IEAE CRP

THE ROLE OF SEDIMENTS IN THE ACCUMULATION AND TRANSPORT OF RADIONUCLIDES IN WATERWAYS

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MODELLING OF RADIONUCLIDES TRANSPORT BY SEDIMENTS IN WATERWAYS

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1. INTRODUCTION

Radionuclides released by nuclear plants in surface waters (lakes, rivers, ...) are not simply transported by the water. Other complex phenomena occur: adsorption/desorption to sediments, sedimentation and accumulation on river's bed, interaction with interstitial water, and eventually resuspension in the water column.

The aim of this work is the description and modelling of the major processes which govern the contamination of surface waters and sediments by radionuclides. This study is necessary to assess transfer of radionuclides to food chain and to man.

The model is a deterministic model, which formulates in a simple way the conservation of water, of sediments and of radionuclides. The model can be used in common rivers and impoundments, and should be easily extended to tidal rivers or estuaries.

The model is applied to the river MEUSE.

The results of the model are verified by comparison with available field measurements realized during routine surveys.

2. STRUCTURE OF THE MODEL

The behaviour and fate of radionuclides in surface waters cannot be described without a preliminary knowledge of the water and sediment transport.

The model is thus organized in three steps:

- $\underline{\text{step 1}}$: description and modelling of hydraulic and hydrodynamic variables of the river;
- step 2: description and modelling of suspended and bottom sediments, including processes of sedimentation and resuspension;

- step 3: description and modelling of the radionuclides concentration in water, in suspended sediment and in bottom sediment, including the physico-chemical interactions between these three compartments.

The model of radionuclides transport is thus composed of three coupled submodels, organized in a hierarchical structure (the output of submodel is the input of submodel 2, and the output of submodels 1 and 2 is the input of submodel 3).

Hydrodynamic processes present basically a three-dimensional non-stationary feature. A three-dimensional description of hydrodynamic and sediment transport is a very difficult task, which needs large computation capacities. The calibration of such three-dimensional models is often impossible (lack of information and measurements: 3D-model = 3D-data).

In the most frequent situations, it is not necessary to achieve a complete 3D description, and a simplification can be made. In shallow waters, a 2D description is generally obtained by integration over the depth; however, a well-suited 2D description, designed to describe explicitly the sedimentation and erosion processes, is obtained by integration over the width (laterally - averaged model). Such a model has been proposed by ONISHI (SERATRA model, ONISHI Y, 1982); there is a good agreement between computed values by this model and field data, but this kind of model is heavy to work: the model incorporates a large amount of unknown parameters; the model needs complicated boundary conditions and numerous specific field data (for calibration).

In the case of a river, the most efficient way is the one-dimensional description, obtained by integration of the equations over the cross-section (computation of cross-section averaged values). The equations are in this case quite simple, the number of parameters is reduced, and more simple boundary conditions are required. Hence in this study, the choice is made to construct a one-dimensional non-stationary model.

3. THE HYDRAULIC AND HYDRODYNAMIC SUBMODEL

This submodel integrates the whole set of data describing the physical geometry of the river (width, slope of the bottom, slope of the banks, location of locks and dams, ...) and the constraints imposed to the river flow (e.g. water levels for navigation, ...).

This hydrodynamic submodel calculates, at each point and each time, the hydrodynamic variables: river flow, depth, cross-section, velocity, bottom stress, ... (cross-section averaged values). The input data are the river flow measurements at one or several stations.

The equations translate in a mathematical form the conservation of water and the equilibrium of the forces :

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

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

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t
          : time (s);
                  longitudinal coordinate along the river axis (m);
A(x,t)
             cross-section area (m<sup>2</sup>);
u(x,t)
                 flow velocity (cross-section averaged) (m/s);
q(x,t)
                  linear lateral inflow (m^2/s);
Q_{i}(t)
                  inflow (m3/s) of the tributary i located at the
                  position x;;
\delta(x-x_i)
                  Dirac function (m^{-1});
           :
                 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);
                 width of the river (cross-section averaged) (m);
L(x,t)
           :
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Chezy coefficient ($C^2 = m/s^2$).

C(x)

4. THE SEDIMENT SUBMODEL

The adsorption/desorption of radionuclides between water and sediment particles appear as the basic process which governs the behaviour of radionuclides in surface waters. However, this process is highly dependent on the physical (and chemical) characteristics of the sediment; particularly, radionuclides are strongly adsorbed by small particles (clay and silt), and few by large particles of sediment.

Hydrodynamic processes of sedimentation resuspension are also influenced by the grain size (and other physico-chemical properties) of the sediment: large particles are characterized by a high settling velocity, while small particles fall slowly, and easily remain in the water column.

For hydrodynamic and radiologic considerations, it is thus necessary to take into account physical properties of the sediments; generally, sediment models consider different grain size classes of particles (see table 1).

The sediment submodel developed in this study is a multiclasses model (N classes, where N is variable). To be able to describe the sedimentation/resuspension mechanisms, it is also necessary to consider two compartments (for each class of particles): the suspended sediment (g/m^3) , and the bottom sediment (g/m^3) .

The different assumptions and parametrizations which underlie the sediment submodel are :

the sedimentation flux is considered proportional to the concentration of suspended sediment, and proportional to the difference between the bed shear stress and a critical deposition shear stress, with one critical deposition shear stress per class of sediment;

- 2) the resuspension flux is considered proportional to the difference between the bed shear stress and a critical resuspension shear stress, with one critical shear stress per class of sediment; the resuspension rate depends on the quantity and cohesiveness of the bottom sediment;
- 3) the cohesiveness of clay and silt bottom sediments is a function of the residence time of the bottom sediments on the bed;
- 4) the source of sediments in the system is the supply of sediments by tributaries, as the result of the global erosion process in the basin; this input of sediment is incorporated in the model via a statistical relation (the sediment supply of the tributary i is related to the flow Q_i of this tributary).

The modelling of sediment supply by tributaries is one of the major problems encountered in the design of the sediment submodel: field measurements of sediment supply by tributaries are generally scarse, and the statistical correlations that can be established between supply of sediment and flows of tributaries present wide variabilities. This point is a factor which contributes to increase the uncertainty associated with the predictions of sediment models.

The equations which govern the sediment concentrations are :

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

$$\frac{\partial}{\partial t} A SS_{j} + \frac{\partial}{\partial x} (AuSS_{j}) = \sum_{i} Q_{i} SS_{j,i} \delta(x-x_{i}) - SED_{j} + RS_{j}$$
 (3)

bottom sediments : (class j, j = 1,N)

$$\frac{\partial}{\partial t} BS_{j} = SED_{j} - RS_{j}$$
 (4)

where

$$SED_{j} (x,t) \begin{cases} = w_{j} \cdot SS_{j} \cdot L \left(1 - \frac{\tau_{b}}{\tau_{S,j}}\right) & \text{if } \tau_{b} < \tau_{S,j} \\ = 0 & \text{if } \tau_{b} \geqslant \tau_{S,j} \end{cases}$$

$$RS_{j} (x,t) = e_{j} \cdot L \cdot (\frac{\tau_{b}}{\tau_{r,j}} - 1) \qquad \text{if} \quad \tau_{b} > \tau_{r,j}$$
$$= 0 \qquad \qquad \text{if} \quad \tau_{b} \leq \tau_{r,j}$$

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

 $SS_{j}(x,t)$: concentration of suspended sediment,

class j (g/m^3) ; cross-section averaged value;

 $SS_{j,i}(t)$: concentration of suspended sediment,

class j (g/m^3) ; cross-section averaged value;

 $BS_{j}(x,t)$: concentration of bottom sediment,

class j (g/m) ;

 Q_i (t) : flow (m³/s) of the tributary i , located at the

point x;;

 $SED_{j}(x,t)$: 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/m².s);

L : width of the river (m) ;

 τ_b (x,t) : bed shear stress (g/m.s²);

 $\tau_{s,j}$: critical sedimentation shear stress, class j (g/m.s 2);

 $\tau_{r,j}$: critical resuspension shear stress, class j (g/m.s²);

 ρ : specific mass of water (g/m³).

5. THE RADIONUCLIDES SUBMODEL

The radionuclides submodel must describe the concentration or activity of radionuclides in water (Bq/m^3), in the suspended sediment (Bq/g), and in the bottom sediment (Bq/g). As the radionuclides released in the river adsorb preferably to small particles, it is necessary to describe the activity of radionuclides in the different sediment classes.

The radionuclides submodel includes then the mechanisms of :

- 1) transport by water;
- 2) transport by sediment;
- 3) adsorption of dissolved radionuclides to sediments (suspended and bottom sediments);
- 4) desorption of adsorbed radionuclides;
- 5) radioactive decay.

The assumptions and parametrizations which underlie the radionuclides submodel are :

- 1) the radionuclides adsorbed on sediments are considered to follow the particles motion: transport, sedimentation, or resuspension;
- 2) the global interaction water/sediment is the sum of the processes of adsorption and desorption between water and each class of particles;
- 3) for each class j of sediments, the adsorption process is considered proportional to the activity in water, the desorption process proportional to the activity in sediment; the kinetics of these reactions are related by a coefficient $(K_{d,j})$ such as that
 - the relative rates of adsorption/desorption decrease if the concentrations in sediment is high;
 - if an equilibrium is reached, the ratio of activity in sediment (Bq/g) to the activity in water (Bq/m 3) is equal to $K_{\rm d,j}$ (m 3 /g).

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

water

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

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

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

$$- \lambda A a_W$$

suspended sediments

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

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

$$+ \frac{SS_{j}}{1 + K_{d,j} \ SS_{j}} \frac{A}{\tau_{j}} K_{d,j} \cdot a_{W}$$

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

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

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

$$(j = 1,N)$$

bottom sediments

$$\frac{\partial}{\partial t} (BS_{j} \cdot a_{BS,j}) = SED_{j} \cdot a_{SS,j} + RS_{j} a_{BS,j}$$
$$- \lambda BS_{j} \cdot a_{BS,j} \qquad (j = 1,N)$$

 a_{lJ} : activity in water (Bq/m³);

 $a_{SS,j}$: 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_i : characteristic time of adsorption process (s⁻¹);

 $K_{d,j}$: distribution coefficient (m³/g);

 λ : characteristic frequency of radionuclide decay (s⁻¹).

6. APPLICATION TO THE CASE OF THE RIVER MEUSE

The model has been applied to assess the transport of Cs-137 released in the river MEUSE by the Nuclear Power Plant SENA (located at CHOOZ, km 473) (see fig. 1).

The model has been used to realize a non-stationary simulation of the year 1975: during the year 75, the Nuclear Power Plant SENA released significant quantities of Cs-137, and a few data are available from routine survey (IHE/CEN, 1965-1983), so that some comparisons can be made between calculated values and field concentrations.

Available information from field measurements

- 1) flow rates: mean daily values of the river flow, measured at two stations (CHOOZ, km 473; AMPSIN-NEUVILLE, km 566);
- 2) sediments:
 - 1 grain size distribution;
 - 24 measurements of suspended sediment concentration :
- 3) radionuclides:
 - 12 monthly mean values of Cs-137 releases by SENA Nuclear Power Plant;
 - 12 measurements of Cs-137 activity in bottom sediments at HASTIERE (km 488);
 - 12 measurmeents of Cs-137 activity in bottom sediments at IVOZ-RAMET (km 579).

Remark : no information is available about Cs-137 activities in suspended sediment and in water.

Hydrodynamic submodel

This model calculates hydrodynamic variables (river flow, depth, velocity, bed shear stres, ...) at each point and at each time (cross-section averaged values). The geographical extension of the numerical scheme covers the French section and the Belgian section of the river MEUSE (km 0 - km 600).

Fig. 2 shows a graph of the river flow during the year 1975 at the station of CHOOZ (km 473) (m^3/s , measured values), and fig.2.b a graph of the river flow at IVOZ-RAMET (km 579) (m^3/s , computed values).

River flows appear to be quite large during the 4 first months of the year 1975 (Q > 250 m 3 /s, u < 1 m/s); from May to November a period of low flow takes place, followed by a flood period in December. This sequence is a typical yearly flow regime of the river MEUSE.

Sediment submodel

In the present application of the sediment model, two particles size classes are routed:

class 1 : clay + fine silt

class 2 : coarse silt + sand.

Parameter calues adopted for settling velocities, critical sedimentation shear stress, critical erosion shear stress are typical values given in the literature.

The model computes the concentration of suspended sediment in the water column (g/m^3) and the concentration of bottom sediment on the bed (g/m^2) (cross-section averaged values) at each point and at each time.

Fig. 3 and fig. 4 show a graph of computed values of suspended sediment concentrations and bottom sediment concentrations at HASTIERE (km 488) during the year 1975. Suspended sediment concentrations vary around 20 g/m 3 , excepted during flood events. Sedimentation cannot take place during high flows, and accumulation rate of bottom sediments is rather constant during low flow period. The flood event of December produces an erosion and a complete wash-out of the bottom sediment.

Fig. 5. and fig. 6 show the same typical behaviour of sediments (computed values) at IVOZ-RAMET (km 579).

Computed concentrations of suspended solids are in rather good agreement with the general trend of field observations (the model is not able to reproduce random fluctuations due to natural variability of sediment concentration, to dredging works or to small scale erosion events in the basin).

Radionuclides submodel

The flux of Cs-137 released in the river at the Nuclear Power Plant SENA (km 473) in 1975 is greater during high flows period than during low flow periods ($\underline{\text{fig. 7}}$, monthly-averaged values).

The radionuclides submodel calculates activities in water (Bq/m³), in suspended sediment class 1 and 2 (Bq/g), and in bottom sediment class 1 and 2 (Bq/g). The adsorption/desorption process between radionuclides in water and radionuclides adsorbed to sediments (class 1 and 2) is determined by the values of the partial distribution coefficients $K_{d,1}$ and $K_{d,2}$.

In the present study, the values of the partial distribution coefficient used are :

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

 $K_{d,2} = 0$.

As the relative concentration of class 1 sediments in the river MEUSE is around 70 %, and the class 2 sediment 30 %, these values of $K_{d,j}$ correspond to a global distribution coefficient

$$K_{d} \sim 0.05 \text{ m}^{3}/\text{g}$$
 (= 50000 ℓ/kg)

according to the most common value given in the literature for Cs-137 under normal physico-chemical conditions.

The Cs-137 activities in water, in suspended sediment, and in bed sediment at the location HASTIERE (km 488, that is 15 km downstream the release point) computed by the model (simulation of the year 1975) are presented at <u>fig. 8</u>, <u>fig. 9</u>, and <u>fig. 10</u>. Fig 10 shows a very good agreement between observed and computed values.

Fig. 11, 12 and 13 present the results of the simulation of Cs-137 in water, suspended sediment and bottom sediment during the year 1975 at IVOZ-RAMET (km 579, that is 106 km downstream the release point). Good agreement is obtained between observed and computed values (fig. 13). Discrepancies can be noted, but these discrepancies reflect the effects of the biased sampling process (sampling of bottom sediment in a dead zone of the flow, where only slow accumulation exists; these samples give thus "smooth" values).

Transport of radionuclides by water and sediment

As the model computes water fluxes, sediment fluxes, and activities in water as in sediment, it is possible to assess (at each point, at each time) the flux of radionuclides transported by water and by sediments through the cross-section of the river.

Fig. 14 shows the transport of Cs-137 in liquid phase and in solid phase at HASTIERE (km 488). The transport of Cs-137 by sediments is more important than the transport by water (55% - 45%), especially during high flows when sediment concentrations are high.

7. CONCLUSIONS

The model of radionuclides transport in rivers by water and sediments presented in this study is able to reproduce the complex processes of sedimentation, accumulation, erosion, adsorption and desorption. Without any calibration, using only values of parameters given in the literature (field or labo experiments), it is possible to obtain a good agreement - at least at the order of magnitude - between computed and observed values.

This kind of model is probably a good compromise between :

- a natural inclination to complexity, to obtain a representation as close as possible to the reality;
- a necessary tendency to simplification, to obtain an utilizable tool.

CLASSIFICATION OF SEDIMENT ACCORDING TO SIZE

	Lower limit (µm)	Upper limit (µm)
,		
Class 1 : CLAY	-	4
Class 2 : FINE SILT	4	16
Class 3 : COARSE SILT	16	64
Class 4 : SAND	64	_

Table 1

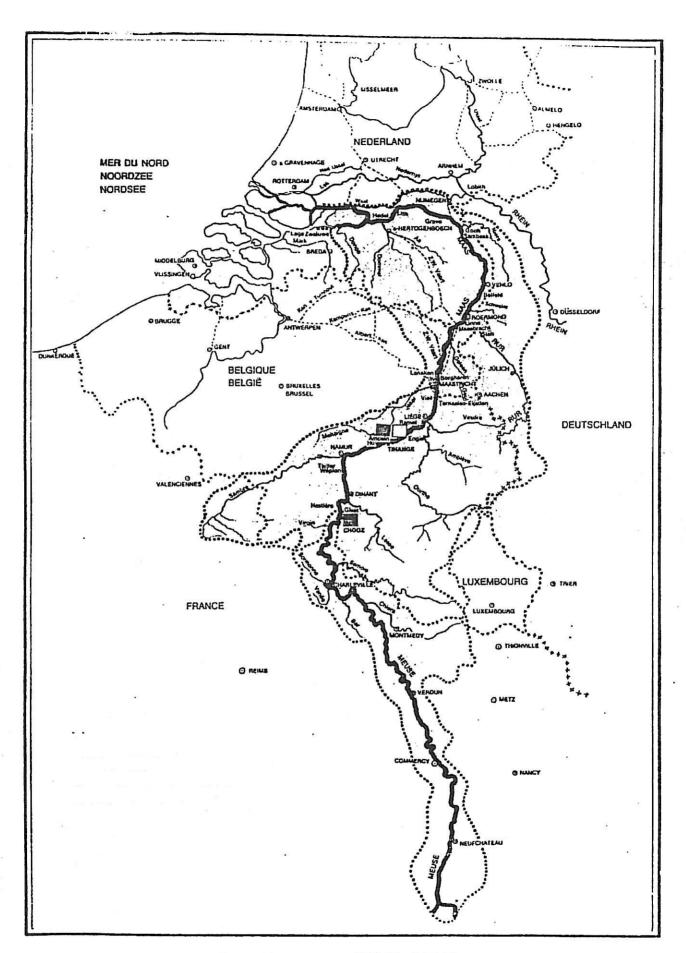
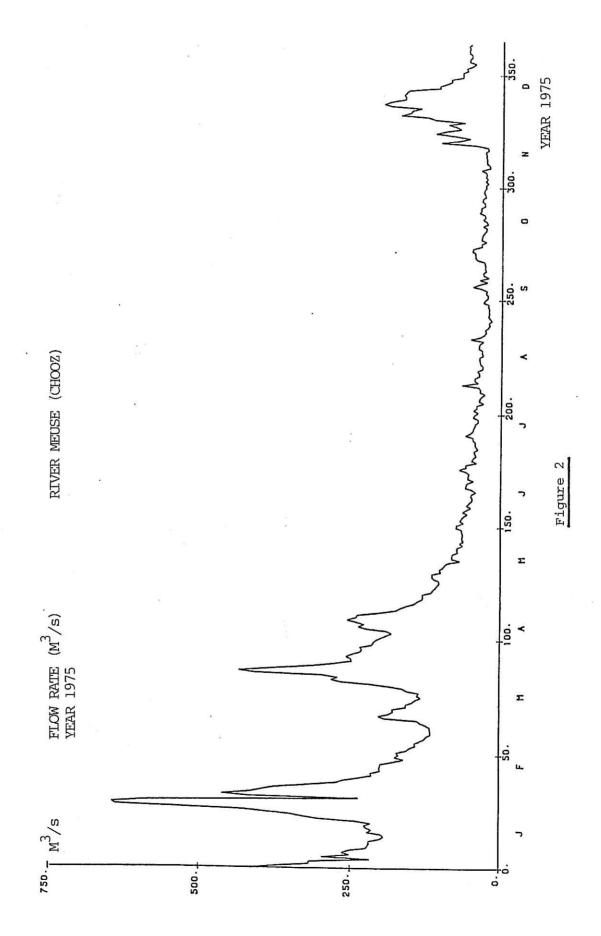
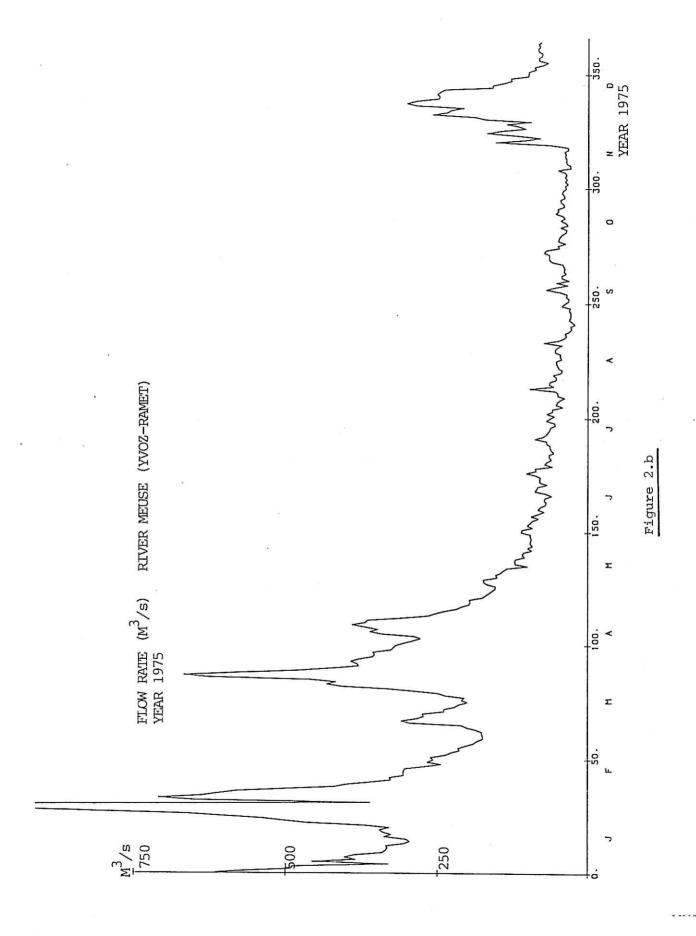
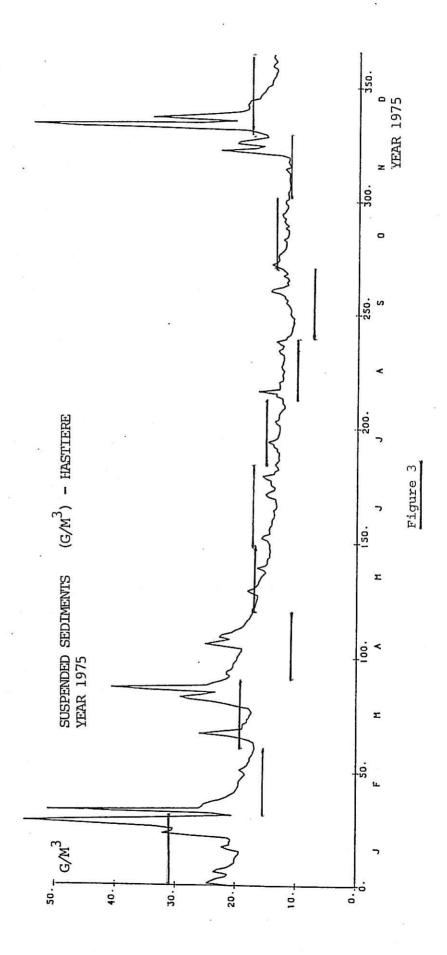


Figure 1

MEUSE BASIN







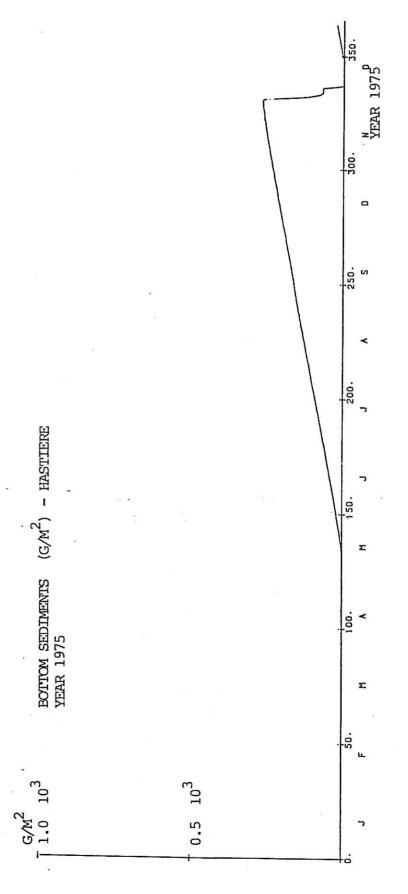
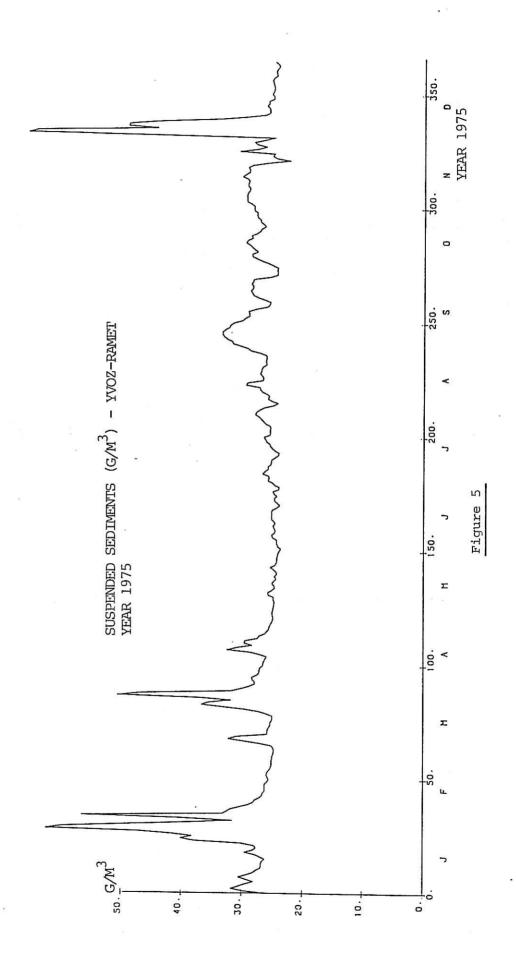
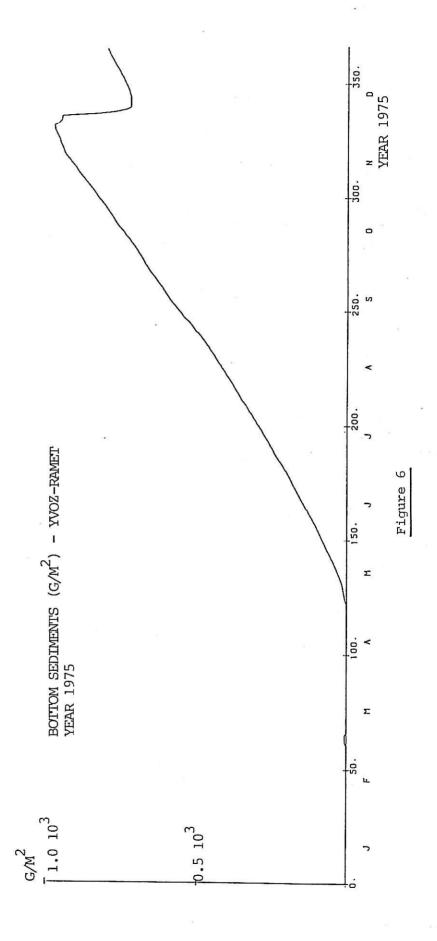
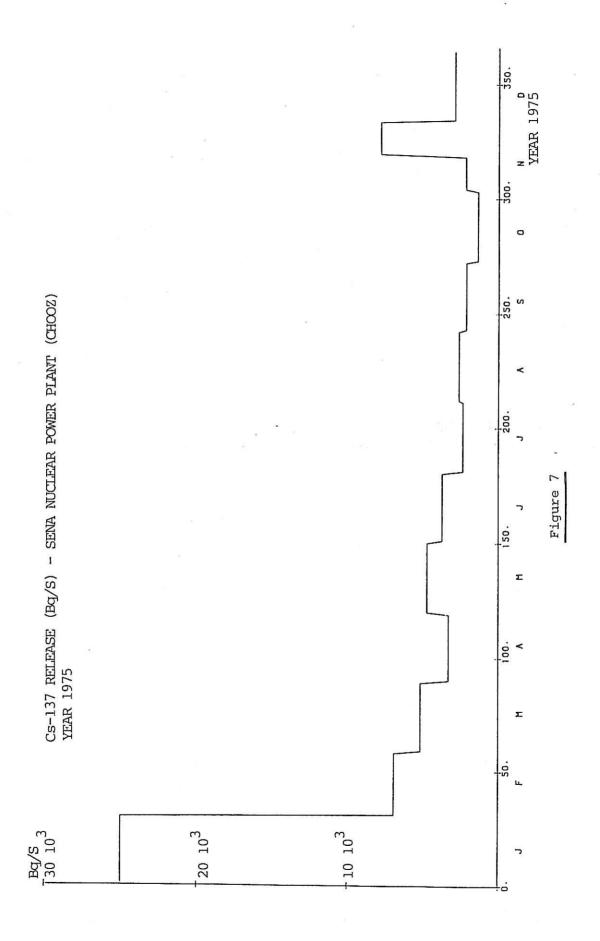
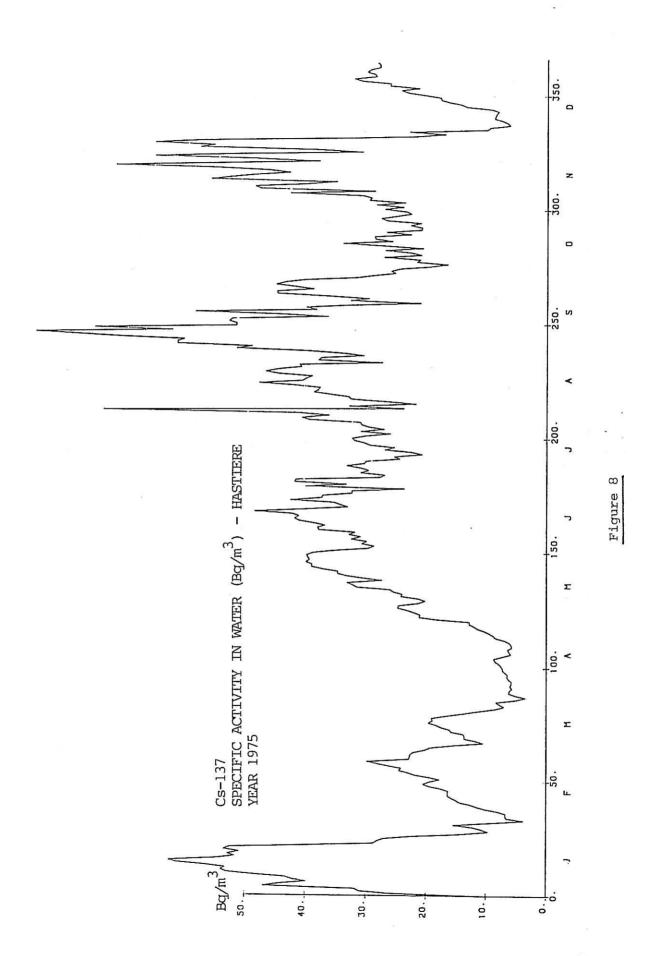


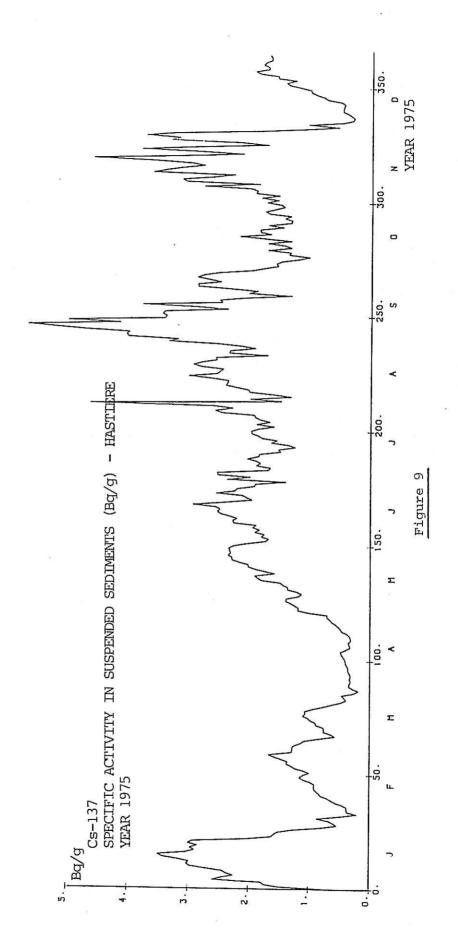
Figure 4

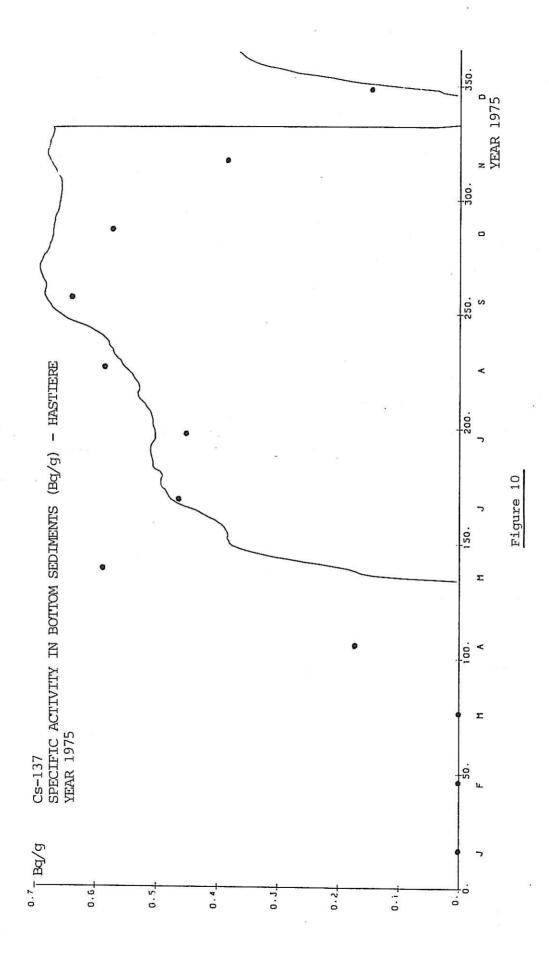


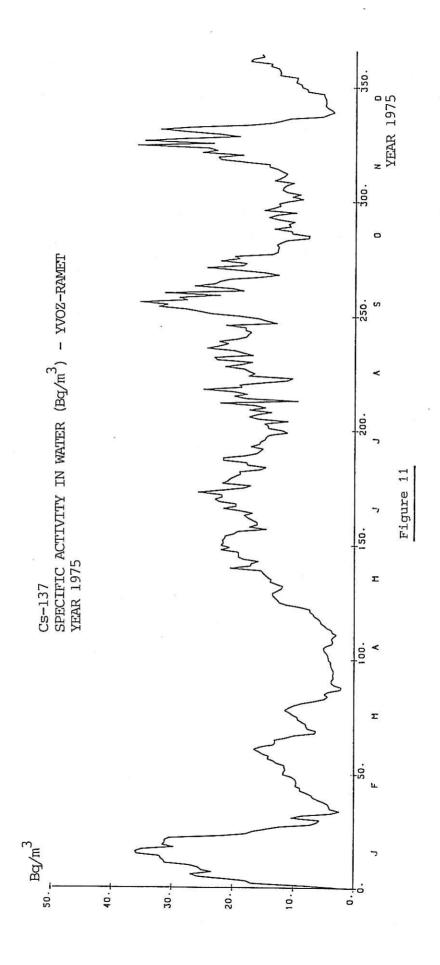


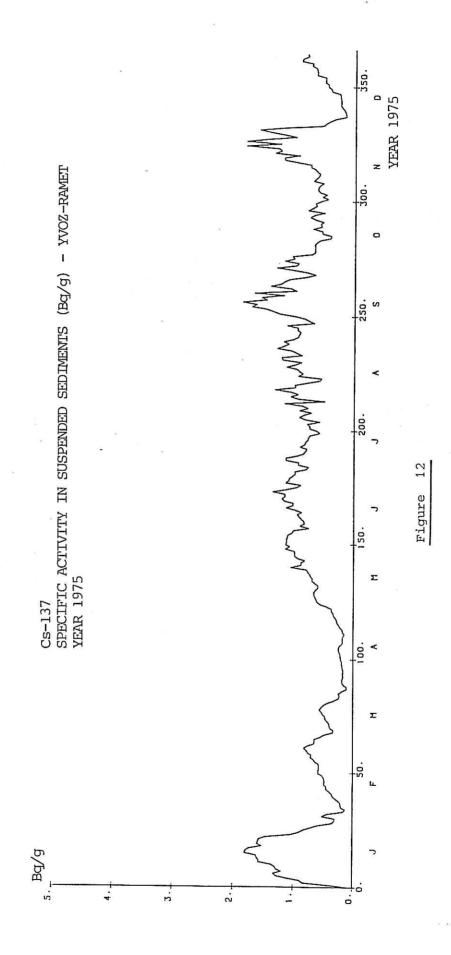


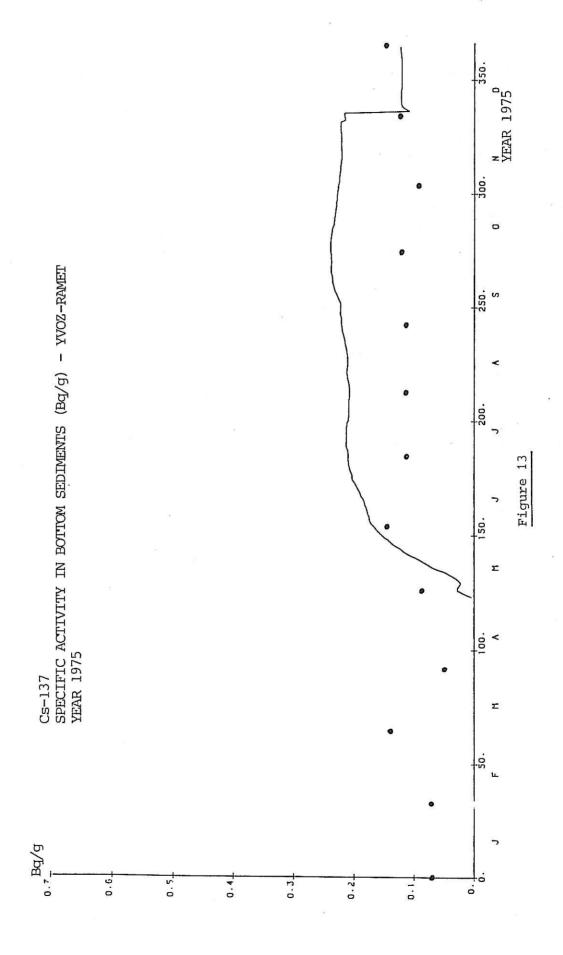


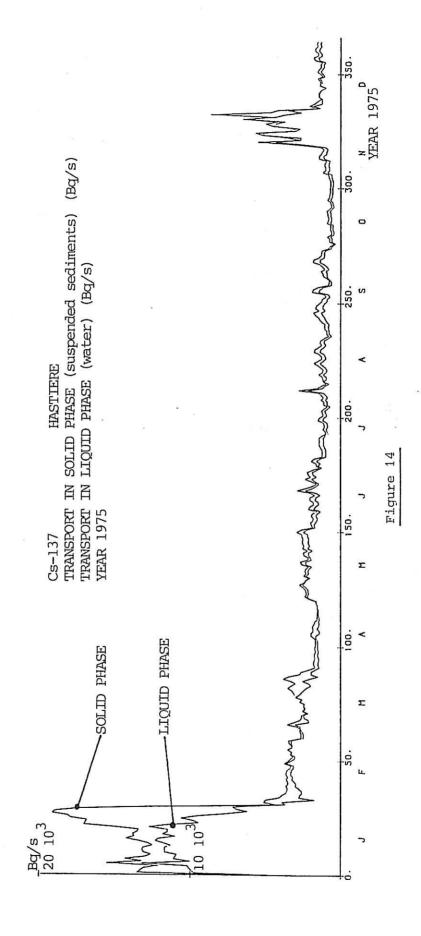












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