

**Source and Behavior Analyses of Dioxins Based on Congener-Specific Information
and Their Application to Tokyo Bay Basin**

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Abstract

Identification of pollution sources and estimation of their contribution to dioxin pollution are important for taking better countermeasures against such sources. The information based on seventeen 2,3,7,8-chlorine-substituted congeners and ten homologues is usually not sufficient for source identification. The usefulness of detailed congener-specific information of tetra- to octachlorinated polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans was examined by principal component analysis and compared with conventional dioxin information. It was revealed that congener-specific information was far more effective for source identification than conventional information. For source apportioning, it was shown that multiple regression analysis with detailed source congener profiles could be carried out successfully when calculations were performed for individual homologues. This was because of the large variation in homologue composition in source dioxin profiles in addition to the difference in environmental behavior among the homologues.

The newly proposed methods for statistical analysis were applied to the estimation of dioxin mass balance in Tokyo Bay basin. It was found that Tokyo Bay sediment core contained dioxins from three major sources: combustion, pentachlorophenol (PCP), and chloronitrophen (CNP). PCP and CNP contained large amounts of dioxin as impurity and were used extensively as paddy field herbicides in Japan in the past. The total dioxin load from the two herbicides to the basin during the past 45 years (1951-1995) was estimated to be five times larger than that from combustion source in terms of toxic equivalents (WHO-TEQ). However, in the surface sediment of the bay, the contribution from the herbicides was nearly equal to that from combustion. The herbicide contribution peaked during the 1960s and 1970s and gradually decreased

thereafter.

Keywords: Dioxin; Source; Mass balance; Congener profile; Principal component analysis; Tokyo Bay

1. Introduction

The identification of pollution source is very important for taking better countermeasures against dioxin pollution. Seventeen 2,3,7,8-chlorine-substituted polychlorinated dibenzo-*p*-dioxin (PCDD) and polychlorinated dibenzofurans (PCDF) congeners and ten PCDD/DF homologues are analyzed during oconventional dioxin monitoring. Such information, however, is not sufficient for source identification. To solve this problem, we have analyzed all the tetra- to octachlorinated PCDD/DF congeners and attempted to identify the behaviors and sources of dioxins based on detailed information. The results obtained are encouraging.

In this study, we compared the source identification potential of different levels of dioxin monitoring information and proposed methods for estimating the contributions of different sources to dioxin pollution. Based on the proposed methods, dioxin mass balances in Tokyo Bay during the past 45 years were estimated.

2. Materials and Methods

We conducted comprehensive monitoring of dioxins in the Kanto region, Japan. The region consists of the Tokyo metropolitan area, agricultural fields on the outskirts, surrounding mountains, and Tokyo Bay. The analyzed environmental samples included air, atmospheric deposition, soil, aquatic sediment, and sediment core. Detailed congener-specific analysis of tetra- to octachlorinated dioxins was conducted using both DB-5 and DB-17 columns (J&W Scientific) on HRGC/HRMS (Micromass Autospec-Ultima coupled with Hewlett-Packard HP6890GC) following conventional pretreatment procedures. The analytical procedures and the obtained data have been presented elsewhere (Sakurai *et al.*, 2000; Yao *et al.*, 2000; Ogura *et al.*, 2001a; Ogura

et al., 2001b). Using the DB-5 column, tetra- to octachlorinated PCDDs and PCDFs congeners were separated into more than 80 peaks. Each peak represented either an isolated congener or a group of congeners.

Statistical analyses, principal component analysis and multiple regression analysis of the data were performed using a software package (STATISTICA 2000 for Windows Release 5.5A, StatSoft, Inc., Tulsa, OK, USA) on a personal computer.

3. Results and Discussion

3.1. Behaviors of dioxins in the environment

3.1.1. Homologue profile

Dioxin homologue profiles for air, atmospheric deposition and soil (which received dioxins only through atmospheric deposition) samples from the Kanto region were monitored. The profiles were different, depending on the media and the location (Fig. 1). Homologue profiles of combustion flue gases varied among solid waste incineration plants (Figs. 1-(a) - 1-(d)). Homologue profiles for air (Fig. 1-(i)) and atmospheric deposition (Figs. 1-(e) - 1-(h)) were similar compared with those for combustion flue gases, but were slightly different between four locations. Error bars in Figs. 1-(e) - 1-(i) show the standard deviations observed during monthly samplings over a one-year period. They indicated that the profiles were similar seasonally in each location. The homologue profile for urban and undisturbed soils, which received dioxins exclusively from atmospheric deposition, resembled those of air and atmospheric deposition except for OCDD (Figs. 1-(j) and 1-(k)). The OCDD level was high in the sediment from Tokyo Bay (Fig. 1-(l)). The relative content of OCDD seemed to increase from air, through atmospheric deposition, to soil. The results of the

monitoring showed that homologue profiles for the samples from the same region were not uniform and indicated that each dioxin homologue was transported in a particular way between environmental media.

Fig. 1 Dioxin homologue profiles for different media and locations.

4F, 5F, 5F, 6F, 7F, 4D, 5D, 6D, 7D and 8D are TeCDFs, PeCDFs, HxCDFs, HpCDFs, OCDF, TeCDDs, PeCDDs, HxCDDs, HpCDDs and OCDD, respectively.

3.1.2. Isomer profile

We have also monitored detailed dioxin congener profiles in addition to the homologue profiles. A comparison of PCDF congener profiles for different media is shown in Fig. 2. Congener profiles are shown in percentage composition in each homologue, namely isomer composition. In the case of combustion flue gas samples, isomer compositions were relatively similar (Fig. 2-(a)). The isomer profiles in air and atmospheric deposition were quite similar irrespective of sampling location and season and resembled to those for combustion flue gas samples (Fig. 3-(b)). The profile for the urban soil sample was also very similar to those for air and atmospheric deposition. However, when dioxin sources other than combustion were present, isomer composition became different, as shown in the case of Tokyo Bay sediment (Fig. 3-(c)). The difference in profile between urban soil and Tokyo Bay sediment may be explained by the contribution of dioxin impurities from agrochemicals such as pentachlorophenol (PCP) and chloronitrophen (CNP), which were used extensively as rice field herbicides

in Japan in the past (Fig. 2-(d)).

The observation of isomer profiles indicated that the isomer profiles remained the same regardless of the environmental media when they had the same origin. This may be because the dioxin isomers, which have the same number of chlorine atoms, have similar physical characteristics and are thus transported and partitioned among environmental media in a similar manner.

Fig. 2 Dioxin isomer profiles for different sources and media from Kanto region.

PCP: pentachlorophenol, CNP: chloronitrophen.

3.1.3. Isomer composition as a key for source identification

As discussed above, isomers have similar physical characteristics and are therefore transported in a similar manner between environmental media. On the other hand, homologues have different physical properties and are therefore transported in different manner between environmental media. Thus, isomer composition should be used as the basis for source identification. It should be noted, however, that isomer composition may also change when biological processes are involved.

3.2. Principal component analysis with different levels of dioxin information

Principal component analysis (PCA) has often been used as a means of source identification (Brakstad, 1991; Evers *et al.*, 1993; Fattore *et al.*, 1997; Grundy *et al.*, 1997; MacDonald *et al.*, 1998). Most studies have used 2,3,7,8-congener or

homologue compositions as input data, but have rarely succeeded in the quantitative estimation of source contribution (Bright *et al.*, 1999). This may be because seventeen 2,3,7,8-congeners belong to ten different homologues and behave differently in the environment, as discussed in the previous section. Thus, in this study, the identification of the source of dioxin pollution was attempted based on the detailed dioxin congener information. Here, we examined how the detailed information of dioxin congeners influenced PCA results. Congener-specific dioxin data of surface sediments and a sediment core from Tokyo Bay and some soil samples in the basin (a total of 23 samples) (Sakurai *et al.*, 2000; Yao *et al.*, 2000) were used as an example data set.

3.2.1. All congeners

Principal component analysis of dioxin congener-specific data (23 samples with 82 individual or groups of congeners) was performed using a software package. The result showed that three major principal components (PCs) and two minor ones were extracted with an eigenvalue larger than 1 (Table 1). The three major PCs could be interpreted as dioxins from combustion, pentachlorophenol (PCP) and chloronitrophen (CNP), based on the characteristic dioxin congeners having high factor loadings in each PC. PCP and CNP have been used extensively as rice field herbicides in the past. These three components accounted for over 93% of the total contribution.

Table 1 Results of principal component analysis with different levels of dioxin information (after varimax rotation)

3.2.2. *2,3,7,8-Chlorine-substituted congeners only*

2,3,7,8-Chlorine-substituted congeners (2,3,7,8-congeners) were selected from the above data set and PCA was performed (23 samples with 17 congeners). Two components were extracted, one with higher chlorinated 2,3,7,8-congeners and one with lower chlorinated ones (Table 1). However, interpretation of the components was difficult. Plotting the 2,3,7,8-congeners in the PCA plot obtained using all congeners showed that there were 2,3,7,8-congeners with factor loadings higher than 0.8 for PC1 (combustion) and PC2 (PCP) (Fig. 3). However, for PC3 (CNP), there were no 2,3,7,8-congeners with factor loadings higher than 0.6 (Fig. 3). Thus, 2,3,7,8-congener data alone were not sufficient to identify CNP as a source of dioxin pollution.

Fig. 3 Factor loading plot of PCA results with all congener data

2,3,7,8-congeners are shown by stars.

3.2.3. *2,3,7,8-Chlorine-substituted congeners and homologues*

Homologue composition data are often available in addition to 2,3,7,8-congener data. To make the variables independent, the 2,3,7,8-congener concentrations were subtracted from their corresponding homologue concentrations and non-2,3,7,8-homologue concentrations were obtained. Then, PCA was performed using this data set (23 samples with 25 variables). The results are shown in Table 1. In this case,

three major PCs were extracted and could be interpreted as combustion, PCP and CNP. Non-2,3,7,8-TeCDDs and non-2,3,7,8-PeCDDs had high factor loadings for CP3 (CNP). This indicated that the addition of homologue composition for PCA analysis was effective only when these three sources were concerned. Its effectiveness, however, was doubtful when other dioxin sources were present because no minor components with an eigenvalue larger than 1 were extracted in this analysis. Two minor PCs were extracted in the PCA with all congener information.

3.2.4. 2,3,7,8-Congeners, 1,3,6,8- & 1,3,7,9-TeCDD, 1,3,6,8- & 1,2,7,8-TeCDF, and homologues

Recently, some of the relatively major dioxin congeners in environmental samples have been quantified in addition to 2,3,7,8-congeners, especially in Japan. Among these are 1,3,6,8- and 1,3,7,9-TeCDDs and 1,3,6,8- and 1,2,7,8-TeCDFs. We examined if the addition of these congeners would improve PCA results (23 samples with 29 variables). The obtained result was almost the same as that from 2,3,7,8-congeners and homologues (Table 1). 1,3,7,9- and 1,3,7,9-TeCDDs were correlated with the CNP source and 1,3,6,8- and 1,2,7,8-TeCDFs were correlated with the combustion source. Thus, although the addition of these congener data may improve the reliability of PCA, it may not help identify other sources.

The results of PCA based on different levels of dioxin monitoring information showed that detailed information of all congeners had significantly high potential for identification of the source of dioxin pollution.

3.3 Estimating the contribution of sources by multiple regression analysis

3.3.1. Multiple regression analysis as a mean of source apportioning

When performing PCA, congener-specific data are transformed to a correlation matrix, and the calculated contribution of each PC does not correspond to the actual dioxin concentration. In addition, factor scores do not correspond to the actual dioxin concentration either. Thus, an understanding of the contributions from different dioxin sources in terms of actual dioxin concentration cannot be gained from PCA. Multiple regression analysis (MRA) can be used to fill this gap. In MRA, congener compositions of dioxin sources are required as independent variables and those of samples are regarded as dependent variables.

When MRA was performed by inputting of all the congener information together, the results depended strongly on the dioxin source composition. In particular, dioxin congener profiles for agrochemicals varied markedly among formulations and production lots that obtaining a reliable average source congener profile was difficult. In the case of PCP for example, the content of OCDD was much higher than those of other congeners and its content varied largely. However, the isomer compositions within each homologue were relatively stable. Thus, in this study, MRA calculations were performed for each homologue and then a summation of the results was made. This method had an additional merit in that it could avoid the influence of homologue composition change across environmental media, as discussed in the previous section. In the present calculation, as there were three major sources, more than four cases (isomers) were necessary to perform MRA. HpCDDs had two isomers and OCDD had only one isomer. Thus, HxCDDs, HpCDDs and OCDD were combined into one group for MRA. Similarly, HpCDFs and OCDF were combined.

3.3.2. Comparison of two MRA methods

In this study, the contributions of various dioxin sources to pollution recorded in a Tokyo Bay sediment core were calculated by means of the two MRA methods, one calculation with all congeners together and the other, with each homologue. The results are shown in Fig. 4. Calculation with all congeners together gave a higher contribution for PCP and a lower contribution for combustion than that with individual homologues. The contributions in terms of TEQ were calculated based on these results and the source congener profiles (Fig. 5). In this case, total TEQ estimated by all congeners together was approximately two times larger than the observed TEQ value in the sediments. In contrast to this, TEQ estimated with individual homologues was similar to the observed TEQ. This indicated that MRA calculation based on each homologue gives highly reliable results.

Fig. 4 Source apportioning by multiple regression analysis calculated with all congeners together and with individual homologues.

Fig. 5 Results of source apportioning in terms of TEQ by the two calculation methods

3.4. Application of source apportioning method: Dioxin mass balance in the Tokyo Bay basin

Using the new analysis methods discussed above, dioxin mass balance in Tokyo

Bay basin was estimated. This results serve as an update of a previous study conducted by the authors, which was based on very limited data at that time (Masunaga *et al.*, 1998). New data included in the present analysis are dioxin congener profiles for Japanese agrochemicals (Masunaga and Nakanishi, 1999) and those for a dated sediment core (Sanada *et al.*, 1999; Yao *et al.*, 2000) from Tokyo Bay. The new data set is exactly the same as the example data set used in previous sections. The sampling points of the soil and sediments used in the present analysis are shown in [Fig. 6](#).

3.4.1. Trend of dioxin input to Tokyo Bay basin and accumulation in sediment

The major sources of dioxin in Tokyo Bay sediment were shown to be combustion, PCP and CNP as discussed above, and the contributions of the three sources to dioxin pollution in surface sediments and in slices of sediment core were calculated ([Figs. 4-\(b\) and 5-\(b\)](#)). The concentrations of PCP- and CNP-originating dioxins in the sediment core peaked during the mid 1960s and beginning of the 1970s, respectively. On the other hand, combustion- originating dioxin was estimated to have increased during the 1960s, reached a plateau at the beginning of 1970s and then leveled off. [Figures 7 and 8](#) show the estimated trends of dioxin input to the Tokyo Bay basin in the form of agrochemical impurities, and the trends of PCP- and CNP-originating dioxin concentrations in slices of the dated sediment core. The dioxin load to the basin was estimated from the annual amounts of agrochemical shipment to Tokyo, Saitama, Chiba and Kanagawa Prefectures (Ministry of Agriculture, Forestry and Fishery, 1959-1996), the percentage of prefecture area belonging to the basin, and the dioxin content in the agrochemicals. The representative dioxin content in PCP (1,120 μg PCDD/DFs/g active ingredient and 1.25 μg WHO-TEQ/g active ingredient) was determined by taking

the arithmetic average of many relevant data reported worldwide. In the case of CNP, dioxin content was assumed to have decreased annually based on the five samples analyzed (Masunaga and Nakanishi, 1999).

Figures 7 and 8 show sharp increases in PCP- and CNP-originating dioxin concentrations immediately after the beginning of use of these chemicals, although significant error in sediment core dating may be present. PCP- and CNP-originating dioxin input to the basin was estimated to have peaked in the middle of the 1960s and the early 1970s, respectively, and decreased rapidly thereafter. On the other hand, PCP- and CNP-originating dioxin concentrations in sediment core peaked in 1970 and 1975, respectively, which were several years after the input peaks, and did not decrease as drastically as the input peaks. The decrease in sediment core dioxin concentrations slowed down in recent years. These phenomena indicated that dioxin loaded to agricultural land ran off gradually and continued to pollute the aquatic environment. This is an important observation because Japanese people ingest dioxins mainly through fish and shellfish consumption.

Fig. 6 Sampling points of soil, surface sediment and sediment core samples

Fig. 7 Estimated annual dioxin load from PCP in Tokyo Bay basin and PCP-originating dioxin concentration in Tokyo Bay sediment core

Fig. 8 Estimated annual dioxin load from CNP in Tokyo Bay basin and CNP-originating dioxin concentration in Tokyo Bay sediment core

3.4.2. Agrochemical-originating dioxin loss rate from terrestrial soil

As a preliminary rough estimate, it is possible to assume that the amount of annual dioxin loss from paddy fields is proportional to the amount of dioxin present in the field and that the annual amount of dioxin deposition in Tokyo Bay is proportional to the amount of dioxin loss from the fields. Thus, it can be assumed that annual deposition is proportional to the amount of dioxin present in agricultural fields in the basin. If the annual loss rate of dioxin from the field were constant over the past 45 years, which is the average annual loss rate, the trend of the amount of dioxin in the field could be simulated using this average annual loss rate. The simulated trends at different annual loss rates (8, 9, and 10%/year) are shown in Fig. 9 for the case of CNP in terms of PCDD/DFs. Although the fitting is not so good, an annual loss rate of 9%/year best fits the trend of dioxin deposition in Tokyo Bay. Therefore, the simulation indicates that the amount of dioxin in the basin decreases at a rate of about 9%/year.

A similar analysis was conducted for PCP and CNP in terms of PCDD/DFs and TEQ. The annual loss rates estimated based on the decrease in dioxin concentration in sediment core were 5 and 5%/year for PCP in terms of PCDD/DFs and TEQ, respectively; and 9 and 7%/year for CNP in terms of PCDD/DFs and TEQ, respectively. These results suggest that dioxin loss in the soil in Tokyo Bay basin were occurs at a rate of about 5 - 9%/year or a half-life of about 7 - 14 years. The loss mechanism may include runoff, volatilization, degradation and burial (covering of surface soil due to a change in land use). As vertical mixing in the sediment may have occurred, it is

possible that the loss rates are underestimated. On the other hand, existence of loss mechanism such as burial may contribute to overestimation of the loss rates. Thus, the estimated values should be taken as rough approximation.

Fig. 9 Estimation of CNP-originating dioxin loss rate from terrestrial soil in Tokyo Bay basin.

3.4.3. Dioxin mass balance in Tokyo Bay basin during the past 45 years

The total dioxin loads from the two agrochemicals to the Tokyo Bay basin during the past 45 years can be estimated by taking the summation of the annual loads shown in Figs. 7 and 8. The total loads from PCP were 11,000 kg PCDD/DFs and 12 kg TEQ. Those from CNP were 6,500 kg PCDD/DFs and 10 kg TEQ. Assuming that annual dioxin deposition from combustion sources did not vary much during the past 45 years (see Figs. 4-(b) and 5-(b)), the estimated recent average deposition in the Kanto region (Ogura *et al.*, 2001a) was used as the average deposition of past years. The total dioxin loads to the basin from the three sources during the past 45 years (1951-1995) are listed in Table 2. On the other hand, the total amount of dioxin that sedimented in Tokyo Bay during the past 45 years was estimated from the sediment core data with horizontal corrections based on surface sediment data. The results are summarized in Table 2.

The amount of PCP- and CNP-originating dioxins lost from terrestrial soil during the past 45 years could be estimated using the loss rates from soil estimated in the previous section and are shown in **Table 2**. In terms of TEQ, the total dioxin load to

Tokyo Bay basin from both PCP and CNP was estimated to be five times larger than that from combustion. The amount of combustion-originating dioxin that accumulated in Tokyo Bay sediment, however, was estimated to be comparable to total of PCP- and CNP-originating dioxin accumulated in the sediment. In the surface sediment, combustion-originating dioxins contributed about half of the contamination (Fig. 5-(b)).

The ratio of loss from soil to input ($[L/I]$ in Table 2) was from 70 to 85 %, indicating that a amount of dioxins loaded to paddy field was already lost. The ratio of the amount of dioxin accumulated in sediment to the amount of dioxin lost from soil ($[A/L]$ in Table 2) was around 10% for PCP and 0.8 - 2.5% for CNP. These values were expected be close to each other if the environmental behaviors of the dioxins from the two sources were similar. The large difference observed may be mainly due to error in the estimation of the average dioxin concentrations in the agrochemicals. This, however, could not be avoided because of the large variation in dioxin concentration among formulations and production lots. The contribution from the different environmental behaviors of dioxins from the two herbicides, however, cannot be ruled out because the homologue profiles of the two herbicides are different.

The ratio of the amount of dioxin accumulated in sediment to the amount of input ($[A/I]$ in Table 2) was relatively large for combustion source. This may be because a large part of combustion-originating dioxins was deposited on land by rain and easily flows into the aquatic environment.

In conclusion, the estimated mass balance showed that dioxins released into the environment more than two decades ago as herbicide impurities remained in paddy fields and gradually flowed into Tokyo Bay.

4. Conclusions

In this study, new source identification and mass balance calculation methods based on detailed congener-specific dioxin data were proposed. They were applied successfully to estimate the dioxin mass balance in Tokyo Bay basin. The obtained mass balance showed that congener-specific analysis data of all the tetra- to octa-chlorinated PCDD/DFs were far more useful for source identification than the conventional data set consisting of only seventeen 2,3,7,8-chlorine-substituted congeners and ten homologues. For source apportioning, it was revealed that MRA with source congener profiles should be performed for individual homologues because of the large variation of homologue composition in source dioxin profiles in addition to the difference in environmental behavior among the homologues.

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Table 1 Results of principal component analysis with different levels of dioxin information (after varimax rotation)

Principal components*	Contribution	Cumulative contribution	Characteristic congener (Factor loading > 0.7)	Interpretation
All congeners				
PC1	0.50	0.50	Most of TeCDFs, half of PeCDDs, most of PeCDFs, some HxCDDs, and most of HxCDFs.	Combustion
PC2	0.25	0.75	Most of HxCDDs, some HxCDFs, HpCDDs, most of HpCDFs, OCDD, and OCDF.	PCP
PC3	0.18	0.93	Some TeCDDs and PeCDDs, especially 1,3,6,8- & 1,3,7,9-substituted TeCDDs and PeCDDs, and 2,4,6,8-TeCDF.	CNP
PC4	0.021	0.95	(1,2,4,7,9-, 1,2,3,4,9- and 1,2,3,8,9-PeCDFs)**	Difficult
PC5	0.016	0.98	(1,2,8,9-TeCDF)**	Difficult
2,3,7,8-congeners only				
PC1	0.50	0.50	HxCDDs, HpCDD, OCDD, 2,3,7,8-TeCDF, 1,2,3,4,7,8-HxCDF, HpCDFs, and OCDF.	Difficult
PC2	0.46	0.96	2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, PeCDFs, and 1,2,3,6,7,8-/2,3,4,6,7,8-/1,2,3,7,8,9-HxCDF.	Difficult
2,3,7,8-congeners+ non-2,3,7,8-homologues				
PC1	0.45	0.45	OCDD, OCDF, HpCDDs, HpCDFs, and some HxCDDs & HxCDFs.	PCP
PC2	0.40	0.85	Most of TeCDFs, PeCDFs and HxCDFs.	Combustion
PC3	0.11	0.96	Non-2,3,7,8-TeCDDs and non-2,3,7,8-PeCDDs.	CNP
2,3,7,8-congeners + some TeCDDs and TeCDFs, residual homologues				
PC1	0.41	0.41	OCDD, OCDF, HpCDDs, HpCDFs, and some of HxCDDs & HxCDFs.	PCP
PC2	0.38	0.79	Most of TeCDFs, PeCDFs, and HxCDFs.	Combustion
PC3	0.18	0.96	1,3,6,8-/1,3,7,9-TeCDDs, residual TeCDDs, and residual PeCDDs .	CNP

* Principal components with eigenvalue > 1 were extracted.

** Factor loadings were lower than 0.7 but higher than 0.3.

Table 2 Dioxin mass balances in Tokyo Bay basin during the past 45 years**(1951-1955)***

Source	Input to the basin [I] (kg) <i>(g TEQ)</i>	Remaining in basin soil (kg) <i>(g TEQ)</i>	Lost from basin soil [L] (kg) <i>(g TEQ)</i>	Accumulation in sediment [A] (kg) <i>(g TEQ)</i>	L/I ratio [L/I] (%) <i>(%)</i>	A/L ratio [A/L] (%) <i>(%)</i>	A/I ratio [A/I] (%) <i>(%)</i>
PCP	11,000	2,800	7,800	760	71	9.7	7.2
	<i>12,000</i>	<i>3,100</i>	<i>8,700</i>	<i>1,200</i>	<i>73</i>	<i>14</i>	<i>10</i>
CNP	6,500	960	5,500	140	84	2.5	2.2
	<i>10,000</i>	<i>2,100</i>	<i>8,200</i>	<i>65</i>	<i>82</i>	<i>0.79</i>	<i>0.63</i>
Combustion	42**			150			40 (33)***
	460**						
	340#			950			23 (14)***
	3,600#						

* Upper row: Amount of PCDD/DFs in kg. Lower row in italics: Amount of TEQ in g.

** Input directly to water surface of Tokyo Bay.

Input to terrestrial surface in Tokyo Bay basin.

*** Run-off ratio for terrestrial input.

Figures

Fig. 1 Dioxin homologue profiles for different media and locations.

4F, 5F, 5F, 6F, 7F, 4D, 5D, 6D, 7D and 8D are TeCDFs, PeCDFs, HxCDFs, HpCDFs, OCDF, TeCDDs, PeCDDs, HxCDDs, HpCDDs and OCDD, respectively.

Fig. 2 Dioxin isomer profiles for different sources and media from Kanto region.

PCP: pentachlorophenol, CNP: chloronitrophen.

Fig. 3 Factor loading plot of PCA results with all congener data

2,3,7,8-congeners are shown by stars.

Fig. 4 Source apportioning by multiple regression analysis calculated with all congeners together and with individual homologues.

Fig. 5 Results of source apportioning in terms of TEQ by the two calculation methods

Fig. 6 Sampling points of soil, surface sediment and sediment core samples

Fig. 7 Estimated annual dioxin load from PCP in Tokyo Bay basin and PCP-originating dioxin concentration in Tokyo Bay sediment core

Fig. 8 Estimated annual dioxin load from CNP in Tokyo Bay basin and CNP-originating dioxin concentration in Tokyo Bay sediment core

Fig. 9 Estimation of CNP-originating dioxin loss rate from terrestrial soil in Tokyo Bay basin.

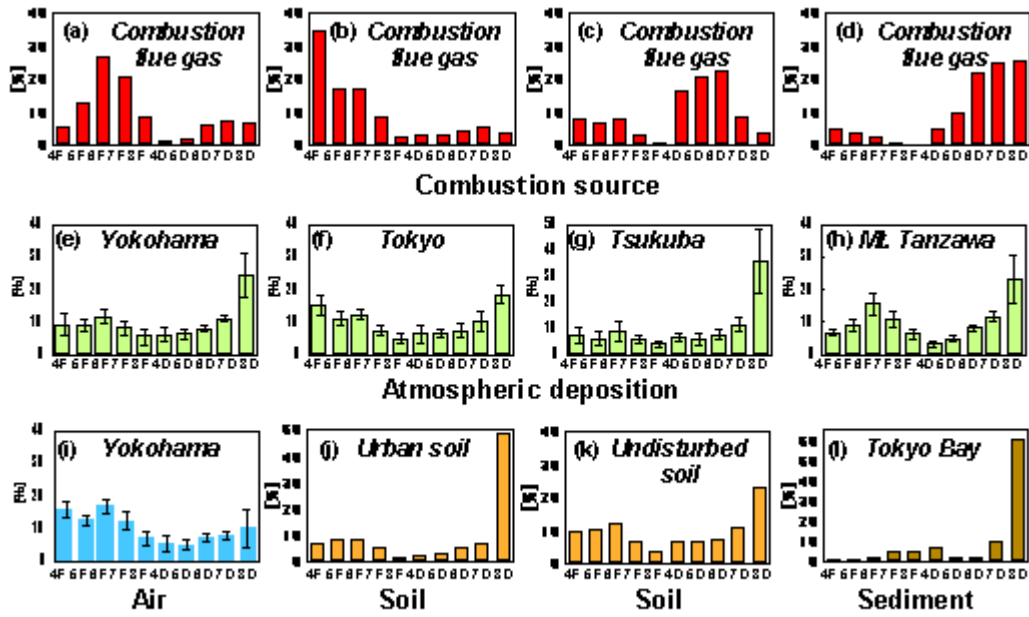


Fig. 1

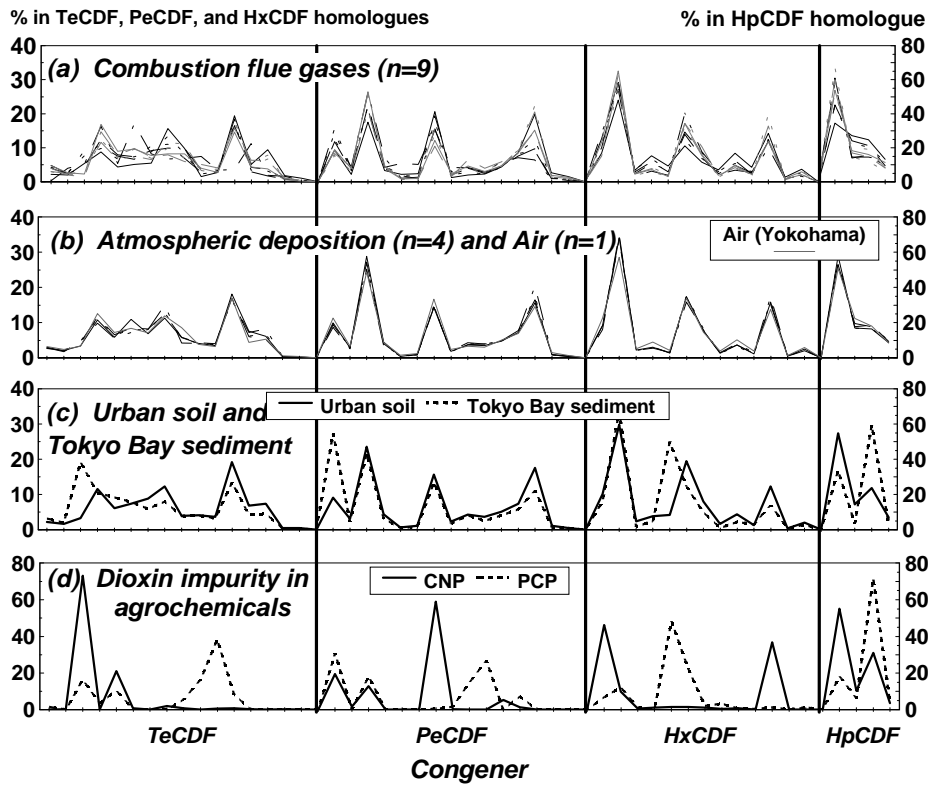


Fig. 2

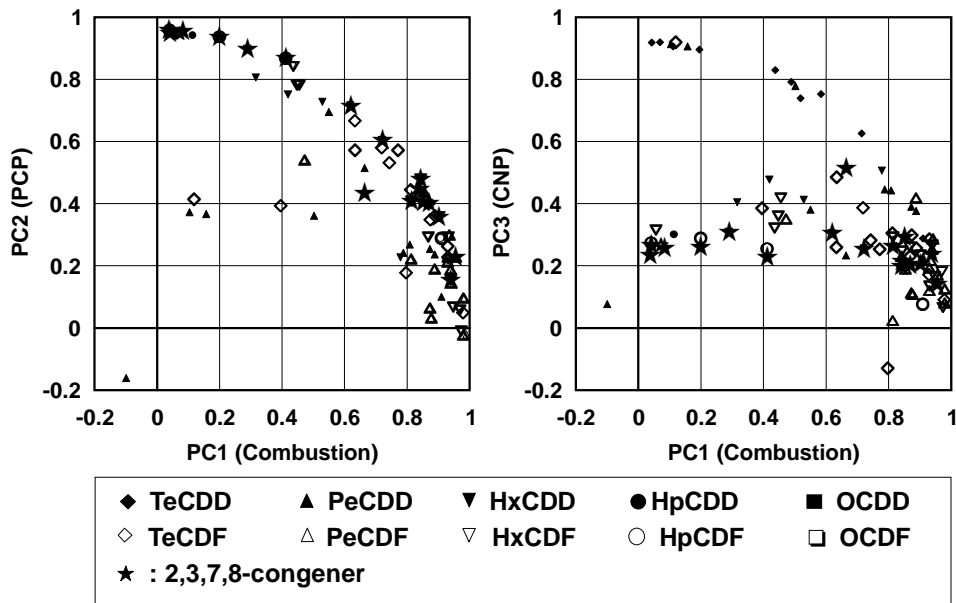


Fig. 3

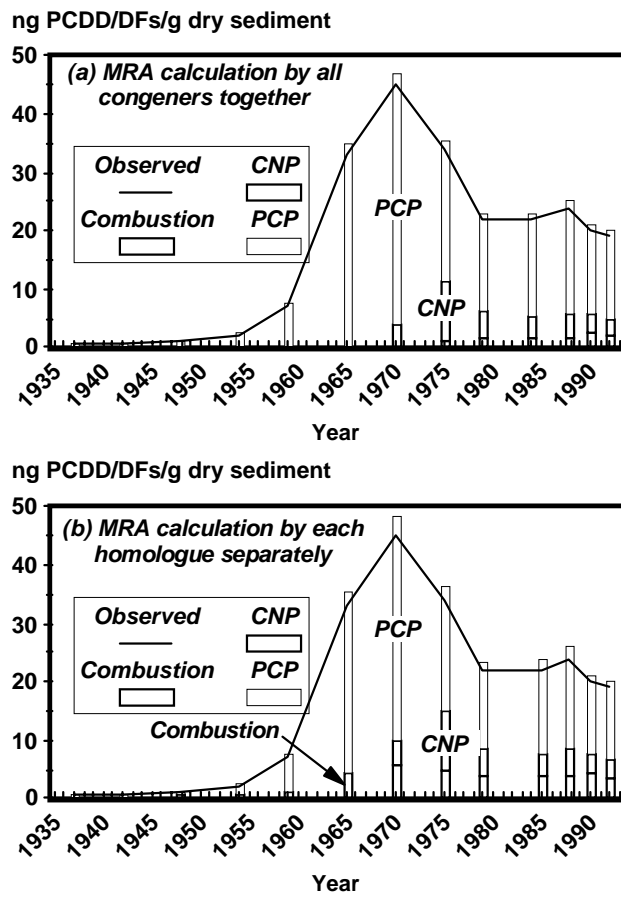


Fig. 4

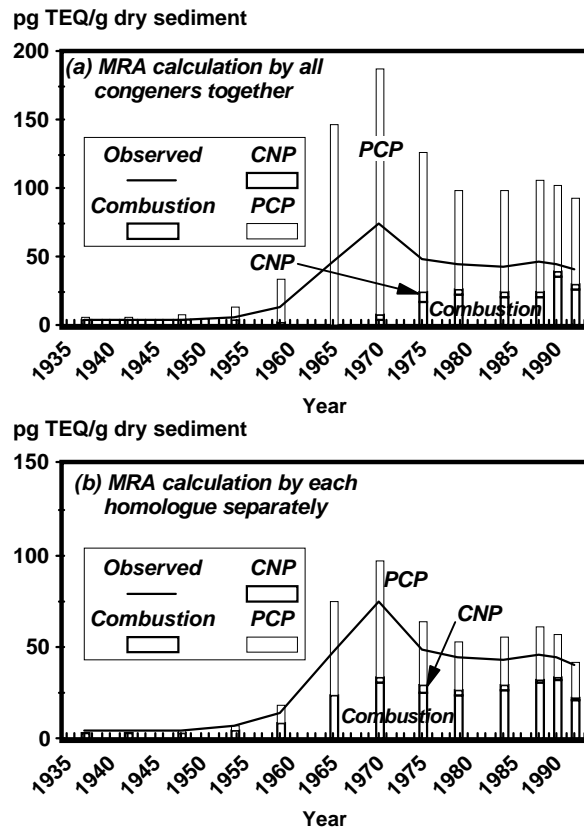


Fig. 5

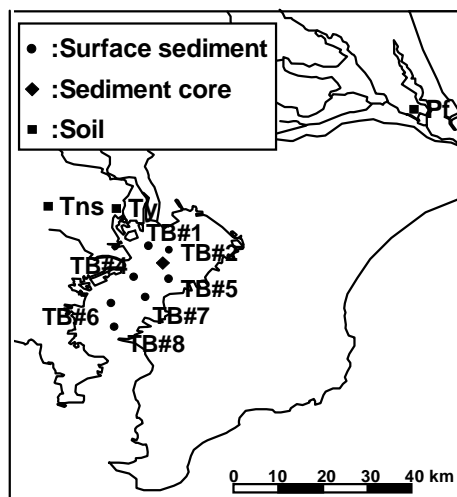


Fig. 6

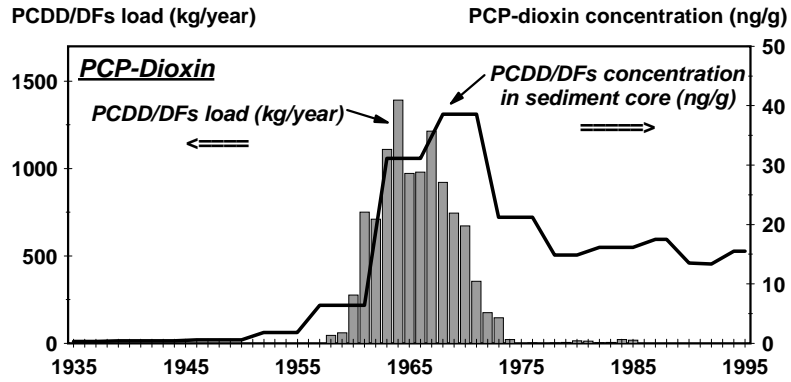


Fig. 7

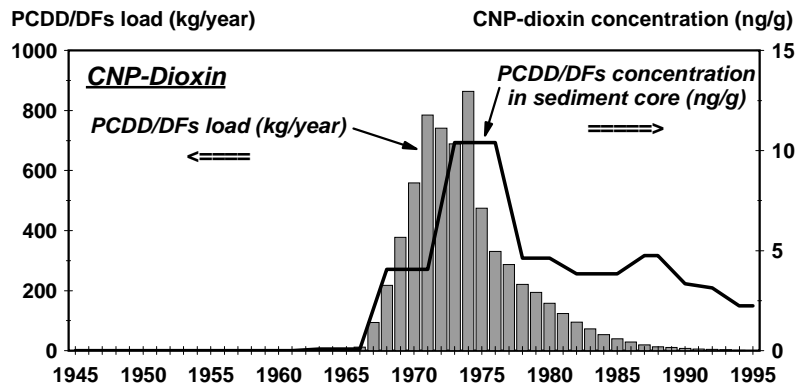


Fig. 8

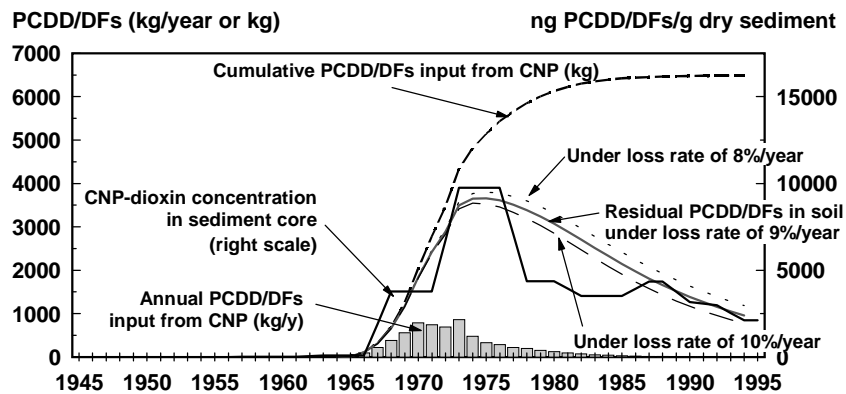


Fig. 9