PCDD/Fs CONCENTRATION IN SOIL OF A JAPANESE LOCAL CITY: POSSIBLE PCDD/Fs SOURCES AND RELATIONSHIP WITH LAND UTILIZATION

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Introduction

Although the Japanese Ministry of Environment has monitored and disclosed PCDD/Fs levels in soil across the country every year, people have much concerned about PCDD/Fs values in soil in their living environment even if there is no prominent susceptible PCDD/Fs sources in their vicinity. This paper presents measurements results of PCDD/Fs levels in soil of a bed-town city of Tokyo, in which 70 soil samples of different land uses were collected throughout the city. PCDD/Fs levels were compared by land use profiles, and possible sources of PCDD/Fs were examined.

Methods and Materials

The city is located in the central Saitama prefecture, northwest to Tokyo, and has a population of 80,000 with 3,600 ha of autonomous area of which a rice paddy, a vegetable growing field, a residential area, and a business-industrial area cover about 800 ha each. Remains are a swamp, forest, and others. There are no centralized municipal waste incinerator or heavy industries including steel or secondary copper smelters that are supposedly major producer of atmospheric PCDD/Fs. However small scale incinerators with a capacity of less than 200 kg/hr, used at the local industries as well as at citizen's home, were once numerous before those incinerator became regulated for PCDD/Fs emission. Bone fires or open burns of household waste or yard clippings including fallen leaves, and agricultural residues at farmland are rather ubiquitous in the area. Soil sampling points include the lane of agricultural farmland (generally used for growing vegetable or flowers) and the rice field, playgrounds at schools or parks, and their shrubbery sites, shrine premises, and green belts of main roads, and others. Samples were undisturbed soils for the last 5 years or more. After removing gravel, wood chips, fallen leaves and plant residue five soil cores with 5 cm diameter and 5 cm depth were sampled at each sampling point. Each core was naturally dried separately and sieved using 2mm mesh sieve. Equal amounts of soil were grabbed from each dried sample and mixed making about 100 g sample for one sampling point in total for a further analysis.

Results and Discussion

In Table 1 are shown PCDD/Fs levels in 70 soil samples by land use patterns together with soil characteristics. Highest level, 157.1 pg-TEQ/g, was recorded in the soil of a mound on which row trees were planted to divide a residential area and the main road. However, on average, soil samples obtained at the lane of rice field showed higher values, 71.0 pg-TEQ/g, followed by the soil of the mound, shrubbery of the shrine, and the lane of farmland. The lowest average value, 9.1 pg-TEQ/g, was obtained at the playground soil of schools and parks.

Principal Component Analysis

Possible major origins of PCDD/Fs in the soil may be: (1) direct application, spill or disposal of PCDD/Fs containing materials to the soil. These include a sort of chemical products or combustion residues, and (2) atmospheric deposition of PCDD/Fs traveled from the sources of industrial and social activities including waste combustions. Some samples showed higher levels of 1,3,6,8-TCDD, 1,3,7,9-TCDD, and OCDD, which are known to characterize isomer or congener pattern of the soil at rice paddy or CNP and PCP[(1),(2)], a herbicide commonly used at the Japanese rice paddy in the past. Meanwhile, as shown in Figures 1 and 2, congeners values reduced by OCDD or OCDF values are different between soil portion (under 2mm sieve) and humus portion (above 2mm sieve) of the same soil sample, extremely different in PCDFs. This means that in the process of soil mineralization the ratio of lower chlorine congeners to Octa-congeners decreases abruptly. PCDD/Fs deposited on the tree

Table 1. PCDD/Fs levels in 70 soil samples

Area	ID No.	Description	PCDD/Fs (pg-TEQ/g)	PCDDs (pg/g)	PCDFs (pg/g)
Lane of farm land	3	Soil loam.	19	8100	710
	18	Fallow land Silt.	4.7	550	160
	A1	Close to rice paddy. Loam	17	5300	1100
	A2	Close to rice paddy. Loam.	10	3500	420
	A3	Former farm land. Silty loam.	240	9800	650
	A4	Close to residential area. Silty loam.	12	4800	300
	A5	Close to rice paddy. Loam.	8.4	12000	430
	A6	Close to residential area. Silty loam.	43	22000	1000
	A7	Close to residential area. Loam	91	68000	1800
Shrubbery of parks or schools	7	Park. Clay.	15	5900	490
	14	Silty skeletal.	22	7900	1000
	19	Park. Sandy-skeletal.	15	2600	540
	20	School. Soil-skeletal.	13	1900	510
Playground of parks or schools	C1	Park. Loam. Herbicide applied in the past.	5	3100	300
	C2	School. Sandy loam.	0.35	69	61
	C3	Park. Silty loam.	0.33	230	38
	C4	Park. Loam	9.1	1900	300
	C5	Park. Loam.	67	19000	3600
	C6	School. Sandy loam.	1.5	140	90
	C7	School. Sandy loam.	4.5	530	150
	C8	School. Sandy loam.	2.7	88	130
	C9	School. Sandy loam.	1.1	69	99
	C10	School. Sand.	0.51	140	73
	C11	School. Sandy silt-loam.	0.17	110	43
	C12	School. Sandy loam.	30	21000	810
	C13	Baseball playground. Loam.	1.8	1700	130
	C14	Park. Herbicide applied.	8.8	3300	250
	C15	Park. Loam.	3.3	370	140
Shrine site	1	Shrine site. Silt.	6.6	350	230
	5	Temple site. Clay.	5.6	700	200
	17	Temple site. Silty skeletal.	25	3500	870
Lane of rice paddy	10	Irrigation channel lane. Humic soil.	46	26000	2500
	11	Spirit house site. Humic.	73	13000	2800
	12	Irrigation channel lane.	92	53000	2500
	13	Humic.	43	15000	1500
	16	Silty-skeletal.	53	28000	1700
	27	Close to rice paddy. Loam.	120	110000	4700
	D1	Silty-loam.	59	30000	2200
	D2	Silty-loam.	110	46000	4000
	D3	Loam.	11	4400	380
	D5	Silty-loam.	150	69000	7400
	D6	Silty-loam.	78	34000	2900
	D7	Silty-loam.	48	22000	2200
	D8	Loam.	41	19000	1300
Side way of main road	2	Sandy loam.	16	1400	640
	6	Clay, Gravel	15	1600	510
	8	Clay, Gravel.	15	1900	430
	15	Silty skeletal.	25	16000	980
Shrubbery of shrines	4	Humic soil.	31	1500	950
	9	Shrine in rice paddy Humic sand	20	5700	690
	25	Humic soil.	38	1600	1400
	B2	Loam. Herbicide applied. Small incinerator	77	7400	2400
	B3	Loam. Herbicide applied. Small incinerator	40	3300	1300
	B4	Humic soil.	41	2600	1500
Mound	21	Skeletal soil.	77	17000	2400
	22	Skeletal soil.	160	22000	3700
	23	Silty-skeletal	53	29000	2200
	24	Silty-skeletal.	15	5500	450
	A8	Loam. Herbicide applied.	0.18	110	51
	B1	Mound, Silty loam. Herbicide applied.	7.1	3000	210
	D4	Silty-loam.	15	6600	690
	E1	Mound. Loam.	71	21000	3300
	E2	Mound. Loam.	46	13000	1500
	E3	Mound. Loam.	35	7900	2500
	E4	Mound. Loam.	49	9400	1500
	E5	Mound. Loam.	72	23000	1900
	E6	Mound. Loam.	14	3500	630
	E7	Mound. Loam.	22	5800	690
	E8	Mound. Loam.	20	4000	700
Others	26	Silty skeletal.	23	5500	670

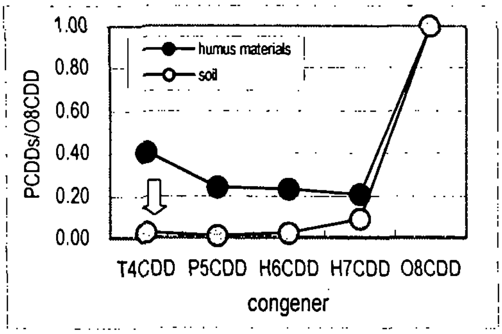

Figure 1. Ratio of PCDDs congener to O8CDD in humus materials and soil.

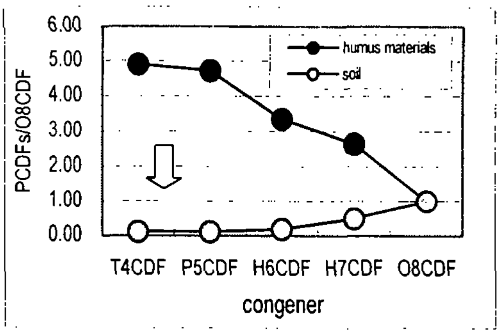

Figure 2. Ratio of PCDFs congener to O8CDF in humus materials and soil.

leaves are considered to follow this fate. Principal Component Analysis, taking these facts mentioned above into consideration, showed the *Eigenvalues* of the first and the second components were 5.597996 and 2.358598, respectively and contributions of each component were 62.2% and 26.2%, respectively. Plus *Eigenvalue* of the first component includes samples obtained at places contaminated by herbicide, while minus means soils mineralized from humus (Figure 3). On the other hand the *Eigenvalue* of the second components expresses contamination levels in general, showing larger value at heavily contaminated sites.

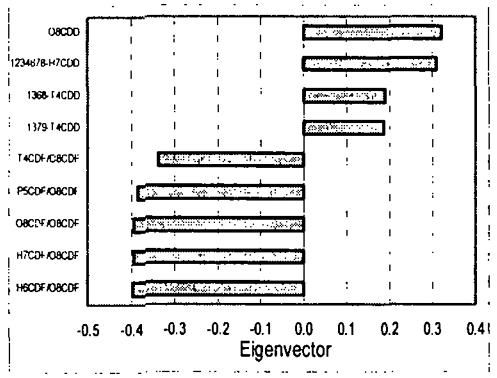

Figure 3. Eigenvector of Component 1

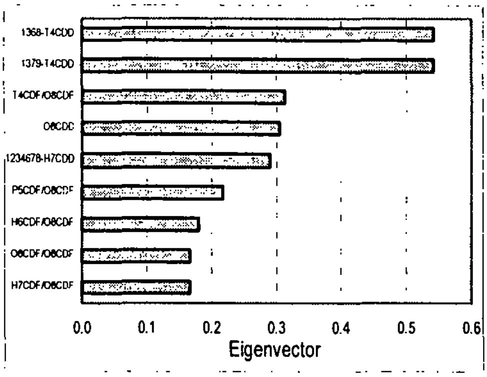

Figure 4. Eigenvector of Component 2

Cluster Analysis

Cluster analysis was conducted for the components values and PCDD/Fs levels (pg-TEQ/g). As shown in Figure 5, the level of PCDD/Fs in Cluster 1, 2 are relatively low and well mineralized, while the soils in Cluster 3,4,5 are relatively contaminated by PCDD/Fs, presumably resulted from PCP or CNP application in the past. The result of cluster analysis and land use profiles are shown in Table 2.

Conclusion

Each cluster is characterized as follows.

(1) *Cluster 1:* Playgrounds, shrine sites, and farmlands are included in this cluster, and PCDD/Fs

levels are relatively low (4.7 pg-TEQ/g), meaning less anthropogenic contamination. Playground is abundant in sand on which PCDD/Fs are unlikely adsorbed.

(2) Cluster 2: Farmlands, mounds, shrubbery of shrines, parks and schools, and sideways of the main road are included in this group, and the soil in these area contain much loam that is likely to adsorb PCDD/Fs. Average value of PCDD/Fs in this cluster is 20.2 pg-TEQ/g.

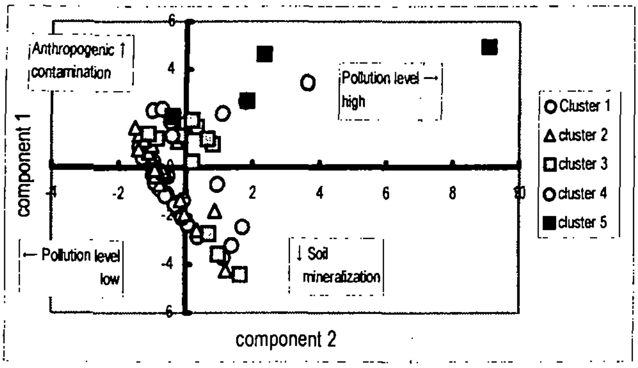

Figure 5. Relationship between component score and cluster

(3) Cluster 3: Average value of PCDD/Fs of this cluster is 46.0 pg-TEQ/g and includes samples of lanes of a rice fields, shrine shrubbery and mounds where PCP or CNP were likely applied in the past.

(4) Cluster 4: Lane of rice paddy and mound areas are included in this cluster, same as Cluster 3. But average value of PCDD/Fs is 77.6 pg-TEQ/g, higher than that of Cluster 3.

(5) Cluster 5: Average PCDD/Fs in this cluster was calculated as 134.4 pg-TEQ/g, abnormally higher, and the usage of PCP and CNP or open burning of agricultural residues in the past were definitely suspected

Table 2. Cluster analysis and land utilization

Area	Cluster					n	TEQ-pg/g
	1	2	3	4	5		
Playground of parks or schools	13	1		1		15	9.1
Shrine site	2	1				3	12.2
Lane of farm land	4	3	1	1		9	25.4
Lane of rice paddy	1		6	3	3	13	71.0
Mound	2	6	3	3	1	15	43.6
Shrubbery of shrines		2	3	1		6	41.1
Shrubbery of parks or schools		4				4	16.2
Side way of main road		4				4	17.6
Others		1				1	22.8
n	22	22	13	9	4	70	Total
average TEQ-pg/g	4.7	20.2	46.0	77.6	134.4		34.0
median TEQ-pg/g	4.6	19.2	45.7	76.8	136.4		21.0
max TEQ-pg/g	11.6	34.5	58.9	92.0	157.1		157.1
min TEQ-pg/g	0.2	13.4	38.2	66.5	107.7		0.2

References

(1) Wakimoto, T., Murakami, T., and Tatsukawa, R. (1986) The 6th International Symposium on Chlorinated Dioxins and Related Compound, Fukuoka, Japan.

(2) Masunaga, A., Nkanishi, J. (1999) The 2nd International Workshop on Risk Evaluation & Management of Chemicals, Yokohama, Japan.