the Gamma Vision® software.

#### 3. Results and discussion

#### 3.1. Sediment characterization

Fig. 2 shows the average sediment grain size distribution (%) at the four sites subdivided into three size classes: clay (<2  $\mu$ m), silt (2–63  $\mu$ m) and sand (63–2000  $\mu$ m). The data for each sediment layer is given in Supplementary Material (SM) 1. Fig. 3 shows the TS, TOC and carbonate content. The data is also given in SM 2 together with the TC (%) content. The sediments in the outer part of the fjord (site 1121) are finer grained and have higher TOC and carbonate content compared to the inner part of the fjord. The differences in grain size are probably related to the proximity/distance from the Vefsna river mouth. The higher carbonate content in the outer part of the fjord implies that the sediments has a biogenic origin and do not originate from eroded rock material. The coarsest sediments in the inner fjord are found in site 1124 located

closest to the river mouth, and there is a gradual fining trend with increasing distance to the river mouth. The higher TOC content in the outermost site 1121 compared to the other sites suggests either higher marine organic matter (OM) input or less detrital sediment discharges in the outer Vefsnfjord compared to the inner part. A higher OM input in the outer part of the fjord can be caused by higher phytoplankton growth in this part of the fjord. Phytoplankton growth in fjords is controlled by e.g. bathymetry, stratification and advection with OM from coastal waters. Various aspects related to primary production and sedimentation in Norwegian fjords are highlighted by e.g. Wassmann (1990). The conditions in the Vefsnfjord need, however, to be further investigated in order to conclude.

## 3.2. Cesium-137 (<sup>137</sup>Cs)

Activity concentrations of <sup>137</sup>Cs in Vefsnfjord (Table 2, Fig. 4, SM 3) are two to three orders of magnitude higher than values typical for sediments in Norwegian sea areas. For comparison, activity concentrations in surface sediments (0–2 cm) from the Norwegian Sea in 2017 range from 0.2 to 8.7 Bq kg<sup>-1</sup> d.w (Skjerdal et al., 2020).

The highest activity concentration of  $^{137}$ Cs in surface sediments (0–2 cm) was found at site 1124, close to the mouth of the river Vefsna (Table 2). This suggests that contaminated particles transported with the river Vefsna settle in or near the estuary before they are potentially



 $\Box < 2 \ \mu m \ (\%)$   $\Box 2 - 63 \ \mu m \ (\%)$   $\Box 63 - 2000 \ \mu m \ (\%)$ 

Fig. 2. Average sediment grain size (GS) distribution (%) at the four sites.



Fig. 3. The vertical distribution (%) of total sulphur (TS), total organic carbon (TOC) and carbonate content at the four sites.

redistributed in the fjord. Brydsten and Jansson (1989) describe a similar situation for the river Öre in northern Sweden. They conclude that river transported material is primarily deposited within the influence area of the river plume and within the estuary, followed by resuspension and transport to deeper parts of the estuary. Higher <sup>137</sup>Cs levels in surface sediments in the head compared to the mouth of the fjord are also found in other fjords in central Norway (e.g. Sværen, 2010; Skjerdal et al., 2017), consistent with riverine discharges of <sup>137</sup>Cs into the fjords.

The <sup>137</sup>Cs activity concentrations in the sediment cores show a similar vertical distribution with a clear maximum-peak between 6 and 16 cm core depth followed by a rapid decline down the cores (Fig. 4). While the profiles of two parallel cores collected at sites 1123 and 1124 agree well with each other, there are few cm vertical offsets between the cores collected at sites 1121 and 1122. These offsets may be related to statistical uncertainties in the radiometric assays, small variations in the sampling and sample preparation procedures or local variations in e.g. sediment composition.

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Fig. 4. Vertical distribution (Bq  $kg^{-1}$ ) of <sup>137</sup>Cs, <sup>40</sup>K, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra in cores from the four sites.

#### Table 2

Activity concentrations (Bq kg<sup>-1</sup> d.w.) of <sup>137</sup>Cs, <sup>40</sup>K and <sup>228</sup>Ra in 0–2 cm surface sediments at the four sites. Analytical uncertainties are given as  $\pm 2\sigma$ .

Sample ID	<sup>137</sup> Cs (Bq kg <sup>-1</sup> ) d.w.			<sup>40</sup> K (Bq kg <sup>-1</sup> ) d.w.			<sup>228</sup> Ra (Bq kg <sup>-1</sup> ) d.w.		
1121-S	164	±	15	525	±	50	25.7	±	3.2
1122-S	161	±	15	659	±	63	48.5	±	5.8
1123-S	159	$\pm$	14	555	±	53	50.6	±	6.1
1124-S	191	$\pm$	17	652	$\pm$	63	45.8	±	5.5

The <sup>137</sup>Cs maximum-peak concentrations at the four sites range from 204 to 432 Ba kg<sup>-1</sup> d.w. The highest is found in the core from site 1121 (Fig. 4), i.e. in the outermost part of the fiord. This is not consistent with the findings for the surface sediments and may suggest resuspension and redeposition of particles settled in the estuary of Vefsnfjord shortly after the Chernobyl accident. Sediments at site 1121 are slightly finer compared to the innermost part of Vefsnfjord (Fig. 2) and suggest that fine-grained particles are important carriers of <sup>137</sup>Cs. A higher input of marine organic matter in the outer part of the fjord as suggested by elevated TOC contents at site 1121 may also have caused scavenging of <sup>137</sup>Cs from the water column, and in this way contributed to higher levels in the sediments. The <sup>137</sup>Cs maximum-peaks in the cores from site 1121 occur at deeper level compared to other sites, indicating the highest sedimentation rates at site 1121. This is confirmed by results from <sup>210</sup>Pb-dating, which indicate a higher post-1986 sedimentation rate at this site compared to the other sites (Heldal et al., 2021).

# 3.2.1. Sediment-seawater distribution coefficient (Kd) and biological Concentration Factor (CF)

According to IAEA (2004), the recommended sediment-seawater distribution coefficient (Kd) (the concentration per unit mass of particulate (Bq  $kg^{-1}$  d.w.) divided by the concentration per unit volume of seawater ( $Bq L^{-1}$ )) for coastal and continental shelf environments for Cs is  $4 \cdot 10^3$ . A theoretical activity concentration in sediments from the Vefsnfjord of 400 Bq kg<sup>-1</sup> thus gives a seawater concentration of 0.1 Bq L<sup>-1</sup>. The Kd concept assumes equilibrium conditions. It is, however, not realistic to assume that <sup>137</sup>Cs levels in sediments in Vefsnfjord are in equilibrium with the overlying water masses, and the equilibriumderived seawater concentration is thus most likely overestimated. IAEA (2004) also recommends a biological Concentration Factor (CF) (the concentration per unit mass of organism (Bq  $kg^{-1}$  wet weight (w. w.)) divided by the concentration per unit volume of seawater (Bq  $L^{-1}$ )) of  $5 \cdot 10^1$ . A seawater concentration of 0.1 Bq L<sup>-1</sup> will give activity concentrations of  $^{137}$ Cs in fish of 5 Bq kg $^{-1}$  w.w. This is far below the maximum permitted level for radioactive cesium in food set by the Norwegian authorities after the Chernobyl accident (600 Bq  $kg^{-1}$  w.w.).

Analyses of surface seawater and fish collected in Vefsnfjord in 2018 show that the  $^{137}Cs$  levels are lower than calculated from Kd and CF values from IAEA. Surface seawater (5 m) collected from site 1124 contained 0.003 Bq L $^{-1}$  (3.0  $\pm$  0.4 Bq m $^{-3}$ ) and Greater argentine (*Argentina silus*) and Norway pout (*Trisopterus esmarkii*) contained 0.19  $\pm$  0.03 and 0.16  $\pm$  0.03 Bq kg $^{-1}$  w.w., respectively (IMR/DSA unpublished data). Any potential health risk caused by such  $^{137}Cs$  levels in seafood will be very low and should be of no concern to the consumer. A little more than one half-life has passed since the Chernobyl accident, and the past  $^{137}Cs$  activity concentrations in the Vefsnfjord water masses have probably been somewhat higher than today.

## 3.3. Potassium-40 (<sup>40</sup>K)

The natural radionuclide  $^{40}$ K is one of the major sources for radioactivity in the environment. Activity concentrations in surface sediments (0–2 cm) and sediment cores from the four sites in the Vefsnfjord are relatively uniform both spatially (across cores) and temporally (down core) (Table 2, Fig. 4 and SM 3). The profiles of the two cores collected from the same box corer agree well. The cores from site 1124 stands out a little, with the activity minimum occurring at 5 to 8 cm core depth, corresponding the slightly coarser sediment interval carrying less K-containing aluminosilicates. The <sup>40</sup>K-levels in the Vefsnfjord are comparable with levels found in the Norwegian Trench (Dowdall and Lepland, 2012; Helvik, 2019). Potassium-40 levels are several times higher than <sup>137</sup>Cs-levels.

The levels of <sup>40</sup>K in marine sediments depend on the bedrock in the catchment area and its mineralogical composition and grain size distribution. Artimieva et al. (2017) show that K is present in granitic rocks together with other radioactive elements including Th and U. The presence of granitic rocks in the catchment area implies that <sup>40</sup>K might be present. Ilus et al. (2007) report levels in Baltic Sea sediments ranging from approximately 300 to 1000 Bq kg<sup>-1</sup> d.w. Further, El-Reefy et al. (2010) report <sup>40</sup>K-levels in samples from different parts of the world ranging from approximately 10 to 1000 Bq kg<sup>-1</sup> d.w. As expected, levels in the Vefsnfjord fall in between these extremes. The minor variations that we observe in the Vefsnfjord are most likely due to local variations in sediment composition with higher activity associated with finer grained sediments containing more K-bearing aluminosilicate minerals.

## 3.4. Lead-210 (<sup>210</sup>Pb)

The activity concentrations of total <sup>210</sup>Pb in sediment cores from the four sites are presented in Fig. 4 and SM 3. See Heldal et al. (2021) for a detailed discussion of the temporal (down core) and spatial (across cores) distribution. Slightly higher values in the 0–1 cm and 1–2 cm layers at site 1123 in the middle of the inner fjord are consistent with a slightly finer grain size in these layers compared to 1122 and 1124 (SM 1). Variations in the temporal distribution of <sup>210</sup>Pb in the fjord are most likely related to variations in sediment accumulation rates over time. One factor that can affect this is variations in precipitation. Periods of heavy rainfall may increase the supply of <sup>210</sup>Pb from runoff from rivers (Appleby, 2001; Appleby et al., 2019). Fresh water supply varies throughout the year and may vary from year to year. Sedimentation rates at the four sites ranged from 0.042 to 0.25 g cm<sup>-2</sup> y<sup>-1</sup> (0.060 to 0.38 cm y<sup>-1</sup> (Table 3) (Heldal et al., 2021)).

The  $^{210}$ Pb-levels in the uppermost layers of the sediment cores are comparable to levels found in other Norwegian fjords including the Sognefjord and Laksefjord (Sværen, 2010).

## 3.5. Radium-226 (<sup>226</sup>Ra) and radium-228 (<sup>228</sup>Ra)

Activity concentrations of <sup>226</sup>Ra and <sup>228</sup>Ra in sediment cores from the four sites are relatively uniform both temporal and spatial (Table 2, Fig. 4 and SM 3), possibly indicating that sediment sources in the Vefsnfjord have remained relatively constant at least during the past 100 years or so (see details in Heldal et al., 2021). The two cores collected from the same box corer agree well. Some of the cores have elevated levels in the upper 0–4 cm layers, which may be due to recent changes at these sites. As for <sup>40</sup>K, levels compare with those found in the Norwegian Trench (Dowdall and Lepland, 2012; Helvik, 2019). Further, the <sup>226</sup>Ra levels compare to those found by Sværen (2010) in the Sognefjord and Laksefjord, but here, the levels do not change with increasing depth. The study by Sværen (2010) does not include analyses of <sup>228</sup>Ra.

As for  $^{40}$ K, the levels of radium isotopes in marine sediments are dependent on the geology of the catchment area. Radium-226 is part of the  $^{238}$ U decay chain while  $^{228}$ Ra is part of the  $^{232}$ Th decay chain. Catchment areas with bedrock rich in uranium and thorium thus will give higher supplies of radium isotopes to marine sediments. Acidic igneous rocks including granites and pegmatites as well as hydrothermally influenced rocks are important carriers of both uranium and thorium, hence elevated  $^{226}$ Ra and  $^{228}$ Ra activity concentrations are expected in marine sediments having provenance in such lithologies (Banks et al., 1995). Granitic rocks are present in the Vefsna drainage area. Ilus et al. (2007) report levels of  $^{226}$ Ra and  $^{228}$ Ra ( $^{232}$ Th) in Baltic

#### Table 3

Mean pre- and post-1986 sedimentation rates (g cm<sup>-2</sup> y<sup>-1</sup> and cm y<sup>-1</sup>). Data taken from Heldal et al. (2021).

Site	Sedimentation rates									
	Pre-1986		Post-1986							
	$\mathrm{g}~\mathrm{cm}^{-2}~\mathrm{y}^{-1}$	${\rm cm}~{\rm y}^{-1}$	$\mathrm{g}~\mathrm{cm}^{-2}~\mathrm{y}^{-1}$	${\rm cm}~{\rm y}^{-1}$						
1121	$0.042\pm0.005$	$\textbf{0.060} \pm \textbf{0.008}$	$\textbf{0.25} \pm \textbf{0.03}$	$\textbf{0.38} \pm \textbf{0.04}$						
1122	$0.12\pm0.01$	$0.13\pm0.02$	$\textbf{0.12} \pm \textbf{0.01}$	$0.17 \pm 0.02$						
1123	$0.11 \pm 0.02$	$0.12\pm0.03$	$\textbf{0.18} \pm \textbf{0.03}$	$0.25\pm0.04$						
1124	$\textbf{0.18} \pm \textbf{0.03}$	$\textbf{0.17} \pm \textbf{0.03}$	$\textbf{0.18} \pm \textbf{0.03}$	$0.21 \pm 0.03$						

Sea sediments ranging from 10 to 90 and 13 to 42 Bq kg<sup>-1</sup> d.w., respectively. Further, El-Reefy et al. (2010) report levels of <sup>226</sup>Ra and <sup>228</sup>Ra in samples from different parts of the world ranging from approximately 5 to 200 and 3 to 220 Bq kg<sup>-1</sup> d.w. As expected, levels in the Vefsnfjord fall in between these extremes. The minor variations that we observe in the Vefsnfjord are most likely due to local variations in sediment composition.

#### 4. Conclusions

- The Vefsnfjord is still heavily affected and still receives significant amounts of <sup>137</sup>Cs-contamination from the Chernobyl accident, more than 30 years after the accident. Activity concentrations of <sup>137</sup>Cs in 0–2 cm surface sediments ranged from 159 to 191 Bq kg<sup>-1</sup> dry weight (d.w.), which are orders of magnitude higher than in open Norwegian sea areas.
- Any potential health risk caused by <sup>137</sup>Cs contamination in the Vefsnfjord is very low and should be of no concern to consumers of seafood from the fjord.
- Levels of the natural occurring radionuclides <sup>40</sup>K, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra in the Vefsnfjord are comparable to levels found in other marine environments. <sup>40</sup>K exhibited the highest activity concentrations, ranging from 525 to 659 Bq kg<sup>-1</sup> d.w. in surface sediments.
- Levels of <sup>40</sup>K show good correspondence with sediment grain size with higher levels occurring in fine-grained sediments carrying K-containing aluminosilicates.
- Elevated levels of <sup>137</sup>Cs in the Vefsnfjord and surrounding catchment area makes this area well suited for studies of transfer processes for <sup>137</sup>Cs from terrestrial to coastal environments.

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### CRediT authorship contribution statement

This work is based on a Master project by Lena Helvik (LH). Hilde Elise Heldal (HEH) was LH's main supervisor and Hallvard Haanes (HH) was co-supervisor.

 $\operatorname{HEH}$  planned the study with input from Henning Jensen (HJ) and HH.

LH and HEH performed the field work.

LH prepared all samples and analysed them using gamma spectrometry with supervision from Andrey Volynkin (AV). LH interpreted the gamma results.

HJ and Aivo Lepland (AL) were responsible for analyses of grain size, total sulphur, total carbon, total organic carbon and calculation of carbonate content.

HEH took the lead in writing the manuscript. HJ and AL wrote chapter 2.2. AV wrote chapter 2.3. HH, HJ and AL contributed to the introduction, results and discussion. HJ and AL wrote the geology parts of the manuscript.

<u>All authors</u> provided critical feedback on the manuscript as a whole and helped shape the research and analyses.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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