**Detection of a hot 137Cs particle in marine sediments from Norway: Potential implication for 137Cs dating**

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**Abstract**

An anomalous value in the 137Cs record from a marine sediment core in Veafjord, Norway, appears to be due to the presence of a micron-size hot radioactive particle. The 137Cs concentration in a subsample from the surficial 0-1 cm sediment slice was over 3 times higher than in the adjoining slice, and double that in a concentration peak significantly deeper in the core dating from the period of Chernobyl fallout. To determine the cause of the anomaly, the sample was divided into two equal parts each of which were then reanalysed. Whereas one had a normal concentration similar to that of the adjoining slice, the concentration in the other was now over 5 times higher. Analyses of three further splits of the high activity subsamples followed a similar pattern with one part having a normal concentration, and the other a concentration that nearly doubled each time. The results are consistent with the presence of a hot 137Cs particle embedded in the sediment matrix. Its activity was estimated to be 15±2 mBq. Possible sources are fallout from the 1986 Chernobyl accident, or marine discharges from UK nuclear installations in the 1970s. The delayed input of such a particle into the sedimentary record highlights the need for care in using 137Cs records for dating sediment accumulations in areas open to contamination from these or similar sources.

Keywords: Marine sediments, geochronology, 137Cs, 210Pb, Chernobyl fallout, hot particles, radioactive contamination

**Declarations**

**Funding**

The study was funded by the Norwegian Public Roads Administration (Statens vegvesen, region vest).

**Conflicts of interest/Competing interests**

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.

**Availability of data and material**

All relevant data is included within the paper.

**Code availability**

Not applicable

**Introduction**

The artificial radionuclide 137Cs is commonly used as a chronological marker for dating lake and marine sediments. In north-west Europe the most common dateable features in 137Cs records from these natural archives are concentration peaks recording the 1963 fallout maximum from the atmospheric testing of thermonuclear weapons, and fallout from the 1986 Chernobyl reactor fire. Fallout from nuclear weapons tests occurred on a global scale and has been detected in all parts of the globe from the Arctic to the Antarctic. Fallout from Chernobyl was more regional and sporadic, and largely dependent on the amount of rainfall when the radioactive cloud was overhead. Results of a nationwide survey (Backe et al. 1986 & 1987) showed that significant levels of Chernobyl fallout occurred over many parts of Norway, including Veafjord in Vestland. However, routine analysis of a sediment core from Veafjord showed that the 137Cs record included a highly anomalous feature present within sediments deposited more than two decades after the Chernobyl accident. The objective of the present paper was to investigate the nature of this anomaly and its possible origins and implications. Although remobilisation of Chernobyl fallout from the surrounding region and redeposition in the fjord some decades later appears most likely, a second potential source of 137Cs in marine sediments along the Norwegian coast such as those in Veafjord is from discharges into the regional marine environment, most notably from Sellafield (Gray et al. 1995) and Dounreay (Dennis et al. 2007) in the UK. Discharges from Sellafield into the Irish Sea reached a very high level in the mid-1970s, and there is evidence that 137Cs from this source has been detected as far away as the Barents Sea (Kershaw and Baxter 1995).

**Physical setting**

In May 2020, the University of Oslo Department of Geosciences carried out a paleoecological study in some western Norwegian fjords using dated sediment cores with the purpose of investigating environmental changes over the last century. Sites from which cores were collected included Veafjord (Fig.1), one of three fjords bordering Osterøy, an island NE of Bergen in Vestland county. Veafjord is a sheltered inland fjord with a N-S orientation bordered by steep mountain slopes and with freshwater inflows. Its deepest basin has a water depth of 385 m. In the North, a shallow sill (water depth ~ 20m) separates Veafjord from the inner Osterfjord. In the South Veafjord is connected to Sørfjord by an approximately 330 m deep sill. Water exchange with coastal waters is limited and takes place through a number of sills and outlets of which the deepest (120 m) and westernmost connects with Hjeltefjord. The exchange is governed by tidal movements, the tidal range is about 1m, and on a more irregular basis by seasonal events and meteorological conditions. The mean water turnover time in the deepest basins of Veafjord is estimated to be between 4-7 years (Johnsen et al. 2017, Todt et al. 2017).



**Fig.1** Study site showing the island of Osterøy in Vestland, Norway, and the surrounding fjords Sørfjord, Osterfjord, and Veafjord. The core location in Veafjord is marked by the star.

**Methods**

**Sample collection and preparation**

The Veafjord core was collected on 7th May 2020 at 60°31′ N, 5°42.8′ E in 370 m water depth using a Gemini corer (80 mm inner diameter), a modified Niemistö corer (Niemistö, 1974). Inspection of the core immediately following retrieval showed that the sediment-water interface was intact and undisturbed, evidenced by the abundance of living benthic foraminifera and presence of Polychaeta tubes on the sediment surface. The sediment color changed from brown at the surface to grey at the base (22 cm). The core was sliced on board, at 1-cm intervals down to a depth of 20 cm, leaving a 2-cm thick slice at the base, and the samples stored on board in a freezer. On return to the laboratory, the samples were weighed before and after freeze-drying to calculate the water content, and the dried samples homogenized prior to subsampling for a range of different analyses.

The sediments consisted mainly of a fine silt. The sand content, measured by washing a ~3 g subsample over a 63µm sieve, varied from 12% in the surficial sediments to 17% at the base of the core. Subsamples of ~4 g of the dried sediment were sent to the Liverpool University Environmental Radioactivity Research Centre for radiometric analysis and dating by 210Pb and 137Cs. Other subsamples were used for geochemical (total organic carbon, 13C isotopes, total nitrogen, 15N isotopes, heavy metals) and micropaleontological analyses of benthic foraminifera. The methodological details and results of these analyses are reported in Hess et al. (2021).

**Radiometric analysis**

Sub-samples from selected slices were analysed by direct gamma assay using Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detectors (Appleby et al. 1986) in the Liverpool University Environmental Radioactivity Laboratory, preparatory to dating by 210Pb and 137Cs. 210Pb was determined via its gamma emissions at 46.5 keV, and 226Ra by the 295 keV and 352 keV γ-rays emitted by its daughter radionuclide 214Pb following 3 weeks storage in sealed containers to allow radioactive equilibration. 137Cs was measured by its emissions at 662 keV. The spectra were also scanned for the presence of other artificial gamma emitting radionuclides including 241Am via its emissions at 59.5 keV. Detection limits were ~8 mBq for 210Pb, ~5 mBq for 226Ra and ~2 mBq for 137Cs and 241Am. The absolute efficiencies of the detectors were determined using calibrated sources and sediment samples of known activity. Corrections were made for the effect of self-absorption of low energy γ-rays within the sample (Appleby et al. 1992). Supported 210Pb in each sample was assumed equal to the 226Ra activity, and unsupported 210Pb calculated by subtracting this from the total 210Pb activity.

**Results**

Fig. 2 plots concentrations of unsupported 210Pb and 137Cs activity versus depth in the core. Unsupported 210Pb concentrations decline more or less exponentially with depth, suggesting relatively uniform sedimentation rates during the period of time spanned by the 210Pb record. The mean sedimentation rate was calculated to be 0.16±0.02gcm-2 y-1 (0.27cmy-1). This places 1986 within the 8-9 cm slice and 1963 between 13-15 cm.

Excluding the surficial (0-1cm) slice, 137Cs concentrations have a broad but relatively distinct peak spanning three slices between 5-8cm, This feature almost certainly records fallout from 1986 Chernobyl accident. Although deposition around Osterøy (2.67 kBq m-2, Backe et al. 1986) was fairly modest much higher levels (11.6 kBq m-2) were recorded nearby in the area of Modalen. The small discrepancy with the 1986 depth suggested by the 210Pb calculations may be due to delayed inputs to Veafjord resulting from its long water residence time. Such discrepancies are not uncommon in marine environments (Heidal et al. 2021) where the relationship between atmospheric fallout and delivery to bottom sediments is more complex than in relatively enclosed systems such as lakes. Although there is no visible record of the nuclear weapons fallout maximum at depths dated by 210Pb to 1963, the absence of such a feature is not unusual due to the mobility of 137Cs in marine sediments (see e.g. Foster et al. 2006). This is a particular problem at sites subject to significant levels of Chernobyl fallout where the weapons test record can be masked by downwards migration of Chernobyl 137Cs.



**Fig.2** Fallout radionuclides in the Veafjord sediment core showing (a) unsupported 210Pb and (b) 137Cs concentrations versus core depth. Fig.2 (b) shows both the anomalous 137Cs value for the initial aliquot from the 0-1 cm sample, and the normal value from a second aliquot

The maximum 137Cs concentration in the Chernobyl peak (7.4 Bq kg-1) is significantly lower than an apparently anomalous value (15.6 Bq kg-1) recorded in the surficial (0-1 cm) sediment slice. As a check on the reliability of this measurement. a second aliquot from the 0-1 cm slice was also analysed for 210Pb and 137Cs. This had a similar 210Pb concentration to the first but a significantly lower 137Cs concentration (4.3 Bq kg-1), comparable to that in the adjoining 1-2 cm slice. Given that each slice had been homogenized after drying, although a number of different biogeochemical processes, including for example preferential absorption onto fine-grained clays, can result in significant levels of 137Cs enrichment, they cannot account for the more than three-fold difference between these two aliquots. In order to determine the cause of the difference, the sample with the anomalously high concentration was divided into two parts both of which were reanalysed. The results showed that they again had comparable 210Pb activities but distinctly different 137Cs activities. One had a 137Cs concentration comparable to the low activity sample from the first partition but the second an even higher anomalous value (23.7 Bq kg-1). This process, a relatively standard method for determining the presence of hot particles (see e.g. Falk 1988, Pöllänen 1999), was repeated several times, and each time one of the splits had a normal 137Cs concentration typical of the matrix, and the other an increasingly high concentration.

**Table 1** 210Pband137Cs concentrations in the matrix and successive splits of the 0-1 cm slice containing the hot particle, and total 137Cs activities in the splits attributable to the matrix and the hot particle. 210Pb activity in the 5th split was below the minimum detectable activity (MDA)

|  |  |  |  |
| --- | --- | --- | --- |
| Sub Sample | Mass | Specific activities | Total 137Cs Activity |
| 210Pb | 137Cs | Matrix | Hot particle |
|  | g | Bq kg-1 | ± | Bq kg-1 | ± | mBq | ± | mBq | ± |
| Matrix |  | 125.5 | 4.9 | 4.3 | 0.9 |  |  |  |  |
| Split 1 | 1.583 | 127.8 | 6.5 | 15.6 | 0.9 | 6.8 | 1.5 | 18.0 | 2.1 |
| Split 2 | 0.779 | 144.6 | 12.3 | 23.7 | 2.1 | 3.3 | 0.7 | 15.1 | 1.8 |
| Split 3 | 0.399 | 135.8 | 20.3 | 35.4 | 4.3 | 1.7 | 0.4 | 12.4 | 1.8 |
| Split 4 | 0.229 | 100.4 | 21.9 | 71.6 | 7.2 | 1.0 | 0.2 | 15.4 | 1.7 |
| Split 5 | 0.124 | Below MDA | 118.1 | 12.9 | 0.5 | 0.1 | 14.1 | 1.6 |
|  |  |  |  |  |  | Mean value | 15.0 | 2.0 |

It appears from these results (shown in Table 1) that the sediment matrix in the 0-1 cm slice has relatively uniform 210Pb (125.5±4.9 Bq kg-1) and 137Cs (4.3±0.9 Bq kg-1) concentrations. Further, since the anomalous 137Cs concentrations nearly doubled each time, the activity in the “hot” splits is mainly due to the presence of a single hot particle. To test this hypothesis, for each of the hot splits the activity of the presumed hot particle was calculated by separating the total activity (mBq) into two parts, the activity due to the matrix, and that due to the hot particle. The activity due to the matrix was calculated by multiplying the sample mass by the 137Cs concentration in the first low activity split. Estimates of the activity of the hot particle determined by these calculation (also shown in Table 1) range from 12.4–18.0 mBq, with a mean value of 15±2 mBq.

In order to determine whether the particle had any other distinguishing radiometric properties, the gamma spectra were in each case examined for the presence of other natural and artificial radionuclides. There were no significant differences between 238U activities in samples containing the hot particle from those containing just the sediment matrix. Differences in 226Ra activities were also insignificant. In all samples, including those containing the hot particle, 241Am activities were below detection limits.

**Discussion and conclusions**

Since the sheltered location of Veafjord and limited water exchange with the open sea would appear to reduce the possibility of a marine source, the most likely origin of the Veafjord particle is fallout from the 1986 Chernobyl reactor fire. Deposition of radioactive particles from the Chernobyl cloud during its passage over Sweden in late April and early May 1986 was reported in Persson et al. (1987). Although many were smaller than 1 m there was a significant number of larger particles with diameters of 5 m or more. A number of these particles were detected in Swedish soil samples collected in 1987, a year after the accident (Falk et al. 1988). Some, composed mainly of uranium oxide, appeared to be fuel fragments from the damaged reactor, though their activity derived mainly from a range of fission products typically found in reactor fuel, together with some transuranic elements including 241Am. These particles (designated Group 1) had total activities (decay corrected to 1986) ranging from 80-350 Bq. Other particles (designated Group 2) were much more active, mainly due to high levels of 103Ru and 106Ru (up to 40,000 Bq), though with small amounts of other fission products, including in some cases 137Cs. Particle sizes ranged from 2.4-11.5 m. Pöllänen et al. (1999) reported the presence of what was most probably a fuel fragment in marine sediments collected in 1995 from Gotland Deep in the Baltic Sea. This particle had a total activity (decay corrected to 1986) of 141 Bq and diameter of around 11 m. Radionuclides present included 137Cs and 241Am as well as a number of short-lived fission products.

By 2020 all of the short-lived fission products in these particles apart from 137Cs would have decayed to values close to or well below limits of detection. Residual 137Cs activities would have fallen to values comparable to but a little higher than that of the Veaford particle, 61 mBq in the case of the Gotland Deep particle, and 181 mBq in the case of the 9 m Haga1 particle reported in Falk et al. (1988). Assuming a similar origin and composition, calculations suggest that the Veafjord particle would have a diameter of between 4-7 m. The absence of 241Am suggests that it might be a Group 2 particle.

Although Chernobyl is the most likely source, discharges of 137Cs into the marine environment from nuclear installations in the UK cannot be excluded. Discharges from the Sellafield plant on the Irish Sea coast increased dramatically in the 1970s and remained at a high level through to the early 1980s (Gray et al. 1995). Whilst the main impact was on estuaries around the Irish Sea (Oldfield et al. 1993), a significant fraction was transported north towards the Arctic via the Norwegian Coastal Current (Kershaw et al. 1995) and would thus have been a potential source of 137Cs in marine sediments along the Norwegian coast. Transit times to western Norway were estimated to be 3-4 years. A second potential marine source is the Dounreay nuclear complex on the north coast of Scotland. A significant number of hot radioactive particles were released from this facility into the North Atlantic Ocean during the mid to late 1960s and early 1970s (Dennis et al. 2007). Particles collected from the nearby shores and seabed ranged in size from sub-millimetre to several millimetres, with 137Cs activities ranging from 105 Bq to more than 107 Bq. Based on these data, micron size particles (discharged directly or created by fragmentation of larger particles) are likely to have activities of no more than a few mBq. Having settling velocities of at most a few cm per day these particles are capable of long-distance transport within the water column.

Whatever the origin, the winnowing effects of transport by atmospheric and/or aquatic pathways are such that the Veafjord particle is almost certainly of micron size. However, regardless of size or origin what is most significant is its presence in the surficial and hence contemporary (2020) sediments of the fjord, decades after injection into the environment. Its presence in these sediments shows that it must have been transported to this site relatively recently.

Features in 137Cs records are widely used as chronostratigraphic markers for dating environmental records in these natural archives. Their value lies not just in the 137Cs dates themselves, but perhaps more importantly in their use for validating 210Pb dates, and correcting those dates when there are significant discrepancies between these two independent dating methods (Heldal et al. 2021). The Veafjord particle was detected largely by chance, and mainly due to the fact that it stood out as an outlier because of its presence in the surficial sediments. Had it been elsewhere in the core its true nature may have gone undetected. Since such particles can have a major impact on the 137Cs record, this finding highlights the need to interpret features in these records with care, particularly when they are used for dating purposes. This is particularly relevant to marine sediments where concentrations are generally lower than in lacustrine environments. Samples containing potentially anomalous features should ideally be split and reanalysed to determine the presence of any significant level of inhomogeneity.

**Acknowledgments**

The authors thank the crew of ‘MS Solvik’, Elisabeth Alve and Aud Helland for their help with field work and sampling. Aud Helland (formerly Rambøll Norway, now COWI Norway) initiated the project. We thank Rambøll Norway (Rambøll Norge) for their constructive cooperation throughout the project. The Norwegian Public Roads Administration (Statens vegvesen region vest) funded the study.

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