# RADIATION PROTECTION PROGRAMME Progress Report

Contractor:

Contract no.: BI6-B-191-NL

Delta Institute for Hydrobiological Research Vierstraat 28 NL-4401 EA Yerseke

Head(s) of research team(s) [name(s) and address(es)]:

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Telephone number: 1131/1920
Title of the research contract:

Transfer processes and modelling of plutonium species and gamma emitters in the Scheldt estuary; redox and organic speciation in relation to aqueous and particulate fractionation.

List of projects:

1. Transfer processes and modelling of plutonium species and gamma emitters in the Scheldt estuary; redox and organic speciation in relation to aqueous and particulate fractionation.

## Title of the project no.: B16-B-191-NL

Transfer processes and modelling of plutonium species and gamma emitters in the Scheldt estuary; redox and organic speciation in relation to aqueous and particulate fractionation.

## Head(s) of project:

Dr. E.K. Duursma, Dr. D. Eisma. Nederlands Instituut voor Onderzoek der Zee (Texel, The Netherlands)

Dr. J.M. Martin. Institut de Fiogéochimie Marine (E.N.S. France)
Dr. J. Pentreath. Fisheries Radiological Laboratory (Lowestoft, England)
Dr. Prof. R. Wollast. Université Libre de Bruxelles (Belgique)
Scientific staff:

Dr. D. Petit. Institut de Biogéochimie Marine (E.N.S. France)
J. Nieuwenhuize. Delta Institut voor Hydrobiologisch Onderzoek
(Yerseke, The Netherlands)

## I. Objectives of the project:

Study of total dissolved and rarticulate Pu-238 and Pu-239,240 and gamma emitters (Co-60, Cs-137,...) in the Western Scheldt area. Determination of Kd's as function of major physicochemical parameters such as dissolved oxygen, pH, Eh, salinity, DOC and POC. Study of redox partitioning of dissolved plutonium (III + IV) and (V + VI) at selected locations of the Western Scheldt.

## II. Objectives for the reporting period:

The experimental procedure has been tested at three locations of the Western Scheldt estuary, where a relative high concentration of total plutonium had been measured in suspended matter and sediments during a preceding cruise.

## III. Progress achieved:

In September 1986, Dr. D. Petit stayed for two weeks at the Fisheries Radiological Laboratory (Lowestoft, England) to learn analytical techniques for dissolved plutonium speciation analysis.

Two cruises have been carried out in the Western Scheldt on board of the R/V LUCTOR from D.I.H.O. (Yerseke, The Netherlands) in October and December 1986, respectively.

The first cruise has allowed us to solve some difficult field manipulations such as large volume of estuarine water samples (500 l) filtration at 0.45  $\mu m$  porosity, storage of subsamples, etc.

During the second cruise, we collected samples (i) for total dissolved and particulate plutonium analysis (ii) for redox speciation of dissolved plutonium and (iii) for gamma emitters measurement in suspended matters. The samples were collected from three stations located along the Western Scheldt estuary: Schaar Van Spijkerplaat (marine reference), Doel (in front of a nuclear power plant, PWR) and Rupelmonde (possible inputs of plutonium coming from the C.E.N. at Mol, via the Rupel river). The major physicochemical parameters have been measured in each sampling station by the Delta Institute (salinity, T, O2, pH, DOC, POC and suspended matters) and by the Free University of Brussels (alkalinity, Eh, dissolved iron and manganese). Large volume of surface waters (200 1 to 500 1) were sampled by pumping and particulate matters were recovered by continuous centrifugation. Water samples were then filtered by pressure filtration on membrane filters of  $0.45 \mu m$ , stored in 25 1 flasks and immediately spiked with Pu-242, after acidification to pH 1.5 with HCl and with both Pu-242 (in reduced form) and Pu-236 (in oxidized form) for total plutonium and redox speciation plutonium samples, respectively. The concentration and oxidation state differentiation of dissolved plutonium have been carried out at the Delta Institute following the Fe(OH) 3 technique used at the F.R.L. (Lowestoft). The radiochemical purification and alpha spectrometry counting of plutonium samples are now performed at the Institut de Biogéochimie Marine (E.N.S., Montrouge).

On the other hand, the Free University of Brussels has installed 4 sediment traps at various locations in the Scheldt (Doel, Antwerp (2), Hemiksem). Samples have been collected once a month since July 1986. Suspended matters have also been collected twice by centrifugation in the Scheldt river (upstream Gent where there is no more salt water intrusion) and in some tributaries.

The analysis of  $\gamma$ -emitters will be performed in the collected samples at ENS.

During the week of 8-12 December 1986 the R/V LUCTOR from the Delta Institute was used by the Netherlands Institute for Sea Research to take water samples in the Western Scheldt (Flushing-Gent). The aim was to study aggregation/de-aggregation of suspended matter, particle formation and the role of oxygen and salinity in these processes in this estuary. Samples were taken and the following analyses were carried out: temperature, salinity, suspended matter, grain-size analyses, plankton, bacteria, POC/DOC, nutrients, oxygen, nitrogen, sugars, pyrolyse, fluorescence, amino-acids, ETS, electronmicroscopy, in-situ photography.

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Study the adsorption/desorption behaviour of plutonium in the estuarine environment. In this context, we plan to investigate redox speciation of dissolved plutonium under oxic or anoxic conditions, which occurred seasonally in some Western Scheldt areas. In particular we shall try to relate both oxidation states and organic speciation of plutonium (and possible other radionuclides such as Co-60) to their distribution between the dissolved and particulate fractions of the water as well as their binding to bottom sediments.

V. Other research group(s) collaborating actively on this project [name(s) and address(es)]:

VI. Publications: