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Contract number: 268-79-1-BION
Head of Research team: Dr. E.K. Duursma (coordinator)
General Subject: Plutonium in the Rhine-Meuse-Scheldt delta; analysis of estuarine sediment, salt-marsh soil and vegetation samples.

The Rhine-Meuse-Scheldt estuary is investigated for plutonium as occurring in representative materials which might act as accumulators for this element. Sediment will adsorb plutonium with a factor of about 10^4 from water, while salt-marsh vegetation is equally known to accumulate heavy metals from estuarine waters.

The delta area might be envisaged as a sink for the three major European rivers, Rhine, Meuse and Scheldt, while the chosen samples are taken from material representative for long-range contaminative effects.

The plutonium measurements are independantly carried out in two laboratories, while the techniques are intercompared within an intercalibration programme.

Two well-chosen intercalibration samples have been sent to equally other laboratories, while simultaneously a characterisation of the sample material is carried out with standard chemical analysis.

The delta area (see Fig. 1) consists of four different sea arms where the influence of Rhine and Meuse is separated from that of the Scheldt. Nuclear installations at short distance are the power stations of Doel (Belgium) and Borssele (Netherlands) at the Scheldt system, and Dodewaard (Netherlands) at the Rhine/Meuse system. Sampling stations are marked in the attached map for sediments and vegetation.

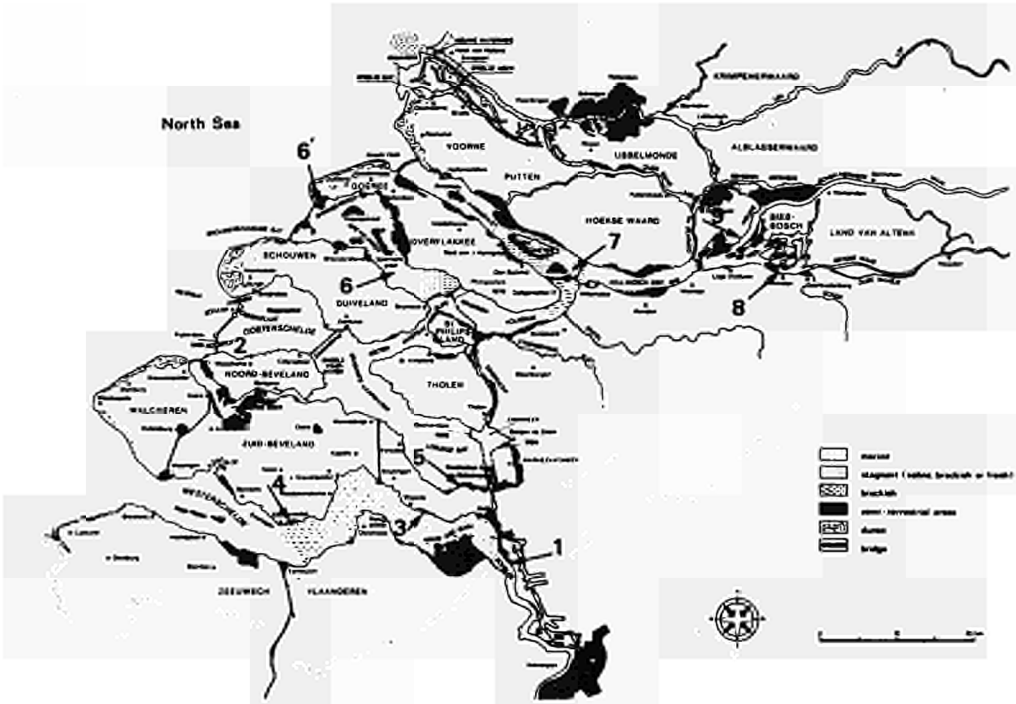

Fig.1. Delta of the rivers Rhine, Meuse and Scheldt.

Table 1: Homogeneity tests of intercalibration sediment samples SD-N-1 (station 1) and SD-N-2 (station 2). A: 12 samples, B: 7/8 samples, C: 6 samples (Lab. Géologique,Paris), D: 1 sample (NIOZ, Texel).

Determined compounds	SD-N-1		SD-N-2	
	Average	Stand.Dev.	Average	Stand.Dev.
A. Organic C (%)	3.50	0.068	0.098	0.007
Organic N (%)	0.207	0.0037	0.0016	lim.detect.
$CaCO_3$ (%)	13.03	0.25	1.84	0.030
Humidity (%)	0.89	0.067	0.12	0.016
B. Total alpha (pCi g^{-1})	21.9	1.2	1.6	0.5
C. Potassium (%)	1.71	0.09	-	-
^{137}Cs (pCi g^{-1})	0.35	0.05	-	-
^{60}Co (pCi g^{-1})	0.44	0.05	-	-
D. Quatrz	60 %		85 %	
Feldspar	6 %		10 %	
Calcite	20 %		4 %	
Clay-minerals	10 %		-	
Pyrite	1-2 %		-	
NaCl	3 %		3%	

Sub-Project 1. Delta Institute for Hydrobiological Research
Head research team: Dr. E.K. Duursma
General subject : Collection and preparation of samples, and additional analysis by chemical methods.

Results:

1) Preparation of samples for intercomparison programme.
Two sediment samples, each of 100 kg weight have been collected from stations 1 and 2, representing one sediment rich and one poor in organic content, respectively. After drying at 105°C, they were grinded to powder and homogenised for a week.

Homogeneity tests were carried out on 12 samples of each sediment for the contents of organic carbon and nitrogen, calciumcarbonate, humidity and total alpha radioactivity (by direct scintillation technique). Additionally within Sub-project 3 tests were made with potassium, ^{137}Cs and ^{60}Co. The results of these tests are given in Table 1.

2) Chemical and total alpha analysis of samples.
(i) Non-radioactive chemical analysis has been carried out on aquatic and salt-marsh sediments, as well as on salt-marsh vegetation (Aster tripoloum), while additionally (ii) the total alpha-radioactivity was determined by direct alpha-scintillation counting according to the technique of Cherry (unpublished).

For 1979 two sampling programmes have been executed, namely one in January and one in September. The chemical parameters so far determined for the sediments are: humidity of wet sediment, salinity in interstitial water and $CaCO_3$, organic carbon, total nitrogen, potassium and clay-content, all per dry sediment. The vegetation samples were analysed for chloride, total nitrogen, sodium, potassium, calcium, magnesium, total phosphorus and sulphate. The results are given in Table 2.

The interpretation of the chemical parameters must be considered later against the obtained plutonium results. At present they are basic informations on the conditions around and the quality of the samples. Concerning the total alpha data, a level between 3 to 20 pCi per gram dry sediment is found where the results are not significantly higher in areas

Table 2: Sediment and vegetation parameters of Delta-region samples (1979).

SAMPLES	humidity/wet %		S-interst.water %		$CaCO_3$ %		POC %		tot.N %		K meq/100g		Clay %		α radioact. $pCig^{-1}$	
Aquatic sediments	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.
9	73.5	64.1	0.53	0.25	12.88	7.92	8.64	3.57	0.68	0.26	1.32	0.42	50.1	25.4	21.4	4.5
8	69.6	24.3	0.20	0.29	13.39	3.33	6.69	1.82	0.50	0.08	0.92	0.24	45.8	12.3	16.1	3.6
7	60.3	64.5	0.67	1.05	10.40	9.16	4.30	3.95	0.25	0.23	0.73	0.73	35.4	48.3	14.6	4.6
6	44.6	22.7	21.62	29.81	7.04	4.99	1.62	0.57	0.10	0.05	1.75	0.68	10.3	4.9	9.3	4.5
Salt-marsh sed.																
5. 0-10 cm	44.3	54.2	18.86	32.47	2.33	1.96	3.54	5.70	0.24	0.35	2.31	2.61	19.7	37.2	10.3	3.2
10-20 cm	46.8	56.7	22.76	28.67	2.54	4.54	3.97	5.60	0.23	0.34	2.33	3.45	26.0	51.4	11.7	4.4
4. 0-10 cm	42.2	43.7	18.31	27.12	18.30	14.23	4.36	5.00	0.27	0.33	2.46	2.10	38.8	45.2	11.1	4.9
10-20 cm	46.7	41.9	21.06	25.10	15.08	12.73	5.78	4.41	0.30	0.28	2.33	1.96	33.3	47.4	12.1	6.4
3. 0-10 cm	65.6	40.6	16.74	20.32	11.82	11.34	7.93	3.76	0.42	0.21	3.95	1.55	53.3	25.2	12.0	4.5
10-20 cm	62.5	42.3	17.31	20.39	14.04	13.07	7.24	4.32	0.33	0.22	3.61	1.44	60.0	32.0	14.2	4.5

Vegetation	Cl $mg\ g^{-1}$		tot.N $mg\ g^{-1}$		Na $mg\ g^{-1}$		K $mg\ g^{-1}$		Ca $mg\ g^{-1}$		Mg $mg\ g^{-1}$		P_2O_5 $mg\ g^{-1}$		SO_4 $mg\ g^{-1}$	
salt-marsh	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept.	Jan.	Sept	Jan.	Sept.
6'	51.4	30.0	29.2	14.1	41.0	23.0	29.4	23.0	6.7	5.0	3.1	2.2	7.7	5.3	5.0	5.7
5	102.0	61.6	34.2	19.5	68.5	49.5	36.2	18.3	4.2	3.8	3.6	2.8	7.4	7.3	4.7	6.2
4	87.8	91.5	36.5	20.0	62.4	68.4	27.4	20.0	4.3	3.2	3.9	3.7	9.8	6.7	4.2	6.9
3	68.2	154.4	25.8	23.7	56.3	102.9	20.8	23.6	4.7	5.7	3.5	5.4	8.8	7.2	3.0	8.9

influenced by Rhine, Meuse or Scheldt. The natural radioactivity probably overlaps that of plutonium which is some orders of magnitude lower (see Sub-projects 2 and 3).

Sub-Project 2. Association Euratom - ITAL

Heads research team: Dr. F.I. Frissel & Drs. P. Poelstra

General subject : Analysis of Pu in field samples.

Results:

From the samples collected in January 1979 analyses for ^{239}Pu and ^{238}Pu have been completed while those for September 1979 are still under treatment. Results are presented in Table 3.

The preliminary results are more or less comparable to those found within Sub-project 3 and do not allow at present a detailed discussion as far as the differences concern. The results as such are shortly discussed in Sub-project 3.

Sub-Project 3. Laboratoire de Géologie, Paris

Head research team: Dr. J.M. Martin

General subject : Homogeneity tests of intercalibration samples, intercalibration programme and analyses Pu in field samples.

Results:

Homogeneity tests were carried out on the two prepared sediment samples for the radionuclides ^{137}Cs and ^{60}Co. From these samples material was sent to other interested laboratories from which the results have not yet been obtained. The major quantity (80 kg) of each sediment is given to Dr. R. Fukai of the IAEA laboratory in Monaco for later use in the IAEA intercalibration programmes. So far the results of the homogeneity tests are given in Table 1.

The results of the field samples are presented in Table 3, where they can be compared with those obtained from the ITAL - Wageningen. In general, both outcomings of $^{239-240}Pu$ are for the sediments matching with those earlier determined by Livingston and Bowen (1979) in the Atlantic

Table 3: Preliminary results of radionuclide analysis in sediments and vegetation.
For sample no. see Fig.1; fCi = 10^{-15}Ci; pCi = 10^{-12}Ci.

Samples	ITAL-Wageningen		Laboratoire Géologique-Paris			
	^{239}Pu	^{238}Pu	^{137}Cs	^{60}Co	^{239,240}Pu	^{238}Pu
	(both fCi g^{-1})		(both pCi g^{-1})		(both fCi g^{-1})	
Aquatic sediments						
1 (Intercalibration)	11.5, 12.7	4.0, 4.4	0.35	0.44	12.38	
2 "	0.1, 0.2	0.1, 0.1	0.05	0.02	0.95	
9 (Influence Rhine/Meuse)	6.2	0.5	0.63		14.6	
8 " "	5.9	1.0	0.66		19.2	1.2
7 " "	5.5	0.9	0.55		6.1	
6 (Saline lake)	4.8	0.3	0.19		5.5	
Salt-marsh sediments						
5 (Influence North Sea)						
0-10 cm	7.2	0.6	0.45		7.9	1.2
10-20 cm	26.2	1.4	0.50		23.1	1.4
4 (Influence Scheldt)						
0-10 cm	18.1	1.8	0.66		30.0	
10-20 cm	16.0	1.0	0.44		5.3	
3 0-10 cm	18.7	1.9	0.64		17.9	4.4
10-20 cm	37.7	2.2	0.99		27.0	
3 0-20 cm			0.03 (n.d.)		6.3	
20-40 cm			0.29		13.0	1.0
40-60 cm			0.11		11.2	
60-80 cm			0.02 (n.d.)		1.3	
Vegetation (Aster tripolium)						
6'			0.11 (n.d.)		0.4	
5			0.07 "		0.3	
4			0.07 "		0.3	
3			0.13 "		0.8 (?)	

and Fukai et al (1974) in the Mediterranean. Equally the ratio of $^{238}Pu/^{239,240}Pu$ is of similar value of that of Livingston and Bowen being in the range of 0.018-0.050.

The $^{239,240}Pu/^{137}Cs$ ratios as found in the Delta-region sediments differ from those of Livingston and Bowen which are a factor of 10 higher.

Conclusions on the origin of the radionuclides have to be preliminary since the investigations are not yet completed. Probably plutonium and ^{137}Cs derive dominantly from fallout sources, where ^{137}Cs might have reacted under estuarine conditions different as in the Atlantic Ocean.

References:

Livingston, H.D. & Bowen, V.T., 1979. Pu and ^{137}Cs in coastal sediments. Earth Planet. Sc. Lett., 43, 29-45.

Fukai, R., Murray, C.N., Statham, G. & Asari, K., 1974. Radionuclide measurements of water, sediment and biota collected from the Ligurian Sea. Tech. Rep. IAEA - 163, Monaco, 134-138.