Final Report

Contractor:

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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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PLUTONIUM AND GAMMA EMITTORS IN THE RHINE-MEUS-SCHELDT DELTA AND ADJACENT NORTH SEA COAST.

With support of the Commision of the European Communities, DG XII, a cooperative research programme has been set up in 1979 to study the presentday artificial radioactivity in the Dutch Delta an the adjacent North Sea coast. This Delta in the southwest Netherlands (Fig. 1) is an important and complex estuarine system, through which three major European rivers the Rhine, Meuse and Scheldt are discharging into the North Sea. Since a few decades, the Delta has been submitted to contamination by artificial radionuclides through atmospheric fallout and industrial discharges of radioactive effluents into the neighbouring seas, the rivers and the estuarine zone itself.

The major objectives of the programme were:(i)-to assess the overall distribution of the artificial radionuclide concentrations in sediments and biota of the different basins and adjacent sea; (ii)-to identify possible sources of radioactive contamination; and (iii)-to make a first assessment on the radioactive behaviour in the various estuarine systems. The analysed materials are: surface bottom sediment, sediment cores, salt-marsh sediments, suspended matter, mussel flesh and mussel shells, lichens, salt-marsh plants and aquatic plants. All analysis of radionuclides have been carried out with proper calibration, while additionally intercalibration on different samples has been excecuted between the two Pu-analytical laboratories in Paris and Bilthoven/Wageningen. Gamma-ray activities have only been analysed by the Paris laboratory, while at Yerseke total alpha activity (including the natural alpha activity) was measured, (Duursma et al, 1985).

Concerning the plutonium concentrations in deposited sediments and suspended matter average values of 12-21 pCi $^{239+240}$Pu/kg and 1.5-6 pCi ^{238}Pu/kg (dry weight) have been found. These activities only represent 0.1 % of natural alpha radioactivity. In the soft parts of mussels, plutonium concentrations are one order of magnitude lower (also on dry-weight basis). Normalization of plutonium concentrations to aluminum, considered as an index of clay concentrations, allows some practical determination of the possible sources in the environment.

Clearly the whole Delta area receives plutonium isotopes from the North Sea, where ^{238}Pu/$^{239+240}$Pu isotopic ratios are high due to the Sellafield and La Hague reprocessing plant effluents. In coastal sediments and mussels, $^{239+240}$Pu contamination decreases from La Hague to the north-eastern Dutch coast, where the influence of industrial effluents can hardly be distinguished from fallout background, contrary to industrial ^{238}Pu contamination which is still detectable so far north.

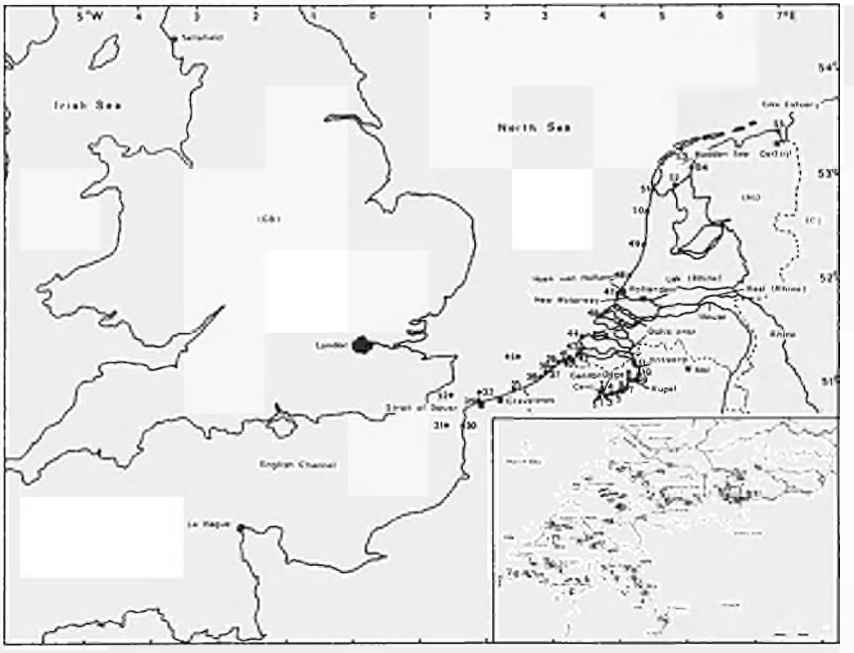

Fig. 1: Map of sampling areas.

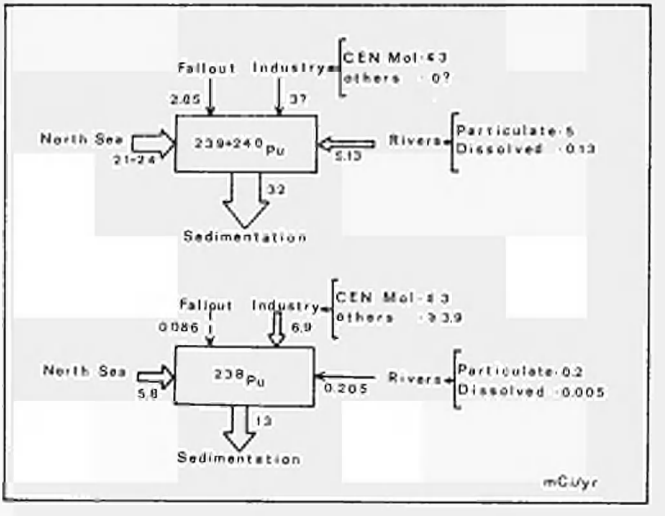

Fig. 2: Tentative budget of average plutonium isotopes annual sources and sinks of the Western Scheldt estuary in the major shoaling area of sediments near Antwerp. All data are in mCi/yr.

Studies of the two estuarine systems of the Delta, the Rhine-Meuse-Eastern Scheldt and the Scheldt-Western Scheldt system, have shown a systematic seaward increase of plutonium concentrations in the sediments, thus confirming earlier findings in French estuaries. Uptake of marine plutonium is probably responsible for this general estuarine pattern.

^{238}Pu distribution in the Western Scheldt estuary is altered by additional inputs in the estuary itself. The study of the distribution of ^{60}Co allows the identification of the area where the highest contamination by nuclear power plant effluents occurs. This area does not correspond with the peak of the $^{238}Pu/^{239+240}Pu$ ratios, showing that excess ^{238}Pu originates partly or completely from another source, located in the watershed of a Scheldt tributary the Rupel river.

The occurrence of relatively high plutonium concentrations and isotopic ratios in the upper estuary, resulting from the landward transport of sediments, emphasizes the possibility of artificial radionuclides as valuable tools for studying estuarine hydrodynamic processes, (Guary et al, in preparation).

A tentative budget calculation for the Western Scheldt estuary (Fig.2) presents an annual input of 'marine' plutonium of 21-24 mCi $^{239+240}Pu$ and 5.8 mCi ^{238}Pu. Radioactive contamination of mussels is far below the annual limit of consumption intake (ALI), being 10^{-5} ALI for 50 kg mussels/yr.

Although a number of results have to be investigated in more detail, the gamma emittors in sedimentary material, if normalized to potassium, show the following behaviour: $^{137}Cs/K$ and $^{134}Cs/K$: Since ^{134}Cs does not exist in fallout, the ratio $^{134}Cs/^{137}Cs$ is indicative for local contamination by nuclear installations. The results for suspended matter and top-layer bottom sediments from Rhine, Meuse, Scheldt, Delta region and southern North Sea towards Boulogne, showed ^{137}Cs levels of 10-50 pCi/gK, and a $^{134}Cs/^{137}Cs$ ratio between 0.05 and 0.10. No clear increase was detectable near the power stations of Doel and Borssele. $^{60}Co/K$: The concentrations of ^{60}Co ranged, equally for the same sedimentary material, from 0.6 to 43 pCi/gK, with the highest values in the English Channel (Gravelines) and the Scheldt (Doel), showing the influence of nuclear power plants. $^{125}Sb/K$: Antimony-125, attached to sedimentary material, is mainly from marine origin and in fact from the La Hague effluents. Values of 33-47.5 pCi/gK were found in the English Channel, descending to 1.5 pCi/gK in the upper Scheldt and 3.6-3.7 pCi/gK in the Rhine and Meuse. $^{106}Ru/K$: ^{106}Ru shows and identical picture as ^{125}Sb, high values in the Channel (130-570 pCi/gK), lower values towards the mouth of the Western Scheldt (100-110 pCi/gK), while the river values of Rhine, Meuse and Scheldt ranged between 6 and 23 pCi/gK. $^{144}Ce/K$: The determined ^{144}Ce concentrations were too close to the detection limit in order to produce reliable features. Approximately 5 pCi/gK was found in river sedimentary material, while this ranged between 10-250 pCi/gK in the Channel.

Concerning lichens, sampled directly at the border of the estuaries, it was expected that these would give additional information on radionuclide contamination caused by spray from the aquatic system. It has been documented (Guary, 1980) that plutonium concentration factors in lichens can range between 3200 and 4300. 15 samples of lichen Xanthoria parientina were collected, giving values (all in pCi/kg) of ^{238}Pu: 1.13 ± 1.03, $^{239+240}$Pu: 26.5 ± 26.2, (ratio: 0.05 ± 0.02) ^{54}Mn: 61 ± 84, ^{60}Co: 17 ± 17, ^{95}Zr: 1060 ± 1020, ^{106}Ru: 457 ± 330, ^{125}Sb: 190 ± 98, ^{137}Cs: 3930 ± 2560, ^{144}Ce: 1270 ± 1060 .

The values are much lower than those measured in Lichina pygmaea near La Hague outfall, but appear, with the exception of ^{106}Ru and ^{95}Zr, close to those of northeast Brittany. The data of the Delta show a slight northward decrease.

Although salt-marsh plants can be usuful indicators for Cd and Hg contaminations (Beeftink et al, 1982), only detectable concentrations have been found for ^{238}Pu, $^{239+240}$Pu and ^{137}Cs. If normalized per g of potassium, the activities appear to be low for $^{239+240}$Pu and high for ^{137}Cs in comparison to those of grass from a fallout area (North Cotentin, Brittany) , and that of La Hague. In this sequence the pCi/g K are for $^{239+240}$Pu: 12.5 (Aster tripolium), 34 and 222 (grass), and for ^{137}Cs: 54 (Aster tripolium), 4 and 17 (grass). A first assesment of the transfer coefficient between soils and Aster tripolium shows that less than 5% can be accumulated by this species. On the basis of dry weight the average concentrations found were: ^{238}Pu: 0.11 ± 0.08 pCi/kg, $^{239+240}$Pu: 0.37 ± 0.21 (ratio: 0.28 ± 0.15) and ^{137}Cs: 95 ± 30 pCi/kg.

A few samples of aquatic plants , sampled in the Delta, have been analysed for ^{238}Pu and $^{239+240}$Pu. Average values in pCi/kg dry weight were found of ^{238}Pu: 0.21 ± 0.23 and $^{239+240}$Pu: 0.95 ± 0.77, giving a ratio of ^{238}Pu/$^{239+240}$Pu of 0.21 ± 0.09). The plants concern Ulva lactica, Zostera marina and Sargassum japonica.

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