

Detection of technetium-99 in *Ascophyllum nodosum* from around the Welsh coast

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Abstract

The presence of the radionuclide ⁹⁹Tc in the marine environment is of concern to environmental scientists because of its conservative nature and high concentration factor in commercially valuable species. The brown seaweed *Ascophyllum nodosum* (Linnaeus) Le Jolis was used to biomonitor the spatial distribution of ⁹⁹Tc around the Welsh coast, an area relatively unstudied with respect to this isotope. Over the course of a year an inverse relationship was observed between the ⁹⁹Tc concentration in *A. nodosum* samples and approximate straight-line distance from Sellafield. These data show that detectable levels of a Sellafield derived radionuclide are reaching the Welsh coast despite the overall northward movement of the Sellafield plume.

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1. Introduction

The increased presence of technetium-99 (⁹⁹Tc) in the marine environment following changes in the operations at European nuclear facilities is of interest to environmental scientists for a number of reasons. ⁹⁹Tc has a high bio-concentration factor in commercially valuable species such as the common lobster, *Homarus gammarus* (Linnaeus) and the Norway lobster, *Nephrops norvegicus* (Linnaeus) that under certain circumstances could be of concern to consumers (Busby et al., 1997; Leonard et al., 2001; Olsen and Batlle, 2003; Coppleson et al., 2004). ⁹⁹Tc has a half-life of 213000 years and in the marine environment forms the soluble pertechnetate ion, TcO₄⁻, which means that once ⁹⁹Tc enters the marine environment it is highly conservative and can be carried long distances from its point of entry (Aarkrog et al., 1988; Bonotto et al., 1988;

McDonald and Busby, 1998; Brown et al., 1999; Kershaw et al., 1999, 2004; Kanisch et al., 2000; Lindahl et al., 2003).

The safe discharge of contaminants such as ⁹⁹Tc into the sea requires an adequate understanding of their fate in the marine environment, especially if there is any possibility of feedback to humans (Bourne and Assinder, 1991). One method employed in many investigations into the biological implications of releasing contaminants into the environment is that of biological monitoring. It is well documented that both *Fucus vesiculosus* (Linnaeus) and *A. nodosum* (Linnaeus) Le Jolis accumulate ⁹⁹Tc to a high degree and are useful indicators of the distribution of this element in the marine environment (Mitchell et al., 1989; Masson et al., 1995; Smith et al., 2001; Ilus et al., 2002). Brown seaweeds (mainly *F. vesiculosus*) are sampled by both the nuclear industry and government agencies such as the Food Standards Agency (FSA) as part of their annual reviews of the impact of Britain's nuclear industry on the environment.

The primary aim of this study was to map the distribution of ⁹⁹Tc around the Welsh coast using the brown seaweed *A. nodosum* as a biomonitor. This is an area that is

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Table 1
Locations, names and sampling frequencies of study sites

Site ID	Site Name	Latitude (°N)	Longitude (°E)	Distance from Sellafield	Sampling Frequency
1	Beachley	51.6141	−2.6477	630	Twice monthly
2	Porthcawl	51.4774	−3.6686	550	Twice monthly
3	Manorbier	51.6430	−4.8061	470	Twice monthly
4	Fishguard	51.9997	−4.9867	400	Twice monthly
5	Aberystwyth	52.4157	−4.0890	320	Twice monthly
6	Porthmadog	52.9145	−4.1356	260	Twice monthly
7	Nefyn	52.9408	−4.5361	200	Twice monthly
8	Cemaes Bay	53.4147	−4.4527	180	Twice monthly
A	Ramsey	54.3118	−4.3624	130	Once
B	Castletown	54.0717	−4.6543	90	Once
C	Walney Island	54.0816	−3.2476	50	Once
D	Bardsey Island	52.7531	−4.7936	45	Once

relatively unstudied with respect to ^{99}Tc despite the proximity of Welsh coastal waters to the principal source of this isotope in the NE Atlantic, the BNFL nuclear reprocessing facility at Sellafield. The commissioning of the thermal oxide reprocessing plant (THORP) and enhanced actinide removal plant (EARP) at Sellafield in 1994 led to a 20-fold increase in the discharges of ^{99}Tc into the Irish Sea. This was partly due to the processing of the backlog of irradiated fuel that had accumulated while THORP and EARP were built; the amounts discharged fell from 190 to 69 TBq per annum between 1995 and 1999 (McCartney and Rajendran, 1998; BNFL, 2002). Discharges increased to 85 TBq in 2002 but fell to 37 TBq in 2003 and new technology introduced by BNFL should continue to decrease discharge of ^{99}Tc to 90% of the 2002 levels in years to come (RIFE 9, 2004).

The geographical distribution of ^{99}Tc has been well documented for the waters immediately surrounding the Sellafield pipeline, the north Atlantic, North Sea and even as far north as the Arctic (Kershaw and Baxter, 1995; McDonald and Busby, 1998; Brown et al., 1999; Kershaw et al., 1999; Lindahl et al., 2003; Kershaw et al., 2004) which reflects the fact that the prevailing currents influencing the waters of the Irish Sea move from south to north (Leonard et al., 2001). There is, however, a certain amount of southward water movement in the Irish Sea (Bailly du Bois and Guegueniat, 1999), which enters the Irish Sea to the north of Anglesey and moves anti-clockwise around the Isle of Man before rejoining the main flow to exit through the North Channel, which means that any radionuclides released from the Sellafield pipeline could potentially have an effect on the coast of Wales. This is borne out by the fact that the FSA has found detectable levels of ^{99}Tc in marine biota from several sites around the Welsh coast (RIFE 9, 2004).

2. Study sites

In the period from August 1999 to June 2000 eight sites around the Welsh coast were visited at two-monthly intervals, from the Severn Estuary in the south, to the northern

tip of Anglesey (see Table 1 and Fig. 1). Sites 4, 6 and 8 were selected due to their being sampled by other monitoring programs (BNFL, 2002; RIFE 9, 2004). In order to place the results from this study into a wider context, the ^{99}Tc content of the samples collected from the two-monthly study sites were compared to those found in plants collected on a 'one-off' basis from a number of other sites including two on the Isle of Man.

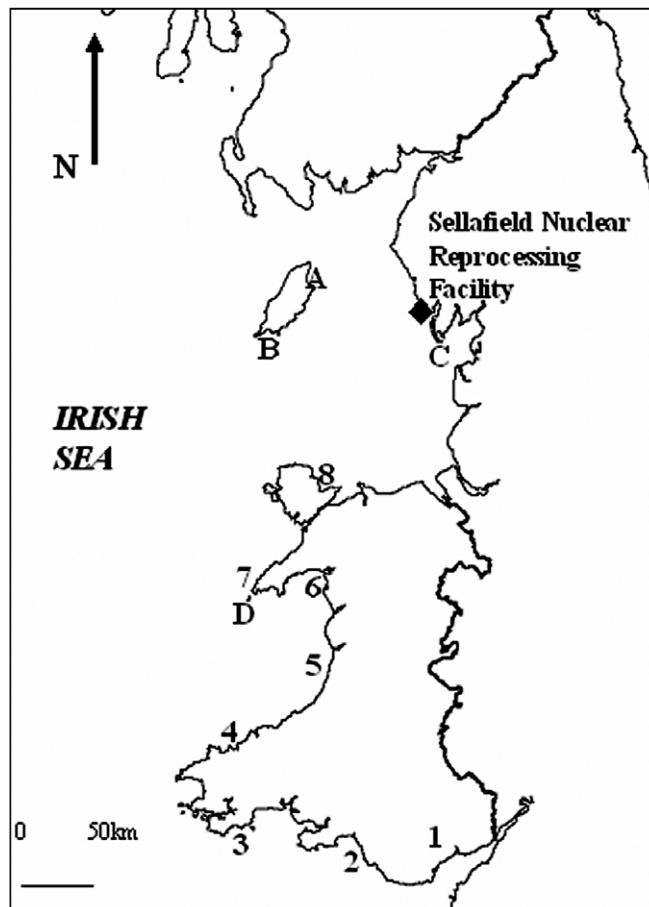


Fig. 1. Map showing location of study sites (map reproduced with kind permission of the Ordnance Survey).

3. Methods

3.1. Sample collection and pre-treatment

Samples of *A. nodosum* were collected at two-monthly intervals from each study site, from the same position on the shore each time. Approximately 1 kg (wet wt.) of seaweed was collected on each occasion from 4 or more individual plants that were within 1 m² of each other. The samples were placed in clean plastic bags and transported back to the laboratory where all visible epiphytes and fauna were removed. Seaweed samples were washed twice in tap water and once in Milli-Q 18 M Ω cm⁻¹ (MQ) H₂O water then dried to constant weight at 60 °C. Samples were then homogenised by passing them through an IKA Technologies MF 10 grinding mill at 4500 rpm using a 2 mm mesh, ensuring that the mill was thoroughly cleaned between samples.

3.2. Sample digestion

Twenty grams of each dried and homogenised seaweed sample were then subjected to a combination of dry ashing followed by a period of wet oxidation as recommended by Wigley et al. (1999). This was necessary to reduce the volume of sample and to separate the inorganic elements present from the large amounts of organic matrix. The need for complete sample digestion was particularly important, as all analytes were required to be present as free ions, or ion complexes, for the ion exchange procedures that were performed prior to analysis and it was thought that this digestion method allowed a large enough sample to be used to ensure that any ⁹⁹Tc present would be detected, without causing considerable loss of technetium (Momoshima et al., 1991; Ihsanullah, 1993; Butterworth et al., 1995; Bettinelli et al., 2002). A procedural blank was treated in exactly the same way as the seaweed samples and any ⁹⁹Tc detected in the blank was subtracted from the sample concentrations.

3.3. Ion exchange separation

The next stage of the sample preparation procedure involved the separation of any ⁹⁹Tc present from the isobaric interference of Ru and Mo (Ihsanullah, 1994; McCartney et al., 1999; Tagami and Uchida, 1999; Mas et al., 2002). However, due to the assumption that ⁹⁹Tc would be present in some of the sample solutions in amounts that were at or near the detection limits of the ICP-MS, it was also important that the Tc was also separated from the greater part of the sample matrix as recommended by Butterworth et al. (1995) and McCartney et al. (1999). The removal of potentially interfering elements was achieved by passing the sample solutions through two stages of ion exchange chromatography. First an anion exchange resin (Amberlite IRA-400) was used to separate the positively charged cationic elements from any elements

which would be present as anions or oxy-anions following the strongly oxidising digestion e.g. Tc, Mo and trace amounts of Ru and Re (see Butterworth et al., 1995). This initial separation was followed by a fine-grained, analytical grade resin (Teva Spec[®], supplied by Eichrom Industries Inc.) to separate the Tc from the remaining traces of Ru and to a lesser extent Mo. This method of separation has been used in the analysis of technetium from a range of environmental samples including seaweed, water and soil (Beals, 1992; Butterworth et al., 1995; Tagami and Uchida, 1999).

In the early stages of this work experiments were conducted to test the ion exchange resins using a series of synthetic mixed element solutions prepared from Aldrich[®] single element standard solutions (1000 mg l⁻¹) for Mo, Ru and Re and from a Tc standard solution from Nycomed Amersham. The tests confirmed that the Amberlite IRA 400 resin produced a first stage separation of pertechnetate (TcO₄⁻) and perrhenate (ReO₄⁻) from the interfering elements Mo and Ru. However, analysis showed that once this first stage separation had taken place Mo and Ru were still present in the test solutions at concentrations which would cause interferences at the low environmental levels of Tc. Tests performed on the second resin (Teva Spec[®]) showed that Mo and Ru were effectively eluted from the resin with 0.5 M HNO₃ whilst Tc and Re was quantitatively retained. Recovery of Tc and Re was better than 99% when 20 ml of 6 M HNO₃ was used to elute these elements.

3.4. Analysis of samples

All samples in this study were analysed using ICP-MS, a method of analysis that has been used for the detection of ⁹⁹Tc in a number of environmental materials (Beals, 1992; Shigemitsu et al., 1993; Butterworth et al., 1995; Tagami and Uchida, 1999; Copplestone et al., 2004). The ICP-MS used in this study was a VG Plasma Quad PQII+ with a deGallen V groove nebuliser. The instrument was tuned with a solution of ¹⁰³Rh to give optimum sensitivity in the region of the ⁹⁹Tc mass without adding significantly to the background. The sample solutions did not contain rhodium after the ion-exchange treatment and it does not interfere with the measurement of technetium so rhodium was chosen as the internal standard for this method. A water blank (a solution of MQ H₂O and 2% HNO₃) was analysed with each set of samples to give a measure of the background level of elements in the ion source. A standard solution consisting of 100 ng ml⁻¹ of ⁹⁹Tc in 2% HNO₃ was also analysed to calibrate the instrument at the start and end of the sample run and at several intervals during the sample analysis, which allowed a drift correction to be made if any instrumental drift had occurred during the sample run. An acid blank consisting of 8 N HNO₃ (the solvent that the samples were dissolved in) was run to ascertain the background levels of elements contained in the acid. The samples were analysed with the

instrument in peak jumping mode to improve the counting statistics.

3.5. Quality assurance

At the time of this study there were no readily available reference materials for Tc to validate the analytical method against an accredited reference material. Synthetic samples were subjected to the full digestion and separation procedures to test for loss of Tc. These studies showed that recovery from synthetic solutions was better than 95%. Typical precision measurements for analyses on replicates of the seaweed samples range from 2.6% RSD at 6701 Bq kg⁻¹ to 8.0% RSD at 770 Bq kg⁻¹.

4. Results

Due to a problem with access during February 2000, no sample was collected from site 7 at this time. All distances quoted are approximate straight-line distances, by open sea, in km.

The spatial distribution of ⁹⁹Tc around the main study area as recorded on each two-monthly sampling trip is shown in Fig. 2 and Table 2. The data show that although the concentration of ⁹⁹Tc recorded at each site varied over the study period, the general trend was for the ⁹⁹Tc concentration to decrease from the north to the south of the study area. It also shows that ⁹⁹Tc was detectable in seaweed samples from every sample site even sites 1, 2 and 3, which are between 470 and 630 km from the point of entry of ⁹⁹Tc into the Irish Sea, the Sellafield marine pipeline.

The data also show that ⁹⁹Tc concentrations varied more throughout the year at sites 1–4 than was the case at sites 5–8. This may indicate that the ⁹⁹Tc concentration in the seawater south of site 5 was more variable throughout the study period particularly at sites 1, 2 and 3, which

are influenced by the Bristol Channel as well as the Irish Sea. It is more probable, however, that the data reflect the relationship between concentration and analytical reproducibility and therefore the lower ⁹⁹Tc concentrations of the samples in the south have a greater margin of error.

The general pattern of spatial distribution of ⁹⁹Tc around the study area shown in Fig. 2 is of the highest ⁹⁹Tc concentrations at the most northerly site (site 8) and the lowest at the southerly sites (i.e. those sites located to the south of site 5), with the ⁹⁹Tc concentration getting progressively greater between sites 4 and 8. The only exception to this pattern was site 6 which had a lower mean ⁹⁹Tc concentration than site 5 at every sampling time except for August 1999 (see Table 2). This may be a result of the fact that site 6 is relatively sheltered from the plume of water coming from the northern Irish Sea by the Lley Peninsula, which forces the southerly moving water outwards as it comes into Cardigan Bay.

Comparison of the site means in Table 2 also shows that the ⁹⁹Tc concentration changed over the study area by two orders of magnitude from the most northerly site (site 8) to the two most southerly sites (sites 1 and 2). This indicates that the amount of ⁹⁹Tc found in seaweed samples decreased by approximately one order of magnitude every 160 km moving from the north to the south of the study area. This pattern was further developed when the results of the ⁹⁹Tc analysis of the 'one off' samples taken from other parts of the UK, were compared with the mean study site data (see Table 2 and Fig. 3). The highest ⁹⁹Tc concentration was recorded at Walney Island (site C), which was the closest site to the Sellafield pipeline. The concentration measured here was also one order of magnitude higher than the average concentration found at site 7, which is approximately 160 km to the south. The ⁹⁹Tc concentration recorded at site D filled in the gap between sites 6 and 7 and followed the general pattern of decreasing ⁹⁹Tc concen-

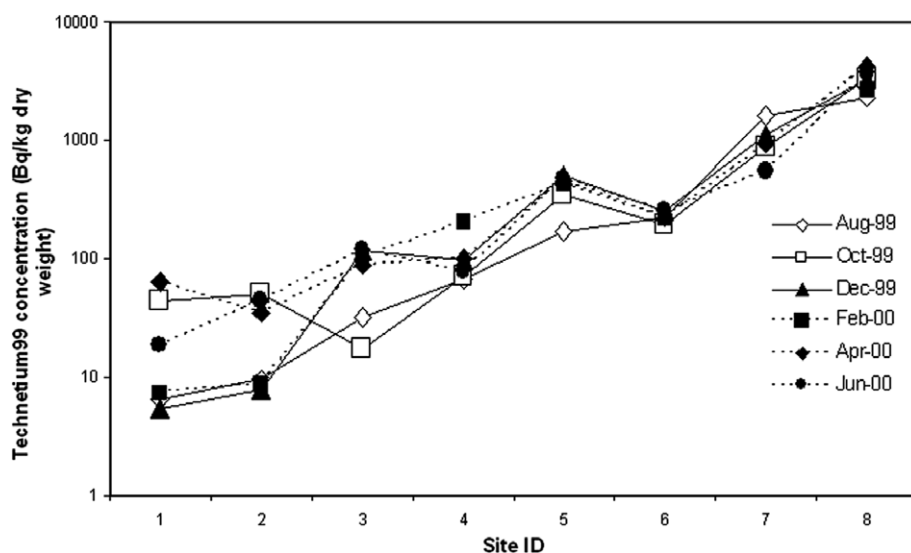


Fig. 2. Results of ⁹⁹Tc analysis of *A. nodosum* samples collected during the study period (⁹⁹Tc concentration in Bq kg⁻¹ dry wt.). The data are plotted on a log₁₀ scale so that all 8 sites can be included on the same graph.

Table 2

Results of ^{99}Tc analysis of *A. nodosum* samples collected during the study period (^{99}Tc concentration in Bq kg^{-1} dry wt.)

Sample site ID	August 1999	October 1999	December 1999	February 2000	April 2000	June 2000	Mean	Standard deviation
1	6.5	44.6	5.5	7.4	64.2	18.6	24.5	24.4
2	9.8	50.2	7.8	9	35.6	44.3	26.3	19.5
3	31.7	17.9	115	103	89.3	120	79.6	43.9
4	68.7	72.8	98.2	204	101	79.8	104	50.6
5	171	354	496	437	466	475	400	123
6	222	191	254	223	225	252	229	25.7
7	1614	891	1140		936	558	1028	388
8	2311	3337	3294	2653	558	3761	3262	699
A			4356					
B			4125					
C			11776					
D						523		

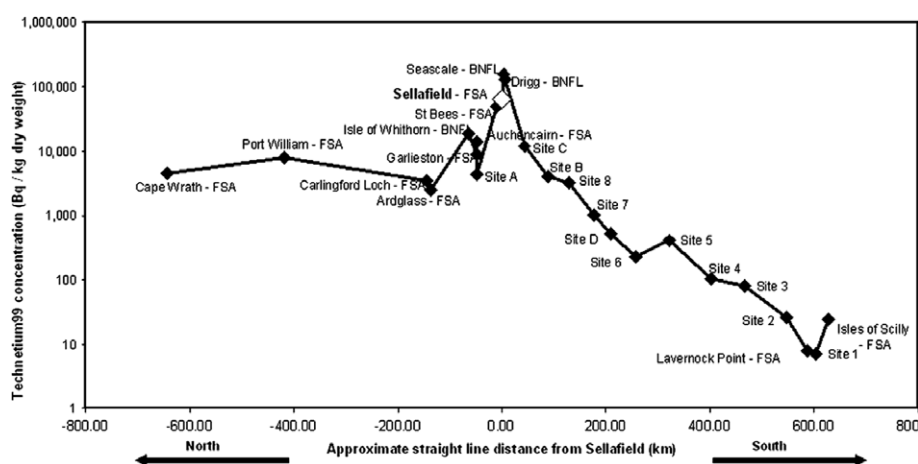


Fig. 3. Graphical representation of the ^{99}Tc concentrations (in Bq kg^{-1} dry wt.) at all sites sampled during 1999 and 2000 by this study (mean values for sites visited more than once) as compared to those gathered by BNFL and the FSA in 2000 plotted against approximate straight-line distance from Sellafield by open sea (after McDonald and Busby, 1998).

tration with increased distance from Sellafield by falling in between the average concentrations of sites 6 and 7.

5. Discussion

There are few published works regarding the spatial distribution of ^{99}Tc around the UK in the period from 1999 to 2000 and the only sources of data with which to directly compare the results gathered during this study are the annual monitoring data published by BNFL and the FSA. It is important to note, however, that the FSA and BNFL sampled *F. vesiculosus* at the majority of sites, which limits the comparability of their data with the data from this study. Also, the results published by BNFL and the FSA are expressed in terms of ^{99}Tc concentration in Bq kg^{-1} wet weight of seaweed, whereas those in this study are expressed in terms of Bq kg^{-1} dry weight of seaweed. The second of these issues has been addressed by conversion of the wet weight results to approximate dry weight ones by multiplying the wet weight results by five as suggested by BNFL (2002) and Kershaw et al. (1999).

Surface seawater concentrations of ^{99}Tc recorded from around the Irish Sea in 1998 as part of a longer-term study by Leonard et al. (2001) fit with the results of this study by showing decreasing concentrations of ^{99}Tc from Anglesey in the north to Fishguard in the south. Concentrations ranged from 40 mBq l^{-1} at the point of discharge, $20\text{--}25 \text{ mBq l}^{-1}$ in the North Channel, $10\text{--}18 \text{ mBq l}^{-1}$ around the north and west coasts of Anglesey, 2.8 mBq l^{-1} off the south coast of the Llyn Peninsula, 1.1 mBq l^{-1} near Porthmadog, 0.7 mBq l^{-1} off Aberystwyth and 0.5 mBq l^{-1} off Fishguard.

Comparison of the results from sites sampled during this study and by the FSA during the same period, i.e. Cemaes Bay (site 8), Porthmadog (site 6) and Fishguard (site 4) shows that at site 8 and site 6, the average ^{99}Tc concentrations found in *A. nodosum* were higher than those found in *F. vesiculosus*, whereas at Site 4 the opposite was the case. The ratios between the *F. vesiculosus* and *A. nodosum* data for Site 8 and Site 6 are approximately 1.2 and 1.8 respectively, indicating that the difference in the ^{99}Tc content of the two species is not consistent, although it appears that

A. nodosum generally has a higher concentration than *F. vesiculosus*.

Fig. 3 shows how the results from this study and those gathered by BNFL and the FSA relate to approximate distance from Sellafield. Samples from Seascale and Drigg have the highest concentrations with levels decreasing with distance from Sellafield, a pattern also shown by McDonald and Busby (1998). The wider range of sampling carried out to the south of Sellafield in this study allows a much better estimate to be made of the rate of decline in concentration with distance than was possible in the work of McDonald and Busby (1998), which showed a symmetrical decrease in the ^{99}Tc concentration of seaweed up to 140 km north and south of Sellafield. Fig. 3 clearly demonstrates that beyond 140 km the relationship is not symmetrical. The samples to the south show a more rapid decline in concentration with distance than those to the north. This reflects the general movement of water in the Irish Sea from south to north. Fig. 3 shows that the rate of decrease in ^{99}Tc concentration from Seascale to Cemaes Bay, which is located approximately 130 km to the south, is $1204 \text{ Bq kg}^{-1} \text{ km}^{-1}$. This is comparable to the rate of decrease from Seascale to Carlingford Loch, located approximately 140 km to the north, which is $1075 \text{ Bq kg}^{-1} \text{ km}^{-1}$. However, the rate of decrease from Cemaes Bay to the Isles of Scilly ($6.8 \text{ Bq kg}^{-1} \text{ km}^{-1}$) is more than three times greater than the rate of decrease between Carlingford Loch and Cape Wrath ($2.1 \text{ Bq kg}^{-1} \text{ km}^{-1}$) resulting in seaweed concentrations at Cape Wrath and the Isles of Scilly, which are both approximately 650 km from Sellafield, differing by more than two orders of magnitude. In addition to this, Lindahl et al. (2003) found a ^{99}Tc concentration of 200 Bq kg^{-1} dry wt. in *F. serratus* collected from Sirdal in Sweden, which is approximately 1800 km north of Sellafield, a comparable concentration to that found at Site 6 in this study, which is approximately 260 km south of Sellafield.

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