HISTORICAL TREND OF PCDD/Fs CONTAMINATION IN MATSUYAMA, JAPAN

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Introduction

Sediment core[1), 2)] and vegetation[3)] are an important media to investigate temporal trend of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs). These results show that artificial PCDD/Fs contamination was started about year 1900. The next PCDD/Fs contamination was clearly observed during 1960s – 1980s. Evan though environmental PCDD/Fs levels has been reduced in recent years, it is important to clarify status of PCDD/Fs contamination in the past for future predictions.

We have been investigated PCDD/Fs in environment components, such as air[4)], atmospheric deposition[5)], water[6)], sediment[6)], fish[7)] and human adipose tissue[8)] from Matsuyama, Japan. However information about historical trend of PCDD/Fs in Matsuyama is meager. Likewise, some congeners, such as 1,3,6,8-TeCDD, 1,3,7,9-TeCDD and OCDD are impurity of 2,4,6-Trichlorophenyl 4-nitrophenylether (CNP) and Pentachlorophenol (PCP), were used at paddy fields as herbicide in the past are widely distributed in aquatic environment. Therefore, It is important to investigate historical trend of PCDD/Fs concentration in paddy soils.

In this study, PCDD/Fs were analyzed in paddy soil, sediment core and human adipose tissue to evaluate historical trend of PCDD/Fs contamination in Matsuyama, Japan.

Materials and Methods

Paddy soil sample collected years were in 1968(n=10), 1977(n=10), 1986(n=10) and 1995(n=36), respectively. A sediment core was taken from the Shigenobu river estuary that located at Matsuyama. Human adipose tissue samples were collected during 1990 to 1995. In this study, we used PCDD/Fs data in human adipose tissues from 1960 to 1985 that was reported by Ono[9)].

All the samples were Soxhlet extracted with toluene or dichloromethane. Purification and separation were carried out by sulfuric acid, silica gel, alumina and activated carbon column chromatography. Samples were analyzed by HRGC/HRMS (HP5890 II /JEOL-102A) equipped with a CP-Sil88 for Dioxins (CHROMPACK) or DB-5ms (J&W SCIENTIFIC).

Results and Discussion

Contamination of PCDD/Fs in paddy soils, sediment core and human adipose tissue were observed during 1960s – 1980s. The contamination has been reduced in recent years (Fig.1).

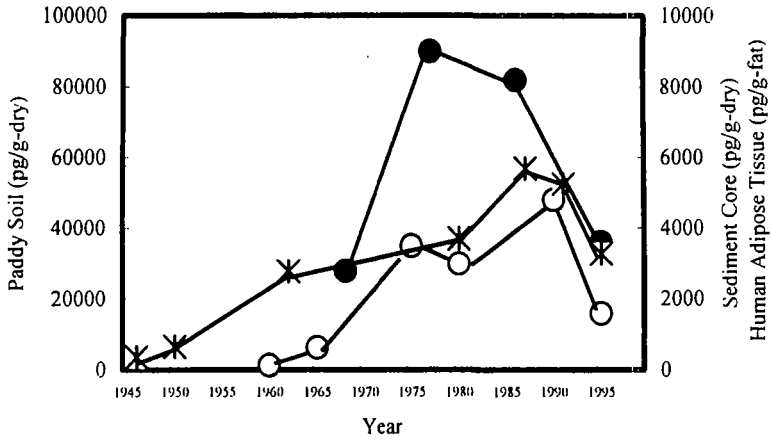

Fig.1 Historical Trend of PCDD/Fs Concentration in Paddy Soil, Coastal Sediment Core and Human Adipose tissue from Matsuyama, Japan

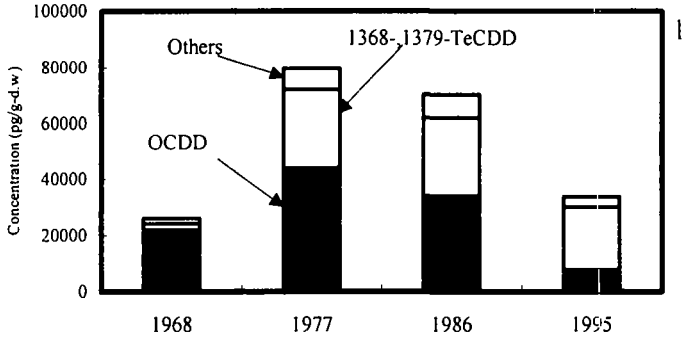

Figure 2. Historical Trend of PCDD/Fs Concentration in Paddy Soil

Paddy Soils

Fig.2 shows that historical trend of OCDD, 1,3,6,8-, 1,3,7,9-TeCDD and other PCDD/Fs in paddy soils. The highest concentration of PCDD/Fs in paddy soil was detected in 1977. OCDD, 1,3,6,8- and 1,3,7,9-TeCDD were accounted nearly 90% of the total PCDD/Fs. Proportion of OCDD was decreased in recently, however 1,3,6,8- and 1,3,7,9-TeCDD were increased. It could be resulted of CNP and PCP usage on paddy soil.

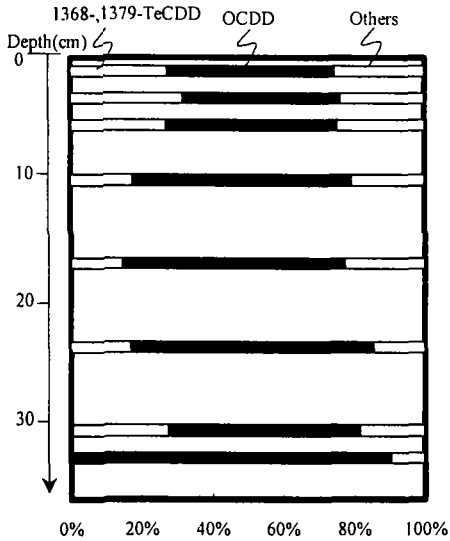

Fig.3 Vertical Distribution of PCDD/Fs Composition in Sediment Core from Estuary of R.Shigenobu

Sediment Core

Fig.3 shows vertical distribution of OCDD, 1,3,6,8-, 1,3,7,9-TeCDD and other PCDD/Fs in a sediment core from estuary of R.Shigenobu. Total PCDD/Fs was accounted for about 80% of OCDD 1,3,6,8- and 1,3,7,9-TeCDD. It is thought be that these congeners could be adsorbed on paddy soil particles and flown into the aquatic environment where finally deposited on the coastal sediment.

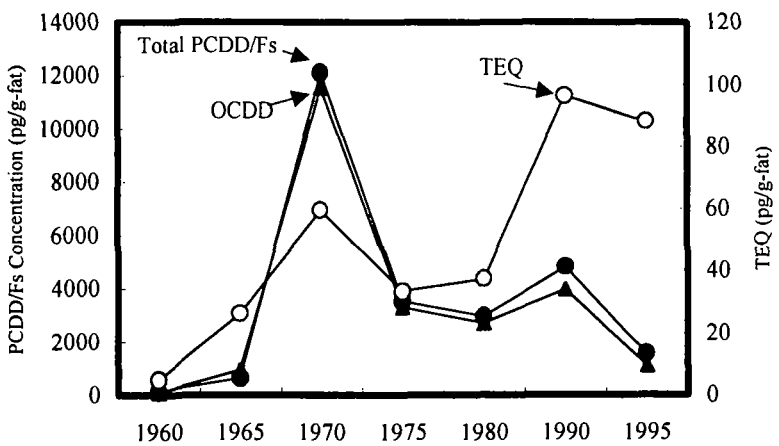

Fig. 4 Historical Trend of OCDD, Total PCDD/Fs Concentration and TEQ in Human Adipose Tissue

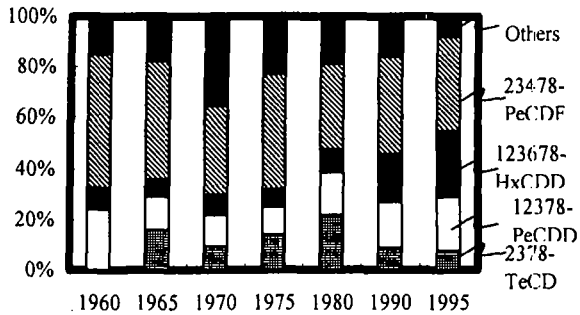

Fig.5 Contribution (%) of Some Toxic Isomers to TEQ in Human Adipose Tissue

Human adipose tissue

Fig.4 shows that historical trend of total PCDD/Fs, OCDD and TEQ in human adipose tissue. OCDD was accounted for over 90% of total PCDD/Fs. The highest concentration of PCDD/Fs in human adipose tissue was detected in 1970. Recently, total PCDD/Fs concentration has been gradually reduced. However, TEQ has been increased for the past ten years.

Fig.5 shows contribution of some toxic isomers, such as 2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PeCDF to TEQ in human adipose tissue. These congeners were mainly contributed to total TEQ. It seems to be in recent years, increase of 1,2,3,7,8-PeCDD and 1,2,3,6,7,8-HxCDD contaminations contributed to increasing of TEQ in human. Moreover, it could be indicated that human exposure of PCDD/Fs has been changed during last 40 years.

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