Progress Report
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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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ARTIFICIAL RADIONUCLIDES IN RHINE-MEUSE-SCHELDT DELTA

I. INTRODUCTION

The studies on the distribution of plutonium isotopes and various gamma emittors in the Rhine-Meuse-Scheldt delta of the south-west Netherlands were continued. Taking into account the limitation caused by the restricted number of plutonium analysis, due to long counting times, only selected samples were analysed. These are chosen on the basis of earlier results and derived hypothesis of distribution patterns.

In former progress reports, various results were presented and discussed on the distribution of radionuclides in the region and related to the type of sample: sediment, suspended matter, mussels, plants and lichens. (Euratom, 1979, 1980, 1981). Here the additional achievements will be presented for each studied radioisotope, and well for Cs-137, Co-60, Pu-238, 239, Sb-125, Ru-106 and Ce-144.

II. RADIOISOTOPE BEHAVIOUR

In general the behaviour of the detected radioisotopes has been investigated by relating their ratios to stable potassium, with the environmental circumstances. Potassium (determined by K-40) is indicative for the clay fractions and most radionuclides are proportionally present in these fractions. Instead of potassium also aluminium can be taken, which is equally related to the clay fractions.

II.1 Cs-137/K and Cs-134/Cs-137

The two available isotopes of cesium, Cs-134 ($t_{\frac{1}{2}}$=2.2 yr) and Cs-137 ($t_{\frac{1}{2}}$=30 yr) have been investigated. Cs-134 does not exist in fallout, thus the ratio Cs-134/Cs-137 is an indication of local contamination by nuclear installations. The results for suspended matter and top-layer sediment from Rhine, Meuse, Delta region, Scheldt and Southern North Sea towards Boulogne, showed Cs-137 levels of 0.1 - 0.5 pCi/g per % K. The Cs-134/Cs-137 ratios in % were about 5-10%.

In general there could not be distinguished a clear pattern for the Delta region, since the levels are relatively constant, while little variation was observed from river to sea. No clear increase was detectable close to the nuclear power stations Doel and Borssele.

II.2 Co-60

The concentrations of Co-60 ($t_{\frac{1}{2}}$=5.24 yr) ranged, equally for top-layer sediments and suspended matter, from 6 to 430 fCi/g per % K The highest values were detected for the Channel (Calais) and Scheldt, while the lowest concentrations were found in the Grevelingen. Thus Co-60 is a clear indicator for inputs from nuclear installations like Doel (Scheldt), and Gravelines (Channel) and those of the English Coast.

II.3 Pu-238, 239 (+240)

The suspended matter and sediment values for Pu-239 ($t_{\frac{1}{2}}$=2.4 x 10^4 yr) mixed with Pu-240 ($t_{\frac{1}{2}}$=6.6 x 10^3 yr) ranged in the Delta region from 0.9 to 43 fCi/g Pu-239,240/%K, with the highest values in the Southern North Sea (26-43 fCi/g/%) with North Sea water. No influences of Doel and Borssele could be detected, probably the surplus Pu-239,240 above fallout is explained from releases of the Windscale and/or La Hague reprocessing plants.

The Pu-238/Pu-239,240 ratios (Pu-238, $t_{\frac{1}{2}}$=86 yr) ranged from 4 to 54%, with again the heighest values (19-43%) in the Southern North Sea,

but also in the upper Scheldt (29-54%). The explanation is difficult to give for the Scheldt values, since the average source ratios are 35% for La Hague, 0.2% for Windscale and 5% for fallout. Abnormal high ratios also have been found in sediments of French rivers downstream of nuclear facilities (Loire, Thomas, pers. comm.) but also in rivers only exposed to fallout (Var-river, Ballestra, 1980 and Gironde, Jeandel, 1981).

II.4 Sb-125

Antimony-125 ($t_{\frac{1}{2}}$=2.8 yr) attached to sedimentary material is mainly from marine origin and in fact from the La Hague effluents. Values in the Channel of 330-475 fCi/g Sb-125/%K were found, descending to 15 fCi/g/%K in the upper Scheldt and 36-37 fCi/g/%K in the Rhine and Meuse.

III.5 Ru-106

An identical picture as Sb-125 was shown by Ru-106 ($t_{\frac{1}{2}}$=1.0 yr). High values in the Channel were found for both suspended matter and top-layer sediment of 1300-5700 fCi/g Ru-106/%K. In the mouth of the Western Scheldt these values were reduced to about 1000-1100, while the river concentrations of Rhine, Meuse and Scheldt were between 60 and 230 fCi/g/%K.

III.6 Ce-144

The Ce-144 ($t_{\frac{1}{2}}$=0.78 yr) data, determined in a similar way as the other gamma emittors, are not very reliable. The values are too close to the detection limit. Approximately 50 fCi/g/%K in the rivers and 100-2500 fCi/g/% in the Channel. Probably the source is equally the La Hague reprocessing plant.

IV. CONCLUSIONS

The observed patterns of the various radionuclide distribution in the Dutch Delta Region become more clear, in particular for radionuclides attached to suspended matter and top-layer sediment. In particular the presentation of the radioactivity per % K gives the opportunity to understand the transport of these radionuclides in the environment as related to their sources.

Additional measurements are proceeding on mussels, as filter-feeders of suspended matter. For this investigation the sampling area will be extended from the Channel and the Delta region to the Dutch North Sea coast northwards.

The lichen studies are completed, while the salt-marsh plant analysis revealed too low concentrations at the level of the detection limits (Euratom, 1980, 1981).

Some core samples are still under investigation in order to get a better view on the fallout maximum of the years 1958-1963.

REFERENCES

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