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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 estuaries might be envisaged as sinks for many substances which are transported by the rivers Rhine, Meuse and Scheldt to the sea. Identically through tidal movements contaminants transported by the North Sea might arrive in these estuaries.

A research was made to determine ^{238}, ^{239} and ^{240}Pu concentrations in sediment, particulate matter, salt-marsh plants and lichens from various locations of the Delta area (Fig. 1). The objectives were to trace the sources of plutonium for this area, either being fallout, up-river nuclear installations or the reprocessing plant in La Hague (transport along the coast).

Not all 1980-samples have been analysed yet, which are in particular the particulate matter samples taken outside the Western Scheldt area and the lichen samples.

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.

For 1980 additional salt-marsh sediments, particulate matter and old salt-marsh sediment samples from our stock have been analysed on humidity, salinity, $CaCO_3$, POC (particulate organic carbon), total nitrogen, potassium (K), clay, total α-radioactivity and pH.

The total α-radioactivity reflects all natural and possible artificial

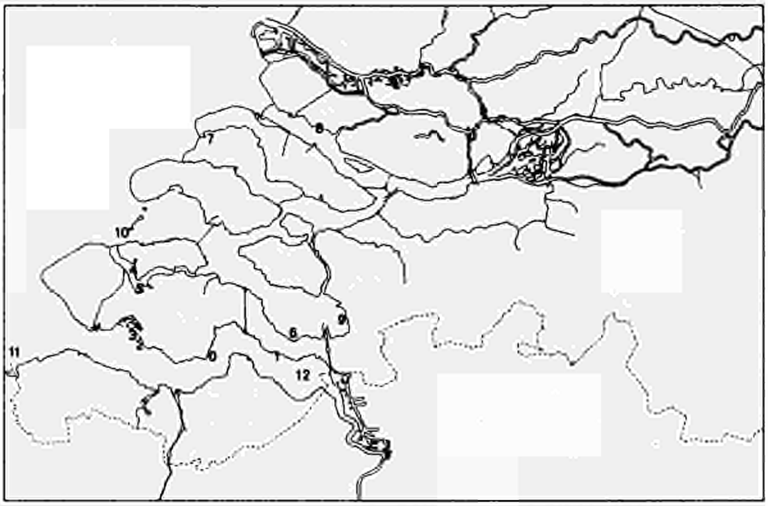


Fig. 1. Rhine-Meuse-Scheldt estuaries.
Sampling stations: (0) Ellewoutsdijk, (1) Waarde, (2) Borssele, Nuclear Power Station, (3) Kaloot), (4) Spieringschor, (5) Noordsloe, (6) Stroodorpepolder, (7) Springersgors, (8) Zuidland, (9) Salt marsh Bergen op Zoom, (10) Mouth Eastern Scheldt, (11) Wielingen, (12) Nauw van Bath.

Table 1 Sediment and suspended matter parameters per dry weight. For sampling stations see Fig. 1.

samples			humidity/ wet weight %	salinity-interst.water ‰	$CaCO_3$ %	POC %	tot.N %	K meq/100 g	Clay %	Cs sediment. pCi g^{-1}	pH-KCl	Cs sed/clay pCi g^{-1}
Salt-marsh sediment												
Waarde	20-40 cm	6'79	47.3	18.5	14.6	1.63	0.18	1.73	15.4	5.5	7.74	33.3
(1)	40-60 cm	6'79	50.0	21.1	13.8	1.86	0.19	1.76	15.8	5.4	7.65	33.1
	60-80 cm	6'79	47.0	19.3	16.3	1.58	0.19	1.89	36.5	7.4	7.64	20.1
	80-100cm	6'79	40.9	18.5	15.8	1.87	0.17	1.72	37.2	4.1	7.60	11.8
Borssele:	0-10 cm	2'80	17.8	13.9	2.61	0.12	0.01	1.34	0.4	3.3	8.65	825.-
(2)	10-20 cm	2'80	19.0	17.5	1.75	0.16	0.20	1.26	1.7	3.1	8.13	188.1
Old salt-marsh sediments (0-10 cm)												
(3) Kaloot		5'62		23.9	0.5	1.16	0.13	1.37	18.5	4.4		23.3
(4) Spieringschor		7'62		3.80	11.0	1.96	0.29	2.23	56.3	5.1		9.2
(5) Noordsloe		8'62		0.30	7.5	1.06	0.19	1.82	36.6	4.1		11.5
(6) Stroodorpepolder		3'65		20.3	3.4	1.34	0.08	1.37	10.7	5.1		47.7
idem		7'66		26.0	5.0	1.42	0.07	1.19	11.4	0.7		5.6
(7) Springersgors		8'62		20.9	7.0	4.13	0.31	2.82	20.1	n.d.		n.d.
idem		7'68		17.3	9.2	6.91	0.46	2.41	18.5	3.3		18.8
idem		3'74		19.4	12.1	3.67	0.41	4.95	60.8	4.0		7.4
idem		10'72		2.24	12.2	1.70	0.10	3.33	34.6	6.1		7.0
(8) Zuidland		8'61		3.3	10.4	2.15	0.13	0.85	15.0	4.7		33.3
(9) Salt marsh Bergen op Zoom		3'64		24.4	1.6	0.10	0.02	0.31	1.5	1.9		85.1
idem		6'75		15.3	5.5	5.53	0.30	2.90	18.0	4.9		26.3
idem		5'78		26.0	4.2	3.13	0.26	2.52	26.6	4.7		17.7
Suspended matter												
(10) Mouth Eastern Scheldt		6'80	55.3	30.8	7.86	4.06	0.54		11.3	3.9	8.04	33.3
(11) Wielingen (high-tide)		7'80	47.3	26.1	27.5	4.29	0.38		29.7	3.7	7.93	9.3
idem (low-tide)		7'80	50.7	29.7	18.3	4.05	0.38		41.8	4.3	7.94	10.2
(12) Nauw van Bath		9'80	33.3	9.95	5.51	4.75	0.49		66.1	10.5	7.76	15.6

Fig. 2. Concentration relationship between ^{137}Cs and $^{239, 240}Pu$ of estuarine sediments and suspended material for three estuaries: Rhine-Scheldt delta (this study), the Seine Bay and estuary and Gironde estuary (other studies Martin and coworkers).

α-emitting radionuclides, where the total α-radiation is about two to three orders of magnitude higher than the plutonium-α-radiation.

The 1980 results are presented in Table I, divided into three sections: Fresh salt-marsh sediment at Waarde and Borssele (Western Scheldt), old salt-marsh samples from Delta Area and seston (suspended matter) obtained by centrifugation of sea water (50 gram requires about 2 days shiptime).

The results are not yet related with the plutonium data since these are not yet complete.

Sub-Project 2. Association Euratom-ITAL
Head research team: Dr. F.I. Frissel
General subject : Analysis of Pu in field samples

The analyses for plutonium have been concentrated in 1980 on a few vegetation samples *(Aster tripolium)* and on a number of salt-marsh sediment samples from the stock of the Delta Institute at Yerseke. The results obtained for ^{239}Pu in *Aster tripolium* (Table II) agree with those obtained in 1979,but the ratio $^{238}Pu/^{239}Pu$ is higher (0.14-0.27) than the values found in salt-marsh sediments (0.08-0.14). At present these results need to be re-evaluated against other results, but the ^{239}Pu (normalized to clay) values for the sediments sampled from 1961 show that at one place (e.g. Springersgors) the concentrations have a maximum at or before 1968, related probably with fallout peaks from the bomb-testing series which ended in 1962.

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

As already mentioned in the introduction not all plutonium- and other radionuclide analyses have been completed for the 1980 samples. On the other hand a comparison can be made of the so-far obtained results with measurements from other areas.

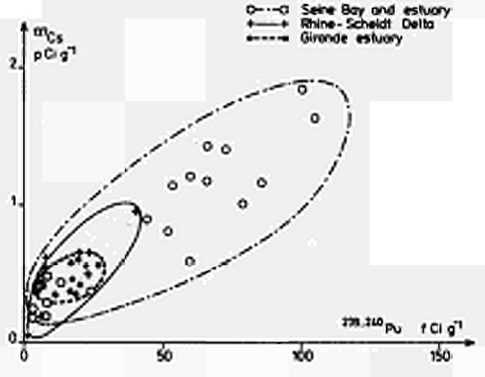
Table II. Results on Pu measurements in *Aster tripolium* and salt-marsh sediments sampled from 1961-1978 (in pCi kg^{-1})

Samples		$^{238}Pu/^{239}Pu$	^{239}Pu	^{240}Pu	Tracer recovery %	^{239}Pu normalised to clay
Aster, Ellewoutsdijk	(9'79)*(0)**	0.14	0.29	0.04	46	
Aster, Springersgors	(9'79) (7)	0.14	0.14	0.02	46	
Aster, Stroodorpepolder	(9'79) (6)	0.18	0.11	0.02	60	
Aster, Waarde	(9'79) (1)	0.27	0.41	0.11	84	
Kaloot	(5'63) (3)	0.00	2.5	0.2	32	13.2
Spieringschor	(7'62) (4)	0.05	17.9	0.9	32	31.8
Noordelee	(8'62) (5)	0.14	0.7	0.1	13	1.9
Stroodorpepolder	(3'65) (6)	0.08	2.3	0.2	18	21.5
idem	(7'68) (6)	0.10	1.8	0.2	32	15.3
Springersgors	(8'62) (7)		18.0	?	43	59.8
idem	(7'68) (7)	0.06	53.7	3.3	47	165.2
idem	(7'70) (7)	0.05	36.4	1.9	22	61.8
idem	(10'72) (7)	0.06	34.3	2.3	33	57.6
Zuidland	(8'61) (8)	0.04	5.1	0.2	20	34.0
Salt marsh Bergen op Zoom	(3'64) (9)	0.5	0.2	0.1	33	13.3
idem	(6'75) (9)	0.1	9.4	0.9	18	52.2
idem	(5'70) (9)	0.09	4.5	0.6	12	24.4

*) 9'79 = Sept. 1979
**) (0) see Fig. 1

Table III. Average results of $^{239, 240}Pu$ and ^{137}Cs measurements in vegetation from different regions. The activities are normalised to the potassium contents of the plants in fCi g^{-1} potassium for $^{239, 240}Pu$ and pCi g^{-1} potassium for ^{137}Cs.

	$^{239,240}Pu$	^{137}Cs
Aster tripolium (Rhine-Scheldt delta)	12.5	54
Grass (Nord Cotentin Brittany)	34	4
Grass (La Hague area)	222	17



Vegetation

Specific activities, normalized to potassium of *Aster tripolium*, a salt-marsh plant also used as vegetable, have been compared to those of grass samples from 2 French areas located in North Cotentin and La Hague. The last location is near to the La Hague Nuclear Reprocessing Plant, while the first one is from an area only contaminated by atmospheric fall-out from the 1958-1962 nuclear bomb-testing.The results are summarized in Table III.

In all samples, the activities appear to be very low, especially those measured in *Aster tripolium* of the Rhine-Scheldt delta. These are very close to grass samples from the Northern Cotentin Brittany area, both primarly contaminated by fall-out from bomb-testing. It is worthwhile to note that the La Hague grass samples Pu activities are higher by more than one order of magnitude. This is mainly due to the atmospheric recycling of marine aerosols (MARTIN, THOMAS and JEANDEL, in press), rather than due to direct contamination.

A first assessement of the transfer coefficient between soils and *Aster tripolium* shows that less than 5% can be accumulated by this species.

Sediment samples

In Fig. 2 ^{137}Cs and $^{239-240}Pu$ activities of deposited and suspended sediments from the Rhine-Scheldt delta are compared to those of other estuarine environments (i) the Gironde estuary (which can be assumed as a background for atmospheric fall-out and (ii) the Seine Bay estuary (which is notably contaminated by the La Hague Centre and possibly by Windscale nuclear plant wastes).

For both isotopes, the Rhine-Meuse-Scheldt estuary samples show in general similar activities as compared to those measured in the Gironde estuary. A very few number of samples appear to have slightly higher concentrations. However, it must be kept in mind that due to the well known affinity of trace metals and radionuclides for clay-size fractions the specific activities representative of suspended sediments (i.e. with a similar matrix to the Seine and Gironde samples) might be higher and therefore probably intermediate between the fall-out level (Gironde) and the contaminated Seine estuary.

It is also expected that the general pattern of the plutonium geochemical behaviour in the Rhine-Scheldt delta might be similar to that observed in French estuaries, i.e. a systematic increase of particulate specific activity along with salinity (JEANDEL, MARTIN and THOMAS, 1980).

References

Jeandel, C., Martin, J.N. and Thomas A.J. (1980). Radionucleides artificielles dans les estuaires français. Coll. Int. sur l'impact des radionucleides rejetées dans le milieu marin. AIEA Vienne SM 248/123.