Application of a mathematical model to predict dioxin concentrations in the Tokyo Bay estuary

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Introduction

In order to assess the ecological risk posed by dioxins in the environment, information regarding the distribution of dioxin concentration under long term monitoring and over a wide area is necessary. However, it is almost impossible to obtain such data because dioxin analysis needs considerable time and effort. Thus, mathematical models that can predict dioxin concentrations in the environment are required. Especially in Japan, where main pathway of human exposure to dioxins is through consumption of fish and shellfish, investigation of aquatic environment is very important. However, data on these environmental mediums, such as river, bay, and inner sea, are very limited. In this study, a 3-D chemical fate prediction model was developed and applied to the Tokyo Bay estuary (see Figure 1) to predict distributions and variations of polychlorinated dibenzo-*p*-dioxins, dibenzofurans (PCDD/Fs), and dioxin-like PCBs in the seawater of the estuary.

Methods and Materials

General Description of the Model

A 3-D chemical fate prediction model (FATE3D), which was developed by the Research Center for Chemical Risk Management, National Institute of Advanced Industrial Science and Technology, Japan, was applied to predict dioxin concentrations in the Tokyo Bay estuary. This model can take into consideration three forms of dioxins (particulate-phase dioxins, dissolved-phase dioxins, and dioxins in sediment) in the estuary, and can simulate the diffusion and sinking processes of dioxins from the loading points of their sources. This model requires meteorological parameters (temperature, wind conditions, tide level, river water discharge, etc.), flow field data (current velocity, water temperature, salinity, coefficients of horizontal and vertical eddy diffusivity, etc.), particulate matter (phytoplankton and detritus) concentrations, loading flux, and physical-chemical property of dioxins as input data for the calculations.

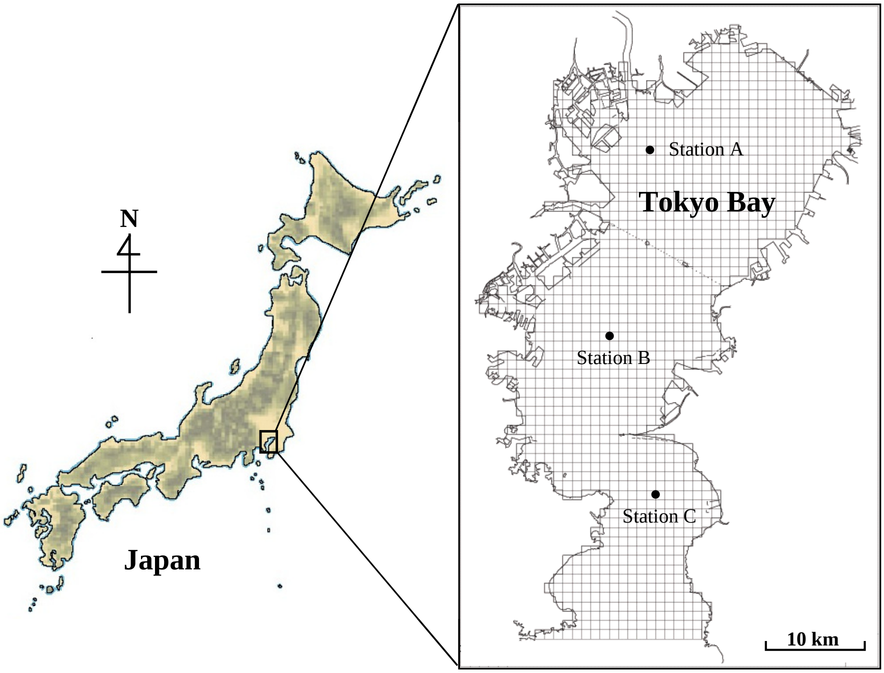

Figure 1: Location of the target area.

Schematic view of the FATE3D model is shown in Figure 2. In this study, the following five processes of dioxin transportation and change were taken into account to predict dioxin concentrations in the estuary.

1. Loading flux of dioxins: Loading fluxes from rivers that flow into the Tokyo Bay estuary and atmospheric depositions were considered to constitute dioxin loading into the estuary. However, in this study, only the loading fluxes from rivers were taken into account as the dioxin loading to the estuary. This is because we concluded that the loading fluxes of rivers were the major source of dioxins in the Tokyo Bay estuary[1)] whereas atmospheric deposition was not. Furthermore, two phases of dioxins (i.e., particulate- and dissolved-phase dioxins) were considered in the flux of dioxins from rivers.

2. Partition between particulate- and dissolved-phase dioxins: Dioxins in the Tokyo Bay seawater were considered to exist as particulate- or dissolved-phase dioxins. We took into account two types of particulate matters (phytoplankton and detritus) in the model. Further, we assumed that the partition between particulate- and dissolved-phase dioxins was determined by the adsorption equilibrium.

3. Horizontal and vertical transportation of dioxins: Particulate- and dissolved-phase dioxins from rivers are diluted and diffused by the seawater, and then transported by tidal flow into the estuary. Vertical transportation was not only determined by tidal flow, but also by the sinking of particulate

matter. We assumed that the sinking rates of particulate-phase dioxins were determined by the sinking rate of particulate matter, which mainly consists of phytoplankton and detritus.

4. Interaction between seawater and sediment: Sinking of particulate matter, resuspension of sediment, adsorption (or desorption) between dissolved-phase dioxins and dioxins in sediment are generally considered to be the interaction between seawater and sediment. However, in this study only the sinking of particulate matter was taken into account.

5. Biodegradation: Biodegradation of dioxins was not taken into account in this study.

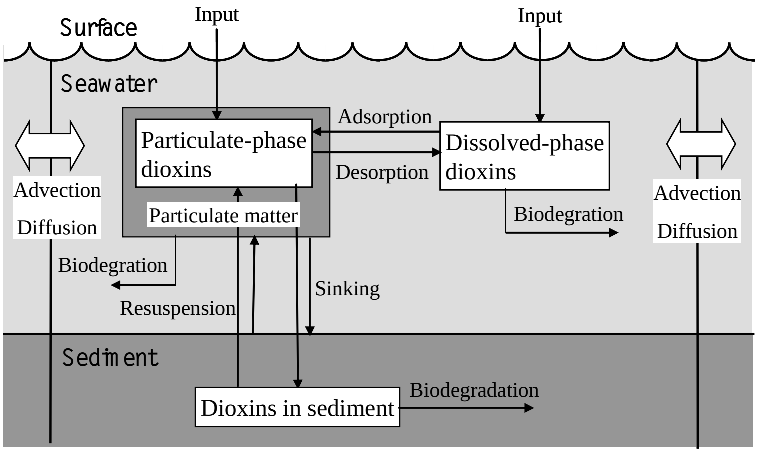

Figure 2: Schematic view of the FATE3D model.

Governing Equations of the Model

Governing equations of the FATE3D model are expressed below.

1. Dissolved-phase dioxins: The transportation and change for the dissolved-phase dioxins in the estuary are expressed below.

$$\frac{\partial G_w}{\partial t} = -(v \cdot \nabla)G_w - w\frac{\partial G_w}{\partial z} + [\nabla \cdot (K_H \nabla)]G_w + \frac{\partial}{\partial z}\left(K_z \frac{\partial G_w}{\partial z}\right)$$
$$-\lambda G_w - \sum_{j=1}^{N} K_j (C_j \cdot K_{dj} \cdot G_w - G_j) + Q_w \ ,$$

where G_w is the concentration of dissolved-phase dioxins, v and w are the horizontal and vertical vector of flow field, respectively, ∇ is the gradient, K_H and K_z are the coefficients of horizontal and vertical eddy diffusivity, respectively, λ is the degradation rate, Q_w is the loading flux of dissolved-phase dioxins from rivers, K_{dj} is the partition coefficient between dissolved-phase dioxins and particulate-phase dioxins bound to the particulate matter j (j = 1: phytoplankton, j = 2: detritus), C_j is the concentration of particulate matter j, G_j is the concentration of particulate-phase dioxins bound to particulate matter j, and K_j is the adsorption rate of dissolved-phase dioxins to particulate matter j.

2. Particulate-phase dioxins: The transportation and change for the particulate-phase dioxins bound to particulate matter j in the estuary are expressed below.

$$\frac{\partial G_j}{\partial t} = -(v \cdot \nabla) G_j - (w - w_{sj}) \frac{\partial G_j}{\partial z} + [\nabla \cdot (K_H \nabla)] G_j + \frac{\partial}{\partial z}\left(K_z \frac{\partial G_j}{\partial z} \right)$$
$$- \lambda G_j - K_j (G_j - C_j \cdot K_{dj} \cdot G_w) + Q_j ,$$

where G_j is the concentration of particulate-phase dioxins bound to particulate matter j, w_{sj} is the sinking rate of the particulate matter j, and Q_j is the loading flux of particulate-phase dioxins from rivers.

Since loading fluxes and physical-chemical properties (e.g., partition coefficient) of dioxins differ widely in each dioxin congener, it is thought that the calculation should be done for each congener. However, it takes considerable time to set these parameters for all congeners and caluculate for each congener. Since congeners in the same homologue have almost the same properties, calculations were done for each homologue. The loading flux and concentration were converted to TEQ value and summed for each homologue. And the average physical-chemical properties in each homologue were used.

Parameters of the Model
Important parameters for the FATE3D model and selected values that were used in this study are summarized in Table 1.

Table 1: Parameters for the FATE3D model.

Parameters	Selected values
Target area	Entire Tokyo Bay area (see Figure 1).
Number of horizontal meshes	71 (north to south) - 45 (east to west) (see Figire 1).
Size of horizontal meshes [km]	1 (north to south) - 1 (east to west) (see Figire 1).
Number of vertical layers	10.
Layer locations [m]	1st layer: surface to -2, 2nd layer: -2 to -4, 3rd layer: -4 to -6, 4th layer: -6 to -8, 5th layer: -8 to -10, 6th layer: -10 to -12, 7th layer: -13 to -16, 8th layer: -16 to -19, 9th layer: -19 to -22, 10th layer: -22 to bottom.
Calculation period	1 year (From Sep. 1, 2002 to Aug. 31, 2003).
Time step of calculation [sec]	200.
Number of loading sources of dioxins	6. (Edo, Naka, Ara, Sumida, Tama, and Tsurumi Rivers)
Meteorological parameters	Meteorological parameters (temperature, wind conditions, tide level, river water discharge, etc.) were obtained from Japan Meteorological Agency (2002, 2003)[2)].
Flow field data	Flow field data (current velocity, water temperature, salinity, coefficients of vertical eddy diffusivity, etc.) were obtained from the results of calculations by the flow field model (COSMOS) reported by Horiguchi et al. (2001)[3)] and Taguchi et al. (1999a)[4)].
Particulate matter concentrations [mg/L]	Particulate matter (phytoplankton, detritus) concentrations were obtained from the results of calculations by the coastal ecosystem model (EUTROP) reported by Taguchi et al. (1999b)[5)].
Initial conditions of dioxin concentrations [fg-TEQ/L]	Average concentrations of measured values in the estuary reported by Kobayashi et al. (2003)[1)] were used.
Boundary conditions of dioxin concentrations [fg-TEQ/L]	0 for all dioxin homologues, both particulate- and dissolved-phase dioxins.
Loading flux of dioxins from rivers [fg-TEQ/L]	It is considered that loading fluxes from rivers vary daily in response to river water discharges. These fluxes were obtained from Kobayashi et al. (2003)[1)].
Sinking rate of particulate matter [cm/s]	Sinking rates of particulate matters were obtained from Taguchi et al. (1999b)[5)]. phytoplankton: 2.0×10^{-4}, detritus: 5.0×10^{-4}
Adsorption rate [1/sec]	Adsorption rates were obtained from Ministry of the Environment, Japan (1992)[6)]. phytoplankton: 2.0×10^{-5}, detritus: 2.0×10^{-5}
Partition coefficient [log K_{oc}]	Partition coefficients were set for each homologue. These values were obtained from Mackay et al. (1992)[7)]. TeCDF: 5.71, PeCDF: 6.11, HxCDF: 6.61, HpCDF: 7.01, OCDF: 7.61, TeCDD: 6.41, PeCDD: 7.01, HxCDD: 7.41, HpCDD: 7.61, OCDD: 7.81, TeCB: 5.61, PeCB: 5.81, HxCB: 6.12, HpCB: 6.37

Results and Discussion

Figure 3 shows the comparison of particulate-phase dioxin concentrations between measured values and values predicted by the model at Station A, Station B, and Station C. In Figure 3, concentrations are expressed in TEQ values of total dioxins (concentrations of each PCDD/F and dioxin-like PCB congener were converted to TEQ and summed). It is shown that concentrations of dioxins, both particulate- and dissolved-phase (dissolved-phase dioxins are not shown), predicted by the model compared favorably with the field measurements in the estuary indicating the validity and predictive capability of the model. It turned out that dioxin concentrations varied in response to the loading fluxes from rivers and seasonal variation of phytoplankton abundance in the bay, especially at Station A. Figure 4 shows the distribution of dioxins concentration in Tokyo Bay predicted by the model (1 year average). In Figure 4, concentrations are expressed in total TEQ values. It was predicted that particulate-phase dioxins ranged from approximately 2.5 fg-TEQ/L to 200 fg-TEQ/L and dissolved-phase dioxins ranged from approximately 1.0 fg-TEQ/L to 20 fg-TEQ/L in the estuary. It was also predicted that particulate-phase dioxins from rivers were immediately deposited after they flowed into the estuary and were not transported outside the estuary. On the other hand, it was predicted that dissolved-phase dioxins transported from the river were immediately diluted with seawater, revealing no marked variation in the estuary.

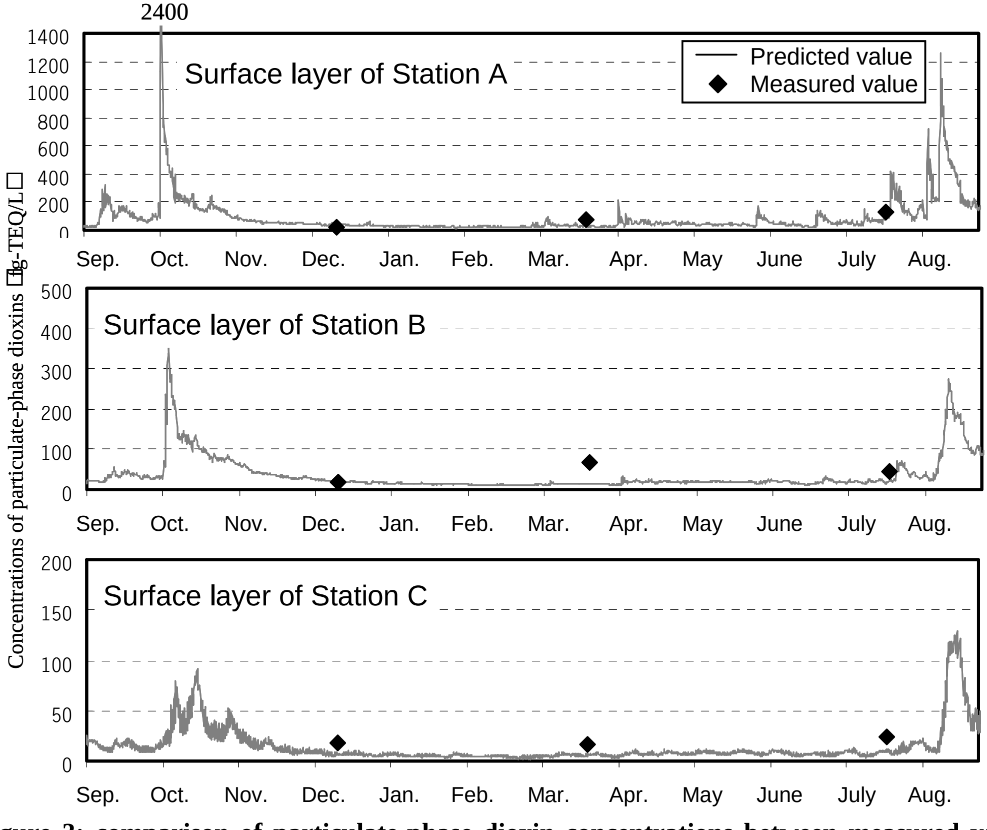

Figure 3: comparison of particulate-phase dioxin concentrations between measured values and predicted values.

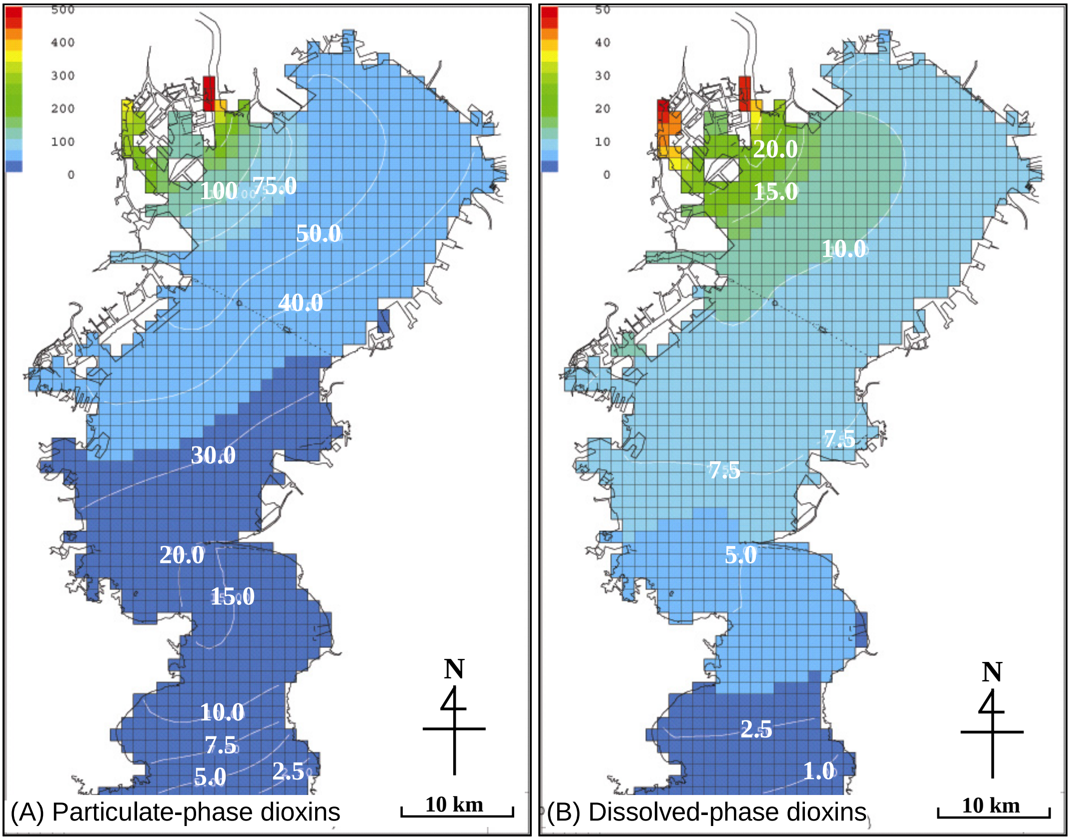

Figure 4: Predicted distributions of dioxin concentrations in Tokyo Bay (1 year average, expressed in fg-TEQ/L).

Acknowledgements

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