Progress Report

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General subject of the contract:

Differential migration of plutonium in the delta estuaries of Rhine, Meuse and Scheldt.

List of projects:

1. Differential migration of artificial radionuclides in the delta estuaries of Rhine, Meuse and Scheldt

Joint proposal of three laboratories.

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ARTIFICIAL RADIONUCLIDES IN RHINE-MEUSE-SCHELDT DELTA

I. INTRODUCTION

In order to improve the knowledge of the distribution of plutonium isotopes and major gamma emitters in the Rhine-Meuse-Scheldt delta, a more detailed examination of the radionuclides sources and their overall concentrations in both suspended and deposited sediments was needed. The studies were focused in 1983 on the western Scheldt estuary in which earlier investigations had shown a complex distribution pattern of radionuclides originating from three distinct sources : atmospheric fallout, nuclear power plants effluents and sea-waters. Sampling was extended to areas which remained insufficiently documented : the outer and lower estuary, and the river section.

II. RADIOISOTOPE BEHAVIOUR

Significant comparisons of results concerning sediment samples ranging from sands to fine muds can only be achieved when radionuclides concentrations are normalized to an appropriate tracer of the fine-grained clay fraction. Comparisons with other studies of environmental radioactivity undertaken in French rivers and estuaries require the use of the classical aluminium normalization rather than potassium normalization which has been applied in the former progress reports (both elements are well correlated in the Scheldt estuary).
All available data concerning the artificial radionuclides in the sediment collected in the Scheldt area from 1979 to 1983 are summarized in table 1.

II.1. Origin of radionuclides

The assessment of the origin of radionuclides is not easy because several elements can be supplied to the Scheldt estuary by different sources. Furthermore important qualitative and quantitative fallout variations were observed in 1981 as a consequence of the nuclear atmospheric test of october 1980 in China. These variations have been well recorded in French river and estuarine sediments (Elbaz-Poulichet et al., 1982 ; Thomas, 1983) and probably also further northwards. The impact of this event became undetectable in 1983.

a. Marine input

The suspended matter samples collected in the vicinity of the Strait of Dover do not allow to determine representative long-terme average concentrations. Considerable enrichment relative to Al was observed for Ru-106, Ce-144, Mn-54 and Zr-95, and to a lesser extent for Sb-125 and Pu-239+240. These high values can be partly ascribed to the enhanced fallout in 1981 but Ru-106, plutonium and possibly Sb-125 essentially reflect the overall marine contamination by La Hague and Sellafield plants. The regular landwards decrease of Pu-239+240, Ru-106 and Sb-125 concentrations inthe Scheldt estuary emphasizes their marine origin. Ce-144, Mn-54 and Zr-95 were hardly measurable in 1982-83 due to their rapid decay.

b. River input

Only Pu-238, Pu-239+240, Sb-125 and Cs-137 could be detected in river sediments near Gent in 1983. Their normalized concentrations fall in the range of the values found in French fresh-water sediments during the same period. It was too late to detect any effect of the last chinese explosion.

c. Input of nuclear effluents in the estuary

In 1980, the nuclear centers at Doel and Borssele discharged respectively 2.64 and 0.12 Ci of radioactive effluents (excluding H-3) in the estuary (Luykx, and Fraser, 1983). Cs-134, Cs-137, Co-58 and Co-60 represent 90% of these effluents. The sharp maximum of Co-60 concentrations near Doel is obviously related to this nuclear centre. The concentrations seawards are much lower and the occurrence of high marine values can be attributed to French and English littoral power plants. Despite significant releases (0.62 Ci in 1980) Co-58 was almost undetectable, due to its shorter half-life (70 days). The Cs-134/Cs-137 ratio rises from 0.05 to 0.20 near Doel and further landwards. Whereas an industrial origin of Cs-134 is not dubious, river and marine inputs of Cs-137 must also be

envisaged. Pu-238/Pu-239+240 ratio exceeds 0.5 near Doel, whereas it is only 0.26-0.37 and 0.06 ± 0.04 in marine and river sediments respectively. Plutonium releases from Doel nuclear center can therefore be envisaged.

II.2. Landward transport of radionuclides

As far as 120 km from inlet, the normalized concentrations of various radionuclides originating from the sea or from the Doel area were still much higher than the river values. This locations seems to correspond to the maximum landward limit of salt intrusion. These concentrations may result either from a pronounced landward transport of estuarine sediments or from the adsorption of dissolved radionuclides in the particles in the brackish-waters.

III. CONCLUSIONS

The origin of radionuclides in the Scheldt estuary is now rather well identified. They can be used as tracers to study the sediment transport and exchange processes in the upper tidal estuary.

IV. REFERENCES

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E/Al pCi/g Al	Scheldt river near Gent	French rivers <1981 ⩾1982	1981	Scheldt estuary (km≤120)	Eastern Scheldt basin	PWR liquid effluents 1980 (Ci)	Channel suspended matters 1979+1981	Origin fallout	PWR	Sea
Ce-144	<(1.0-1.6)	<3	7-84	(<1.5)-6	(<2.4)-6.6	0.0054	20 - 100	+?	-	+?
Cs-137	4.7 - 4.9	6-20	6-20	6.5-16	4 - 14	0.64	7 - 16	+	+	+
Cs-134	nd	nd	nd	≤2.1	<(0.4-0.9)	0.58	<(0.5-4)	-	+	+?
Co-60	nd	nd	nd	(<0.3)-16	(<0.4)-0.9	0.66	3 - 14	-	++	-
Co-58	nd	nd	nd	≤2.4	nd	0.62	-	-	(++)	-
Mn-54	<(0.4-0.8)	<0.5	<(0.5-2.0)	(<0.3)-0.6	-	0.038	(<1)-25	?	+?	?
Pu-239+240	0.072	0.11-0.34	-	0.2-1.3	0.1-0.6	no data	0.8-2.6	+	+?	++
Ru-106	<(0.9-2.1)	<(5-13)	5-26	(<2)-42	(<14)-20	≤0.002 ?	60-300	+?	-	++
Sb-125	0.5-1.1	∿ 1	1.5-6.0	0.6-3.7	<2.2	≤0.003?	11 - 39	+	-	+
Zr-95	<(2.9-3.3)	< 0.6	4-40	<(1.3-8)	<2.8	∿0.002	(<14)-100	+?	-	+?

Table 1. Artificial radionuclides in the deposited and suspended sediments of the Scheldt river and estuary, and the Eastern Scheldt basin (1979-1983). All concentrations are normalized to Al content. French river data only concern areas submitted to fallout only. Nuclear effluents of pressurized water reactors (PWR), including Doel (Belgium) and Borssele (Netherlands) nuclear centers, after Luykx and Fraser, 1983. Channel suspensions were sampled near Strait of Dover, mainly in 1981.