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PERFORMANCES OF MODELS FOR PREDICTING MERCURY  
CONCENTRATIONS IN FRESH-WATER FISH AFTER  
CHRONIC RELEASES INTO RIVERS  
— APPLICATIONS TO A TEST SCENARIO OF THE BIOMOV5 STUDY —

January 1992

Orihiko TOGAWA

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Performances of Models for Predicting Mercury Concentrations  
in Fresh-water Fish after Chronic Releases into Rivers  
- Applications to a Test Scenario of the BIOMOVs Study -

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(Received December 3, 1991)

The performances of assessment models for estimating the transfer and bioaccumulation of mercury in fresh-water ecosystems were tested by being applied to a test scenario proposed in an international cooperative study BIOMOVs. Two kinds of models have been developed to estimate mercury concentrations in fish after chronic releases into rivers. One uses a bioaccumulation factor approach which is applied to ecosystems in equilibrium, whereas the other is a dynamic model which considers the change of the concentrations in water and the metabolism in fish.

The success of the models tested by three different scenarios depended upon whether mercury was in equilibrium in the environment. For the scenario where mercury concentrations reached equilibrium, the first model performed satisfactorily. For the scenario where equilibrium was not attained, the first model was not adequate but the second model could predict more accurately. The limitations of applications were suggested for the two models employed here.

Keywords: Environmental Transfer Model, Model Performance, BIOMOVs,  
Fresh-water Ecosystem, Mercury Concentration,  
Bioaccumulation Factor Approach, Dynamic Model,  
Equilibrium Condition

水銀の河川への長期的な放出に伴う淡水魚中濃度を予測するモデルの性能  
—— BIOMOV5 計画のテストシナリオへの参加 ——

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(1991年12月3日受理)

国際共同研究 BIOMOV5で提案されたテストシナリオに参加することによって、淡水の生態系における水銀の移行・蓄積を推定する評価モデルの性能を検証した。水銀の河川への長期的な放出に伴う魚中濃度を予測するために2種類のモデルを開発した。1つは平衡状態の系に適用される濃縮係数を使用した方法であり、他方は水中濃度の変化と魚における代謝を考慮したダイナミックモデルである。

3つの異なったシナリオによるモデルの検証結果は環境における水銀が平衡状態に達しているかどうか依存した。平衡状態のシナリオに関しては、第1のモデルは満足な予測をした。平衡状態に達していないシナリオに関しては、第1のモデルによる予測は十分でなかったが第2のモデルはより正確な予測をした。ここで使用された2つのモデルについて適用限界が示唆された。

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## 1. Introduction

### 1.1 Background

An international cooperative study BIOMOV<sup>1),2)</sup>, BIOSpheric Model Validation Study, was initiated by the Swedish National Institute of Radiation Protection, SSI, to test models designed for calculating the environmental transfer and bioaccumulation of radionuclides and other trace substances. The primary objectives of BIOMOV are threefold, namely:

- to test the accuracy of the predictions of environmental assessment models for selected contaminants and exposure scenarios,
- to explain differences in model predictions due to structural deficiencies, invalid assumptions and/or differences in selected input data,
- to recommend priorities for future research for improvement of the accuracy of model predictions.

A secondary objective of the study is to act as a forum for the exchange of ideas, experience and information in order to improve the confidence with which the environmental behavior of trace substances in the biosphere can be assessed quantitatively.

BIOMOV involves two different approaches to fulfill the objectives, namely:

- Approach A, involving the formulation of test scenarios based on suitable data and a comparison of model predictions against these independent data sets,
- Approach B, involving the comparison of model predictions and associated estimates of uncertainty for specific test scenarios selected on the basis of assessment priorities in the case that such independent data sets are not available.

BIOMOV is concerned with terrestrial and aquatic pathways of importance to the assessment of exposure to critical groups and human populations.

Several test scenarios were proposed for each approach. Titles of test scenarios within the BIOMOV study are given in **Appendix 1** and detailed descriptions of the scenarios are presented in BIOMOV Progress Reports<sup>2)</sup>. A scenario on a release of mercury into a river, which is called Scenario A1, was provided as one of the test scenarios included in the Approach A.

This is the scenario for which participants were asked to predict mercury concentrations in fresh-water fish after chronic releases into rivers located in three different sites.

The Japan Atomic Energy Research Institute, JAERI, participated in this scenario to test the performances of assessment models for estimating the transfer and bioaccumulation of trace elements in aquatic ecosystems. This paper describes assessment models and parameter values employed in the predictions, and discusses the results of comparisons between the predictions and measurements.

## 1.2 Scenario descriptions

The Scenario A1 was provided to test the performances of models for predicting the behavior of mercury in fresh-water ecosystems. This scenario is the first that used independent data sets to test model predictions within the BIOMOV5 study and is the only scenario that considered a non-radioactive pollutant. Detailed descriptions for the scenario are shown in **Appendix 2**. A summary of the descriptions is as follows.

Three different sites in the United States and Canada were selected for this scenario in order to derive general conclusions. These sites are:

- Scenario A1-1: a small stream known as East Folk Poplar Creek<sup>3)</sup> at Oak Ridge, Tennessee in the southeastern United States,
- Scenario A1-2: a small river called North Folk Holston River<sup>4)</sup> at Saltville, Virginia in the southeastern United States,
- Scenario A1-3: a river system known as Wabigoon River and Clay Lake<sup>5)</sup> at Dryden, Ontario in eastern Canada.

At each site, mercury had been chronically released from an industrial plant into a river but the mercury release ceased before the time of sampling. Sediment had been highly contaminated during the release and this was the contamination source of water at the time of sampling.

Participants were requested to predict mercury concentrations in the edible tissue of fish sampled in the rivers or the lake, using information on site descriptions and measured data. Model predictions must provide the following results for each scenario:

- Scenario A1-1: mercury concentrations in largemouth bass, bluegill,



redear sunfish and the common carp as a function of distance upstream from the mouth of the stream,

- Scenario A1-2: mercury concentrations in rock bass, hog sucker and shiners as a function of distance downstream from a release point,
- Scenario A1-3: mercury concentrations in northern pike and walleye sampled in Clay Lake.

Uncertainty estimates associated with the predictions were also asked for all three scenarios.

The following information on site descriptions and measured data was presented for participants to use in the predictions:

- fish to be assessed: the species, feeding habits, migratory behavior and body size of fish to be assessed,
- morphological features: locations of an industrial plant, a river, a lake and so on, configuration and water flow of a river,
- mercury release conditions: a period and the total amount of the mercury release, chemical forms of mercury at the time of releases and the detection in fish,
- measured data: mercury concentrations in sediment at several points, those in water and benthic macroinvertebrates at the specified point, mean water quality parameters, mean densities of benthic macroinvertebrates for the Scenario A1-1, mercury concentrations in fish in the past for the Scenario A1-3.

## 2. Model Descriptions

The pathway shown in Fig.1 was considered to estimate the transfer of mercury in fresh-water ecosystems. Water is contaminated by inflow from an industrial plant or the upper stream, and the processes of resuspension and desorption from sediment. Mercury is removed from water by outflow down the stream, and the processes of deposition and sedimentation onto sediment. The mercury in water is transferred to fish via gills and skin, including also via foodchains.

Two kinds of models, JAERI-I and JAERI-II models, have been developed to estimate mercury concentrations in fresh-water fish. The JAERI-I model uses a bioaccumulation factor approach which is applied to ecosystems in

redear sunfish and the common carp as a function of distance upstream from the mouth of the stream,

- Scenario A1-2: mercury concentrations in rock bass, hog sucker and shiners as a function of distance downstream from a release point,
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- mercury release conditions: a period and the total amount of the mercury release, chemical forms of mercury at the time of releases and the detection in fish,
- measured data: mercury concentrations in sediment at several points, those in water and benthic macroinvertebrates at the specified point, mean water quality parameters, mean densities of benthic macroinvertebrates for the Scenario A1-1, mercury concentrations in fish in the past for the Scenario A1-3.

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Two kinds of models, JAERI-I and JAERI-II models, have been developed to estimate mercury concentrations in fresh-water fish. The JAERI-I model uses a bioaccumulation factor approach which is applied to ecosystems in

equilibrium, whereas the JAERI-II model is a dynamic model which considers the change of the concentrations in water and the metabolism in fish.

At first, the JAERI-I model was applied to the three scenarios, assuming that the mercury concentrations were in equilibrium for all scenarios. However, it was afterward known that equilibrium was not attained in the ecosystems for the Scenarios A1-2 and A1-3 because only a few years had passed since the mercury release ceased. Also was found the fact that the metabolism of mercury in fish is very slow though the high concentrations of mercury in water in the past have rapidly decreased. Therefore, the JAERI-II model was then applied to the two scenarios. Two kinds of the models are explained below in detail.

## 2.1 JAERI-I model

A compartment model shown in Fig.2 has been developed to estimate mercury concentrations in stream water. Water and sediment are divided into several compartments with the length of the stream. The changes of mercury inventories in  $i$ -th water compartment with time are expressed by the following differential equations:

$$\frac{dY_1}{dt} = Q + p_1 \cdot X_1 - (f_1 + d_1) \cdot Y_1 \quad (1)$$

$$\frac{dY_i}{dt} = f_{i-1} \cdot Y_{i-1} + p_i \cdot X_i - (f_i + d_i) \cdot Y_i \quad i = 2, 3, 4, \dots \quad (2)$$

where  $Y_i$ : mercury inventory in water (g),  $X_i$ : mercury inventory in sediment (g),  $Q$ : mercury release rate from an industrial plant (g/y),  $p_i$ : transfer rate for resuspension and desorption ( $y^{-1}$ ),  $f_i$ : removal rate for stream flow ( $y^{-1}$ ),  $d_i$ : removal rate for deposition and sedimentation ( $y^{-1}$ ). The subscript,  $i$ , denotes the parameter on  $i$ -th compartment hereinafter.

The mercury inventories in sediment are considered to be constant since the inventories are large enough compared with the loss from sediment. The mercury inventories in water can be then presented by the following equations when the inventories reach equilibrium:

$$Y_1 = \frac{Q + p_1 \cdot X_1}{f_1 + d_1} \quad (3)$$

$$Y_i = \frac{f_{i-1} \cdot Y_{i-1} + p_i \cdot X_i}{f_i + d_i} \quad i = 2, 3, 4, \dots \quad (4)$$

It is assumed that equilibrium is attained between concentrations of mercury dissolved in water and those adsorbed on suspended soil. The concentrations of mercury dissolved in water are calculated by using a distribution coefficient approach:

$$W_i = \frac{\bar{Y}_i}{1 + K_d \cdot SS_i} \quad (5)$$

where  $W_i$ : concentration of mercury dissolved in water (g/l),  $\bar{Y}_i$ : total mercury concentration in water (g/l),  $K_d$ : distribution coefficient (l/kg),  $SS_i$ : suspended soil in water (kg/l).

The transfer or removal rates,  $p_i$ ,  $f_i$  and  $d_i$ , for a river can be estimated by the following equations (refer to **Appendix 3**):

$$p_i = \frac{10^3 \cdot TR_i \cdot D_i \cdot U_i}{\rho_i \cdot L_i} \quad (6)$$

$$f_i = \frac{U_i}{L_i} \quad (7)$$

$$d_i = \frac{V_i}{D_i} \quad (8)$$

where  $TR_i$ : transfer ratio from sediment to water (kg/l),  $D_i$ : mean water depth (m),  $U_i$ : flow velocity of stream (m/y),  $\rho_i$ : surface density of sediment ( $\text{kg/m}^2$ ),  $L_i$ : stream length of  $i$ -th compartment (m),  $V_i$ : deposition velocity onto sediment or benthic organisms (m/y).

The values of the above parameters except  $L_i$  are not given in the scenario description. Furthermore, the values of  $TR_i$  and  $V_i$  are difficult to estimate because measured data are few and considerably specific to

sites. If the values of  $TR_i$ ,  $D_i$ ,  $\rho_i$  and  $V_i$  are constant at any position, the following equations can be derived:

$$\frac{p_i \cdot L_i}{U_i} = \frac{p_1 \cdot L_1}{U_1} \quad (9)$$

$$\frac{d_i}{f_i} = \alpha_i \cdot L_i \quad \text{where} \quad \alpha_i = \frac{d_i}{U_i} \quad (10)$$

Measured data of mercury concentrations in water and sediment at a release point are prepared in the scenario description. Using the data, the parameter,  $p_i$ , can be calculated as follows:

$$p_i = \frac{L_1 \cdot U_i}{L_i \cdot U_1} \cdot p_1 = \frac{L_1 \cdot U_i}{L_i \cdot U_1} \cdot \frac{Y_1 \cdot (f_1 + d_1) - Q}{X_1} \quad (11)$$

When the length of the first compartment,  $L_1$ , is set to be sufficiently small, the value of  $f_1$  is much bigger than that of  $d_1$ . If the mercury release from an industrial plant is zero at the time of sampling and water flow of the stream is constant at any position, Eq.(5) is then re-written as follows:

$$W_i = \frac{1}{(1 + \alpha_i \cdot L_i) \cdot (1 + K_d \cdot SS_i)} \left[ \bar{Y}_{i-1} + \frac{\bar{Y}_1}{\bar{X}_1} \cdot \bar{X}_i \right] \quad (12)$$

where  $\bar{X}_i$ : mercury concentration in sediment (g/kg).

Although the same model as described above is applied to a lake, lake water and sediment are modeled as one compartment, respectively. The mercury inventory in lake water is expressed as follows:

$$Y = \frac{Q' + p \cdot X}{f + d} \quad (13)$$

where  $Q'$ : mercury inflow rate into lake (g/y).

The transfer or removal rates,  $p$ ,  $f$  and  $d$ , for a lake can be evaluated by the following equations (refer to **Appendix 3**):

$$p = \frac{I_w}{K_d \cdot \rho \cdot S} \quad (14)$$

$$f = \frac{I_w}{10^3 \cdot D \cdot S} \quad (15)$$

$$d = \frac{K_d \cdot R}{10^3 \cdot D \cdot (1 + K_d \cdot SS)} \quad (16)$$

where  $I_w$ : water inflow into lake (l/y),  $S$ : area of lake ( $m^2$ ),  $R$ : sedimentation rate ( $kg/m^2/y$ ).

Using the above equations, Eq.(13) can be re-written as follows:

$$W = \frac{I_w}{I_w \cdot (1 + K_d \cdot SS) + K_d \cdot S \cdot R} \left[ Q_{in} + \frac{\bar{X}}{K_d} \right] \quad (17)$$

where  $Q_{in}$ : mercury concentration in water supplied into lake (g/l).

Mercury concentrations in fresh-water fish are obtained by using a bioaccumulation factor approach, assuming that equilibrium is attained between the concentrations in water and those in fish, and also that mercury adsorbed on suspended soil is not transferred to fish body:

$$F_i = CF \cdot W_i \quad (18)$$

where  $F_i$ : mercury concentration in fish (g/kg),  $CF$ : bioaccumulation factor (l/kg).

For a river scenario, the values of  $L_i$  are determined as the distance between the points where measured concentrations in sediment are given. The values of  $\bar{Y}_i$ ,  $\bar{X}_i$  and  $SS_i$  are prepared in the scenario description and the mean values are calculated for each compartment. The values of  $\alpha_i$ ,  $K_d$  and  $CF$  have to be provided by the modeler. For a lake scenario, the values of  $I_w$ ,  $SS$ ,  $Q_{in}$  and  $\bar{X}$  are apparently given in the scenario description. The value of  $S$  can be roughly estimated from Fig. A2-3. The values of  $K_d$ ,  $R$  and  $CF$  must be prepared by the modeler.

## 2.2 JAERI-II model

It is assumed that mercury concentrations in water had been in equilibrium before the mercury release ceased and the concentrations have exponentially decreased after the end of the release. The mercury concentrations in  $i$ -th water compartment are presented by the following function:

$$W_i = W_{i0} \cdot \exp(-k_i \cdot t) \quad (19)$$

where  $W_{i0}$ : equilibrium concentration of mercury dissolved in water before the mercury release ceased (kg/l),  $k_i$ : decay rate from water ( $y^{-1}$ ),  $t$ : time after the release ceased (y).

The following differential equation is used to estimate mercury concentrations in fresh-water fish:

$$\frac{dF_i}{dt} = q \cdot W_i - \lambda \cdot F_i \quad (20)$$

where  $q$ : transfer rate from water to fish (l/kg/y),  $\lambda$ : removal rate from fish due to metabolism ( $y^{-1}$ ).

Because the mercury concentrations in water and in fish are assumed to be in equilibrium before the mercury release ceased, the following equation can be derived:

$$\frac{F_{i0}}{W_{i0}} = \frac{q}{\lambda} = CF \quad (21)$$

where  $F_{i0}$ : equilibrium concentration of mercury in fish before the mercury release ceased (g/kg).

Using the above equations, the following equation is derived to estimate the mercury concentrations in fish for the Scenario A1-2:

$$F_i = \frac{\lambda \cdot CF \cdot W_{i0}}{\lambda - k_i} \left[ \exp(-k_i \cdot t) - \exp(-\lambda \cdot t) \right] + CF \cdot W_{i0} \cdot \exp(-\lambda \cdot t) \quad (22)$$

Although the same model as described above is applied to a lake, lake water is modeled as one compartment. For the Scenario A1-3, measured data of the concentrations in fish before the mercury release ceased are given in the scenario description. The following equation is used to estimate the concentration in fish for the Scenario A1-3:

$$F = \frac{\lambda \cdot F_0}{\lambda - k} \left[ \exp(-k \cdot t) - \exp(-\lambda \cdot t) \right] + F_0 \cdot \exp(-\lambda \cdot t) \quad (23)$$

For the Scenario A1-2, the values of  $W_{i0}$ ,  $k_i$ ,  $\lambda$  and CF must be prepared by the modeler. For the Scenario A1-3, the mercury concentrations in fish sampled in 1976 are used as the values of  $F_0$ . The value of  $k$  and  $\lambda$  must be prepared by the modeler. The value of  $W_0$  needs to be also provided to estimate the value of  $k$ .

### 3. Parameter Values and Uncertainty Analysis

#### 3.1 Parameter values employed in predictions

Among the parameters employed in the two models, the values of the following parameters have to be prepared by the modeler:

- $\alpha_i$ ,  $K_d$  and CF in the JAERI-I model for a river scenario,
- $K_d$ , R and CF in the JAERI-I model for a lake scenario,
- $W_{i0}$ ,  $k_i$ ,  $\lambda$  and CF in the JAERI-II model for a river scenario,
- $W_0$ ,  $k$  and  $\lambda$  in the JAERI-II model for a lake scenario.

The above parameter values employed in the predictions for the Scenarios A1-1, A1-2 and A1-3 are shown in **Table 1**, **Table 2** and **Table 3**, respectively. The values were estimated as follows.

Since the values of  $\alpha_i$  are quite different from rivers<sup>6)</sup>, the values must be determined for each river. When a non-radioactive pollutant is continuously released into a river, the concentration in stream water can be empirically estimated by the following equation<sup>7)-9)</sup>:

$$W_0(x) = \frac{Q}{I_v \cdot (1 + K_d \cdot SS)} \exp(-\alpha \cdot x) \quad (24)$$



Although the same model as described above is applied to a lake, lake water is modeled as one compartment. For the Scenario A1-3, measured data of the concentrations in fish before the mercury release ceased are given in the scenario description. The following equation is used to estimate the concentration in fish for the Scenario A1-3:

$$F = \frac{\lambda \cdot F_0}{\lambda - k} \left[ \exp(-k \cdot t) - \exp(-\lambda \cdot t) \right] + F_0 \cdot \exp(-\lambda \cdot t) \quad (23)$$

For the Scenario A1-2, the values of  $W_{i0}$ ,  $k_i$ ,  $\lambda$  and CF must be prepared by the modeler. For the Scenario A1-3, the mercury concentrations in fish sampled in 1976 are used as the values of  $F_0$ . The value of  $k$  and  $\lambda$  must be prepared by the modeler. The value of  $W_0$  needs to be also provided to estimate the value of  $k$ .

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- $K_d$ , R and CF in the JAERI-I model for a lake scenario,
- $W_{i0}$ ,  $k_i$ ,  $\lambda$  and CF in the JAERI-II model for a river scenario,
- $W_0$ ,  $k$  and  $\lambda$  in the JAERI-II model for a lake scenario.

The above parameter values employed in the predictions for the Scenarios A1-1, A1-2 and A1-3 are shown in **Table 1**, **Table 2** and **Table 3**, respectively. The values were estimated as follows.

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$$W_0(x) = \frac{Q}{I_v \cdot (1 + K_d \cdot SS)} \exp(-\alpha \cdot x) \quad (24)$$

where  $W_0(x)$ : concentration of a pollutant dissolved in stream water (g/l),  $x$ : distance downstream from a release point (m),  $Q$ : release rate of a pollutant (g/y),  $I_V$ : water flow of stream (l/y),  $\alpha$ : the parameter defined by Eq.(10). The values of  $\alpha_i$  were estimated by the distribution of mercury concentrations in sediment, assuming that the high concentrations in stream water in the past when a large quantity of mercury had been released were reflected on the concentrations in sediment at the time of sampling.

The values of  $K_d$  for suspended soil in fresh water are different according to the water quality and the nature of the soil. Therefore, the values were basically evaluated by the relationship between concentrations of mercury dissolved in water and those adsorbed on suspended soil. Unless these data were available, the value obtained for a river located in the northern part of Japan<sup>10)</sup> was used here.

The majority of mercury detected in fish was found to be in the form of methylmercury though most of mercury released from an industrial plant was in the inorganic form. Methylation of mercury rarely takes place in organisms but mainly by bacteria in sediment<sup>11)-14)</sup>. Therefore, it is understood that mercury released in the inorganic form is changed to methylmercury in sediment and the latter is accumulated into fish body. The values of CF were surveyed for the bioaccumulation of methylmercury into fresh-water fish. Fig.3 presents the relationship between mercury concentrations in water and the CF values collected<sup>10),12)-16)</sup>. It is obvious that the logarithmic value of CF decreases linearly with increasing the logarithmic value of the concentration in water. This relationship was taken into account when using the CF values and the values were assumed to be independent of the species of fish.

The value of R is different from lakes but a smaller value obtained for a big lake<sup>17)</sup> was used here to estimate the concentration in water conservatively. The values of  $W_{10}$  were calculated by Eq.(24) for a river scenario. For a lake scenario, the value of  $W_0$  was evaluated by the relationship between the CF parameter shown in Fig.3 and the mercury concentrations in fish before the mercury release ceased. The values of  $k_i$  were calculated by Eq.(19) and were assumed to be constant at any position and time. The value of  $\lambda$  for northern pike was obtained for both

laboratory and field experiments<sup>12)</sup>, and was also used for other species of fish.

### 3.2 Uncertainty analysis

Uncertainty analysis was carried out by a parameter perturbation technique. Extreme maximum and minimum values were selected for each of the following several parameters which had to be prepared by the modeler and were considered to be important in each model:

- $\alpha_i$ ,  $K_d$  and CF in the JAERI-I model for a river scenario,
- $K_d$ , R and CF in the JAERI-I model for a lake scenario,
- $W_{10}$ ,  $\lambda$  and CF in the JAERI-II model for a river scenario,
- $W_0$  and  $\lambda$  in the JAERI-II model for a lake scenario.

Since the distributions of the parameter values were not sufficiently known, the uncertainty ranges of the values were determined by a subjective judgement as follows:

- two factors of magnitude for  $\alpha_i$  and  $\lambda$ ,
- five factors of magnitude for  $K_d$ , R and CF,
- one order of magnitude for  $W_{10}$  and  $W_0$ .

Upper and lower estimates were then undertaken by putting various combinations of the extreme values into the models. Correlations between the parameters were not taken into account in choosing sets of the combinations of the values. Equal weight was given to all combinations of the values, including the combinations of the extreme values which in fact were highly unlikely. The uncertainty ranges produced by this technique would therefore tend to be large. The both estimates were assumed to define the confidence interval for the results although this would not be statistically true.

## 4. Results and Discussions

### 4.1 Scenario A1-1

The model separated the species of fish according to migratory behavior or movement up and down the stream. Predictions were made for the common carp that are known to move up and down the stream, and for the other

laboratory and field experiments<sup>12)</sup>, and was also used for other species of fish.

### 3.2 Uncertainty analysis

Uncertainty analysis was carried out by a parameter perturbation technique. Extreme maximum and minimum values were selected for each of the following several parameters which had to be prepared by the modeler and were considered to be important in each model:

- $\alpha_i$ ,  $K_d$  and CF in the JAERI-I model for a river scenario,
- $K_d$ , R and CF in the JAERI-I model for a lake scenario,
- $W_{i0}$ ,  $\lambda$  and CF in the JAERI-II model for a river scenario,
- $W_0$  and  $\lambda$  in the JAERI-II model for a lake scenario.

Since the distributions of the parameter values were not sufficiently known, the uncertainty ranges of the values were determined by a subjective judgement as follows:

- two factors of magnitude for  $\alpha_i$  and  $\lambda$ ,
- five factors of magnitude for  $K_d$ , R and CF,
- one order of magnitude for  $W_{i0}$  and  $W_0$ .

Upper and lower estimates were then undertaken by putting various combinations of the extreme values into the models. Correlations between the parameters were not taken into account in choosing sets of the combinations of the values. Equal weight was given to all combinations of the values, including the combinations of the extreme values which in fact were highly unlikely. The uncertainty ranges produced by this technique would therefore tend to be large. The both estimates were assumed to define the confidence interval for the results although this would not be statistically true.

## 4. Results and Discussions

### 4.1 Scenario A1-1

The model separated the species of fish according to migratory behavior or movement up and down the stream. Predictions were made for the common carp that are known to move up and down the stream, and for the other

fish, that is, largemouth bass, bluegill and redear sunfish that tend to be territorial and stay in the same stretch of the stream. Predictions and uncertainty estimates of mercury concentrations in the common carp and the other fish by the JAERI-I model are shown in **Fig.4**, compared with mean measured values for the two species of fish. The distance is defined by km beginning at the mouth of the stream and proceeding upstream to a release point at 22.9 km.

The predictions by the model are in relatively good agreement with the measured values for the two species of fish. The measured values are included in the uncertainty estimates. Upper and lower estimates vary by one order of magnitude above and below the best estimate resulting in a range of two orders of magnitude for the uncertainty estimates.

The model predictions for the other fish considerably agree with the measured mercury concentrations. The best estimate is within the measured values by three factors of magnitude but is always an overestimate. The predictions for the common carp were obtained by using an average mercury concentration in water for the entire stream. The best estimate is relatively close to the observed values but is always an overestimate by factors of two to six, especially the degree of the overestimate is larger in the upper parts of the stream. It is difficult to determine which part of the concentration in water should be used to predict the concentrations in fish such as carp that move up and down the stream.

Mercury concentrations in stream water and in fish were considered to be in equilibrium because more than twenty years had passed since the mercury release ceased. For this scenario, a bioaccumulation factor approach was very useful to predict the concentrations in fish if the factor was adequately selected.

#### **4.2 Scenario A1-2**

Predicted mercury concentrations in fish by using the JAERI-I and the JAERI-II models are shown in **Fig.5**, compared with mean measured values for the three species of fish, namely, rock bass, hog sucker and shiners. The distance is defined by km beginning at a release point and proceeding down the stream. The predictions were made for all species combined, which included fish with different feeding habits and migratory behavior.

Uncertainty estimates are also shown with the both predicted values. Upper and lower estimates vary by over one order of magnitude above and below the best estimate resulting in a range of more than two orders of magnitude for the uncertainty estimates.

The best estimate by the JAERI-I model is less than the measured values by factors of two to eight. The measured values are always within the range of the uncertainty estimates but are closer to the upper estimate than the best estimate. This underestimate is explained as follows: Since the metabolism of methylmercury in fish is very slow, the fish keep relatively high concentrations of mercury reflecting on the earlier high concentrations in water though the latter have rapidly decreased. On the contrary, the best estimate by using the JAERI-II model is in considerably good agreement with the measured values within two factors of magnitude.

Mercury concentrations in stream water and in fish were not in equilibrium for this scenario because only a few years had passed since the mercury release ceased. For this scenario, a bioaccumulation factor approach was not adequate but a dynamic model could accurately predict the concentrations in fish.

#### 4.3 Scenario A1-3

Model predictions of mercury concentrations in fish by the JAERI-I and the JAERI-II models are shown in **Fig.6**, compared with mean measured values for northern pike and walleye sampled from Clay Lake in 1979 and 1980. Uncertainty estimates are also shown with the both predicted values. The uncertainty of the JAERI-I model results in a range of more than two orders of magnitude but that of the JAERI-II model is within one order of magnitude. This is because the measured data of concentrations in fish before the mercury release ceased, which is an important parameter for the JAERI-II model, are given in the scenario description.

The best estimate by the JAERI-I model is from one to two orders of magnitude less than the measured values. Even the uncertainty range includes only the measured values for northern pike collected in 1980. This underestimate is explained by the same reason as that for the Scenario A1-2. On the contrary, the JAERI-II model predicts the measured values more accurately than the JAERI-I model does. However, the JAERI-II

model prediction is still less than the measured values and the uncertainty range does not include the measured values for walleye. It is considered that the value of  $\lambda$  used here is not adequate for walleye since the value is for northern pike.

Because only a few years had passed since the mercury release ceased, the mercury concentrations were not in equilibrium. Also for this scenario, not a bioaccumulation factor approach but a dynamic model could predict the concentrations in fish accurately. However, a difference of the two models applied here was reflected on the predictions more remarkably than the Scenario A1-2.

## 5. Concluding Remarks

The performances of assessment models for predicting the transfer and bioaccumulation of mercury in fresh-water ecosystems were tested by being applied to the test scenario proposed in the international cooperative study BIOMOVs. Two kinds of models, the JAERI-I model and the JAERI-II model, have been developed to estimate mercury concentrations in fish. The JAERI-I model uses a bioaccumulation factor approach which is applied to ecosystems in equilibrium, whereas the JAERI-II model is a dynamic model which considers the change of the concentrations in water and the metabolism in fish. The success of the models tested by the three different scenarios depended upon whether mercury was in or near equilibrium in the environment. For the Scenario A1-1 where mercury concentrations reached equilibrium, the JAERI-I model performed satisfactorily. In the Scenarios A1-2 and A1-3 where equilibrium was not attained, the JAERI-I model was not adequate but the JAERI-II model could predict more accurately. The limitations of applications were suggested for the two models employed here.

When the transfer of trace elements in aquatic ecosystems is predicted by an assessment model, it is important to select an appropriate model by understanding fully the characteristics of the ecosystems in advance. In the case of these scenarios, it would be significant to grasp whether mercury concentrations in ecosystems were in equilibrium or not.

model prediction is still less than the measured values and the uncertainty range does not include the measured values for walleye. It is considered that the value of  $\lambda$  used here is not adequate for walleye since the value is for northern pike.

Because only a few years had passed since the mercury release ceased, the mercury concentrations were not in equilibrium. Also for this scenario, not a bioaccumulation factor approach but a dynamic model could predict the concentrations in fish accurately. However, a difference of the two models applied here was reflected on the predictions more remarkably than the Scenario A1-2.

## 5. Concluding Remarks

The performances of assessment models for predicting the transfer and bioaccumulation of mercury in fresh-water ecosystems were tested by being applied to the test scenario proposed in the international cooperative study BIOMOVs. Two kinds of models, the JAERI-I model and the JAERI-II model, have been developed to estimate mercury concentrations in fish. The JAERI-I model uses a bioaccumulation factor approach which is applied to ecosystems in equilibrium, whereas the JAERI-II model is a dynamic model which considers the change of the concentrations in water and the metabolism in fish. The success of the models tested by the three different scenarios depended upon whether mercury was in or near equilibrium in the environment. For the Scenario A1-1 where mercury concentrations reached equilibrium, the JAERI-I model performed satisfactorily. In the Scenarios A1-2 and A1-3 where equilibrium was not attained, the JAERI-I model was not adequate but the JAERI-II model could predict more accurately. The limitations of applications were suggested for the two models employed here.

When the transfer of trace elements in aquatic ecosystems is predicted by an assessment model, it is important to select an appropriate model by understanding fully the characteristics of the ecosystems in advance. In the case of these scenarios, it would be significant to grasp whether mercury concentrations in ecosystems were in equilibrium or not.



In the BIOMOVs study, three other organizations participated in this scenario; Institute of Radiation Hygiene in Federal Republic of Germany, Chalk River Nuclear Laboratory in Canada and Studsvik Nuclear in Sweden. Comparisons of predictions by these organizations are given in BIOMOVs Technical Report<sup>18)</sup>, together with models and parameter values employed in the predictions.

### **Acknowledgement**

The author would like to express appreciation to all members of the Committee on the Environmental Behavior of Radionuclides established within JAERI, for their helpful discussions and suggestions. The author also wishes to thank Mr. H. Matsuzuru and Mr. T. Homma of Department of Environmental Safety Research, JAERI, for valuable comments.

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## Nomenclature

- $Y_i$  : mercury inventory in water (g)  
 $X_i$  : mercury inventory in sediment (g)  
 $Q$  : mercury release rate from an industrial plant (g/y)  
 $p_i$  : transfer rate for resuspension and desorption ( $y^{-1}$ )  
 $f_i$  : removal rate for stream flow ( $y^{-1}$ )  
 $d_i$  : removal rate for deposition and sedimentation ( $y^{-1}$ )  
 $W_i$  : concentration of mercury dissolved in water (g/l)  
 $\bar{Y}_i$  : total mercury concentration in water (g/l)  
 $K_d$  : distribution coefficient (l/kg)  
 $SS_i$  : suspended soil in water (kg/l)  
 $TR_i$  : transfer ratio from sediment to water (kg/l)  
 $D_i$  : mean water depth (m)  
 $U_i$  : flow velocity of stream (m/y)  
 $\rho_i$  : surface density of sediment ( $kg/m^2$ )  
 $L_i$  : stream length of i-th compartment (m)  
 $V_i$  : deposition velocity onto sediment or benthic organisms (m/y)  
 $\alpha_i = d_i/U_i$   
 $\bar{X}_i$  : mercury concentration in sediment (g/kg)  
 $Q'$  : mercury inflow rate into lake (g/y)  
 $I_w$  : water inflow into lake (l/y)  
 $S$  : area of lake ( $m^2$ )  
 $R$  : sedimentation rate ( $kg/m^2/y$ )  
 $Q_{in}$  : mercury concentration in water supplied into lake (g/l)  
 $F_i$  : mercury concentration in fish (g/kg)  
 $CF$  : bioaccumulation factor (l/kg)  
 $W_{i0}$  : equilibrium concentration of mercury dissolved in water before the mercury release ceased (kg/l)  
 $k_i$  : decay rate from water ( $y^{-1}$ )  
 $q$  : transfer rate from water to fish (l/kg/y)  
 $\lambda$  : removal rate from fish due to metabolism ( $y^{-1}$ )  
 $F_{i0}$  : equilibrium concentration of mercury in fish before the mercury release ceased (g/kg)

Table 1 Parameter values employed in predictions for Scenario A1-1

Model	Parameter	Value
JAERI-I	$\alpha_i$	$2 \times 10^{-4} \text{ (m}^{-1}\text{)}^*$
	$K_d$	$5 \times 10^4 \text{ (1/kg)}$
	CF	$1 \times 10^3 \text{ (1/kg)}$

\* This value is used for all compartments.

Table 3 Parameter values employed in predictions for Scenario A1-2

Model	Parameter	Value
JAERI-I	$K_d$	$2 \times 10^5 \text{ (1/kg)}$
	R	$0.2 \text{ (kg/m}^2\text{/y)}$
	CF	$2 \times 10^4 \text{ (1/kg)}$
JAERI-II	$W_0$	$50 \text{ (}\mu\text{g/l)}$
	k	$2.4 \text{ (y}^{-1}\text{)}$
	$\lambda$	$0.35 \text{ (y}^{-1}\text{)}$

Table 2 Parameter values employed in predictions for Scenario A1-2

Model	Parameter	Value
JAERI-I	$\alpha_i$	$1.2 \times 10^{-4}$ ( $m^{-1}$ ) for 0 - 3.7 km
		$1.2 \times 10^{-4}$ ( $m^{-1}$ ) for 3.7 - 9.7 km
		$6.7 \times 10^{-5}$ ( $m^{-1}$ ) for 9.7 - 21 km
		$2.7 \times 10^{-5}$ ( $m^{-1}$ ) for 21 - 43 km
	$K_d$	$2 \times 10^5$ (1/kg)
	CF	$1 \times 10^4$ (1/kg)
JAERI-II	$W_{i0}$	19 ( $\mu g/l$ ) for 0 - 3.7 km
		14 ( $\mu g/l$ ) for 3.7 - 9.7 km
		10 ( $\mu g/l$ ) for 9.7 - 21 km
		7.3 ( $\mu g/l$ ) for 21 - 43 km
	$k_i$	1.9 ( $y^{-1}$ )*
	$\lambda$	0.35 ( $y^{-1}$ )
	CF	$3 \times 10^2$ (1/kg)

\* The value of  $k_i$  is assumed to be constant at any position and time.

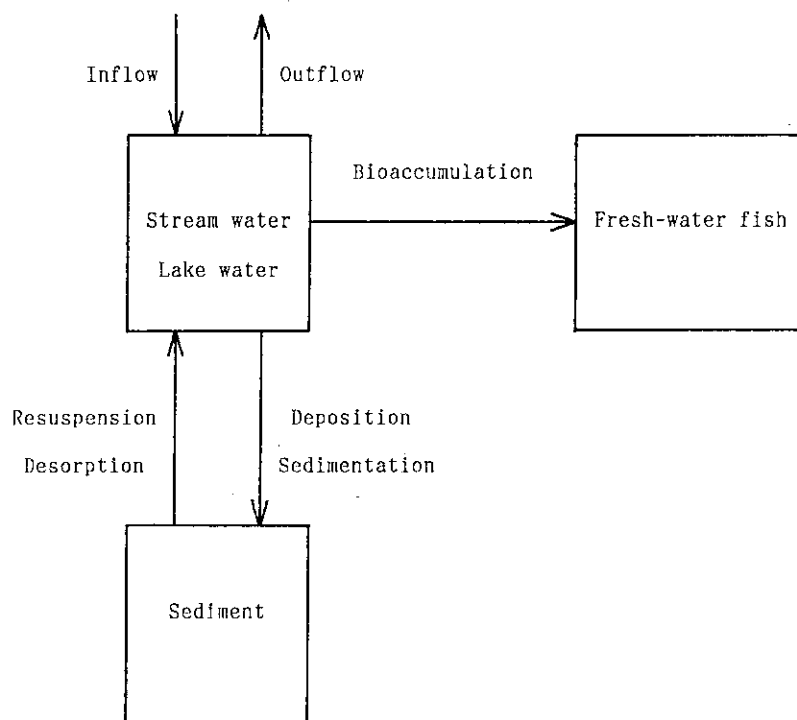
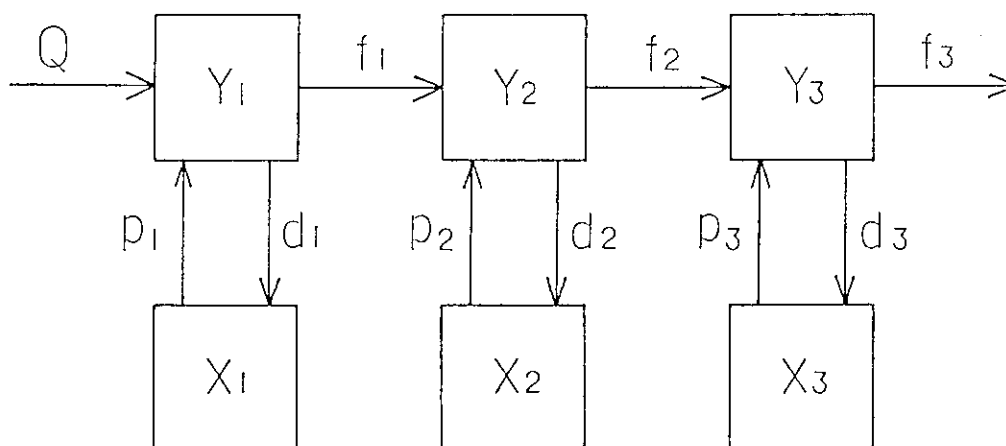


Fig.1 Transfer pathway of mercury in a fresh-water ecosystem



$Y_i$ : mercury inventory in stream water (g)  
 $X_i$ : mercury inventory in sediment (g)  
 $Q$ : mercury release rate from an industrial plant (g/y)  
 $p_i$ : transfer rate for resuspension and desorption ( $y^{-1}$ )  
 $f_i$ : removal rate for stream flow ( $y^{-1}$ )  
 $d_i$ : removal rate for deposition and sedimentation ( $y^{-1}$ )

Fig.2 Compartment model for estimating mercury concentrations in stream water



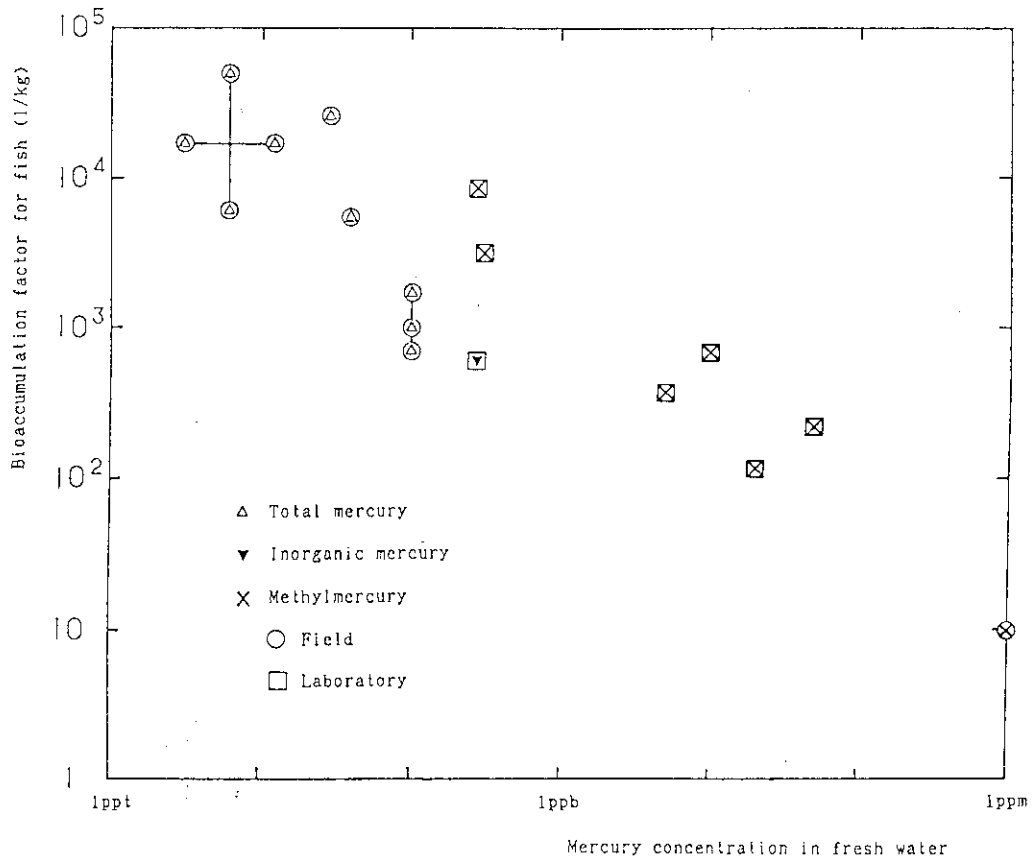


Fig.3 Relationship between mercury concentrations in fresh water and bioaccumulation factors

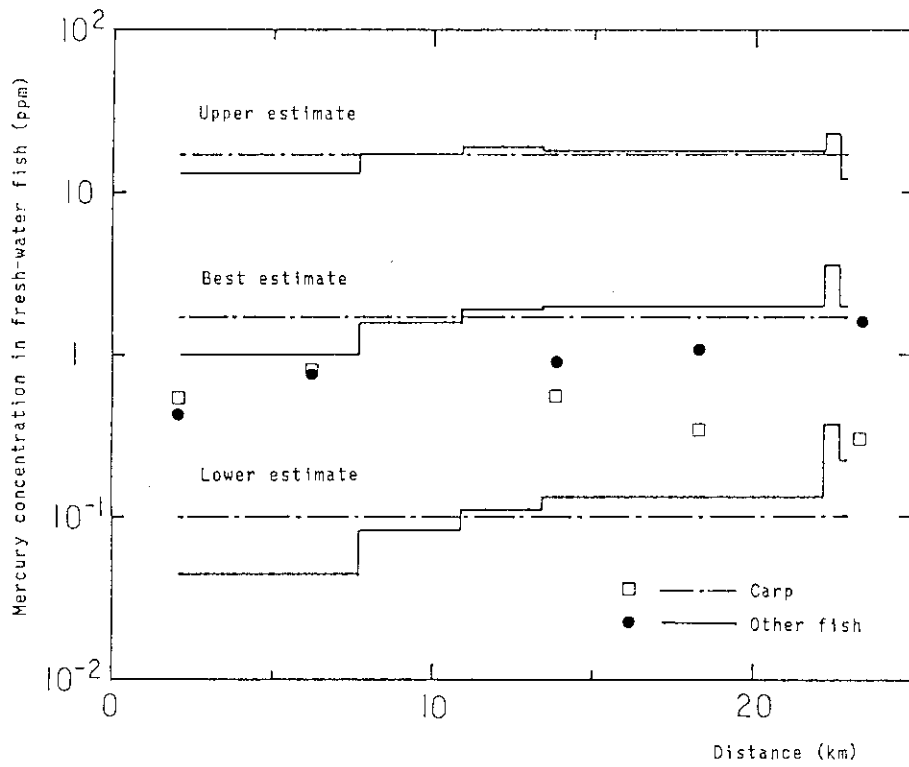


Fig.4 Comparison between predictions and measurements of mercury concentrations in fish for Scenario A1-1

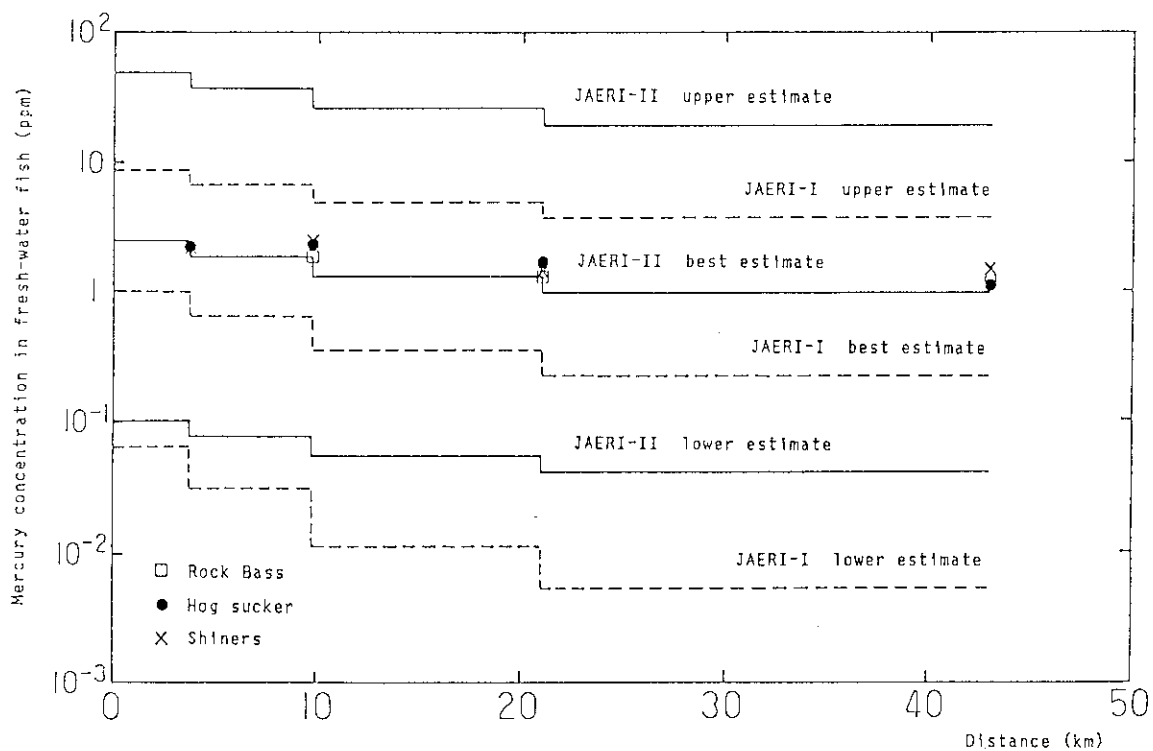


Fig.5 Comparison between predictions and measurements of mercury concentrations in fish for Scenario A1-2

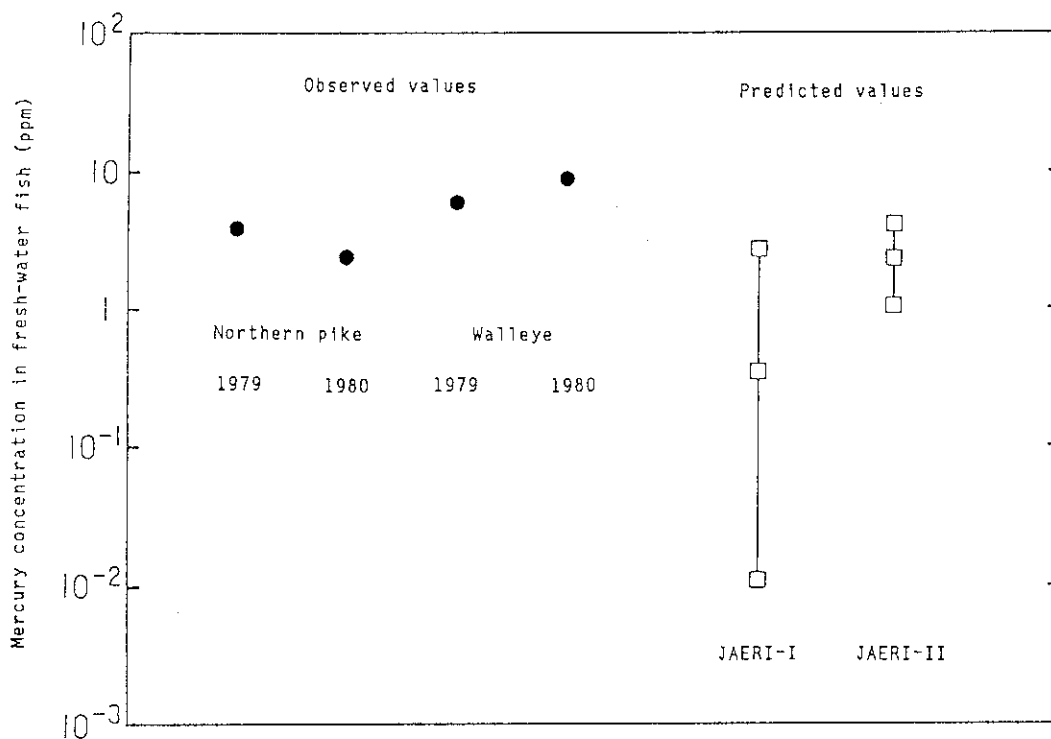


Fig.6 Comparison between predictions and measurements of mercury concentrations in fish for Scenario A1-3

**Appendix 1 Scenarios within the BIOMOVs Study**

Approach A scenarios:

- A1 Release of mercury into a river
- A4a I-131 in milk after the Chernobyl accident
- A4b Cs-137 in milk, beef and barley after the Chernobyl accident
- A5 Dynamics within a lake ecosystem

Approach B scenarios:

- B1 Atmospheric deposition
- B2 Irrigation with contaminated groundwater
- B3 Release into a lake from a river
- B5 Aging of a lake
- B6a Transport of contaminated groundwater to soil
- B6b Transport of contaminated groundwater to soil, specific sites
- B7 Transport of contaminated groundwater to a river
- B8 The importance of different pathways on radiological assessment

## Appendix 2 Detailed Scenario Descriptions for Scenario A1

### A2.1 Scenario A1-1

This is a scenario of a small stream in the southeastern United States. The stream has received a chronic input of mercury from an industrial complex since 1955. Most of the mercury was released into the stream during the late 50's and early 60's. Approximately 360000 kg have been released into the stream to date and low levels of mercury are still being released from an industrial holding pond. Relatively high concentrations of mercury are found in the sediment and flood plain of the stream. The concentration of mercury in the sediment ranged from <0.1 to 1800 mg/kg with a mean concentration of 77 mg/kg.

More than 90 % of the mercury that is detected in fish from the stream is methylmercury; however, most of the mercury that was released to the stream was in the inorganic form. The following results are desired:

- Concentration of mercury in the edible tissues of fish as a function of distance down stream from the holding pond (in  $\mu\text{g}/\text{kg}$ ).

The upper portion of the stream originates in an industrial complex that discharges into a holding pond. The holding pond is used for equalizing the pH of the effluents from the industrial complex, sediment retention and spill control. From the holding pond the stream flows 22.9 km to its confluence with a larger stream that is a tributary of a large river (refer to **Fig.A1**). The holding pond may contribute as much as 849.6 l/s to the flow rate of the stream. Above the pond the flow rate averages 297.4 l/s. The mean average flow of the stream at 8.5 km was 1472.6 l/s (1960-1984); Maximum and minimum daily flows over the same period were 116112.0 and 339.8 l/s, respectively.

Aquatic biota, sediment and water quality parameters have been sampled in the stream. The sampling stations are identified by km beginning at the mouth of the stream (km "0") and proceeding upstream to the holding pond at km 22.9.

The fish population is composed of largemouth bass (*Micropterus salmoides*), bluegill (*Lepomis macrochirus*), redear sunfish (*Lepomis microlophus*) and the common carp (*Cyprinus carpio*). The largemouth bass is piscivorous, bluegill and redear sunfish are omnivorous and the carp is

an omnivorous bottom feeder. Largemouth bass, bluegill and redear sunfish usually remain in same section of the stream; however, carp move up and down the stream from the mouth to the holding pond.

The mean densities of benthic macroinvertebrates are given in **Table A1**. Very high sediment loading could account for the low number of benthic invertebrate species that were observed in a quantitative survey.

Mean water quality parameters and the concentration of total mercury in the water below the holding pond are listed in **Table A2**.

Mercury concentrations in the sediment decrease down the 22.9 km length of the stream. As the contaminated sediment is transported downstream, it is diluted with uncontaminated sediment. The sediment size that is subject to downstream transport, and also contains the highest concentration of mercury, is the <0.125 mm-particle size. The size class consists of fine sand, silt and clay. The total mercury in sediment for the <0.125 mm-particle size at the different sampling stations is given in **Table A3**.

## **A2.2 Scenario A1-2**

A chlor-alkali plant operated near the bank of a small river in the southeastern United States from 1952 until 1972 (see **Fig.A2**). During plant operation, salt wastes were placed in two disposal ponds adjacent to the river. Total mercury concentrations measured in the solid waste range from 4.4 to 350  $\mu\text{g/g}$  (average = 92), and from 11 to 230  $\mu\text{g/l}$  (average = 68) in the leachate. Approximately 39 kg of mercury are estimated to be leached from the ponds each year. The concentration of mercury measured in 1975 in sediments downstream from the plant site was typically 20 times higher than the mercury concentrations measured upstream. The fish species and invertebrates collected immediately downstream, and at progressive distances downstream from the former chlor-alkali plant, contained elevated concentrations of mercury, decreasing with distance from the plant. The majority of the total mercury measured in fish muscle was determined to be in the form of methylmercury (up to 92 %).

Determining the following:

The concentration of mercury in fish axial muscle determined in 1975 as a function of distance downstream from the holding ponds.

### Site description and sampling

The study river originates 77 km above the former chlor-alkali plant site and flows 133 km beyond the site to the confluence with a second river. The two rivers flow an additional 50 km into a man-made reservoir, created in 1942. Mean daily discharge at a point 2.7 km upstream from the former plant is  $8.5 \text{ m}^3/\text{s}$  (drainage area =  $575 \text{ km}^2$ ). At the confluence of the two rivers the mean daily discharge is  $25.4 \text{ m}^3/\text{s}$  (drainage area =  $1740 \text{ km}^2$ ).

**Table A4** presents total mercury concentrations measured in water, bed sediment, and benthic invertebrates collected above and below the plant site. Sampling sites are labeled according to the river distance in kilometers above (-) and below (+) the chlor-alkali plant.

The species of fish sampled were rock bass (*Ambloplites rupestris*), hog sucker (*Hypentelium nigricans*) and shiners (*Notropis* spp.). Rock bass are typically non-migratory fish, feeding on benthic invertebrates, insects and small fish. Shiners are also non-migratory fish, and feed on small insects and benthic invertebrates. Hog sucker are bottom feeders and are often migratory. Rock bass sampled ranged in size from 6 to 165 grams (average = 54), hog sucker ranged from 15 to 536 grams (average = 140) and shiners ranged from 2 to 13 (average = 7.8).

Mean water quality parameters are listed in **Table A5**.

### **A2.3 Scenario A1-3**

It is estimated that between 1962 and 1969 ten metric tons of mercury were released from a mercury cell chlor-alkali operation to a river system in eastern Canada. Fish sampled from a lake below the plant site were observed to contain mercury concentrations of up to  $10 \text{ }\mu\text{g/g}$ . Approximately 85 % of the mercury in the edible portion of the fish was determined to be in the methylated form. In 1970, measures were taken to curtail the mercury releases from the plant and in 1975 the mercury cells were dismantled. In 1971, the mean levels of mercury measured in northern pike (*Esox lucius*) and walleye (*Stizostedion vitreum*) were  $9.09 \text{ }\mu\text{g/g}$  and  $8.71 \text{ }\mu\text{g/g}$ , respectively. After the mercury cells were dismantled (in 1975), northern pike and walleye were measured to have mean mercury concentrations of  $5.84 \text{ }\mu\text{g/g}$  and  $7.83 \text{ }\mu\text{g/g}$ , respectively. A two year

study, beginning in late 1987, was undertaken to help find methods for alleviating the long-term problem.

#### Objects for model calculations

Predict the mercury concentrations measured in lake fish in 1979 and 1980. The concentration of mercury in fish sampled in 1971 and 1976, as well as the concentration of mercury in water and sediment measured in 1979 and 1980, are presented for your use in making the calculations.

#### Site description and sampling

The former chlor-alkali plant is situated on a river, below the dam of one lake and approximately 90 km above a second lake in the river system (the study lake). A second river joins the main river about 45 km below the former plant site (refer to **Fig.A3**). The total drainage area of the river system to the mouth of the study lake is approximately 6800 km<sup>2</sup> and has a mean annual flow of 44.5 m<sup>3</sup>/s.

The concentration of mercury measured in water and sediment from the study lake in 1979 and 1980 is provided in **Table A6**. Mean water quality parameters for the study lake are listed in **Table A7**.

Two species of fish were sampled, northern pike (*Esox lucius*) and walleye (*Stizostedion vitreum*). The northern pike and the walleye are both predacious fish. Total mercury concentrations measured in the axial muscle of northern pike and walleye collected from the study lake in 1971 and 1976 are presented in **Table A8** and **A9**.

**Appendix 3 Estimates of the Transfer or Removal Rates,  $p_i$ ,  $f_i$  and  $d_i$**

**A3.1 Transfer rate,  $p_i$**

The transfer rate,  $p_i$ , was evaluated assuming that mercury in sediment was transferred to water by the processes of resuspension and desorption, and that equilibrium between the mercury concentration in water and that in sediment was attained after the transport time or the residence time. The rate was presented by the following equation:

$$p_i = \frac{TR_i \cdot M_i}{N_i \cdot t_i} \quad (A1)$$

where  $TR_i$ : transfer ratio from sediment to water (kg/l),  $M_i$ : volume of water in i-th compartment (l),  $N_i$ : weight of sediment in i-th compartment (kg),  $t_i$ : transport time or residence time (y).

In the case of a river, Eq.(A1) was re-written as follows:

$$p_i = \frac{10^3 \cdot TR_i \cdot (L_i \cdot D_i \cdot B_i)}{(\rho_i \cdot L_i \cdot B_i) \cdot (L_i / U_i)} = \frac{10^3 \cdot TR_i \cdot D_i \cdot U_i}{\rho_i \cdot L_i} \quad (A2)$$

where  $L_i$ : stream length of i-th compartment (m),  $D_i$ : mean water depth (m),  $B_i$ : width of stream (m),  $\rho_i$ : surface density of sediment (kg/m<sup>2</sup>),  $U_i$ : flow velocity of stream (m/y).

Because no resuspension was assumed in the case of a lake, Eq.(A1) was re-written:

$$p_i = \frac{S \cdot D}{K_d \cdot (\rho \cdot S) \cdot (S \cdot D / I_w)} = \frac{I_w}{K_d \cdot \rho \cdot S} \quad (A3)$$

where  $K_d$ : distribution coefficient (l/kg),  $S$ : area of lake (m<sup>2</sup>),  $I_w$ : water inflow into lake (l/y).

**A3.2 Removal rate,  $f_i$**

The estimate of the removal rate,  $f_i$ , was based on renewal of water. The rate was described by the following equation:



$$f_i = \frac{I_i}{M_i} \quad (A4)$$

where  $I_i$ : water flow (l/y).

In the case of a river, Eq.(A4) was re-written as follows:

$$f_i = \frac{D_i \cdot B_i \cdot U_i}{L_i \cdot D_i \cdot B_i} = \frac{U_i}{L_i} \quad (A5)$$

On the other hand, Eq.(A4) was re-written in the case of a lake:

$$f_i = \frac{I_w}{10^3 \cdot S \cdot D} \quad (A6)$$

### A3.3 Removal rate, $d_i$

In the case of a river, mercury was considered to be removed from water by the processes of deposition and sedimentation on to the bottom. The removal rate,  $d_i$ , was estimated by using a deposition velocity concept:

$$d_i = \frac{V_i}{D_i} \quad (A7)$$

where  $V_i$ : deposition velocity onto sediment or benthic organisms (m/y).

In the case of a lake, it was assumed that mercury was removed from water by only sedimentation. The removal rate,  $d_i$ , was obtained by a particle scavenging model which assumed that mercury dissolved in water was adsorbed on resuspended soil and the soil was settled down to bottom sediment. The rate was expressed by the following equation:

$$d_i = \frac{F_w \cdot K_d \cdot R}{10^3 \cdot D} = \frac{K_d \cdot R}{10^3 \cdot D \cdot (1 + K_d \cdot SS)} \quad (A8)$$

where  $F_w$ : fraction of mercury in water which is in solution,  $R$ : sedimentation rate ( $\text{kg}/\text{m}^2/\text{y}$ ),  $SS$ : suspended soil in water ( $\text{kg}/\text{l}$ ).

Table A1 Mean densities (numbers/m<sup>2</sup>) of benthic macroinvertebrates collected in riffle areas of the stream

Taxon	Sampling site (km)		
	22.8	13.0	10.9
Coleoptera			
<u>Optioservus</u> sp.	-	3.6	-
Decapoda			
<u>Cambarus</u> sp.	1.2	2.4	-
Diptera			
Chironomidae	1666.1	357.6	3589.3
<u>Dicranota</u> sp.	9.6	-	-
<u>Simulium</u> sp.	-	2.4	-
Ephemeroptera			
<u>Baetis</u> sp.	1.2	-	-
Gastropoda			
<u>Physa</u> sp.	35.8	-	-
Nematoda			
Lumbriculidae	-	3.6	-
Oligochaeta	-	2.4	-
Tubificidae	78.9	-	159.1
Odonata			
<u>Enallagma</u> sp.	20.3	-	-
<u>Ischnura</u> sp.	16.7	-	-
Tricoptera			
<u>Cheumatopsyche</u> sp.	3.6	-	-
Number of taxa	9	6	2
Total density	1833.4	372.0	3748.4

Table A2 Mean water quality parameters at 22.8 km under base flow conditions

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Temperature (°C)	22.4
Dissolved oxygen (mg/l)	8.3
pH (standard units)	8.1
Conductivity (µmhos/cm)	454.0
Alkalinity (mg/l as CaCO <sub>3</sub> )	115.0
Turbidity (NTU)	3.5
Total suspended solids (mg/l)	5.0
Total volatile suspended solids (mg/l)	2.0
Hardness (mg/l as CaCO <sub>3</sub> )	170.0
Aluminium (µg/l)	60.0
Organic nitrogen (mg/l)	0.57
Total ammonia nitrogen (mg/l)	0.11
Non-ionized ammonia nitrogen (mg/l)	0.007
Nitrate + nitrite nitrogen (mg/l)	3.8
Total phosphorous (mg/l)	0.66
Total mercury (µg/l)	2.5

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Table A3 Total mercury concentration in sediment (&lt;0.125 mm size fraction)

Station number	Distance (km)	Total mercury ( $\mu\text{g/g}$ dry wt)
1	2.1	19
2	7.7	32
4	10.9	30
5	13.4	55
6	22.2	127
7	22.7	62
8	22.8	90

Table A4 Total mercury concentration measured in 1975 in water, suspended particles, bed sediment and benthic invertebrates at sampling stations above and below the chlor-alkali plant

Station	In water ( $\mu\text{g/l}$ )	Dissolved in water ( $\mu\text{g/l}$ )	Bed sediment ( $\mu\text{g/g}$ dry wt)	Benthic invertebrates ( $\mu\text{g/g}$ wet wt)
-43.0	0.070	0.001	0.14	0.052
-9.0	0.034	-	0.16	0.043
3.7	0.210	0.056	18.5	2.028
9.7	0.074	0.040	9.2	0.876
21.0	0.067	0.027	4.3	0.556
43.0	0.032	0.021	2.4	0.416

Table A5 Water quality parameters measured at five of the sampling stations

Station	pH	DOC <sup>a</sup> (mg/l)	SS <sup>b</sup> (mg/l)	Cond <sup>c</sup> ( $\mu$ mhos/cm)	Cl <sup>d</sup> (mg/l)
-43.0	8.0	2.16	12.0	248	1.8
-9.0	8.1	2.32	7.8	251	4.2
9.7	8.2	2.36	4.2	1179	2.4
21.0	8.4	3.83	6.2	1250	3.8
43.0	8.4	2.60	4.0	821	2.6

a Dissolved organic carbon

b Suspended solids

c Conductivity

d Chlorine

Table A6 Total mercury concentration measured in water and sediment samples collected from the study lake in 1979 and 1980

Site	Water (ng/l)		Sediment ( $\mu$ g/g)
	Mean	Range	0-5 cm depth
	Inflow to study lake	29	17 - 46
Eastern side of lake	25	10 - 46	3.0
Western side of lake	16	7.5 - 22	-
Outflow from study lake	19	12 - 26	2.8

Table A7 Mean water quality parameters

Site	Temp ( $^{\circ}$ C)	Cond <sup>a</sup> ( $\mu$ mhos/cm)	SS <sup>b</sup> (mg/l)	Cl (mg/l)	SO <sub>4</sub> (mg/l)	BOD <sup>c</sup> (mg/l)	pH
Lake inflow	12	126	2.7	11	10	1.8	7.2
Lake outflow	11	125	2.7	9.8	9.7	1.2	7.2

a Conductivity

b Suspended solids

c Biochemical oxygen demand

Table A8 Mercury concentration measured in the axial muscle of northern pike in 1971 and 1976

Year	Mercury	Length		Weight	
	( $\mu\text{g/g}$ )	(cm)		(g)	
	Mean	Mean	Range	Mean	Range
1971	9.09	50.8	36.0 - 62.0	974	312 - 1870
1976	5.84	65.0	50.1 - 86.2	1793	879 - 4423

Table A9 Mercury concentration measured in the axial muscle of walleye in 1971 and 1976

Year	Mercury	Length		Weight	
	( $\mu\text{g/g}$ )	(cm)		(g)	
	Mean	Mean	Range	Mean	Range
1971	8.71	42.6	33.0 - 50.0	1064	454 - 1850
1976	7.83	49.8	40.8 - 59.4	1283	482 - 2013

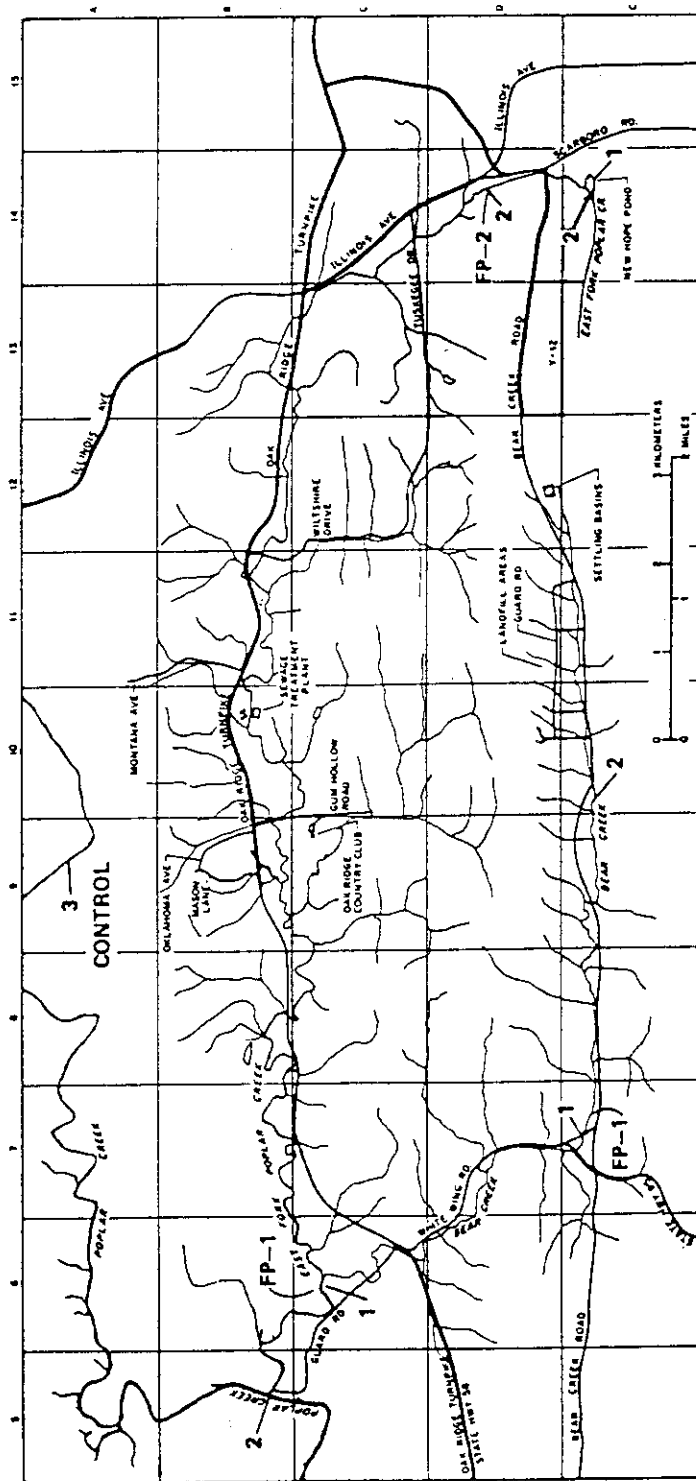


Fig.A1 Map of East Fork Poplar Creek drainage area



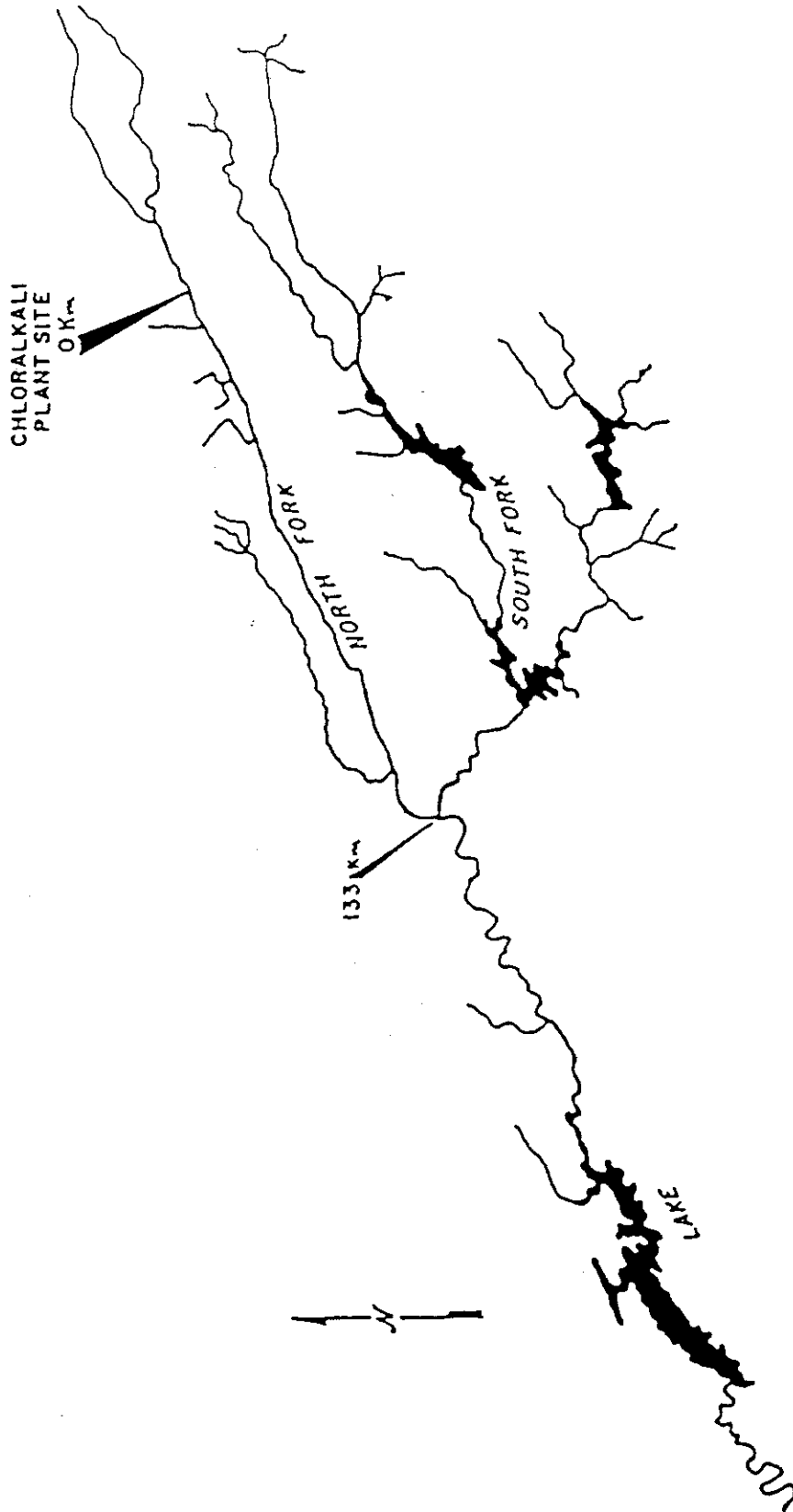


Fig.A2 Map of North Fork Holston River drainage system

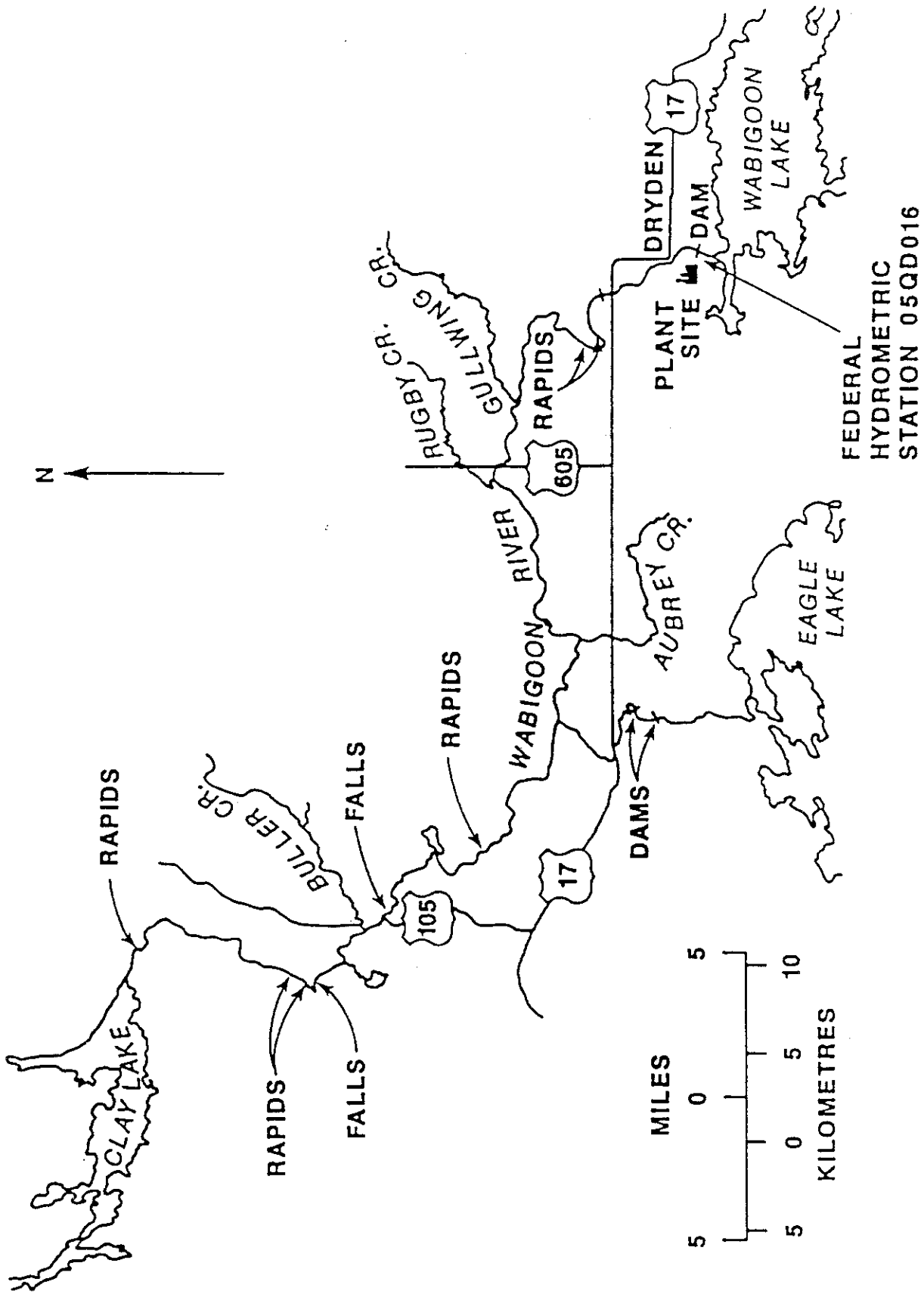


Fig.A3 Major morphological features of Wabigoon River and Clay Lake