Levels of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Soil and Pine needle near Industry Complex in Changwon City

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Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) were investigated in soil and pine needle samples taken from 16 sites of industrial and residential areas in Changwon, Korea to assess their distribution levels and to suggest the influence of industrial activities. PCDDs/DFs levels in the soil samples ranged from 0.57 to 20.79 pg I-TEQ/g dry weight with a mean value of 4.20 pg I-TEQ/g dry weight. PCDDs/DFs levels in the pine needle samples ranged from 0.39 to 8.75 pg I-TEQ/g dry weight with a mean value of 4.09 pg I-TEQ/g dry weight. In both soil and pine needle samples, the PCDDs/DFs concentrations in the industrial area sites were higher than those in the residential area sites. Homologue profiles in pine needle samples showed different patterns compared with soil samples. Based on the results of principal component analysis (PCA), it was confirmed that pine needles reflected a direct influence from local potential sources of PCDDs/DFs, showing a much higher degree of reflection than in soils. Pine needles are very useful as an indicator for monitoring or estimating the contamination of PCDDs/DFs in other areas which have been impacted by point pollution sources.

Key Words : PCDDs/DFs, Soil, Pine needle, Industrial and residential area, PCA, Local potential sources

1. Introduction

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs/DFs) are well-known as the serious toxic environmental contaminants that are never intentionally manufactured from artificial sources by human activities. These compounds are caused severe defects for ecological system and human health due to their toxicity such as carcinogenicity, immunotoxicity, neurotoxicity and reproductive and developmental toxicity etc[1~3]. Especially, 2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin (2,3,7,8-TeCDD) included in Group 1 which is carcinogenic to humans according to the International Agency for Research on Cancer[4]. PCDDs/DFs are ubiquitous and persistent in the environment and the main sources are classified as combustion processes and chemical-industrial processes (i.e., municipal waste incinerators, iron and steel production, cement kilns, secondary copper smelters, medical waste incinerators, automobile emissions, manufacture of chlorinated chemicals, pulp and paper industry, and dry cleaning)[5~7]. PCDDs/DFs emitted from these sources are transported through the atmosphere over long distances and precipitated into soil, sediment, vegetation and biota by wet and dry deposition processes[8~10], and eventually accumulate in the human body[11].

Measuring atmospheric levels of PCDDs/DFs in local regions directly is difficult and thus many researchers have studied to evaluate the air pollution level of PCDDs/DFs by using soil and plant samples as an environmental indicator instead of direct air monitoring[12]. For example, Paustenbach et al.[13] studied for PCDDs/DFs persistence in soil and concluded that 2,3,7,8-TCDD has a half-life of 25 to 100 years in the subsurface soil and 9 to 15 years at the soil surface. Due to the high organic carbon partition co-

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efficient (K_{oc}), PCDDs/DFs are accumulated in the organic matter of soil, which is one of the final reservoirs. Meneses and co-workers[14] reported that soils reflect cumulative PCDDs/DFs deposition during rather long periods of time and the most important pathway contributing to accumulation of PCDDs/DFs in soil is the background concentration itself. Moreover, several studies have demonstrated that the waxy outer surfaces of plants will accumulate lipophilic organic pollutants from the atmosphere[15,16]. Reischl and co-workers[17] have utilized the outer waxy surfaces of pine needles to investigate the levels, distribution and sources of PCDDs/DFs and other organochlorine pollutants in the atmospheric environment.

The objective in this study was to investigate the levels and distribution characteristics of PCDDs/DFs in soil and pine needle samples collected from both industrial district and residential area for assessment of environment impact of PCDDs/DFs followed by industrial activities.

2. Material and method

2.1. Sampling location

Changwon is an industrialized city with ca. 500,000 inhabitants, located at the southeast extreme of the Korean Peninsular (Fig. 1). A huge machinery industrial zone was allocated in Changwon in 1978 and 632 companies are currently within this zone. Therefore, Changwon is a representative city for the study of stationary source of PCDDs/DFs followed by industrial activities. In addition, this study can provide background data of PCDDs/DFs in an industrial zone in Korea.

Sampling was carried out in September, 1999. The map of sampling sites in this study is shown in Fig. 1. Soil (n=15) and Pine needle (n=15) samples were collected from 16 sites in the vicinity of the industrial and residential areas in Changwon (soil sample at site C and pine needle sample at site P were not taken). sites A-H are located in the area of the south-westward arrow on the sampling map belonging to the industrial area and sites I-P are located in the area of the north-eastward arrow belonging to the residential area.

At each sampling site, soil samples were taken from the surface (a size of 30 cm×30 cm×5 cm) and pine needles were also obtained by cutting at a height of 1-2 m above the surface soil. All samples were immediately packed in aluminum foil and sealed in a clean plastic bag, and then shipped to the laboratory. In the laboratory, all samples were stored at -20℃ until analysis.

2.2. Analytical procedures
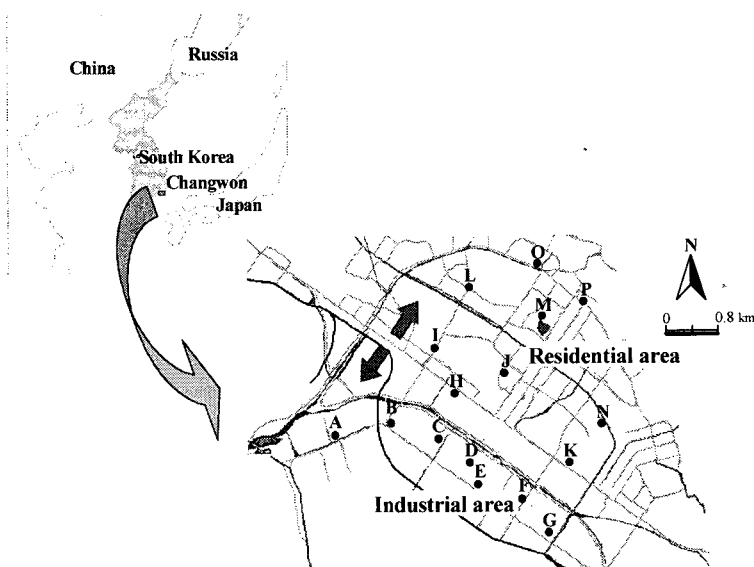
Soil and pine needle samples were dried at room

Fig. 1. Map of sampling sites in this study.

temperature. Soil samples were sieved with a 250 μm sieving screen after breaking up the lumps of soils and removing the large particles such as stones and leaves of plants. About 20 g of each soil sample was reflux-extracted with 150 mL of toluene for 6 hours and then filtered. The extracts were reduced to 1-2 mL in a rotary evaporator.

Pine needle samples were homogeneously cut into approximately 1 cm length. About 50 g of each pine needle sample was reflux-extracted with 500 mL of toluene for 5 hours. After extraction, extracts were filtered to remove the bulk of chlorophyll using silica gel. The extracts were then reduced to 1-2 mL in a rotary evaporator.

Each extract was adjusted to 10 mL of toluene, and 8 mL of the extract was used to analyze for PCDDs/DFs. After adding 0.5 mL of n-nonane as a keeping solvent, extracts were concentrated to less than 0.3 mL under a gentle stream of nitrogen gas. Subsequently, solutions were transferred to 10 mL of n-hexane, ensuring the complete removal of toluene, and an internal standard solution of 15 $^{13}C_{12}$-labeled PCDDs/DFs (EDF-8999, CIL Inc.) was spiked.

Solutions were pre-cleaned up on a multi-layer silica gel column chromatography; $AgNO_3$-silica gel, H_2SO_4-silica gel and KOH-silica gel loaded into a 1.5 cm i.d.×25 cm glass column and eluted with n-hexane. Eluates were concentrated to 10 mL, and cleaned up on an alumina column (1 cm i.d.×20 cm glass column) chromatography activated for 24h at 600℃ and successively eluted with 0.4% methylene dichloride in n-hexane, 4% methylene dichloride in n-hexane and 50% methylene dichloride in n-hexane. The third fraction was concentrated to less than 1mL and left at a clean room for one or two days. The purified extracts were dissolved with 50 L of n-nonane and analyzed for each of the 5 chlorinated dibenzo-p-dioxin and dibenzofuran congener groups (with 4 to 8 chlorines) by HRGC/HRMS[18~21].

HRGC/HRMS analysis was performed using a HP 6890 gas chromatograph coupled to a Jeol JMS-700 mass spectrometer at a resolution of moer than 10,000 (10% valley) in selected ion monitoring (SIM) mode. Each sample was analyzed on two different GC capillary columns. PCDDs/DFs with 4-6 chlorines were quantified on a 60 m SP2331 (0.25 mm i.d., 0.2 um film thickness, SUPELCO) and PCDDs/DFs with 7-8 chlorines on a 60 m DB-5MS (0.25 mm i.d., 0.1 um film thickness, J & W Scientific). Data acquisition by the mass spectrometer was controlled on a HP work station and the data process by using Jeol DioK program on a general PC. Quantitative determination of PCDDs/DFs was performed by the relative response factors of the native PCDDs/DFs standard concentration to the $^{13}C_{12}$-labeled internal standard concentration obtained from standard mixtures (EDF-9999, CIL Inc.), as recommended by the US EPA[22].

3. Results and discussion

3.1. PCDDs/DFs levels

Results of all 2,3,7,8-chlorine substituted congeners, congener groups (homologues), total PCDDs/DFs, the ratio of PCDDs/DFs and I-TEQs for soil and pine needle samples are summarized in Table 1. I-TEQs were calculated by the international toxicity equivalency factors (I-TEFs) recommended by NATO.

I-TEQ levels of PCDDs/DFs in soil samples ranged from 0.57 to 20.79 pg I-TEQ/g dry weight with a mean value of 4.20 pg I-TEQ/g dry weight. The highest value was found at site H in the industrial area, and the lowest value at site O in the residential area. The mean I-TEQ value of the soil samples was 7.12 pg I-TEQ/g dry weight for the industrial sites and 1.65 pg I-TEQ/g dry weight for the residential sites, showing that the industrial area was 4.84 times higher than the residential areas. Schuhmacher et al.[23] reported PCDDs/DFs levels in soil samples from the vicinity of an old municipal solid waste incinerator (San Adri del Bess, Barcelona, Spain) ranged from 1.22 to 34.28 pg I-TEQ/g dry weight with a mean value of 12.24 pg I-TEQ/g dry weight. These values were rather higher than those in this study.

With regard to pine needle samples, I-TEQ levels of PCDDs/DFs ranged from 1.20 to 8.75 pg I-TEQ/g dry weight with a mean value of 4.09 pg I-TEQ/g dry weight. Both the highest and lowest values were found at site I and site M in the residential areas, respectively. The mean value of the industrial area sites was 5.17 pg I-TEQ/g dry weight, and the mean value of the residential area sites was 2.86 pg I-TEQ/g dry weight. The mean I-TEQ level of the industrial pine needles was 1.81 times higher than that of the residential pine needles. Safe et al.[15] reported that PCDDs/DFs levels in pine needles collected in two

Table 1. Summary of PCDDs and PCDFs in soil and pine needle[a]

| | Soil (n=15) | | | | Pine needle (n=15) | | | |
| | Industrial area | | Residential area | | Industrial area | | Residential area | |
	Mean	SD[b]	Mean	SD	Mean	SD	Mean	SD
2378-TeCDD	0.15	0.08	0.08	0.07	0.26	0.16	0.09	0.05
Total TeCDDs	26.94	13.49	17.9	10.28	40.25	27.42	17.57	10.03
12378-PeCDD	0.75	0.55	0.22	0.2	0.98	0.67	0.37	0.32
Total PeCDDs	22.86	15.22	8.09	4.98	38.72	27.22	14.06	8.91
123478-HxCDD	0.76	0.69	0.2	0.15	0.59	0.31	0.46	0.38
123678-HxCDD	1.6	1.26	0.39	0.24	0.99	0.57	0.65	0.35
123789-HxCDD	28.29	28.43	5.09	2.79	16.97	13.08	7.25	11.76
Total HxCDDs	33.94	30.54	8.63	4.62	34.87	21.41	16.28	11.21
1234678-HpCDD	11.8	11.56	4.65	3.06	3.68	1.87	2.47	1.69
Total HpCDDs	23.94	23.57	9.18	5.79	8.61	3.99	5.7	3.86
OCDD	32.91	55.91	21.57	19.71	2.12	1.27	1.71	1.46
2378-TeCDF	1.45	0.81	0.45	0.35	2.17	1.36	0.87	0.51
Total TeCDFs	50.56	29.94	16.02	13.79	61.3	33.22	32.12	18.25
12378-PeCDF	3.8	2.88	1.09	1	3.17	1.83	2.19	1.63
23478-PeCDF	3.17	2.84	0.78	0.68	2.74	1.65	1.76	1.33
Total PeCDFs	49.87	39.04	14.12	12.6	48.56	28.76	29.2	18.78
123478-HxCDF	4.03	3.35	0.93	0.82	2.24	1.56	1.83	1.36
123678-HxCDF	3.4	2.96	0.78	0.78	2.27	1.45	2.03	1.43
123789-HxCDF	1.59	1.63	0.35	0.18	0.46	0.36	0.34	0.26
234678-HxCDF	3.92	4.63	0.8	0.76	2.13	1.36	1.66	1.21
Total HxCDFs	39.97	39.65	9.42	8.18	26.01	16.51	19.49	13.29
1234678-HpCDF	14.64	19.09	4.02	3.73	6.41	3.43	5.54	4.48
1234789-HpCDF	1.52	1.55	0.5	0.48	0.6	0.37	0.44	0.29
Total HpCDFs	24.51	30.1	7.01	6.04	10.1	5.51	8.15	6.57
OCDF	6.81	9.01	2.57	2.08	1.22	0.7	0.82	0.68
I-TEQ	7.12	6.22	1.65	0.8	5.17	2.96	2.86	2.65
Total PCDD/DFs	312.3	235.02	114.52	61.98	271.76	156.1	145.12	91.14
Ratio PCDDs/PCDFs	0.85	0.28	1.66	0.7	0.82	0.27	0.64	0.1

[a]Results are average at each city and given in pg/g dry weight. [b]SD is standard deviation.

wood-preserving chemical sites were 7.7 and 5.4 pg I-TEQ/g dry weight, respectively. These values were similar to those in this study. However, Schuhmacher et al.[23] reported PCDDs/DFs levels in herbage from the vicinity of an old municipal solid waste incinerator ranged from 0.33 to 1.98 pg I-TEQ/g dry weight with a mean value of 0.70 pg I-TEQ/g dry weight. Weiss et al.[24] also reported PCDDs/DFs levels (0.3-1.9 pg I-TEQ/g dry weight and mean 0.70 pg I-TEQ/g dry weight) in spruce needles of background forest sites in Austria. These I-TEQ levels were lower than the results found here.

In this study, the difference of I-TEQ levels between the industrial area sites and the residential area sites showed that pine needles (1.8, mean value of industrial pine needles/mean value of residential pine needles) was about 2-3 times lower than those soils (4.3, mean value of industrial soils/mean value of residential soils). However, the total mean value of soil and pine needle samples were similar in two areas as 4.39 and 4.02 pg I-TEQ/g dry weight, respectively. Although the distribution of PCDDs/DFs concentrations in soil samples were rough in both areas, PCDDs/DFs concentrations in pine needle samples were regularly

distributed. These results predict that pine needles properly showed the influence of local potential sources, exhausting the lipophilic organic pollutants such as PCDDs/DFs, more than in soils.

Fig. 2 shows the correlation between total and I-TEQ concentrations of PCDDs/DFs in soil and pine needle samples. Results showed significant correlations both in soils (R^2=0.72) and pine needles (R^2=0.82). However, no significant correlations between soil and pine needle samples were found for PCDDs/DFs homologues (Fig. 3). This was in accordance with the results previously reported by Schuhmacher et al.[23].
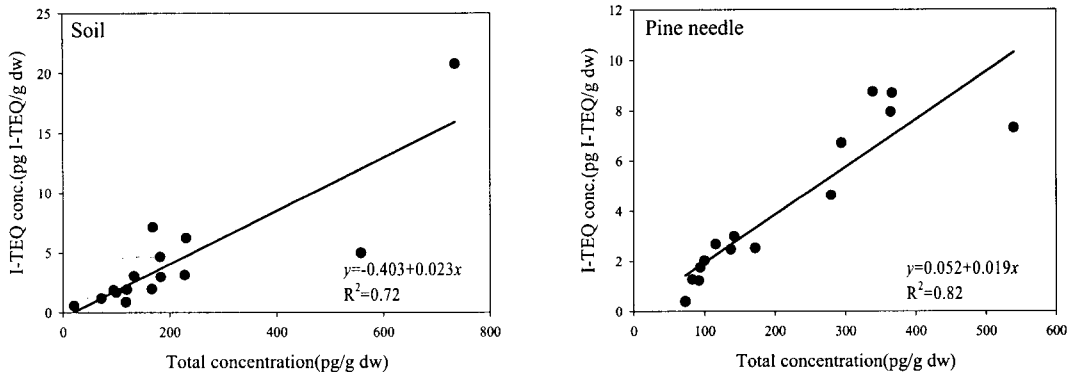

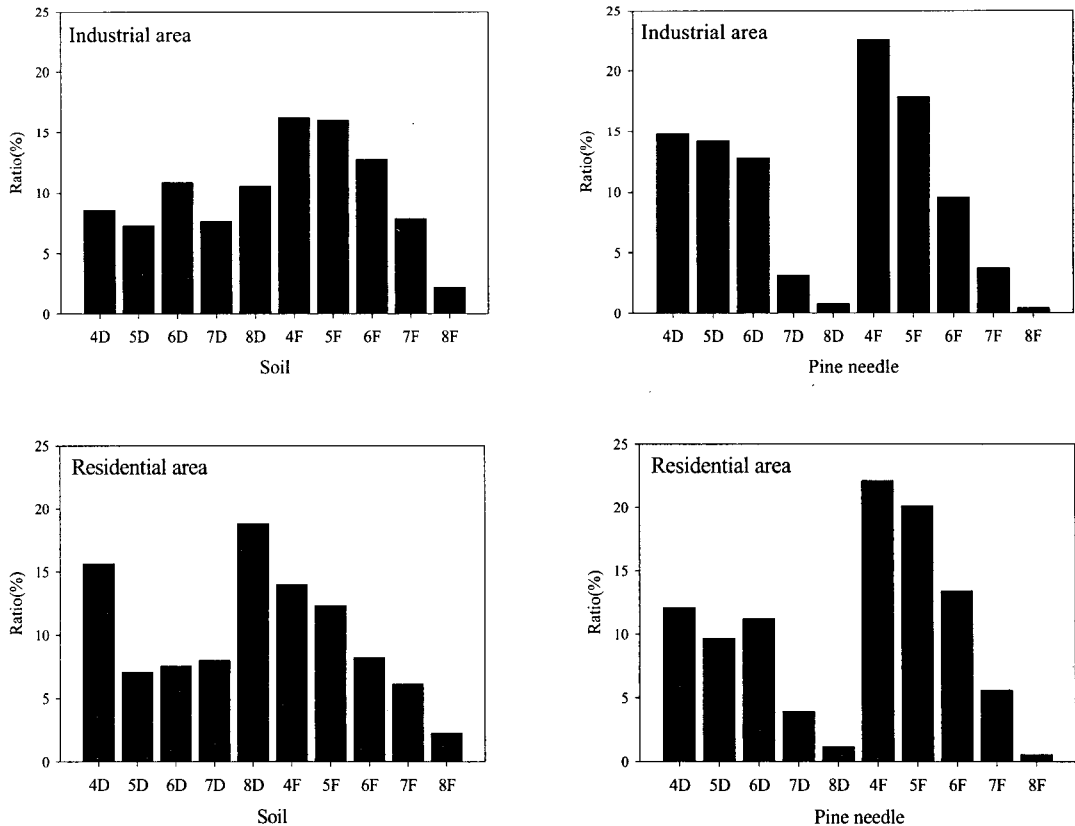Fig. 2. Correlation between total and I-TEQ concentrations of PCDDs/DFs.

Fig. 3. Average PCDDs/DFs homologue contribution profiles to total PCDDs/DFs concentrations. (4D: TeCDDs, 5D: PeCDDs, 6D: HxCDDs, 7D: HpCDDs, 8D: OCDD, 4F: TeCDFs, 5F: PeCDFs, 6F: HxCDFs, 7F: HpCDFs, 8F: OCDF)

3.2. PCDDs/DFs profiles

The PCDDs/DFs homologue profiles normalized to total concentration are shown in Fig. 3. The average homologue profiles showed different patterns between soil and pine needle samples. The contributions of the lower chlorinated PCDDs/DFs in pine needles are predominant in both industrial and residential areas. However, soil samples showed that the contributions among PCDDs homologues in the industrial area were similar each other, whereas the contributions of OCDD and TeCDDs in the residential area were predominant. The contributions of PCDFs homologues between the industrial and residential area were similar. These results could also be confirmed in 2,3,7,8-substituted congener profiles of soil and pine needle samples (Fig. 4). These results for the different patterns of PCDDs/DFs homologue profiles between soil and pine needle samples can be explained by considering that the higher chlorinated PCDDs/DFs have a lower pressure vapor, a higher K_{ow} constant and a higher half-life than the lower chlorinated PCDDs/DFs[24]. Also, it can be assumed by Schuhmacher et al.[22] that homologue profiles found on vegetation samples are determined by deposition of different proportions of vapor-phase PCDDs/DFs and particle-bound PCDDs/DFs from the ambient air.

In order to statistically distinguish the contamination source and evaluate the difference between the profiles of soil and pine needle samples, principal component analysis (PCA) of multivariate statistic analysis was applied. PCA is a useful method to identify specific PCDDs/DFs sources[26]. Statistical analysis in this study was carried out using a SPSS-10.0 statistical software package. Fig. 5 shows the score plot resulting from PCA based on the PCDDs/DFs homologue profiles (A) and 2,3,7,8-substituted congener profiles
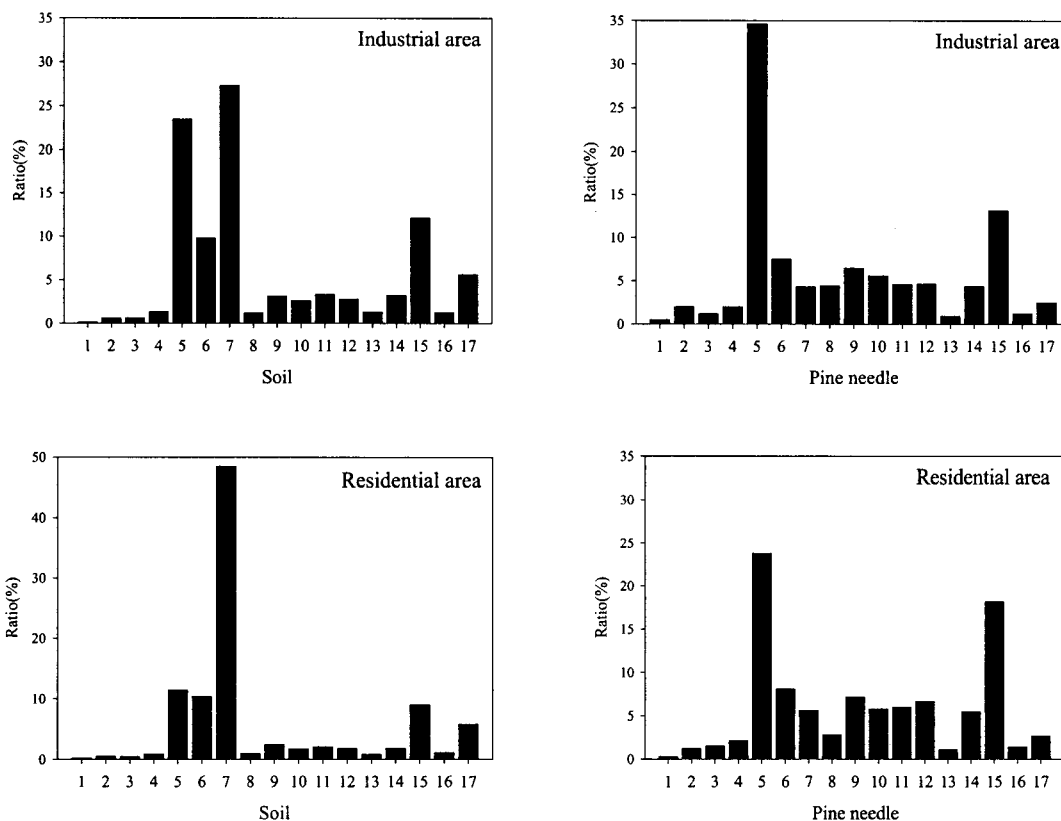

Fig. 4. Average 2,3,7,8-substituted congener contribution profiles to total PCDDs/DFs concentrations. (1: 2378-TCDD, 2: 12378-PCDD, 3: 123478-HxCDD, 4: 123678-HxCDD, 5: 123789-HxCDD, 6: 1234678-HpCDD, 7: OCDD, 8: 2378-TCDF, 9: 12378-PCDF, 10: 23478-PCDF, 11: 123478-HxCDF, 12: 123678-HxCDF, 13: 123789-HxCDF, 14: 234678-HxCDF, 15: 1234678-HpCDF, 16: 1234789-HpCDF, 17: OCDF)

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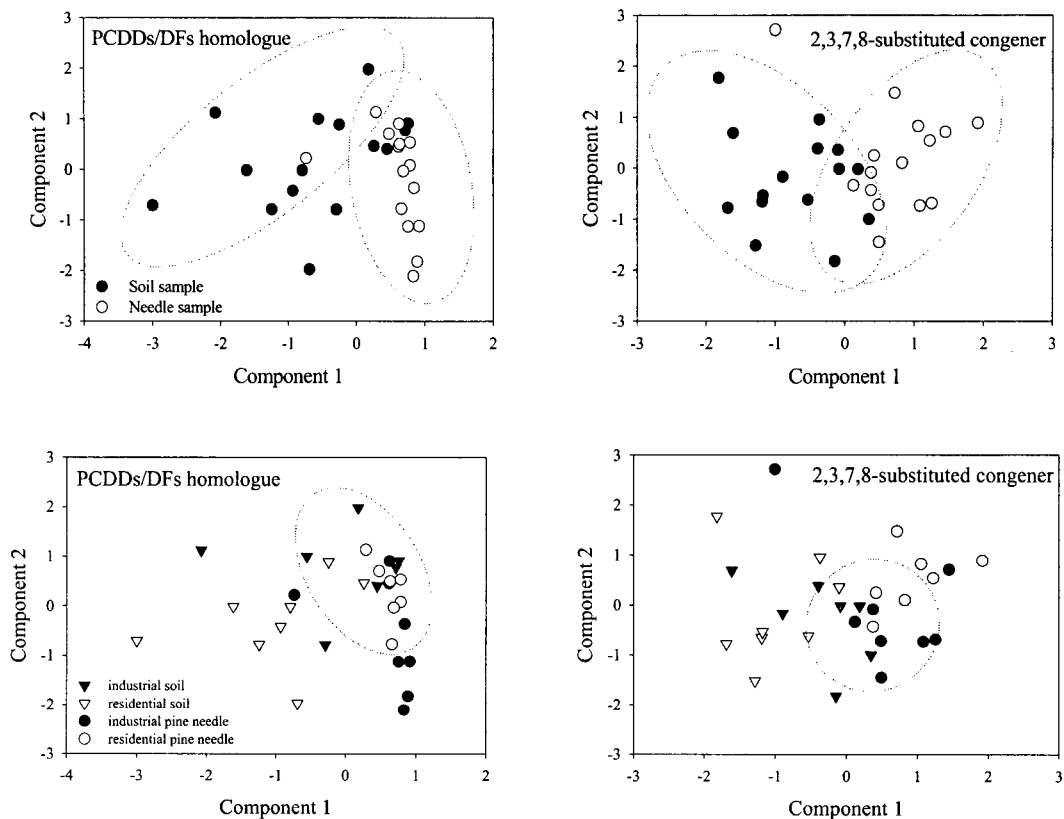

Fig. 5. Score plot of principal component analysis of PCDDs/DFs.

(B) of soils and pine needles at each sampling site. In the score plots, the first principal component (PC 1) explained 54.96% of A and 50.18% of B for the total variance, and the second principal component (PC 2) explained 17.39% of A and 20.59% of B for the total variance. Both PC 1 and PC 2 accounted for 72.35% (A) and 70.77% (B) of the total variance in the data. Therefore it is considered that Fig. 5 includes most of the original information about the PCDDs/ DFs distribution characteristics. Both the upper score plots for soil and pine needle samples showed that two groups could be identified. These groups showed that the first one contained all soil samples and the second one of pine needle samples. It could be also observed from both the lower score plots that pine needle samples in this study were relatively closer to soil samples of the industrial area than the residential soil samples. This apparently indicated that the dis-

tribution characteristics of PCDDs/DFs in pine needle samples were similar to those in soil samples of the industrial area. However, it is not accurately shown by the PCDDs/DFs homologue profiles and the correlations of PCDDs/DFs concentrations between soil and pine needle samples. According to the studies of Ok et al.[25] and Domingo et al.[12], PCDDs/DFs of vapour-phase rather than particle-bound in vegetation samples were absorbed by air-to-leaf. It is suggested that PCDDs/DFs of vapour-phase produced and emitted at factories directly contribute to contamination of PCDDs/ DFs in pine needle within a short-term period[28,29]. Accordingly, the results described above suggested that pine needles reflected the influence of PCDDs/ DFs contamination better than soils by point sources, because we can consider that industrial soil is directly affected, as opposed to residential soil by the production activities of the industrial zone.

4. Conclusions

The investigation on PCDDs/DFs in soils and pine needles in Changwon city revealed that, as expected, PCDDs/DFs concentrations in industrial area were higher than those in the residential area. In this city, PCDDs/DFs levels in soil and pine needle samples varied from 0.57 to 20.79 pg I-TEQ/g dry weight and from 1.20 to 8.75 pg I-TEQ/g dry weight, respectively. However, the correlation of PCDDs/DFs concentrations between soils and pine needles was not significant. It was confirmed that the distribution of PCDDs/DFs between soil and pine needle samples differed, and that pine needles reflected the influence from local potential sources of PCDDs/DFs and showed a much larger degree of reflection than soils. Therefore, it is suggested that pine needles are very useful as an indicator for monitoring or estimating the contamination of PCDDs/DFs in other areas which have been impacted by point pollution sources. In order to more accurately assess the total effects of local potential sources of PCDDs/DFs, further study including the analysis of the atmospheric air in these regions is needed.

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