Helgoländer wiss. Meeresunters. 17, 280–290 (1968)

Fission-product radionuclides in sediments from the North-East Irish Sea

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KURZFASSUNG: **Spaltprodukt-Radionuklide in Sedimenten der nordöstlichen Irischen See.** Als Teil einer Untersuchung über die Verbreitung von radioaktivem Abwasser, das von der radiochemischen Aufarbeitungsanlage in Windscale stammt, wurden Messungen der Gamma-Dosisleistung auf Schlickböden in Ästuaren vorgenommen. Es zeigte sich, daß die Dosisleistungen hier mindestens um eine Größenordnung höher sind als die, welche auf reinem Sandboden gemessen wurden. Oberflächensedimente, die hinsichtlich ihrer Radioaktivität untersucht wurden, enthalten als wichtigste Radionuklide die Spaltprodukte $^{95}Zr + {}^{95}Nb$, ^{106}Ru, ^{144}Ce und ^{137}Cs. Die Konzentrationen der einzelnen Radionuklide stehen in Beziehung zur Abwassermenge. Probebohrungen in Sedimenten zeigten, daß sich die Konzentration der Radioaktivität exponentiell mit der Tiefe ändert. Der Anreicherungsfaktor von $^{95}Zr + {}^{95}Nb$, ^{106}Ru in Oberflächensedimenten von Ästuaren beträgt ungefähr $1{,}5 \times 10^4$, für ^{137}Cs dagegen $1{,}0 \times 10^3$. Die Konzentrationen von $^{95}Zr + {}^{95}Nb$, ^{106}Ru und ^{144}Ce in Oberflächensedimenten sind abhängig vom Abstand der Abwassereinleitung bei Windscale.

INTRODUCTION

Large volume, low-activity aqueous radioactive wastes are discharged to the Irish Sea from the premises of the United Kingdom Atomic Energy Authority at Windscale, Cumberland. These discharges are controlled through authorizations issued jointly by the Minister of Housing and Local Government (M.H.L.G.) and the Minister of Agriculture, Fisheries and Food (M.A.F.F.). A programme of environmental monitoring is conducted on a regular basis by the U.K.A.E.A. and M.A.F.F., to provide data for the estimation of internal and external radiation exposure of members of the public affected by the discharges.

Measurements of the external gamma radiation dose-rate are made by M.A.F.F., on behalf of M.H.L.G., over beach sand and mud flats in the intertidal zone within a few miles of the discharge point. Sand and silt samples are taken for radiochemical analysis, to determine the principal radionuclides contributing to the external radiation dose.

AQUEOUS RADIOACTIVE WASTE DISCHARGES

Liquid wastes are discharged to sea by means of two parallel pipelines, 1.5 miles long, and 60 feet below the sea surface at the discharge point. Details of the isotopic composition of the effluent have been given by HOWELLS (1966). Throughout the period 1955 to 1963 the effluent was characterized by a high percentage of ^{106}Ru. The introduction of a new radiochemical re-processing plant in 1964 has resulted in a slight decrease of ^{106} Ru and a sharp increase in the amounts of ^{95}Zr and ^{95}Nb.

ENVIRONMENTAL MONITORING

An extensive sampling programme in this area has been conducted by the U.K.A.E.A. and M.A.F.F. for more than a decade (LONGLEY & TEMPLETON [1963], DUNSTER et al. [1964], HOWELLS [1966], PRESTON & JEFFERIES [1967]). Samples of seaweed, fish, shellfish, seabed, seawater, shore sand and silt are collected regularly from areas up to 25 miles north to 25 miles south of the site and analysed for radioactivity content.

The main objective of the monitoring programme has been to ensure that the radiation exposure of individual members of the public who are affected by the release of radioactivity to the marine environment is less than that regarded as acceptable by the INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (1966).

The route back to man that results in the greatest degree of human exposure to radiation from this site has been shown to be the reconcentration of discharged fission product radionuclides, principally ^{106}Ru, by the edible seaweed *Porphyra*, which is used in the manufacture of the foodstuff laverbread (PRESTON & JEFFERIES 1967). Ruthenium-106 and *Porphyra* are the critical radionuclide and material respectively.

Monitoring of sub-critical materials at Windscale can provide data that may be applicable to the evaluation of other sites where different conditions of human exposure to radiation may obtain.

EXTERNAL GAMMA RADIATION EXPOSURE

The importance of the radionuclides ^{95}Zr and ^{95}Nb lies in their contribution to the external gamma-radiation dose. BECK (1966) has calculated the gamma ray dose-rate conversion factors $\frac{(\mu\text{R/hr})}{(100\ \text{mCi/mile}^2)}$ at a height of 1 metre above soil for ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{137}Cs and ^{144}Ce as 0.22, 0.23, 0.063, 0.17 and 0.014 respectively, assuming an exponentially distributed source with respect to depth. The gamma dose from ^{95}Zr or ^{95}Nb relative to that from ^{106}Ru/^{106}Rh is 0.22/0.063, or approximately 3.5 : 1.

With the introduction of the new re-processing plant it was decided to examine in more detail the sub-critical materials – beach sand and silt. DUNSTER et al. (1964) have already shown that the mean concentration of gross beta radioactivity of the

fine silt which very occasionally settles out on beaches after spells of calm weather is 40 to 160 times higher than that of beach sand. An area in which fine silt was a more permanent feature was therefore sought in the vicinity of the pipeline. The area selected was the Ravenglass Estuary: three small rivers, the Irt, Mite and Esk, join at

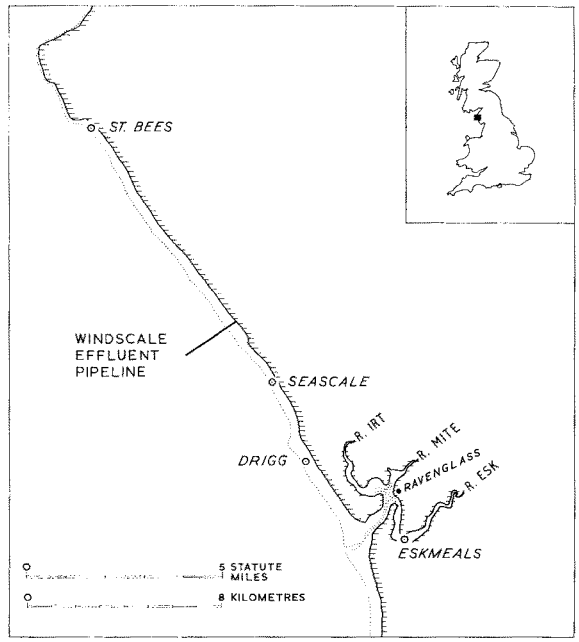

Fig. 1: The Ravenglass Estuary, positions for gamma dose-rate measurements, and the Windscale effluent pipeline

Ravenglass and enter the sea as the Esk about 6 miles south of the Windscale pipeline (Fig. 1). Here, in the intertidal zone, large expanses of mud flats are exposed at low tide. Elsewhere within 6 miles of the pipeline the beaches are flat and sandy with some outcrops of small boulders.

From January 1965, measurements of the gamma dose-rate have been made at regular intervals over mud flats in the estuary at Eskmeals and at three locations over beach sand at St. Bees, Seascale and Drigg. All measurements have been taken at a height of 1 metre using a Geiger instrument (U.K.A.E.A. Type 1368A). On one occasion checks were made over mud flats with a high-pressure ion chamber. There was good agreement between the two sets of measurements.

The data in Figure 2 show the dose-rates, measured in μR/hr, and the amounts of $^{95}Zr + {}^{95}Nb$ and ^{106}Ru released to the sea (the discharge data have been provided by U.K.A.E.A. at Windscale). The dose-rates over mud flats at Eskmeals, as expected from BECK (1966), reflect the trend in the amounts of $^{95}Zr + {}^{95}Nb$ discharged, but with a time lag of approximately two to three months. This is probably due to times of transport. The average dose-rate measured during the period January 1965 to

June 1967 was 140 μR/hr. The background level measured over a similar type of area remote from Windscale is 10 to 15 μR/hr. The levels over sandy beaches have ranged from 11 to 16 μR/hr compared with background measurements over similar areas of from 8 to 12 μR/hr. It can be seen that external gamma radiation over mud flats at Windscale is at least ten times that over beach sand in the same vicinity.

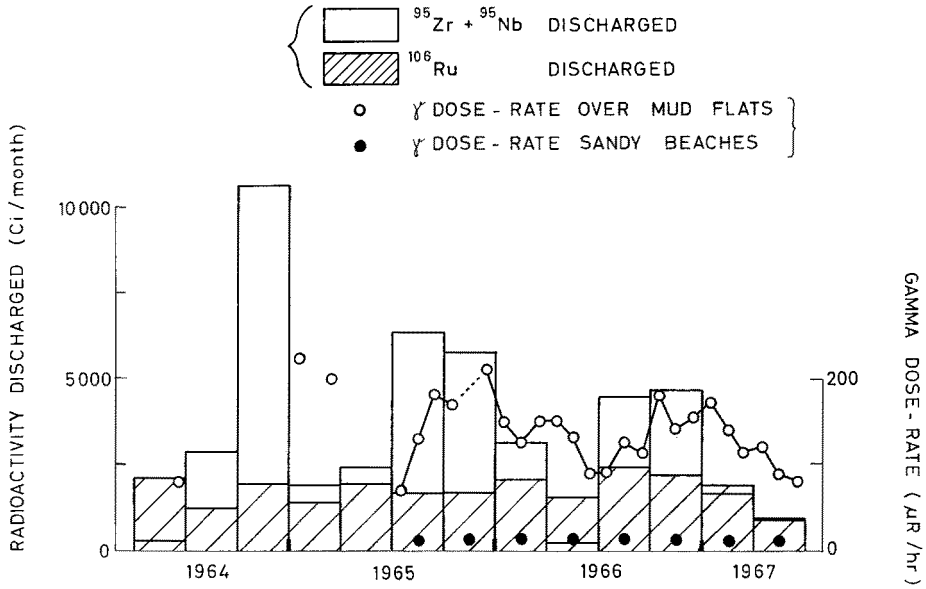

Fig. 2: Gamma dose-rates over mud flats and sandy beaches in the Ravenglass Estuary, and radioactivity discharged from the Windscale effluent pipeline

Table 1

Gamma dose-rates at a height of 1 m over mud flats in the Ravenglass Estuary (μR/hr). The values are normalized to 100 at position 1

Location	Position		February 1966	July 1966	July 1967
River Esk	Eskmeals Reference Area	1	100	100	100
		2	140	100	120
		3	70	100	90
River Mite		4	140	80	70
		5	90	80	90
		6	100	70	90
		7	100	80	90
River Irt		8	80	70	90
		9	90	50	70
		10	150	100	100

Gamma dose-rates at nine other mud-flat areas in the estuary have been compared with those at Eskmeals on three occasions. The results, normalized to 100 μR/hr at the reference area, are shown in Table 1. Similar dose-rates apply at any point on the estuary where mud flats are a feature.

Fission-product radionuclides in silt

In order to examine the contribution of individual radionuclides to the gamma dose and to make an estimate of the surface beta dose, surface silt samples have been taken from the reference area. The silt sample is obtained by scraping off the top centimetre layer at approximately 10 locations within a circle of radius 5 yards, resulting in a weight of silt of approximately 2.5 kg.

For analysis, the sample is freeze dried and the wet and dry weights noted. The dry sample is roughly ground and well mixed; 500 g are then placed in 14 polythene tubes, surrounding a 3 inch × 3 inch sodium iodide (Tl) crystal, and subjected to gamma-spectrometric analysis.

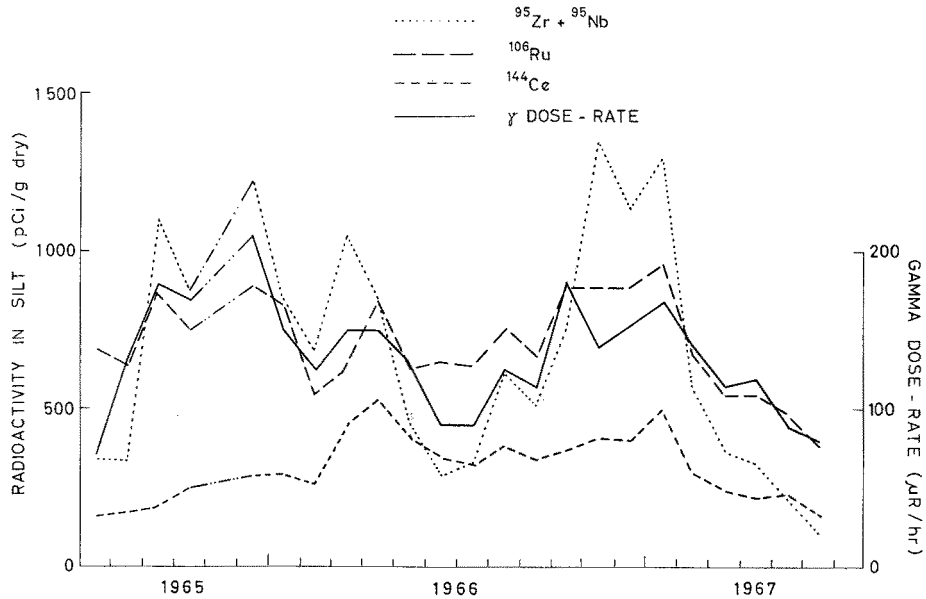

Fig. 3: Concentrations of radioactivity in silt and the gamma dose-rate (right scale) at the Ravenglass Estuary

The results of the analyses of reference area silt samples are given in Figure 3. The main contributors to the gamma activity are shown to be $^{106}Ru/^{106}Rh$, ^{144}Ce and $^{95}Zr + ^{95}Nb$ (these latter two radionuclides are not separated by the method of analysis). The ^{137}Cs concentration has always been < 29 pCi/g. The gamma dose-rate is again shown in Figure 3 and here reflects the trend of both the $^{95}Zr + ^{95}Nb$ and the ^{106}Ru concentrations.

Table 2

Radioactivity in samples of surface silt from position 1

Year	Concentration of radioactivity in surface silt							
	pCi/g dry				pCi/g dry per curie discharged per month			
	$^{95}Zr+^{95}Nb$	^{106}Ru	^{144}Ce	^{137}Cs	$^{95}Zr+^{95}Nb$	^{106}Ru	^{144}Ce	^{137}Cs
1965	755	750	213	12	0.19	0.43	1.1	0.14
1966	742	742	377	18	0.23	0.38	0.59	0.11
1967 (first 6 months)	485	604	278	18	0.21	0.36	0.61	0.24

The mean levels of radioactivity for 1965, 1966 and the first six months of 1967 are shown in Table 2. Also shown in the Table are the results of combining the mean annual concentrations with the mean monthly discharge rates, suitably adjusted for the transport time-lag, in the form pCi per g silt/curie discharged per month.

In an attempt to calculate the contribution of each of the main radionuclides to the gamma dose, the data for the period January 1966 to May 1967 were put in the form

$$\underset{(\mu R/hr)}{\gamma \text{ dose-rate}} = b_{Zr} \times \underset{(pCi/g)}{^{95}Zr + ^{95}Nb} + b_{Ru} \times \underset{(pCi/g)}{^{106}Ru} + b_{Ce} \times \underset{(pCi/g)}{^{144}Ce} + b_{Cs} \times \underset{(pCi/g)}{^{137}Cs}$$

and analysed by multiple correlation techniques. The best fit for the data is obtained by the equation

$$\gamma \text{ dose-rate} = 92 + (0.058 \pm 0.012) \times {}^{95}Zr + {}^{95}Nb \text{ (pCi/g)}.$$

Values for b_{Ru}, b_{Ce} and b_{Cs} were rejected as non-significant. The normal background level in areas remote from the effects of Windscale is 10 to 15 μR/hr measured using the Geiger type of instrument. The remaining 75 to 80 μR/hr in the constant must be due to contributions from the main radionuclides, but the variability of all nuclides other than $^{106}Ru/^{106}Rh$ suggests that the major portion may be due to this nuclide. The collection of data is continuing for further investigation of this point. The result can also mean that the surface layer is not truly representative of the radionuclide composition of the total silt layer contributing to the gamma dose.

Templeton and Preston (1966) have shown different penetrations with depth of ^{137}Cs and ^{106}Ru in seabed cores taken in the Windscale area. Lowton et al. (1966) have also shown a vertical distribution of radioactivity in a core sample of estuarine silt from the Solway Firth. The depth-radioactivity relationship in the Ravenglass situation has been examined by taking core samples at the reference area. The samples were obtained by pressing vertically into the mud a thin-wall P.V.C. domestic soil pipe, measuring 10.5 cm in diameter and 30 cm in length. The pipe was then dug out intact and frozen and, in the frozen state, sawn into sections. The sections were freeze-dried and analysed by the standard procedure for surface layer samples. Figures 4a and 4b show the results of the analysis of two cores collected in the first and second quarters of 1967. The depth-radioactivity profiles are similar: below the top inch (2.5 cm) ^{106}Ru, $^{95}Zr + ^{95}Nb$ and ^{144}Ce decrease exponentially with depth. In the two cores analysed the radioactivity levels in the top 2 cm layer do not fall exponentially

with depth and it is suggested that these values may reflect recent changes in discharge rates.

The relative penetrations are $^{137}Cs > {}^{106}Ru > {}^{144}Ce > {}^{95}Zr + {}^{95}Nb$. The radionuclide half-lives are also $^{106}Ru > {}^{144}Ce > {}^{95}Zr + {}^{95}Nb$, and the relative penetrations may be a simple function of the half-lives.

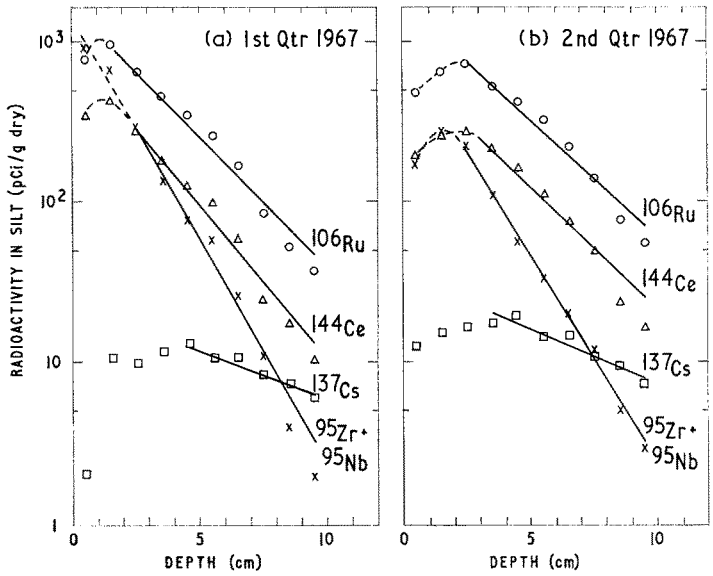

Fig. 4: Distribution of radioactivity in core samples of silt from the Ravenglass Estuary

The remainder of the cores, 10–15 cm and 15–25 cm, have been analysed and the total radioactivity content below a square centimetre has been calculated and shown in Table 3.

Table 3

Total radioactivity in silt under 1 sq. cm (pCi)

Core Sample	$^{95}Zr + {}^{95}Nb$	^{106}Ru	^{144}Ce	^{137}Cs
1st quarter 1967	2351	4314	1792	186
2nd quarter 1967	933	4067	1559	249

The ratios of total $^{106}Ru : {}^{95}Zr + {}^{95}Nb$ in the two cores are 1.8 : 1 and 4.4 : 1, compared with 0.8 : 1 and 2.8 : 1 in the surface layer. The contribution to the gamma dose from nuclides below the surface layer, particularly $^{106}Ru/{}^{106}Rh$, may account for the large value of the constant in the equation relating gamma dose-rate to surface activity, and this is currently being examined.

Surface beta radiation dose-rate

The relationship $D = 1.07$ C.E, where D is the dose-rate in millirads per hour, C is the specific activity of the surface layer in microcuries x 10^{-3} per g, and E is the mean energy per disintegration in MeV, can be used as a close approximation to the beta dose at the air-silt interface (DUNSTER 1957). $^{106}Ru/^{106}Rh$ and $^{144}Ce/^{144}Pr$ have effective beta energies of about 1 MeV, whereas an equilibrium mixture of $^{95}Zr/^{95}Nb$ has one of 0.08 MeV. From Table 2 the mean beta radiation dose-rate can be calculated as approximately 1 mR/hr.

Radionuclide accumulation factors

Shore seawater samples were taken at weekly intervals in 1965 and 1966, on the seaward side of the estuary at approximately the same distance from the pipeline as the mud-flat reference area. 10-litre samples were filtered through 0.22 micron milli-

Table 4

Accumulation factors for radionuclides in surface silt and surface sand

Radionuclide	Accumulation Factor (A.F.)	
	Surface silt	Surface sand
$^{95}Zr + ^{95}Nb$	15×10^3	5×10^2
^{106}Ru	14×10^3	4×10^2
^{137}Cs	0.8×10^3	0.6×10^2

pore filters, dried, and analysed by gamma spectrometry for $^{95}Zr + ^{95}Nb$, ^{106}Ru and ^{137}Cs; ^{144}Ce was below the limit of detection. Combining the results of the analyses with the data in Table 2, accumulation factors (A.F.) in the form $\frac{\text{(pCi/g dry silt)}}{\text{(pCi/g seawater)}}$ were calculated and are given in Table 4. Values of A.F. for beach sand are also shown in the Table. Silt accumulation factors for $^{95}Zr + ^{95}Nb$ and ^{106}Ru are seen to be about 30 times greater than those for beach sand.

Radioactivity in surface silt related to distance from the discharge point

The relationship of radioactivity in surface sediment with distance from the pipeline has been examined in 1966–1967. Samples of similar estuarine surface sediment have been taken at regular intervals at four other positions up to 75 miles from the discharge point. The results of the analyses are plotted as a function of distance in Figure 5. A simple power relationship fits the data well and is similar to that already established for seaweeds (TEMPLETON & PRESTON 1966). It is suggested that the curve

for $^{95}Zr + ^{95}Nb$ can be explained by the comparatively short half-lives of these radionuclides compared with ^{106}Ru and ^{144}Ce and the transit times involved in the greater distances.

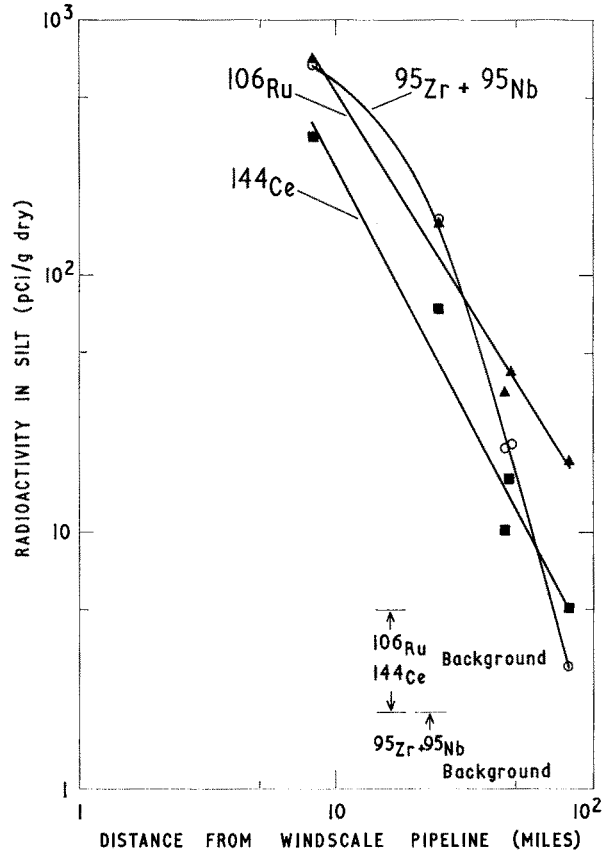

Fig. 5: Relationship between the radioactivity of surface silt and distance from the Windscale effluent pipeline

SUMMARY

1. Gamma radiation dose-rates over mud-flat areas are shown to be at least ten times those measured over sandy beaches.
2. Analysis of the surface sediment layer in an estuarine area 6 miles from the Windscale effluent pipeline shows the major gamma-emitting radionuclides to be ^{95}Zr, ^{95}Nb and ^{106}Rh.
3. Core sampling has indicated that the radioactivity concentration in silt decreases exponentially with depth from the surface layer.

4. It is shown that the gamma dose-rate above silt banks is largely accounted for by the concentration of $^{95}Zr/^{95}Nb$ in the surface silt layer. The residual gamma dose is probably accounted for by $^{106}Ru/^{106}Rh$ distributed beneath the surface layer.
5. The accumulation factors of $^{95}Zr + ^{95}Nb$ and ^{106}Ru in the surface silt layer are approximately 1.5×10^4.
6. The relationship of radioactivity concentration in surface silt to distance from the effluent pipeline can be described by a simple power function of distance.

Acknowledgements: I am grateful to Mr. A. PRESTON for his most helpful advice and encouragement, to Mr. K. FIRMAN for the measurements of gamma radiation and collection of sediment samples, and to all my other colleagues for the radiochemical analyses and preparation of data.

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Discussion following the paper by JEFFERIES

CARRUTHERS: Interest might be attached to the great mobility of the sediments at their surface as having a bearing on what you have said, and perhaps the work of Miss C. A. M. KING (*J. Sedim. Petrol.* 21, 131, 1951) on rapid bed changes might be relevant.

JEFFERIES: I think it would be most useful to know more about the reworking, deposition, and exchange of sediment, in estuarine environments, in relation to the capacity of an estuary to receive radioactive waste discharges.

WERNER: Ich möchte die Frage nach den biologischen Konsequenzen dieser Ablagerung der Radionuklide auf dem Meeresboden stellen. Werden Untersuchungen durchgeführt, in welchem Ausmaß diese Radionuklide in die lebenden Bodentiere eingehen? Wenn ich daran denke, daß z. B. *Arenicola marina* einen großen Umsatz von Bodensedimenten hat, dann wäre zu prüfen, ob diese Radionuklide in solchen Organismen angehäuft werden und damit praktisch in die Nahrungsketten eingehen.

JEFFERIES: I have no information on the radioactivity of bottom living organisms at the moment; my primary objective has been to answer a problem of public radiation exposure. I do have accumulation factors for the flesh of *Mytilus edulis*, found in the estuary. The A. F. for ^{106}Ru is 2×10^3 and the A. F. for $^{95}Zr + {}^{95}Nb$ is 1×10^3.

FONTAINE: The data presented during this session were diverse and very interesting. I should like to stress the necessity for developing the experimental aspect of pollution research. We must strive for a better understanding of the mechanisms of the noxiousness of each kind of pollution. Undoubtedly descriptive research is very important, but it must be associated – in my opinion – with more experimental investigations. Moreover, I should have liked the following aspect to be debated: the need to attempt exploitation of pollution, wherever possible. I think this aspect of our problems should retain our attention in the future and that the best way to struggle against pollution is to utilize it.