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MODELLING THE BEHAVIOUR OF RADIONUCLIDES IN THE AQUATIC ECOSYSTEM

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1. INTRODUCTION

The aim of this work is to model the behaviour of radionuclides released in a surface water ecosystem : contamination of water, sediment, food chain and (finally) fish.

The model is applied to the river MEUSE, to assess the concentrations of CS-137 in the major compartment of the river ecosystem due to releases of Cs-137 by the Nuclear Power Plant SENA (located at CHOOZ).

Radionuclides released in surface water are not simply transported by the flow : physical transfers (adsorption/desorption to sediment) and biological transfers occur and contribute to the contamination of the upper level of the river food chain.
The radiocontamination of the different compartments (water, sediment, biomasses) is a pathway to the contamination of man.

The behaviour of radionuclides in surface waters cannot be described without a preliminary knowledge of the water and sediment transport. The model of radionuclides transfers in the ecosystem is thus organized in successive submodels :

1) the hydrodynamic submodel : to assess river flow, depth, velocities, ... ;

2) the sediment submodel : to assess the concentration of suspended sediment in the water column and the concentration of bottom sediment on the bed of the river, coupled by the sedimentation and resuspension processes ;

3) the radiological submodel : to assess the radionuclides concentrations in water (liquid phase), the radionuclides concentrations adsorbed to suspended and bottom sediments, and the concentration of radionuclides incorporated in the different biological compartments.

The processes considered present basically a three-dimensional non-stationary feature. A 3D description of hydrodynamic, sediment and radionuclides transport is generally useless and unnecessary in rivers, so that the most efficient way is the one-dimensional description (computation of cross-section averaged values, obtained by integration of the equations over the cross-section). Non-stationary effects (flood events, ...) play however an important role in river sediment transport, so that the model proposed is a one-dimensional non-stationary model.

2. THE HYDRODYNAMIC SUBMODEL

This submodel calculates the hydrodynamic variables : river flow, depth, cross-section, velocity, bed shear stress, ... (cross-section averaged values). The input data are the river flow measurements at one or several stations, and the geometry of the river (slope of the bed, slope of the banks, location of locks and dams, ...).

The equations express the conservation of water and the equilibrium of forces :

$$\frac{\partial}{\partial t} A + \frac{\partial}{\partial x}(A u) = q(x,t) + \Sigma Q_i \, \delta(x-x_i) \tag{1}$$

$$\frac{\partial}{\partial t}(A u) + \frac{\partial}{\partial x}(A u u) = - A g \frac{\partial}{\partial x}(Z_b+H) - \frac{g}{C^2} L u^2 \tag{2}$$

t	:	time (s) ;
x	:	longitudinal coordinate along the river axis (m) ;
$A(x,t)$	:	cross-section area (m^2) ;
$u(x,t)$	:	flow velocity (cross-section averaged) (m/s) ;
$q(x,t)$	:	linear lateral inflow (m^2/s) ;
$Q_i(t)$	:	inflow (m^3/s) of the tributary i located at the position x_i ;

$\delta(x-x_i)$	:	Dirac function (m^{-1}) ;
g	:	acceleration of gravity (m/s^2) ;
$Z_b(x)$	:	altitude of the river bed (m) ;
$H(x,t)$	:	depth of the river (cross-section averaged (m) ;
$L(x,t)$	:	width of the river (cross-section averaged) (m) ;
$C(x)$	:	Chezy coefficient ($C^2 = m/s^2$).

3. THE SEDIMENT SUBMODEL

A detailed description of the sediment submodel has been presented in a previous paper (SMITZ & al., 1985).

For hydrodynamic and radiological considerations, it is necessary to take into account the grain size distribution of the sediment particles. The sediment submodel developed is a variable multi-class model. For the application of the model to the river MEUSE simulation, two particles size classes are routed :

class 1 : clay + fine silt
class 2 : coarse silt + sand.

For each class of particles, the sediment content is calculated in two sub-compartments : the suspended sediment and the bottom sediment.

The equations which express the sediment concentrations in water column and on the river bed are :

<u>suspended sediments</u> : (class j, j = 1,N)

$$\frac{\partial}{\partial t} A\, SS_j + \frac{\partial}{\partial x}(A\, u\; SS_j) = \sum_i Q_i\, SS_{j,i}\, \delta(x-x_i) - SED_j + RS_j \qquad (3)$$

<u>bottom sediments</u> : (class j, j = 1,N)

$$\frac{\partial}{\partial t} BS_j = SED_j - RS_j \qquad (4)$$

where SED_j = sedimentation flux (class j) :

$$SED_j\,(x,t) \begin{cases} = w_j \cdot SS_j \cdot L\,(1 - \dfrac{\tau_b}{\tau_{s,j}}) & \text{if } \tau_b < \tau_{s,j} \\ = 0 & \text{if } \tau_b \geq \tau_{s,j} \end{cases}$$

RS_j = resuspension flux (class j) :

$$RS_j(x,t) \begin{cases} = e_j \cdot L\,(\dfrac{\tau_b}{\tau_{r,j}} - 1) & \text{if } \tau_b > \tau_{r,j} \\ = 0 & \text{if } \tau_b \leq \tau_{r,j} \end{cases}$$

$$\tau_b = \rho \cdot \frac{g}{C^2} \cdot u^2$$

$SS_j\,(x,t)$	:	concentration of suspended sediment, class j (g/m^3) ; cross-section averaged value ;
$SS_{j,i}\,(t)$	:	concentration of suspended sediment in tributary i ; class j (g/m^3) ;
$BS_j\,(x,t)$	:	concentration of bottom sediment, class j (g/m) ; cross-section averaged value ;
$Q_i\,(t)$	:	flow (m^3/s) of the tributary i, located at the point x_i ;
$SED_j\,(x,t)$	:	sedimentation flux (g/m.s) ;
$RS_j\,(x,t)$	:	resuspension flux (g/m.s) ;
W_j	:	settling velocity, class j (m/s) ;
e_j	:	erosion rate, class j ($g/m^2.s$) ;
L	:	width of the river ;
$\tau_b\,(x,t)$	:	bed shear stress ($g/m.s^2$) ;
$\tau_{s,j}$	:	critical sedimentation shear stress, class j ($g/m.s^2$) ;
$\tau_{r,j}$	:	critical resuspension shear stress, class j ($g/m.s^2$) ;
ρ	:	specific mass of water (g/m^3).

The source of sediments in the river is the supply of sediments by tributaries (erosion process in the basin). The description of the sediment supply by tributaries is an important problem in the sediment submodel : the input of sediments is incorporated via a statistical relation, but the field measurements are scarse, and the statistical relation present high variabilities.

The model has been applied to the case of the river MEUSE (fig. 1).

During the year 75, the Nuclear Power Plant SENA released significant quantities of Cs-137, and a few data from routine survey (IHE/CEN, 1965-1983) give some information about the river contamination. The model has thus been used to realize a non-stationary simulation of the year 1975.

The extension of the model covers the French and Belgian section of the river MEUSE (km 0 - km 600).

The river flow (m^3/s) during the year 1975 is presented at fig. 2.

The sediment submodel computes the concentration of suspended sediment and the concentration of bottom sediment at each point and at each time (cross-section averaged values). Fig. 3 and fig. 4 present the computed values of suspended sediment and bottom sediment at HASTIERE (km 488) during the year 1975. Computed values are in good agreement with the general trend of field measurements.

4. THE RADIONUCLIDES SUBMODEL

The radionuclides submodel describes the activity of radionuclides in water, in the suspended sediments, in the bottom sediments, and in different compartments of the food chain. A complex dynamic equilibrium governs the exchanges of radionuclides between these compartments of the river system.

However, the contamination of the food chain can be evaluated separately, because the quantity of radionuclides contained in biological compartments is extremely small compared to the quantity of radionuclides contained in the water and sediment compartments.

In a first step, the radionuclides submodel describes the concentrations or activities of radionuclides in water and in the different classes of sediments, and includes the processes of :

- transport by water ;
- transport by suspended sediment ;
- adsorption of dissolved radionuclides to sediments (suspended and bottom sediments) ;
- desorption of adsorbed radionuclides ;
- radiactive decay.

A detailed description of this model has been presented in a previous paper (SMITZ & al., 1985).

The equations which express the conservation of radionuclides in water, in suspended sediment, and in bottom sediment are :

<u>water</u>

$$\frac{\partial}{\partial t}(A\ a_W) + \frac{\partial}{\partial x}(A.u\ a_W) = \sum_i R_i\ \delta(x-x_i)$$

$$+ \sum_j \frac{SS_j}{1 + K_{d,j}\ SS_j} \frac{A}{\tau_j} \cdot a_{SS,j}$$

$$- \sum_j \frac{SS_j}{1 + K_{d,j}\ SS_j} \frac{A}{\tau_j} K_{d,j} \cdot a_W$$

$$- \lambda A\ a_W$$

suspended sediments

$$\frac{\partial}{\partial t}(A\ SS_j\ a_{SS,j}) + \frac{\partial}{\partial x}(A.u.SS_j\ a_{SS,j})$$

$$= - \frac{SS_j}{1 + K_{d,j}\ SS_j}\ \frac{A}{\tau_j} \cdot a_{SS,j}$$

$$+ \frac{SS_j}{1 + K_{d,j}\ SS_j}\ \frac{A}{\tau_j}\ K_{d,j} \cdot a_W$$

$$- SED_j \cdot a_{SS,j}$$

$$+ RS_j \cdot a_{BS,j}$$

$$- \lambda\ A\ SS_j\ a_{SS,j}$$

$(j = 1,N)$

bottom sediments

$$\frac{\partial}{\partial t}(BS_j \cdot a_{BS,j}) = SED_j \cdot a_{SS,j} + RS_j\ a_{BS,j}$$

$$- \lambda\ BS_j \cdot a_{BS,j}$$

$(j = 1,N)$

a_W	:	activity in water (Bq/m^3) ;
$a_{SS,j}$	:	activity in suspended sediment (Bq/g), class j ;
$a_{BS,j}$	:	activity in bottom sediment (Bq/g), class j ;
R_i	:	release of radionuclides in the river (Bq/s) at the location x_i ;
δ	:	Dirac function (m^{-1}) ;
τ_j	:	characteristic time of adsorption process (s^{-1}) ;
$K_{d,j}$	:	distribution coefficient (m^3/g) ;
λ	:	characteristic frequency of radionuclide decay (s^{-1}).

The adsorption/desorption processes between radionuclides in water and radionuclides adsorbed to sediments (class j) are determined by the values of partial distribution coefficients $K_{d,j}$.

This model has been applied to the river MEUSE to assess the radiocontamination of water and sediment by Cs-137 released by the Nuclear Power Plant SENA (located at CHOOZ, km 473). The fluxes (monthly - averaged values) of Cs-137 released in the river MEUSE during the year 1975 are presented at the fig. 5. The radionuclides submodel calculates activities in water, in suspended sediments (class 1 and 2), and in bottom sediments (class 1 and 2). The values of the partial distributions coefficient used in this study are :

$K_{d,1} = 0.08 \ m^3/g \quad (= 80000 \ \ell/kg)$

$K_{d,2} = 0$

These values lead to a global distribution coefficient :

$K_d \sim 0.05 \ m^3/g \quad (= 50000 \ \ell/kg)$

(class 1 $\sim$ 70 % , class 2 $\sim$ 30 % in the MEUSE)
which is a value often given in the literature for Cs-137.

The Cs-137 activities in water, in suspended sediment, and in bed sediment, computed by the model (simulation of the year 1975) at HASTIERE (km 488, that is 15 km the release point) are presented at fig. 6, fig. 7 and fig. 8. Fig. 8 shows a good agreement between observed and computed values.

The model can easily assess the flux of radionuclides transported by water and by sediment through any cross-section of the river ; the transport of Cs-137 by sediment appear to be more important ($\sim$ 55 %) than the transport by water ($\sim$ 45 %).

Contamination of the fish

Experimental studies realized at the CEA/CADARACHE (LAMBRECHTS A., 1983 ; FOURQUIER L., LAMBRECHTS A., 1983) show that the contamination of the fish by Cs-137 is the result of multiple exchanges (adsorption/desorption ; ingestion/excretion) between fish and

- water,
- sediment,
- benthic organisms (food),
- macroplankton organisms (food).

A simplified food chain and contamination chain of the carp has been established (LAMBRECHTS, 1983) ; the main results of these laboratory studies are :

- benthic and macroplankton organisms present a short renewal time, and reach quickly an equilibrium state of contamination ;
- transfer of Cs-137 towards carps is continuous ; no sturation effect can be observed ;
- the carp fixes about 13 % of the activity of the ingested benthic organisms ;
- the carp fixes only 3 % of the activity of the ingested macroplankton organisms ;

- the cumulative contamination of the carp by water, sediment and food shows that the transfers are cumulative ;
- each transfer (contamination/decontamination) can be represented by a transfer coefficient.

It is thus possible to assess the dynamic contamination of fish expressed as a function of the radionuclides concentration in water, in suspended sediment, in bottom sediment : the contamination of the food (benthic and macroplankton organisms) is directly incorporated as a linear function of water and sediment contamination.

The equations which govern the fish contamination are :

$$\frac{\partial}{\partial t} M = \begin{cases} a \quad t^b \quad (\text{APR - NOV}) \qquad (\text{g/s}) \\ 0 \qquad (\text{DEC - MARCH}) \end{cases}$$

$$\begin{aligned} \frac{\partial}{\partial t} M\, a_F = \; & TF_{W,F} \; a_W \; M \\ & + \alpha \; TF_{S,F} \; a_{BS} \; M + (1 - \alpha) \; TF_{S,F} \; a_{SS} \; M \\ & + TF_{BS,F} \; a_{BS} \; M + TF_{SS,F} \; a_{SS} \; M \\ & - DF \; a_F \; M \end{aligned}$$

$M(t)$	:	mass of the fish (g) ;
a,b	:	parameters of growth law ;
a_F	:	specific activity in fish (Bq/g fresh) ;
a_W	:	activity in water (Bq/m^3) ;
a_{SS}	:	activity in suspended sediment (Bq/g) ;
a_{BS}	:	activity in bottom sediment (Bq/g) ;
$TF_{SS,F}$	:	partial transfer factor from suspended sediment to fish ;
$TF_{BS,F}$	:	partial transfer factor from bottom sediment to fish ;

$TF_{W,F}$	:	partial transfer factor from water to fish (includes transfer factor due to ingestion of benthic and macroplanktonic organisms) ;
$TF_{S,F}$	:	partial transfer factor from sediment to fish (includes transfer factor due to ingestion of benthic and macroplanktonic organisms) ;
DF	:	desorption coefficient ;
α	:	unknown factor, which represents the relative influence of suspended sediment activity and of bottom sediment activity in the contamination of the fish (α varies from 0 to 1).

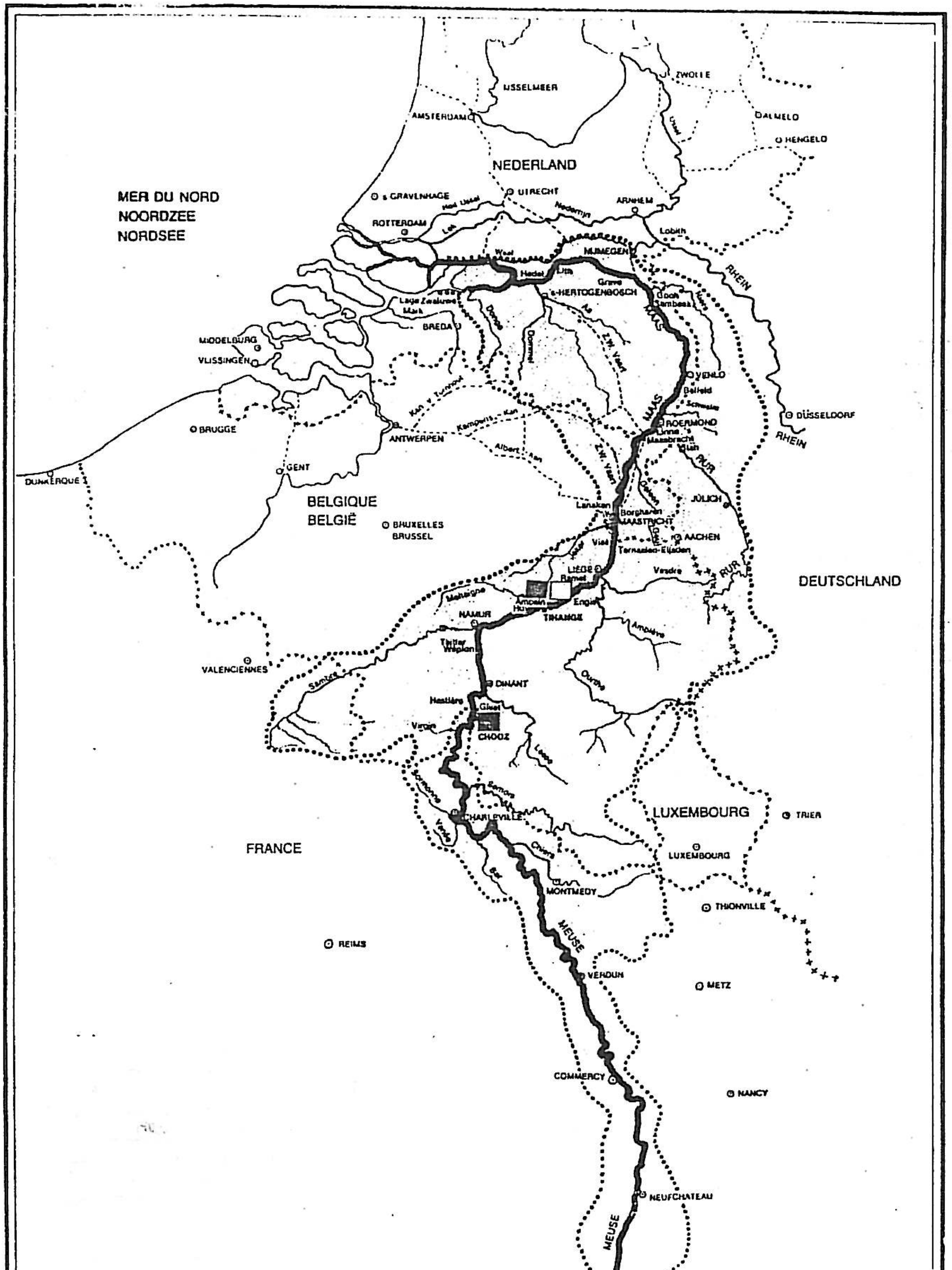
This simplified model of fish contamination is applied to a fish (carp) of the river MEUSE. The numerical values of the different parameters are estimated by the laboratory experiments realized at the CEA/CADARACHE (LAMBRECHTS, 1983).

Fig. 9 presents the computed values of the mass of a river MEUSE typical carp, as a function of his age. Fig. 10 presents the values of the specific activity in a carp, computed by the fish contamination submodel ; the model simulates the evolution of the fish contamination during five successive yearly cycles (each year is assumed to be the same as the year 1975). The activities in water, suspended sediment and bottom sediment are calculated by the first step of the radiological submodel. Case 1 (α = 0) is the simulation of the preferential contamination pathway through bottom sediments ; case 2 (α = 1) is the simulation of the preferential contamination pathway through suspended sediment.

The results obtained by the model show a cyclic contamination of the fish (extra contamination by food during summer months, decontamination of fish during winter months, when the decontamination flux is greater than the contamination fluxes). Fig. 11 presents the time evolution of the different contamination fluxes. Contamination by food appear to be an important way in the mechanism of contamination.

The computed activities in a typical carp of the MEUSE (10-30 Bq/kg fresh) are in good agreement with the observed values : measurements of fish contamination by Cs-137 in the river MEUSE during the year 1975 lie between 8 and 66 Bq/kg fresh (IHE/CEN , 1965-1983). The histogram of measured values and computed values is presented at fig. 12.

The fish contamination model consider the contamination of a typical carp of the MEUSE river, living in the sourroundings of HASTIERE (km 488)n while field measurements concern a lot of fishes sampled in the whole river ; these fishes present different growth rates and diet habits, and thus different contamination rates. These reasons explain that fish contamination observations present a wider variability than the contamination values computed by the model.


MER DU NORD
NOORDZEE
NORDSEE
NEDERLAND
IJSSELMEER
AMSTERDAM
ZWOLLE
DALMELO
HENGELO
ARNHEM
UTRECHT
s GRAVENHAGE
ROTTERDAM
Lobith
RHEIN
NIJMEGEN
Waal
Hedel
's-HERTOGENBOSCH
Grave
BREDA
MIDDELBURG
VLISSINGEN
VENLO
Belfeld
ROERMOND
Linne
Maasbracht
MAAS
DÜSSELDORF
RHEIN
RUR
ANTWERPEN
BRUGGE
GENT
DUNKERQUE
BELGIQUE
BELGIË
BRUXELLES
BRUSSEL
JÜLICH
Lanaken
Borgharen
MAASTRICHT
AACHEN
Visé
LIÈGE
DEUTSCHLAND
NAMUR
Mehaigne
Amay
Huy
TIHANGE
Engis
VALENCIENNES
DINANT
Hastière
Givet
CHOOZ
Viroin
Semois
CHARLEVILLE
LUXEMBOURG
TRIER
LUXEMBOURG
FRANCE
Chiers
Bar
MONTMEDY
THIONVILLE
MEUSE
REIMS
VERDUN
METZ
COMMERCY
NANCY
NEUFCHATEAU
MEUSE

<u>Figure 1</u>

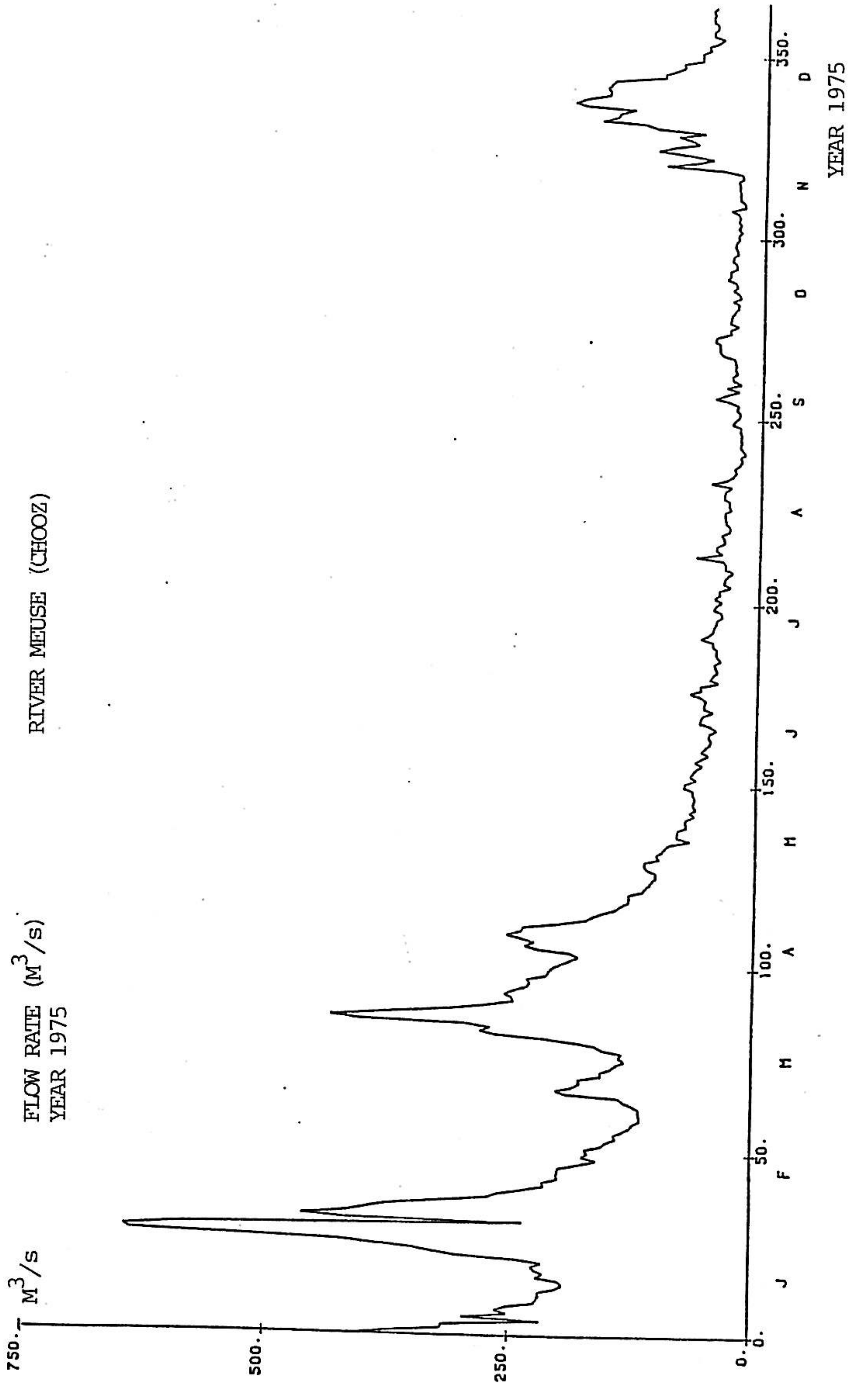

RIVER MEUSE (CHOOZ)
FLOW RATE (M^3/s)
YEAR 1975
M^3/s
750.
500.
250.
0.
0.
50.
100.
150.
200.
250.
300.
350.
J
F
M
A
M
J
J
A
S
O
N
D
YEAR 1975

Figure 2

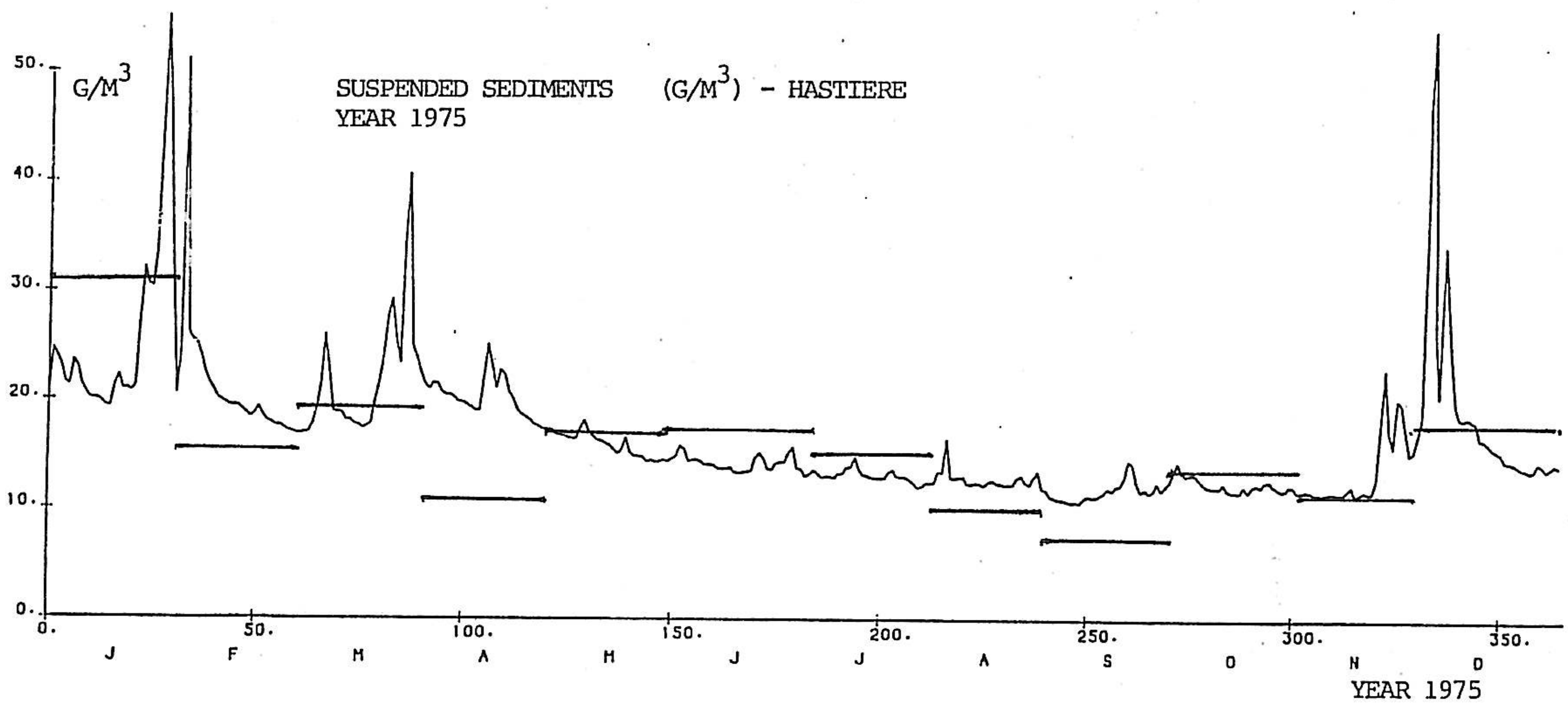

G/M3
SUSPENDED SEDIMENTS (G/M3) - HASTIERE
YEAR 1975
50.
40.
30.
20.
10.
0.
0.
50.
100.
150.
200.
250.
300.
350.
J
F
M
A
M
J
J
A
S
O
N
D
YEAR 1975

Figure 3

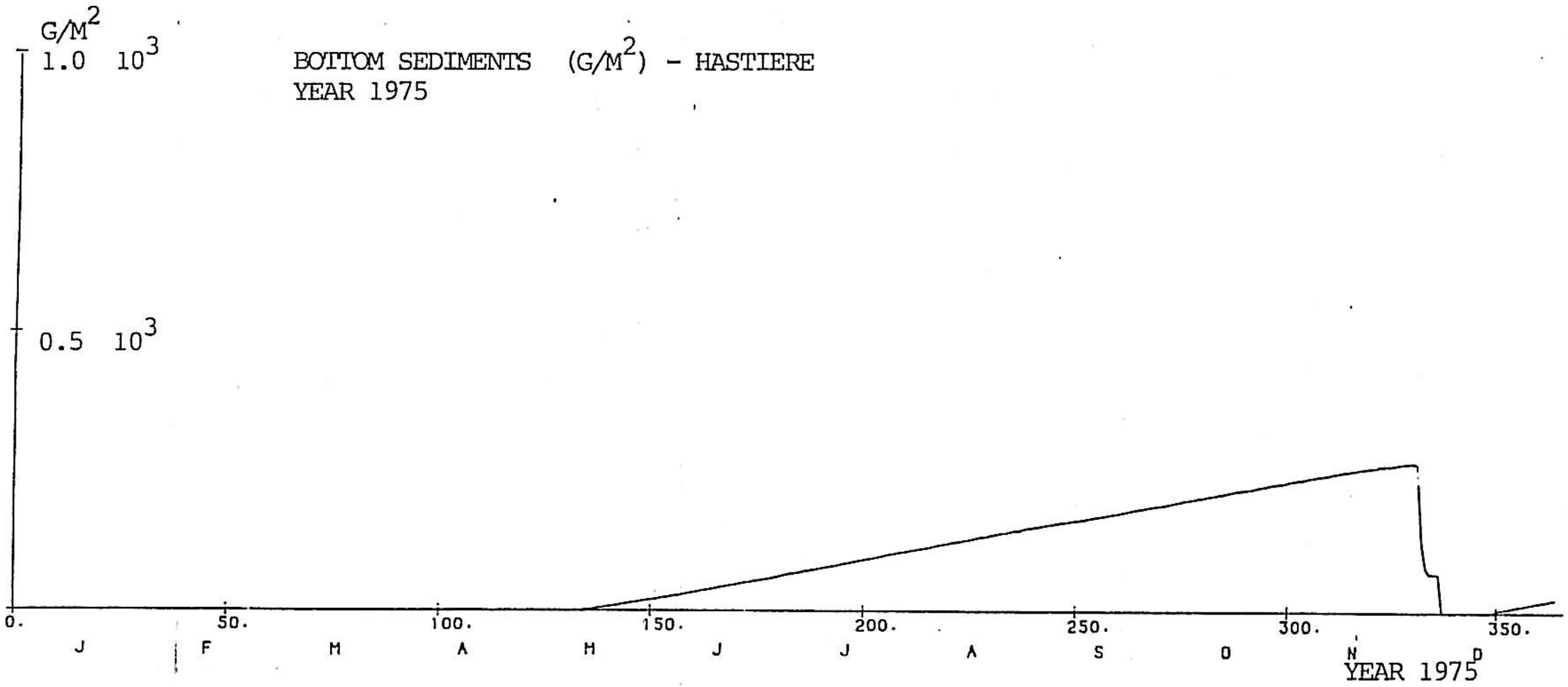

G/M²
1.0 10³
0.5 10³
BOTTOM SEDIMENTS (G/M²) - HASTIERE
YEAR 1975
0.
50.
100.
150.
200.
250.
300.
350.
J
F
M
A
M
J
J
A
S
O
N
D
YEAR 1975

Figure 4

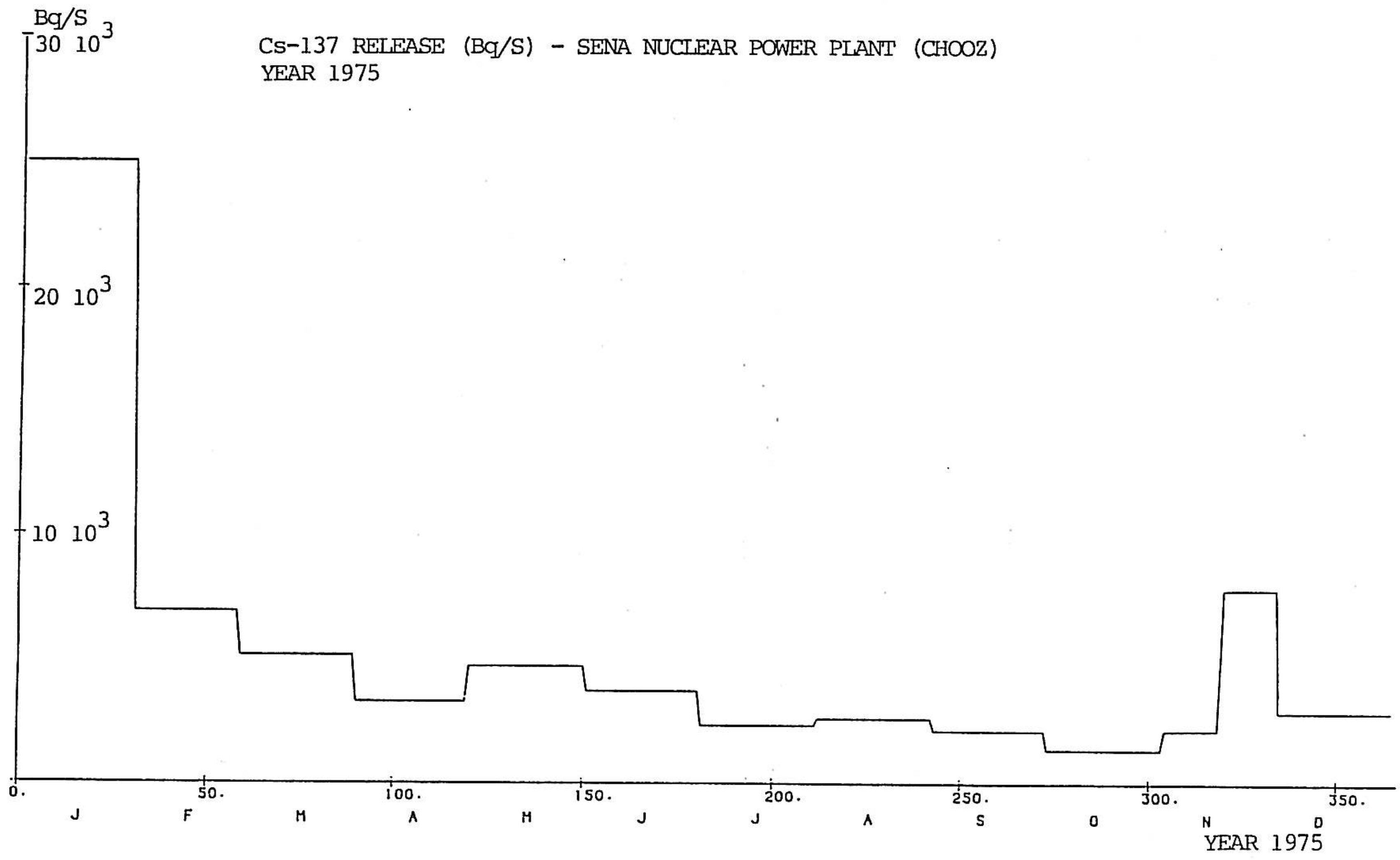

Bq/S
30 10^3
20 10^3
10 10^3
Cs-137 RELEASE (Bq/S) - SENA NUCLEAR POWER PLANT (CHOOZ)
YEAR 1975
0.
50.
100.
150.
200.
250.
300.
350.
J F M A M J J A S O N D
YEAR 1975

Figure 5

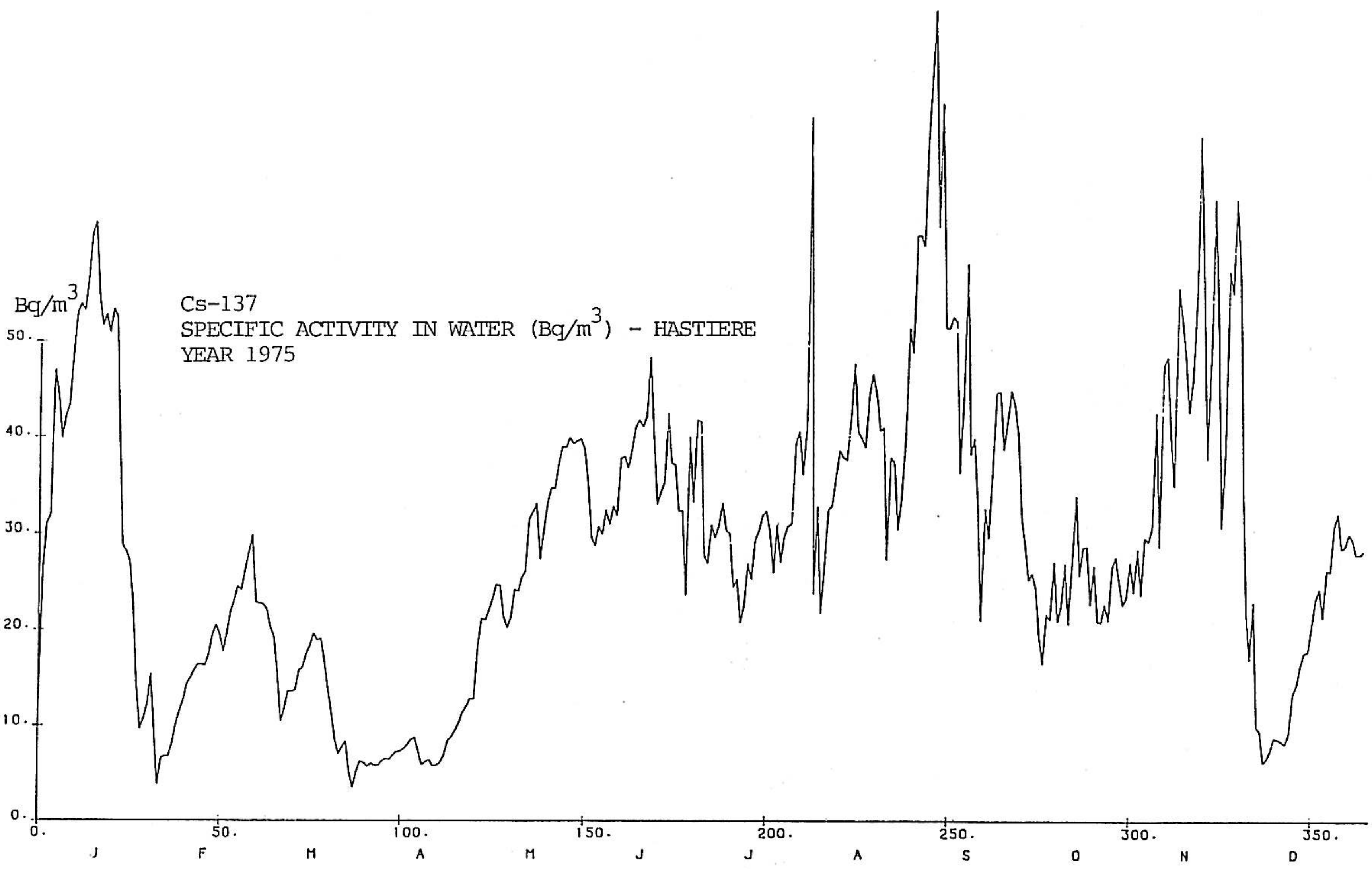

Bq/m3
Cs-137
SPECIFIC ACTIVITY IN WATER (Bq/m3) - HASTIERE
YEAR 1975
50.
40.
30.
20.
10.
0.
0.
50.
100.
150.
200.
250.
300.
350.
J
F
M
A
M
J
J
A
S
O
N
D

Figure 6

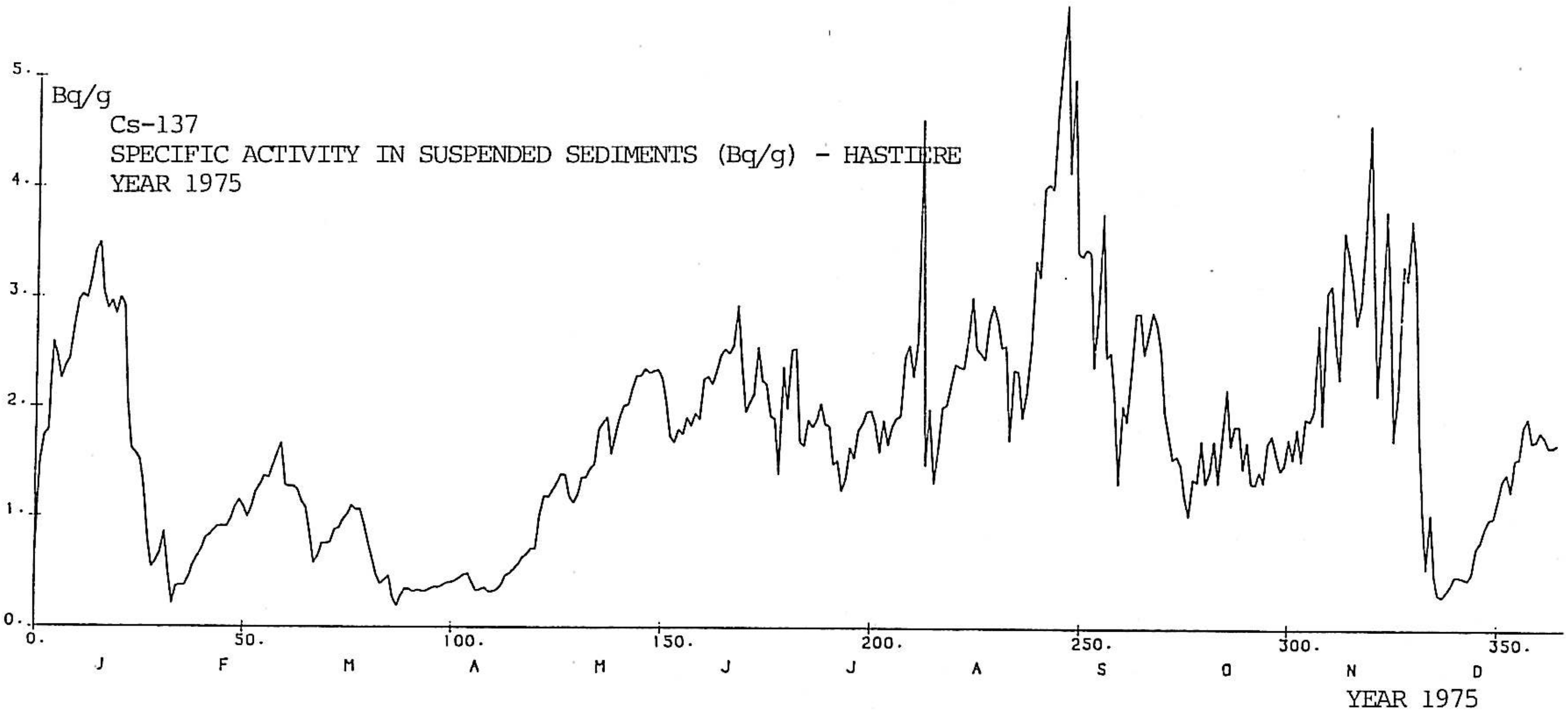

Bq/g
Cs-137
SPECIFIC ACTIVITY IN SUSPENDED SEDIMENTS (Bq/g) - HASTIERE
YEAR 1975
5.
4.
3.
2.
1.
0.
0.
50.
100.
150.
200.
250.
300.
350.
J
F
M
A
M
J
J
A
S
O
N
D
YEAR 1975

Figure 7

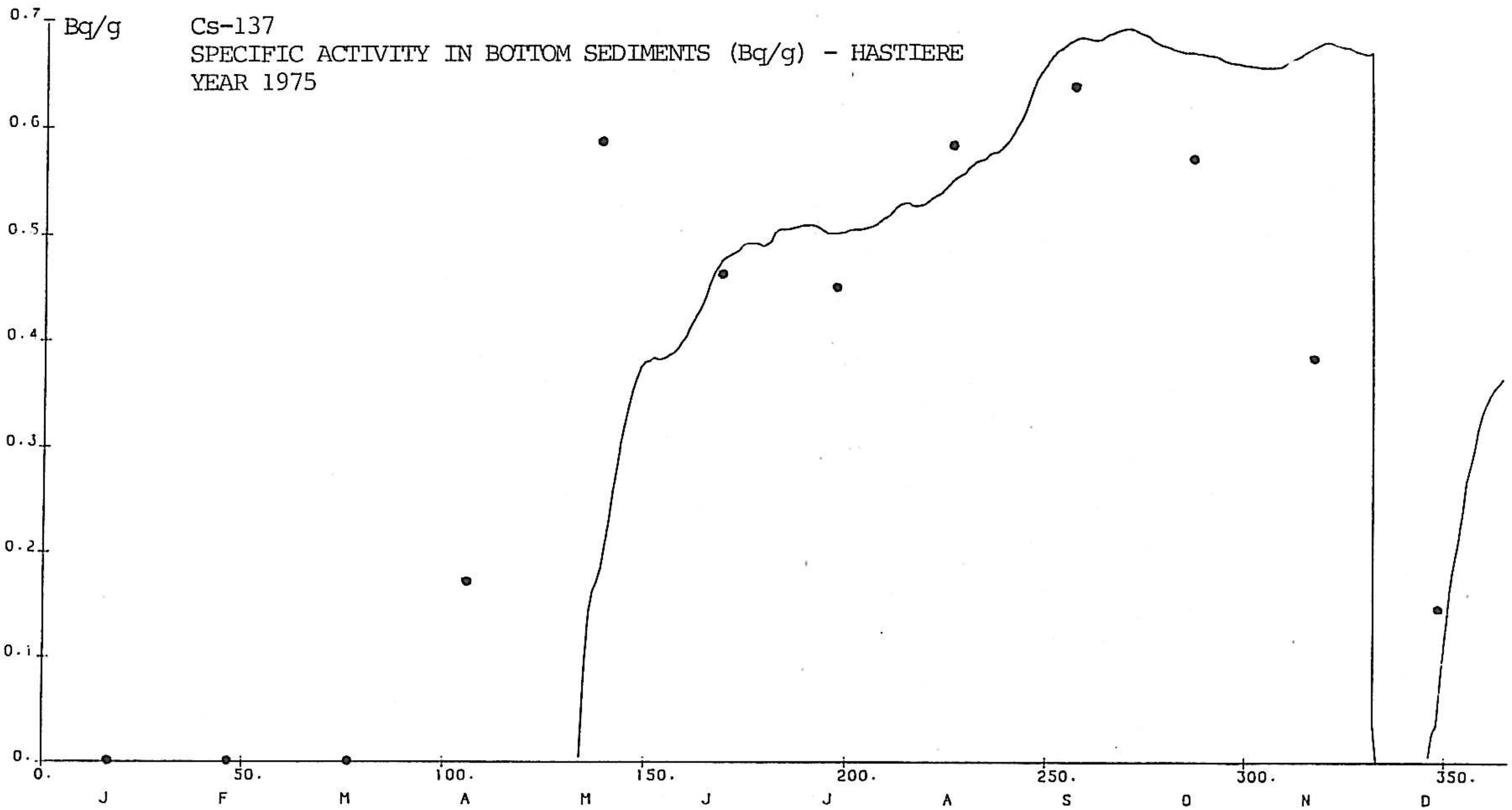

Bq/g
Cs-137
SPECIFIC ACTIVITY IN BOTTOM SEDIMENTS (Bq/g) - HASTIERE
YEAR 1975
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.
0.
50.
100.
150.
200.
250.
300.
350.
J F M A M J J A S O N D
YEAR 1975

Figure 8

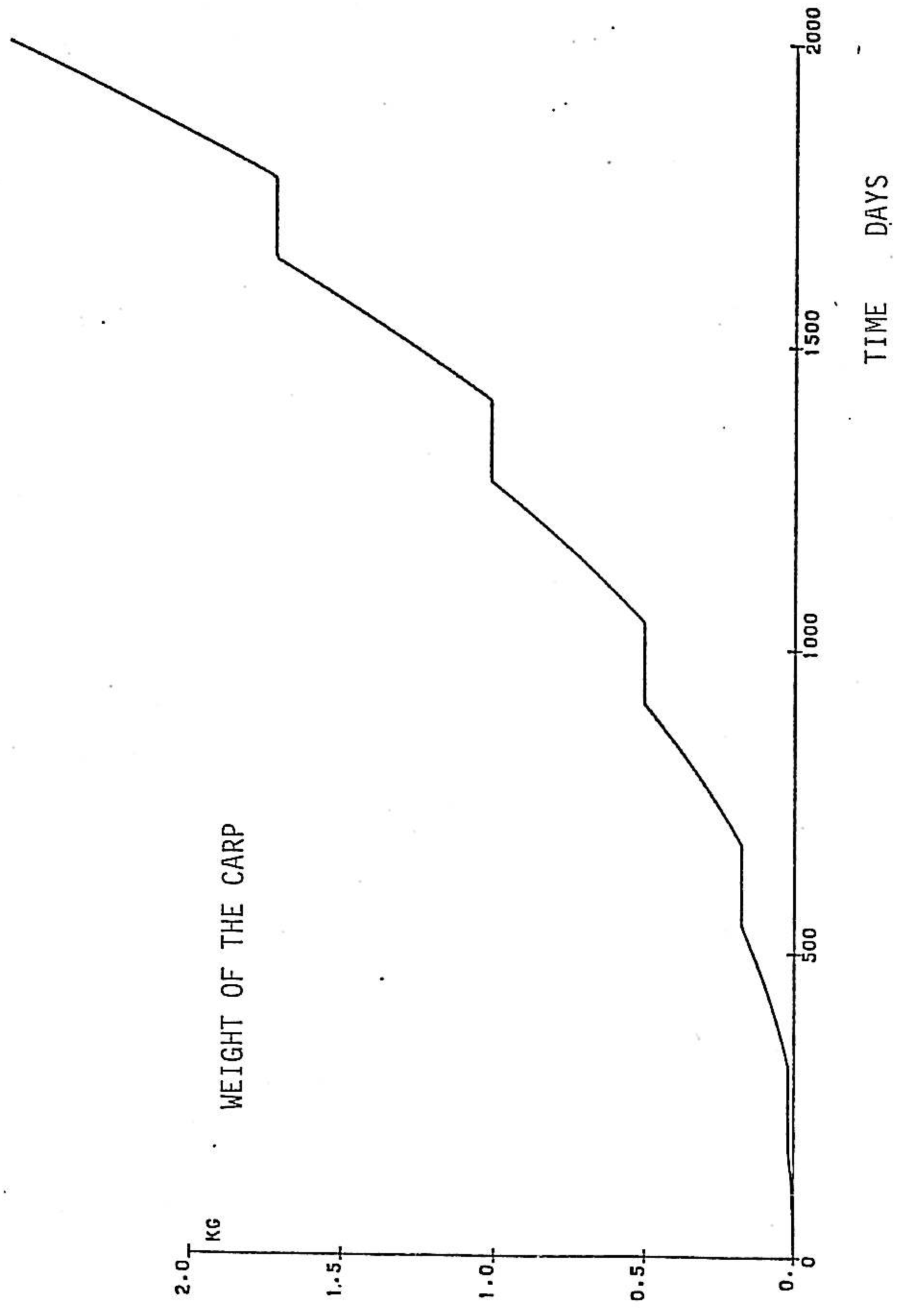
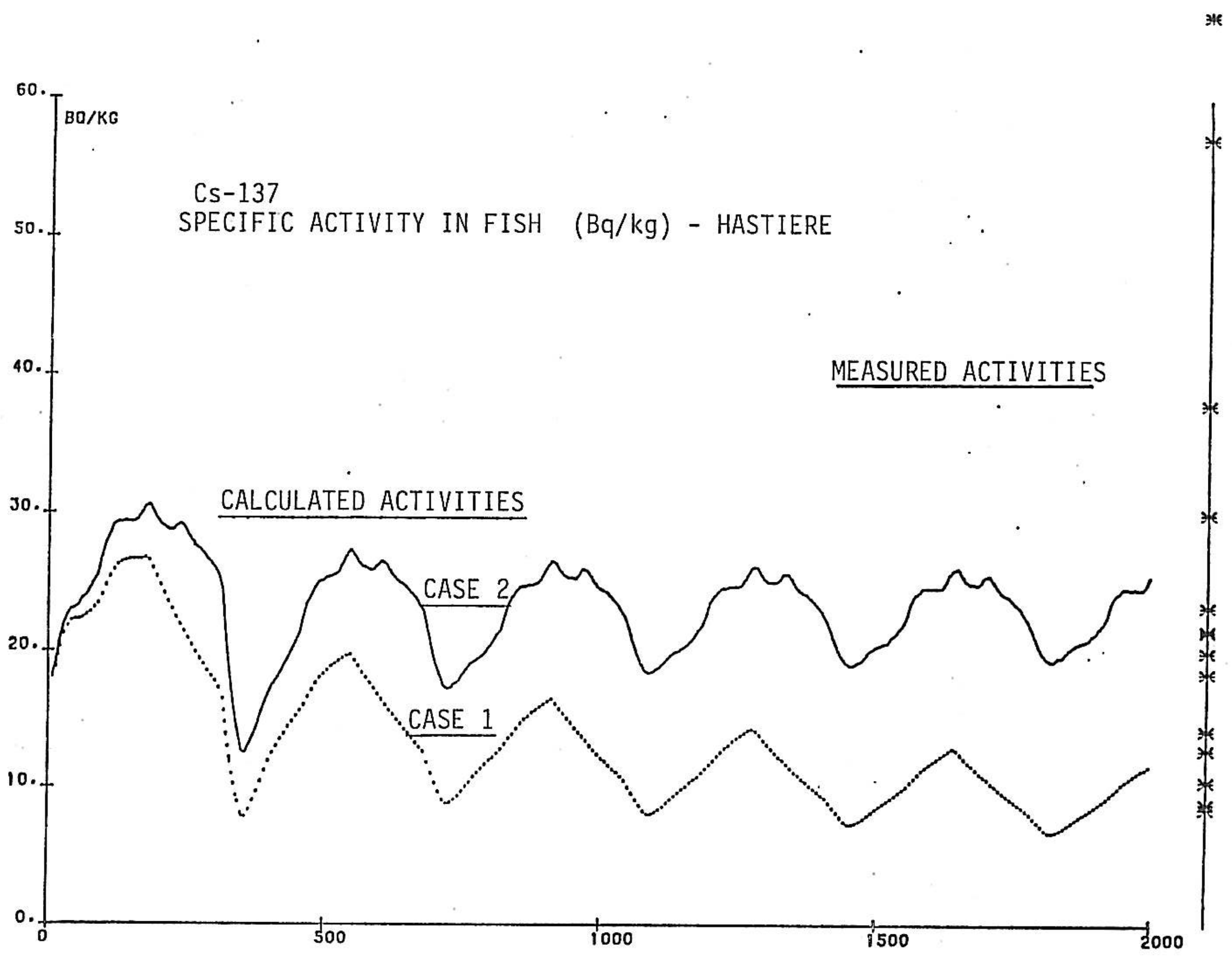

WEIGHT OF THE CARP
KG
2.0
1.5
1.0
0.5
0.
0
500
1000
1500
2000
TIME DAYS

Figure 9



Cs-137
SPECIFIC ACTIVITY IN FISH (Bq/kg) - HASTIERE
BQ/KG
60.
50.
40.
30.
20.
10.
0.
CALCULATED ACTIVITIES
CASE 2
CASE 1
MEASURED ACTIVITIES
0
500
1000
1500
2000
TIME DAYS

Figure 10

Cs-137 CONTAMINATION FLUXES OF FISH (Bq/D.kg) - HASTIERE

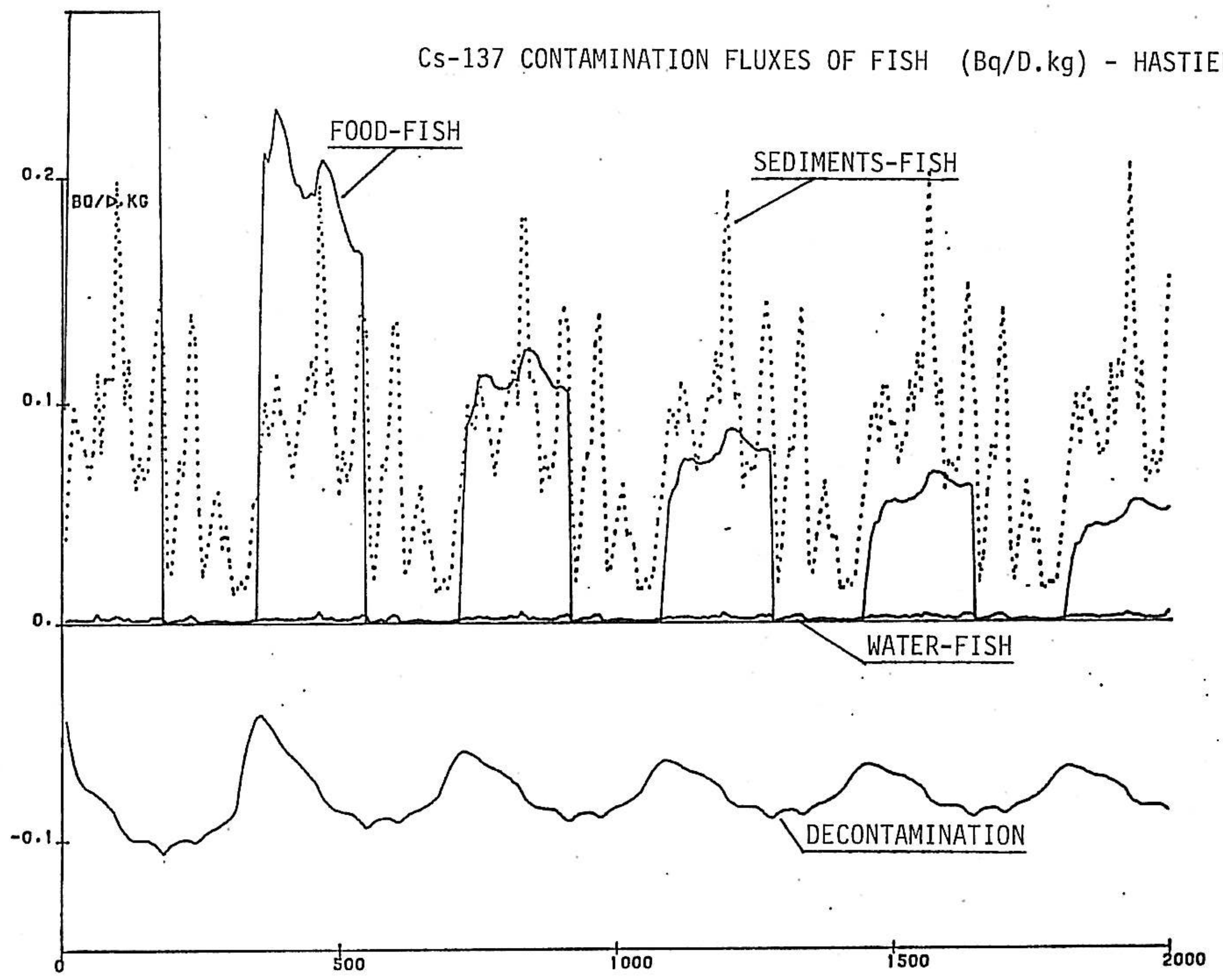


Figure 11

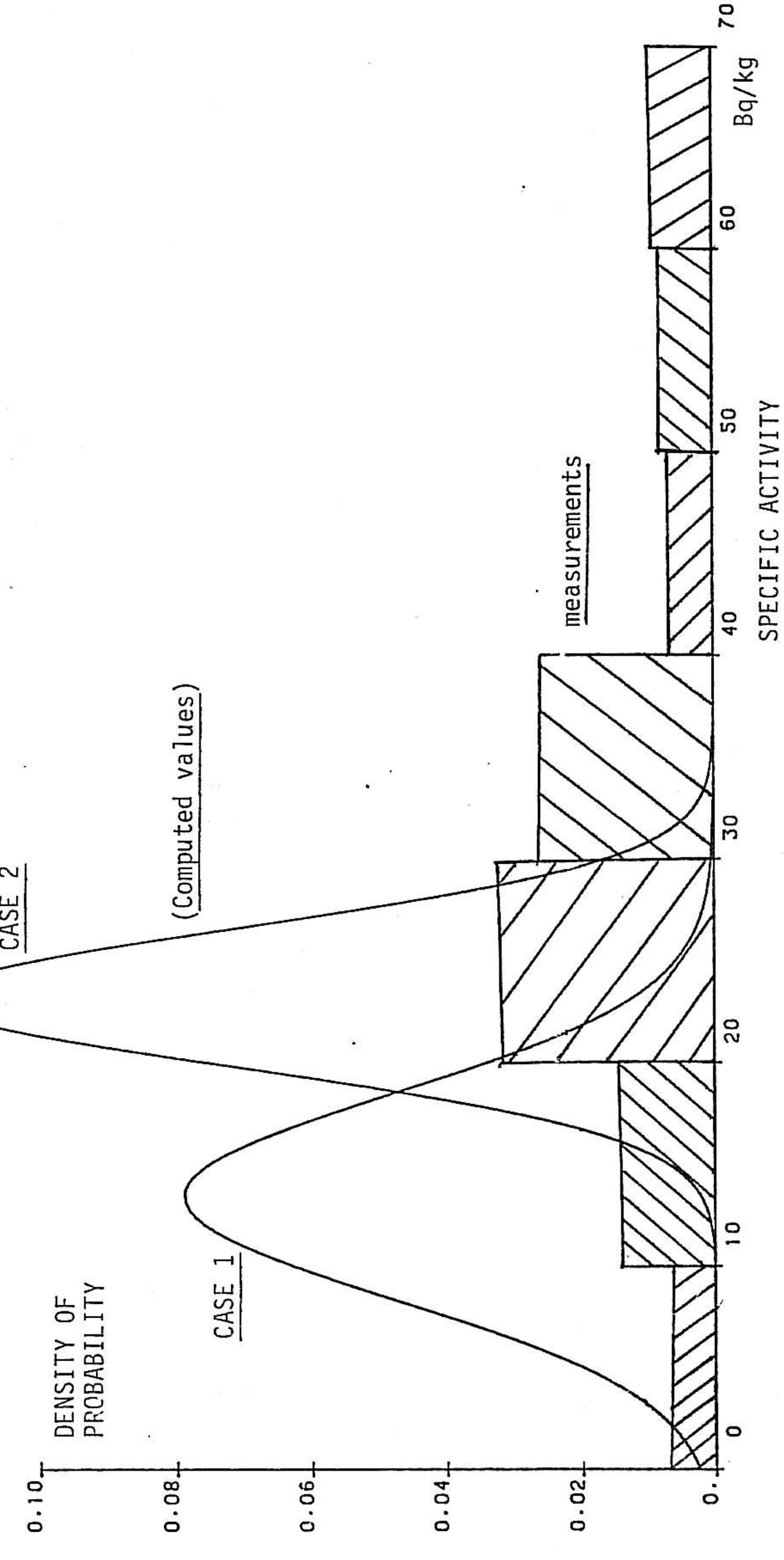

DENSITY OF PROBABILITY
0.10
0.08
0.06
0.04
0.02
0.
CASE 1
CASE 2
(Computed values)
measurements
0
10
20
30
40
50
60
70
SPECIFIC ACTIVITY
Bq/kg

Figure 12

BAYER, A., 1982, The radiological exposure of the population in the Rhine-Meuse region, CCE; Luxemburg.

BILLEN G.,DESCY J.P., EVERBECQ E., SERVAIS P., SMITZ J., 1985, Etude écologique de la Haute Meuse et modélisation du fonctionnement de l'écosystème aquatique,250 pp. Ministère de la Région Wallonne pour l'eau, l'environnement et la vie rurale

BOVARD P., GRAUBY A., FOULQUIER L., PICAT P., 1973, Etude radioécologique d'un bassin européen. Le Rhône. Stratégie et bilan. Colloque sur le comportement dans l'environnement des radioéléments rejetés par l'industrie nucléaire. Aix en Provence, France, 14-28 mai 1973, IAEA SM-172/60

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