Mass Balance of Dioxins in Tokyo Bay and Kasumigaura Lake Basins in Japan

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Introduction

A number of severe soil and air pollution cases around municipal and industrial waste incinerators have recently been found in Japan. It is true that the amount of waste incinerated in Japan is very high compared with other countries; however, it must be clarified whether the incinerators are the sole cause of the present dioxin pollution problem. In this study, the mass balance of dioxins in Tokyo Bay and Kasumigaura Lake basins in Japan is estimated and discussed based on environmental monitoring data.

Materials and Methods

The Japanese consume much fish; therefore, sediment on sea and river beds is a very important environmental medium because it accumulates and transfers pollutants to fish through the aquatic food chain. Furthermore, sediment and soil are conservative media for dioxins and provide a record of past pollution phenomena. Thus, we statistically analyzed dioxin isomer profiles in sediment and soil to determine the contributions of different origins of dioxins. Then the mass balance of dioxins in the study areas was estimated based on both atmospheric deposition measurement and the use of chlorinated chemicals.

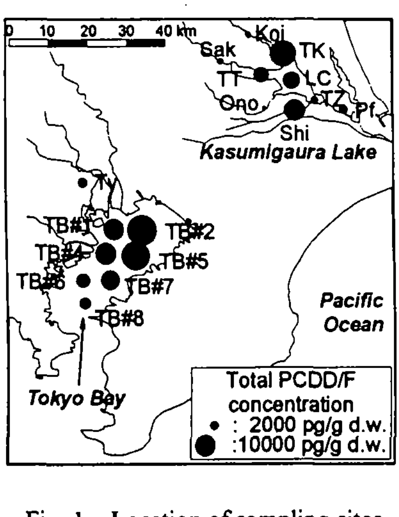

Fig. 1 Location of sampling sites and total PCDD/F concentration

Results and Discussion

Principal component analysis of dioxin isomer profile in soil and sediment samples

Many different processes have been cited as the sources of dioxins; however, there are few reliable data on the relative importance or dioxin composition of these sources. Thus, dioxin concentration data obtained for surface soil and sediment samples from Tokyo Bay and Kasumigaura Lake basin (Fig. 1) were studied by principal component analysis (PCA) to estimate the contribution of different origins of dioxins. The data consisted of seventy-three gas chromatographic peaks from 17 samples. The variance of each peak was normalized to unity and the obtained correlation matrix was used as the input for PCA. Details of the analysis are described elsewhere[1]. The results showed that four principal components (PCs) accounted for over 94% of the total variance. The obtained PCs were interpreted by comparing their isomer profiles with the reported profiles from different sources (Table 1). Both 1,3,5-trichloro-2-(4-nitrophenoxy) benzene (CNP) and pentachlorophenol (PCP) were extensively used as herbicides in paddy fields in Japan.

Table 1 Results of the principal component analysis

PC	Contribution	Characteristic compound[a]	Origin[b]
PC-1	52 %	most of the tetra- to hexa-CDFs	atmospheric deposition[2]
PC-2	17 %	penta- to hepta-CDDs with 1269-Cl substitution	unknown
PC-3	16 %	tetra- to penta-CDD/Fs, especially with 1368- or 1379-Cl substitution	CNP[3]
PC-4	9 %	some of the hexa- to octa-CDD/Fs	PCP[4]

[a] Isomer or isomer-cluster peaks with high loading values (r>0.7).
[b] Reference shows the literature in which the isomer profile is reported.

Contribution of different origins of dioxins at each sampling site

The PC scores calculated by PCA only show the deviation from the mean and do not represent the actual dioxin concentration. Since PC-1 to -4 accounted for over 94% of the total contribution, it is safe to assume that contributions from other origins are negligible. Thus, the contributions of the four origins in terms of concentration were calculated by multiregression analyses (MRA) using the dioxin isomer profiles of atmospheric deposition[2], CNP[5], PCP[4] and PC-2. The isomer profile of PC-2 was reconstructed from the result of PCA. The MRA results are shown in Fig. 2. Atmospheric deposition was found to be very important for urban soil and CNP was important for paddy soil and river sediment, which are very reasonable results.

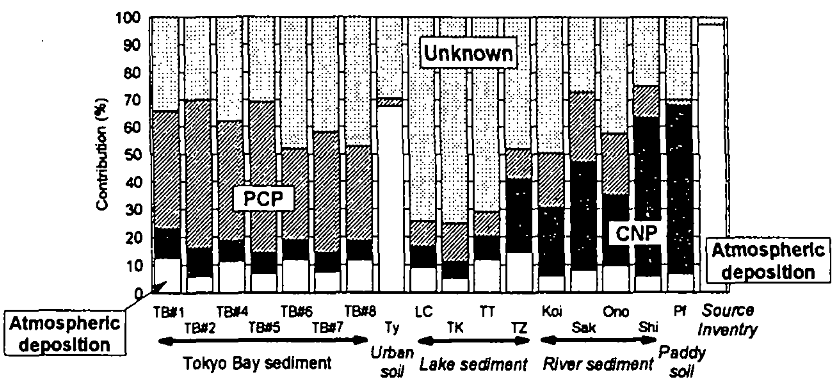

Fig. 2 Contribution of different origins of dioxins to the total PCDD/F concentration in soil and sediment at each sampling site

Total amount of dioxins present in the sediment from Tokyo Bay and Kasumigaura Lake

The sediment samples analyzed in this study were mixtures from 0 to 10 cm depth from the surface. They are considered to represent one to two decades of sedimentation. Taking the rate of sediment deposition into account (around 0.2[6] and 0.14[7] g/cm^2/y for Tokyo Bay and Kasumigaura Lake, respectively), they also represent half to most of the period after the start of mass use of chloroaromatic compounds in Japan [35 years from 1960 to 1994]. Thus, dioxin concentrations in those samples together with the contribution ratios of different origins and rate of sediment deposition were used to calculate the total dioxins accumulated in the bottom sediment of the two water bodies during the past 35 years. The results for total PCDD/Fs and TEQ are shown in Fig. 3. Since CNP hardly contained any 2378-congeners, its contribution in terms of TEQ was negligible.

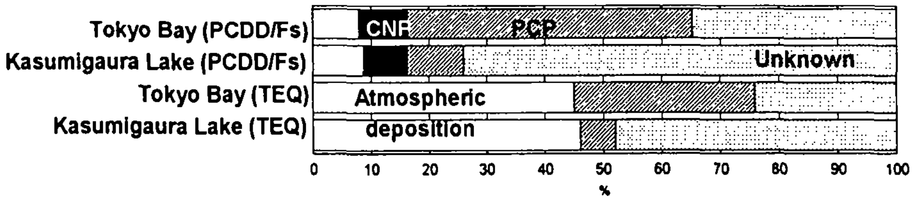

Fig. 3 Contribution of different origins to the accumulated dioxins in the bottom sediment of Tokyo Bay and Kasumigaura Lake for the past 35 years

Potential dioxin sources in Japan

The dioxin loads from different sources were estimated. As dioxins from combustion processes such as waste incineration, metal manufacturing and automobiles can be categorized into one large group of atmospheric dioxin emitters, they can be represented by atmospheric deposition. On the other hand, the sources of dioxin emission to soil and water include the production and use of chloroaromatic compounds and pulp and paper bleaching. Among these, the ones considered to be important are listed and their amount used and dioxin emission to the environment were estimated (Fig. 4). Although the concentration of dioxin impurity in chloroaromatics varies depending on the manufacturer, production lot and year, the number of reported data is limited. Therefore, the amounts of estimated dioxin emissions are uncertain. The relative importance of these sources, however, is estimated to be in the order of CNP ≅ PCP > PCB and may not change in spite of the large uncertainty. This is in agreement with the results of PCA which revealed CNP and PCP as important sources.

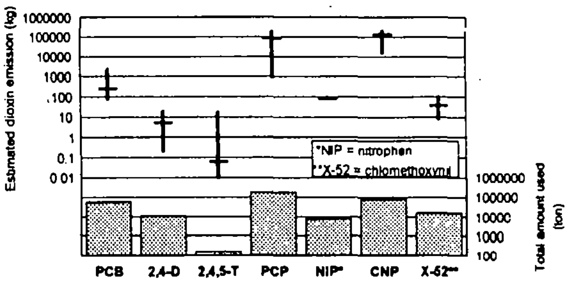

Fig. 4 Dioxin emission from the use of chloroaromatic compounds in Japan

The vertical and horizontal bars show the range and center of the estimates, respectively.

Dioxin mass balance in Tokyo Bay and Kasumigaura Lake basins

The dioxin mass balance in the two basins during the past 35 years was estimated based on herbicide usage and atmospheric dioxin deposition monitoring data[2]. The results for Tokyo Bay basin in terms of central estimate are shown in Fig. 5. For both basins, the amount of dioxin entering directly into the water bodies through atmospheric deposition is comparable to the amount accumulated in the sediment. The run-off ratios of terrestrial dioxin load from herbicides, on the other hand, ranged from 0.2 % (CNP in Kasumigaura Lake) to 10 % (PCP in Tokyo Bay). The run-off ratio of PCP in Tokyo Bay basin was much larger than that of CNP. A possible explanation for this might be the use of PCP other than herbicides. There are timber ponds in the bay and direct input of PCP used as wood preservative may have occurred.

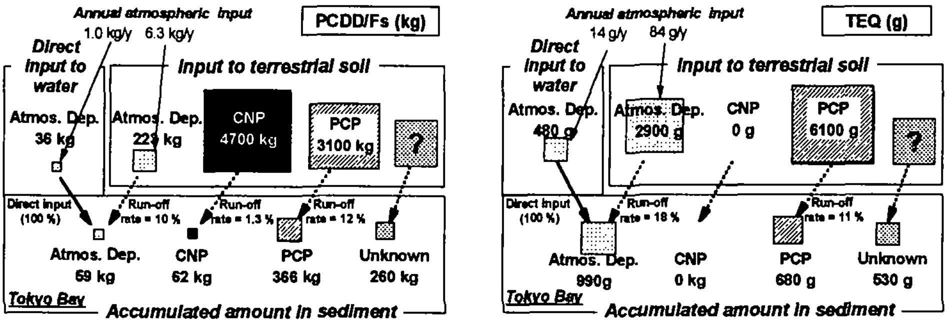

Fig. 5 Dioxin mass balance in Tokyo Bay basin (left: PCDD/Fs weight base, right: TEQ base)

Conclusions

Although the dioxin mass balance obtained here has large uncertainty due to insufficient data, we can still draw some useful conclusions:

1. A large part of the present dioxin pollution in aquatic sediment is caused by the past herbicide use in paddy fields. This is quite different from the current dioxin source inventory, in which combustion is estimated to contribute over 90% of the load.
2. Most of the dioxins emitted in the past as pesticide impurities still exist in agricultural soil. They will continue to flow into water bodies and contaminate the sediment in the future.
3. It is true that control of dioxin emission from incineration processes will reduce the degree of dioxin exposure near these sources, but the reduction of the degree of dioxin exposure in general public may take a long time because a large part of the dioxin intake by the Japanese is by consuming fish.

Acknowledgment

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