Estimation of Dioxin-Levels in Japanese by Mathematical Models: Time Course from the Past to the Future

Kikuo Yoshida[1,2,3], Shino Ikeda[1] and Junko Nakanishi[1,2]

[1] Yokohama National University, 79-7 Tokiwadai, Hodogaya-ku, Yokohama 240-8501, JAPAN
[2] CREST, Japan Science and Technology Corporation
[3] Mitsubishi Chemical Safety Institute Ltd.

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Abstract

Transport pathways of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) from sources to humans are simply modeled to estimate the time course of the background levels in intake and body burden in Japanese from the past to the future. The results of our approaches in terms of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin toxic equivalents (TEQs) seem to represent reasonably well the background levels of PCDD/Fs in the environment, intake, and body burden. Furthermore, the results quantitatively support the fragmentary evidence for a tendency toward lower levels in intake and body burden of Japanese. By combining mathematical models to effectively estimate on-site and off-site exposure, the modeling approaches can contribute to the determination of the major exposure pathways of residents to dioxins at assessment sites and to select quantitatively the most effective measure to reduce the exposure of the residents concerned.

1. Introduction

In the latter half of 1990s, various types of monitoring of dioxins in environmental and biological media were carried out in Japan, because of the great social concern about risk posed by this chemical group. On the basis of the monitoring results which have been made public, the present level of risk to human health due to dioxins has been evaluated, and the Japanese Government has established countermeasures to reduce dioxin levels in emissions from incinerators and to determine Tolerable Daily Intake of dioxins.

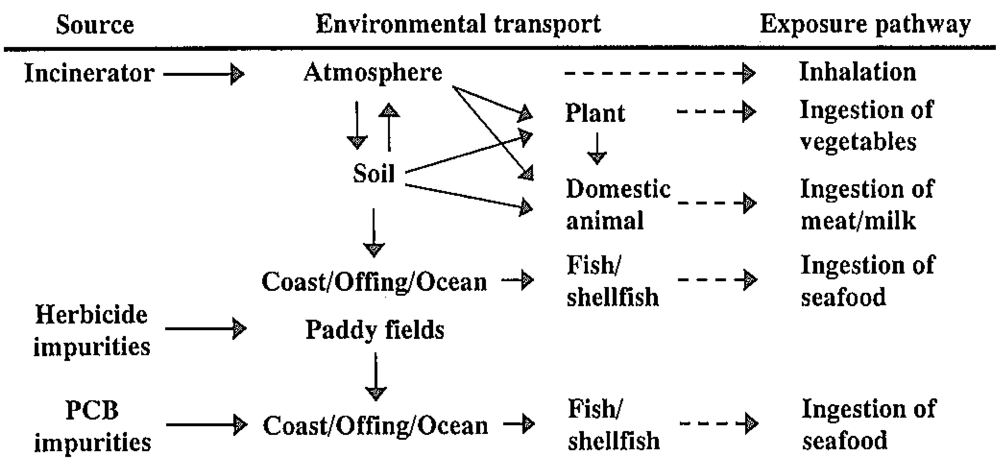
Unfortunately, there has been less study on past dioxin levels in the Japanese environment and in the human body. However, it has been suggested that the intake of dioxins via foods and the body burden of dioxins in Japanese were much higher in the past, based on the results of a recent analysis of breast milk and foods stored in the past by the Ministry of Health and Welfare (MHW) (MHW, 1998a, 1999). Ingestion rates of various foods have remained almost unchanged during the past 30 years in Japan (MHW, 1998b). Therefore, the results of these surveys suggest that there were other sources of dioxins in addition to solid waste incinerators in the past, because the quantity of solid waste is anticipated to increase yearly. In this context, it has been reported that rather large quantities of dioxins were released into Japanese paddy fields as impurities in some herbicides in the past (Masunaga, 1998, 1999). Indeed, the concentrations of dioxins in paddy fields were found to be significantly higher than those in other agricultural fields (Environment Agency, 1999a).

Mathematical modeling is one effective approach to consistently integrating various types of data, analyzing them quantitatively, and assessing changes in exposure-levels from the past to the future. Furthermore, the models are also valuable for evaluating the effectiveness of risk reduction measures.

In this study, we simply modeled the transport pathways of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) from sources to the human body and estimated the time course of background levels of PCDD/Fs in Japanese from the past to the future in terms of 2,3,7,8- tetrachlorodibenzo-*p*-dioxin toxic equivalents (TEQs), based on estimated rates of release from major sources (Masunaga, 1999).

2. Description of Modeling Approaches

To estimate the exposure of the general Japanese population to PCDD/Fs, we considered the following sources, environmental transport pathways, and exposure pathways.



2.1 *PCDD/Fs emitted from incinerators*

The following two-compartment model was applied to estimate the environmental dynamics of PCDD/Fs from incinerators in the air and soil other than in paddy fields:

$$\frac{dMa}{dt} = E - k_{11} \bullet Ma + k_{12} \bullet Ms \tag{1}$$

$$\frac{dMs}{dt} = k_{21} \bullet Ma - k_{22} \bullet Ms \tag{2},$$

where Ma and Ms are the mass in air and soil, respectively. E is for the emission rate into air. k_{11} and k_{22} are the lumped 1st-order rate constants for the transport, transfer, and transformation processes of PCDD/Fs in air and soil, respectively. k_{12} and k_{21} are the lumped 1st-order rate constants for transfer between air and soil, respectively. A detailed description of this model has been presented elsewhere (Yoshida et al., 1999a).

The mass transported from the soil, other than in paddy fields, to the coast is calculated by

$$I_{sea} = (k_{runoff} \bullet fs_w + k_{erosion} \bullet fs_s)Ms \tag{3},$$

where k_{runoff} and $k_{erosion}$ are rate constants for surface runoff and erosion, respectively. f_{sw} and f_{ss} are mass distribution fractions for dissolved and adsorbed phases in soil, respectively.

Another two-compartment model was applied to estimate the dynamics of PCDD/Fs transported into the aquatic environment:

$$\frac{dMw}{dt} = I_{sea} - k_{11} \bullet Mw + k_{12} \bullet Mse \tag{4}$$

$$\frac{dMse}{dt} = k_{21} \bullet Mw - k_{22} \bullet Mse \tag{5},$$

where Mw and Mse are the mass in water and in sediment, respectively. k_{11}, k_{12}, k_{21}, and k_{22} are the lumped 1st-order rate constants calculated as

$$k_{11} = k_{adv} + k_{degw} + (k_{vol} + k_{wse}) \bullet fw_w + k_{dep} \bullet fw_{SS} \tag{6}$$

$$k_{12} = k_{sew} \bullet fse_w + k_{resusp} \bullet fse_s \tag{7}$$

$$k_{21} = k_{wse} \bullet fw_w + k_{dep} \bullet fw_{SS} \tag{8}$$

$$k_{22} = k_{degse} + k_{sew} \bullet fse_w + (k_{resusp} + k_{burial}) \bullet fse_s \tag{9},$$

where k_{adv}, k_{degw}, k_{vol}, k_{wse}, and k_{dep} are 1st-order rate constants for advection, degradation in water, diffusive transfer to sediment, and deposition of suspended solids, respectively. k_{degse}, k_{sew}, k_{resusp}, and k_{burial} are also 1st-order rate constants for degradation in sediment, diffusive transfer to water, resuspension, and burial in sediment, respectively. fw_w and fw_{SS} are mass distribution fractions of dissolved and sorbed phases in water, respectively. fse_w and fse_s are mass distribution fractions of dissolved and sorbed phases in sediment, respectively. These fractions are calculated as

$$fw_w = \frac{Vw}{Vw + Koc \bullet OC_{SS} \bullet W_{SS}} \tag{10}$$

$$fw_{SS} = \frac{Koc \bullet OC_{SS} \bullet W_{SS}}{Vw + Koc \bullet OC_{SS} \bullet W_{SS}} \tag{11}$$

$$fse_w = \frac{\phi}{\phi + (1-\phi) \bullet Koc \bullet OC_{se} \bullet Dses} \quad (12)$$

$$fse_s = \frac{(1-\phi) \bullet Koc \bullet OC_{se} \bullet Dses}{\phi + (1-\phi) \bullet Koc \bullet OC_{se} \bullet Dses} \quad (13),$$

where Vw and W_{SS} are the volume of water and weight of suspended solids, respectively. ϕ, OC_{se}, and $Dses$ are the porosity of sediment, organic carbon content, and density of sediment solids, respectively.

This model was also applied to estimate the dynamics of PCDD/Fs transported from paddy fields and directly released into the aquatic environment as PCB impurities.

2.2 *PCDD/Fs released into paddy fields*

The following one-compartment model was applied to estimate the dynamics of PCDD/Fs in paddy fields:

$$Mrf = APPL \bullet \exp(-krf_{total} \bullet t) \quad (14),$$

where $APPL$ is the mass of released PCDD/Fs. krf_{total} is the total 1st-order rate constant of runoff, erosion, and degradation in paddy soil. These processes contribute to the removal of PCDD/Fs from the top layer of the soil (0 – 5 cm), but only degradation contributes to the removal from the lower layer of soil (5 – 20 cm). The two soil layers are tilled once a year, and concentrations of PCDD/Fs in the two soil layers become uniform.

The mass transported from the paddy fields to the coast is calculated by

$$I_{sea} = (k_{runoff} \bullet frs_w + k_{erosion} \bullet frs_s)Mrf \quad (15),$$

where frs_w and frs_s are mass distribution fractions for dissolved and adsorbed phases in paddy soil, respectively.

2.3 *PCDD/Fs in intake media*

Average daily intakes via various pathways are individually calculated as products of concentrations of PCDD/Fs in each intake medium and its ingestion or inhalation rate. The concentration in coastal fish is calculated based on the assumption of equilibrium between the concentrations in water and fish. The concentrations in offshore and oceanic fish are assumed to be half of that in coastal fish (MHW, 1996). The concentrations in crops, meat, and dairy products are estimated according to the methods described by the U.S. EPA (U.S. EPA, 1994).

To calculate the average daily intake, the following two assumptions are made; ingestion rates of various foods are constant, and the ratio of ingestion rates of coastal fish and other fish was 0.4 before 1970 and has been 0.3 since then.

2.4 *Body burden of dioxins*

The body burden of PCDD/Fs is estimated by the following one-compartment model:

$$M_{human} = \frac{I}{ke}\{1 - \exp(-ke \bullet t)\} \quad (16),$$

where M_{human} is the body burden of PCDD/Fs, and the I is the internal dose. ke is the elimination rate constant. The predictability of this model and bioavailability for calculating internal doses are described elsewhere (Yoshida et al., 1999b).

2.5 *Sensitivity analysis of model parameters*

The sensitivity of environmental concentrations and body burden of PCDD/Fs to the input parameters was analyzed by Monte Carlo simulation. For each parameter analyzed, we defined a normal distribution, and the average and standard deviation of the distribution were fixed as the input value and 10 % of the input value, respectively.

3. Results and Discussion

3.1 *Environmental levels of PCDD/Fs*

The estimated time course of the background levels of PCDD/Fs in the Japanese environment is shown in Figure 1. Because our modeling approaches do not focus on site-specific assessment at each monitoring station, the concentrations at each station are not estimated. However, as shown in Table 1, the estimated concentrations for recent years are within the range of the measured concentrations (Environment Agency, 1996a, 1996b, 1998a, 1998b, 1999a, 1999b).

Furthermore, it is estimated that even now, 75 % of PCDD/Fs transported into the aquatic environment and 84 % of PCDD/Fs in sediment originate from paddy fields.

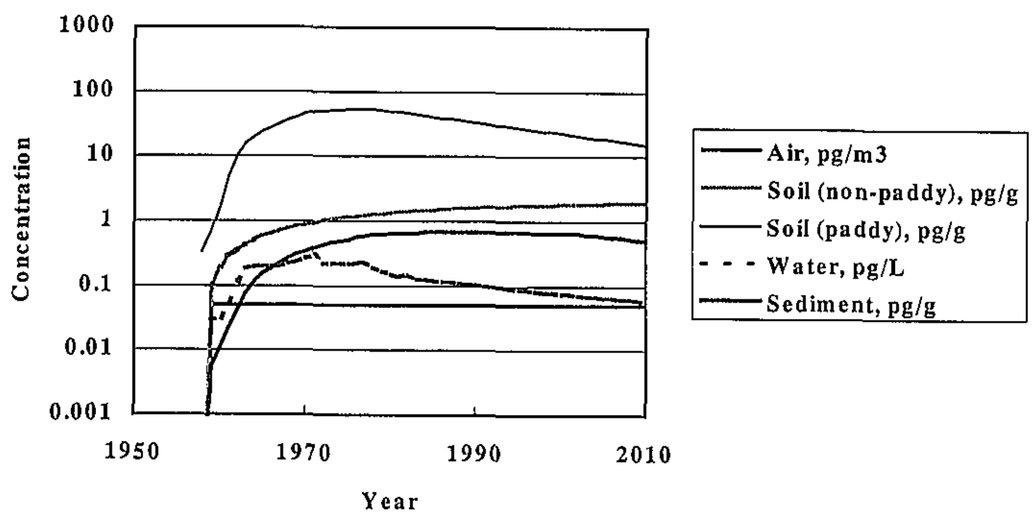


Fig. 1 Estimated time course of background levels of PCDD/Fs in the Japanese environment

Table 1. Comparison of measured and calculated environmental concentrations of PCDD/Fs

Environmental medium	Year	Calculated	Measured
Air, pg/m^3	1997	0.052	0.01 – 0.18*
	1998	0.052	N.D. – 0.067**
Soil other than in paddy fields, pg/g	1998	1.8	0.13 – 5.6**
Paddy soil, pg/g	1998	25	15 – 130**
Seawater, pg/L	1995	0.09	N.D. – 0.3***
	1997	0.09	0.005 – 0.18*
Sediment, pg/g	1995	0.68	0.26 – 75*
	1997	0.67	0.012 – 49*
Fish, pg/g	1997	0.43	N.D. – 2.80*

*: I-TEQ **: WHO(97)-TEQ ***: NATO-TEQ

3.2 *Daily intake of PCDD/Fs*

Estimated average daily intakes via food ingestion are compared with those reported (MHW, 1999). As shown in Figure 2, the average daily intakes via ingestion of fish and vegetables are in good agreement with those measured. However, for meat and dairy products, average daily intakes stored in 1977, 1982, and 1988 were significantly higher than those calculated. The major transport pathway of PCDD/Fs to green vegetables and grass is thought to be absorption and deposition from air. Therefore, the origin of PCDD/Fs, which contaminate the feed of domestic animals, may not be due to emission into air, because the average daily intakes via green vegetables during the past 20 years are more stable than those via ingestion of meat and dairy products.

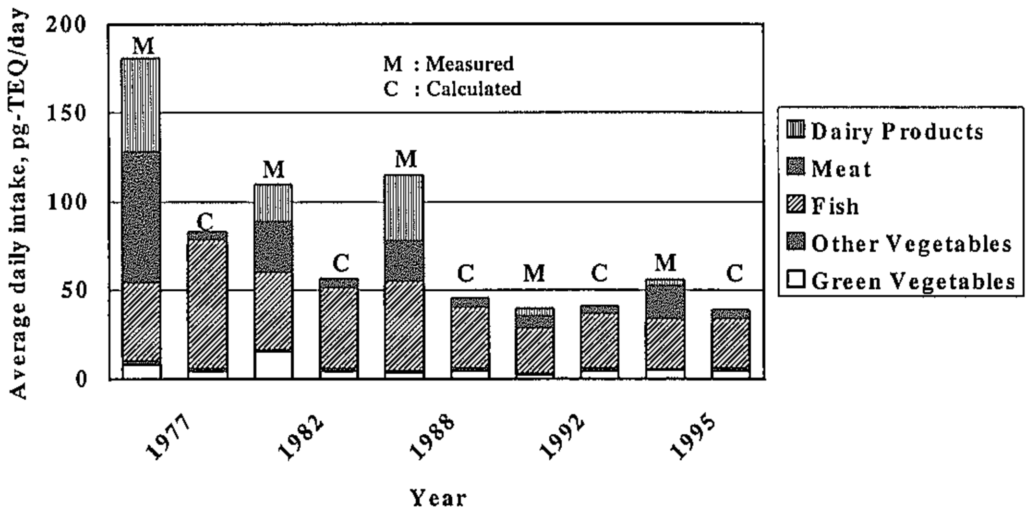


Fig. 2 Comparison between measured and calculated average daily intakes

3.3 *Concentration of PCDD/Fs in fat*

Figure 3 shows estimated concentrations of PCDD/Fs in the fat of 27-year-old females with those measured in the breast milk of 25 – 29-year-old females (MHW, 1998a). As shown in this figure, tendencies of measured and calculated concentrations toward lower levels of PCDD/Fs in fat after 1980 are in good agreement. A slight underestimate of the concentrations before 1980 may be due to the following reasons: no consideration of the intake of PCDD/Fs before 1958 and an unexpectedly high intake via the ingestion of meat and dairy products, as described above.

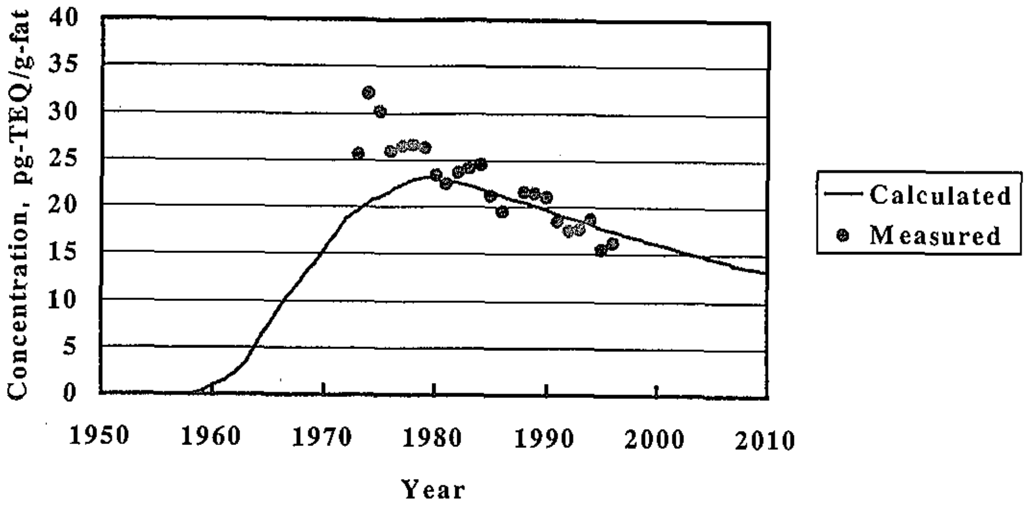


Fig 3 Estimated concentrations of PCDD/Fs in the fat of 27-year-old females

3.4 *Sensitivity analysis of model parameters*

The body burden of PCDD/Fs in Japanese is mainly due to the ingestion of fish which have accumulated PCDD/Fs from paddy fields. Therefore, the body burden is affected by the model parameters which describe the dynamics of PCDD/Fs in paddy fields and the aquatic environment. As shown in Figure 4, parameters related to the adsorption to suspended solids and advective outflow of PCDD/Fs in water have a primary effect on the current concentrations in human fat, and parameters related to persistency in soil and transport to the aquatic environment have a secondary effect. For site-specific or isomer-specific assessment of exposure to dioxins, these effective parameters must be specified as valid values or appropriate probability density functions.

4. Conclusions

Although the final goal of this study is the isomer-specific estimation of internal exposure levels to dioxins taking account of transport from sources to humans, the results of approaches in terms of TEQs seem to estimate reasonably well the current and past background levels of PCDD/Fs in the environment, intake, and body burden. Furthermore, the results quantitatively support the fragmentary evidence for a tendency toward lower levels in intake and body burden of Japanese.

By combining mathematical models to effectively estimate on-site and off-site exposure to dioxins, the modeling approaches will be able to contribute to the determination of the major exposure pathway of residents to dioxins at assessment sites and to select quantitatively the most effective measure to reduce the exposure of the residents concerned.

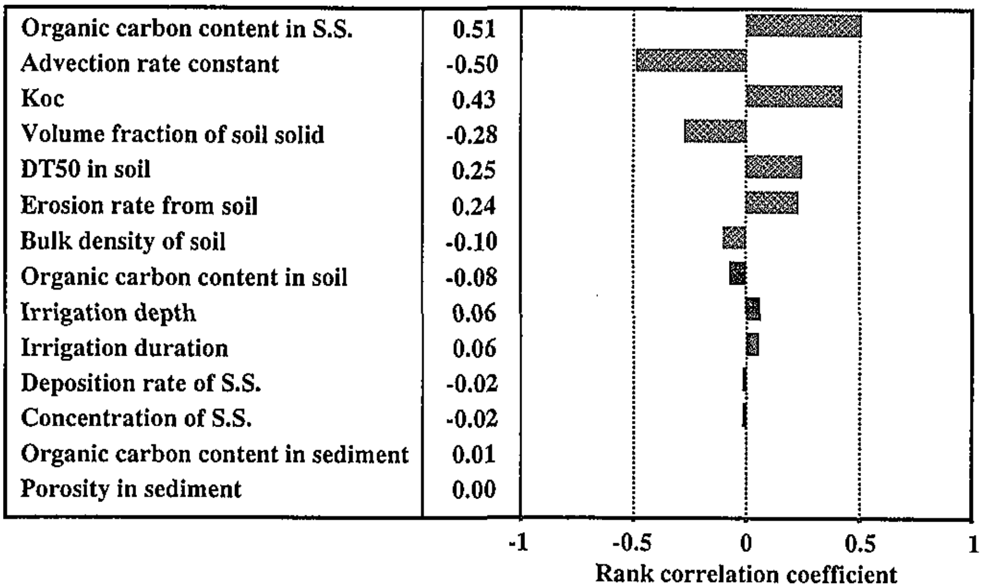


Fig 4 Results of sensitivity analysis of model parameters for paddy fields and aquatic environment

5. Acknowledgments

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