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Long-Term Monitoring of Dioxin and Furan Level in Soil Around Medical Waste Incinerator

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1. Introduction

The annual generation of solid waste is quite huge in China. For instance, approx. 157 million tons of municipal solid waste (MSW) and 2.04 billion tons of industrial solid waste (14.29 million tons of hazardous waste) were produced in 2009 (National Bureau of Statistics of China, 2010). These wastes would contaminate green land, drinking water and even air, ultimately threatening human health, so they must be treated in scientific methods. Waste treatment is a big challenge for every country. At present, the conventional disposal system according the hierarchy of methodologies includes recycle, compost, combustion and landfill. Combustion has noticeable advantages in volume and weight reduction, disinfection and short time cost, can also realize energy recovery by using waste to energy plants. Thermal treatment (pyrolysis and incineration) is the widely applied technology for waste treatment, for instance, accounting for 18.2% of MSW treatment in China and 11.9% in USA (2009). There are over 300 central incinerators for hazardous solid waste (HSW) in China (National Development and Reform Commission of China, 2003) and 93 municipal solid waste incinerators (National Bureau of Statistics of China, 2010). The present Chinese regulations prohibit the co-combustion of HSW and MSW (Ministry of Environment Protection, 2001).

However, waste incineration is still a controversial issue among social and scientific communities due to its secondary pollution, especially after the observation of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in incinerators (Olie et al., 1977). Waste incineration is thought a major source of PCDD/Fs in the environment. UNEP (UNEP Chemical, 2005) published the standardized toolkit for identification and quantification of dioxin and furan, including the emission factor of PCDD/Fs from combustion and incineration. Research (Gao et al., 2009; Ni et al., 2009) shows the emission factor of PCDD/Fs from medical waste incinerators (MWI) is nearly 63.3 μg I-TEQ/ton refuse into the atmosphere and 1.73 μg I-TEQ/ton from municipal solid waste incinerators (MSWI) in China, respectively. There are 135 dioxins and 175 furans, each with a different number and position of the chlorine atoms. 17 congeners of PCDD/Fs with 2,3,7,8 positions substituted by chorine are very toxic, which can induce a variety of adverse health problems, such as sarcomas, lymphomas and stomach cancer (Mitrou et al., 2001). These toxic pollutants can be formed by de novo synthesis and from precursor compounds (McKay, 2002), be emitted into the air through the stack, and transported to the ambient air,

then deposited over a wide area of earth surface (Wu et al., 2009). It's essential to control pollutant emission to minimize the environmental and health impact. A lot of relevant researches on dioxin determination, formation and emission control have been conducted in last decades. Unfortunately, all of this work still can not completely eliminate the public concern. Incinerators construction and operation is opposed by public and environmental protection organizations for PCDD/Fs exposure risk. Public protests happened a couple of times in last two years, and the constructions of several plants were halted in China.

In order to clarify dioxin exposure risk, surveys and monitoring programs have been carried out via detecting PCDD/Fs concentration in environmental media including soil, water, air, food and bio-tissues. On one hand, there are remarkable influences of waste incinerators on the environment. Kim et al. (2005) measured PCDD/Fs concentrations in ambient air, soil, pine needles and human blood in order to assess the relationships between incinerator sources and environment. It was observed the incinerator operation had directly influenced the observed PCDD/F congener profiles of soil and pine needles. Further, the difference between the levels of PCDD/Fs in the blood of office and plant workers demonstrates that human exposure to PCDD/Fs occurs as a result of the operation of the incinerator. By the Korea national monitoring of PCDD/Fs in the environmental media around incinerators (Kim et al., 2008), the average PCDD/Fs levels in soils decreased with increasing distance from the incinerator. From the PCDD/Fs level gradient away plant, a distance of 500 m is suggested as being under the influence of an incinerator. After introduction of technical improvement in MSWI, a reduction of 40% was observed in the median PCDD/Fs level in soil around the facility (Domingo et al., 2002). On the other hand, no significant impact of a waste incinerator on the neighborhood was reported too. In the research of a 10-year surveillance program of a hazardous waste incinerator (HWI) (Vilavert et al., 2011), the median value of PCDD/Fs in soil samples decreased 44% (from 0.75 to 0.42 ng I-TEQ Kg^{-1}) between 1999 and 2009 year survey. In order to establish the temporal variation after 6 years regular operation, the concentrations of PCDD/Fs in blood and urine of 19 workers employed at a HWI were measured in 1999 and 2005 (Mari et al., 2007). The analyzed results indicate that the workers at the HWI are not occupationally exposed to PCDD/Fs in their workplaces. In our previous research (Xu et al., 2009), the overall PCDD/F levels in the soil collected from the vicinity of the MSWI increased significantly, i.e., 39% for I-TEQ (median value) between 2006 and 2007, though the impact of MSWI on this study area is limited by congener-specific factor analysis. By the above review of the environmental impact of incinerators, this topic is still not resolved. The main potential reason is the different operation condition and pollutant emission level.

PCDD/Fs emission factor of MWI is much higher than the value of MSWI (UNEP Chemicals, 2005), so it is presumed that MWI has worse environmental influence than MSWI. In this study, PCDD/Fs level in soil in the vicinity of a MWI was monitored since April 2007, before this plant started operation (May 2007), and continued this determination every year after operation (2008-2010). This studied MWI is a typical central incinerator in China, with a capacity of 20 ton/day. The detailed sampling/analysis methods and experimental results are introduced along with discussion in this chapter.

2. Method and material

2.1 Study region and MWI

This studied MWI locates in the north of Zhejiang province, China. The designed capacity is 20 tons waste per day. The combustion technology is a rotary kiln combined with a

secondary combustor, as well as, an off-gas cleaning system that contains a quencher, a semi-dry scrubber and a fabric filter. There is another pyrolysis furnace (5 tons/day) in this factory, and its exhaust gas is emitted through the same stack as the incinerator. So the stack position is defined as this MWI location. The height of this stack is 35 m, and it is still lower than the near hills (Fig.1).

Fig. 1. Outside view and internal view of the medical waste incinerators.

2.2 Soil sampling method

Twelve soil samples for each year were collected in the vicinity of the MWI as shown in Fig.2. The exact sampling points were determined and recorded within 10 m of accuracy by a handheld GPS device (Meridian Color, Thales Navigation, USA), then transformed each point into the Geographic Information System (GIS) software packages of Google Earth (2003).

Fig. 2. Soil sample sites around the studied MWI.

The background sample (SB) was collected in a farmland southeast of the stack, 2400 m away. The local climate is featuring distinct seasons, typical to a subtropical weather condition. The seasonal wind is from the southeast direction in summer and northwest in winter. The sampling sites are mainly distributed in southeast and northwest. The MWI is built in a valley area, so that the choice of sampling sites must consider the site-condition. As some sites were frequently cultivated by farmer, the sampling was carried out by inserting a cylindrical steel corer (24cm × 4cm, length × internal diameter, Eijkelkamp,

Holland) down to a 10 cm depth. To obtain composite samples for each sampling point, soils were collected by mixing five different components (four main directions of 2 m radius and the center) within a 12.6 m² area. Approx. 1.5 kg of soil was taken at each site. Soil samples were air-dried in a ventilated room until reaching constant weight, and bio-material (roots, leaves) was manually removed. Then they were skived and sieved to < 0.25 mm. They were refrigerated until analysis, within two weeks. The first survey as PCDD/Fs baseline was conducted at April 2007, before this MWI started operation (May 2007). And soil samples were collected every year (2008 to 2010) in the same sites as the first survey after this facility operation began. During this period, fly ash and stack gas samples were collected from this MWI.

2.3 Clean procedure and analysis technology

About 10 g (dry mass) of soil samples were used for PCDD/Fs analysis. A selective pressured liquid extraction (SPLE) method was used for sample extraction by using a fully automated ASE 300 system (Dionex, Sunnyvale, CA, USA) (Fig.3). The extraction condition and procedure was referred to the SPLE method with a slight modification. Briefly, a 100-ml extraction cell was used and the ratio of soil:alumina:copper was 5:5:1. Each sample was spiked with a mixture of $^{13}C_{12}$-labelled PCDD/Fs compound stock solution (5 µl) and clean-up standard (5 µl) before extraction. The extracts from ASE were subsequently followed by rotary evaporation and multilayer silica gel column clean-up procedure following the Method of USEPA 1613. The extracts were blow-down to 20 µl under a gentle stream of nitrogen (N_2), and 5µl of $^{13}C_{12}$-labelled PCDD/Fs internal standard solution were added before sample were subjected to PCDD/Fs analysis by using high-resolution gas chromatography with high-resolution mass spectrometry (HRGC/HRMS) (JEOL JMS-800D) with a DB-5MS column (60 m × 0.25 mm × 0.25 µm). The toxic 2,3,7,8-substituted PCDD/Fs (referred to as congeners) as well as Tetra- to Octa-chlorinated homologues were identified based on isotope, and quantification of PCDD/Fs was performed by an isotope dilution method using relative response factors previously obtained from the five calibration standard solutions. In order to check the duplicate results, two soil samples are analyzed twice each year survey. If there is a wide variation in samples results, it also will be analyzed again. All isotope standards were purchased from the Cambridge Isotope Laboratories, Inc. (USA).
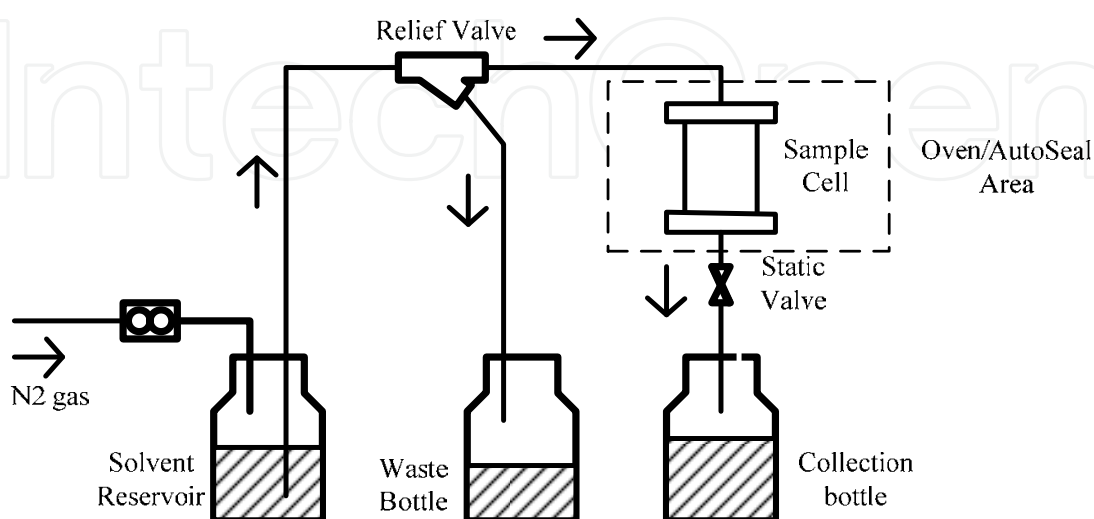

Fig. 3. ASE 300 Schematic System.

For source identification by comparison of PCDD/Fs homologue/congener patterns between soil and MWI emissions, stack gas and fly ash were collected from this MWI. The stack gas samples were collected with an isostack sampler (M5, KNJ Engineering, Korea) according to USEPA method 23A. The sample collection components included a glass fiber filters, in line with a condenser, the sorbent (XAD-2 resin) module and four impingers. The sampling labelled-$^{13}C_{12}$ standard was spiked into the XAD-2 resin before the sampling of flue gas. And the clean procedure was conducted as EPA23 method, including Soxhlet extraction by toluene for 24 h, wash with sulfuric acid (H_2SO4), a multi-layer silica gel column and an alumina column. The final clean extracts were blow-down to 20 μl under a gentle stream of nitrogen (N_2).The fly ash was collected at the exit of the bag filter. The clean procedure was conducted as EPA1613. The difference between EPA23 and EPA1613 is just using different labeled-$^{13}C_{12}$ standard solution as EPA1613 without sampling standard solution, and the clean process is generally the same. All of these samples were analyzed by HRGC/HRMS. The more detailed procedure of clean-up flue gas and fly ash samples can be found in the previous report (Chen et al., 2008).

2.4 Data analysis

All the experimental results were expressed on a dry weight basis. The 2,3,7,8-TeCDD toxic equivalents (I-TEQ) were calculated using NATO/CCMS factors (1988). Data was normalized before comparison of homologue and the multivariate analysis. Principal component analysis (PCA) was used to evaluate the similarities and differences of the PCDD/Fs homologue patterns and HxCDF isomer profile in soil samples, flue gas and fly ash. Each sample was assigned a score after PCA, allowing the summarized data to be further plotted and analyzed. PCA was performed using the SPSS 16.0 software package.

3. Results and analysis

The analysis results are present in table 1, including amount and TEQ concentration. Amount refers the concentration of total PCDD/Fs homologue from Tetra- to Octa-chlorinated species. PCDD/Fs level displays significant variation during these four years.

Sites	Amount, pg·g⁻¹				TEQ, pg I-TEQ·g⁻¹			
	2007	2008	2009	2010	2007	2008	2009	2010
S1	58.26	439.84	258.96	290.41	0.78	2.21	3.17	4.74
S2	848.34	1981.89	1155.45	1279.39	2.63	5.78	3.54	5.11
S3	397.04	465.10	374.05	669.21	1.78	3.51	2.37	6.07
S4	78.44	626.59	170.45	293.11	0.97	4.83	2.55	4.35
S5	433.77	546.01	551.95	1012.10	1.04	1.04	1.84	3.34
S6	66.48	89.55	123.51	164.73	0.64	0.94	1.34	1.41
S7	44.34	175.91	66.82	97.59	0.46	1.77	0.85	0.98
S8	263.18	273.81	252.84	329.64	1.91	1.99	1.47	3.30
S9	81.64	133.31	125.84	159.62	1.08	1.25	0.91	1.07
S10	57.18	78.51	67.49	92.80	0.45	0.88	0.69	1.12
S11	76.71	163.60	106.04	269.31	0.71	0.98	1.01	1.87
SB	55.94	55.72	79.42	85.01	0.60	0.53	0.73	0.65
Mean	205.11	419.15	277.73	395.24	1.09	2.14	1.70	2.83
Median	77.57	224.86	148.14	279.86	0.87	1.51	1.40	2.59

Table 1. PCDD/Fs amount and I-TEQ concentration in soil samples.

3.1 Baseline of PCDD/Fs concentration in soils

In the baseline survey (2007), PCDD/Fs concentration in this studied region is in the range of 44.34 to 848.34 pg g^{-1} (0.45 - 2.63 pg I-TEQ g^{-1}) with a mean of 205.11 pg g^{-1} (1.09 pg I-TEQ g^{-1}). It is lower than 4.0 pg I-TEQ g^{-1}, which is PCDD/Fs limit standard for cultivation land soil (GB15618-2009) in China (Ministry of Environment Protection, 2009), and this reflects there is no remarkable PCDD/Fs contamination. The German guideline (Federal Ministry for the Environment, 1992) recommends a limit of 5 pg I-TEQ g^{-1} for unrestricted agricultural use. US EPA (1998) recommends 1 pg I-TEQ g^{-1} in residential soil and 5 pg I-TEQ g^{-1} in commercial soil. Zheng et al. (2008) did a review of PCDD/Fs source and level in China, and found 0.09 to 2.4 pg I-TEQ g^{-1} in mountain and 0.14 to 3.7 pg I-TEQ g^{-1} in farmland. According to the survey (Jou et al., 2007), it is observed that PCDD/Fs range from 0.10 to 8.48 pg I-TEQ g^{-1} with an average of 2.20 pg I-TEQ g^{-1} in soil collected from a nature preserve area in Taiwan. Dioxin level in a urban surface soil in Norway is in the range of 0.16 to 14 pg I-TEQ g^{-1} (Andersson & Ottesen, 2008), and PCDD/Fs baseline in rural soil in Spain is 0.17 – 8.14 pg I-TEQ g^{-1} (Schuhmacher et al., 2002). Therefore, PCDD/Fs level in this survey is lower or generally comparative with the value of other places, beyond remarkable pollution. Further, the highest concentration is in S2, which is obviously abnormal from other sites. Actually, the surface and soil character in S2 is quite special, where is completely bare without any plant or herb, the soil is like limestone, which is commonly used in construction. So it is presumed that this point was polluted by some unknown historic activity, especially during the MWI construction.

3.2 PCDD/Fs concentration and variation after MWI operation

After this MWI started operation, a significant variation of PCDD/Fs concentration in soil is observed. In 2008, PCDD/Fs concentration ranges from 55.72 to 1981.89 pg g^{-1} (0.53 – 5.78 pg I-TEQ g^{-1}) with an average value of 419.15 pg g^{-1} (2.14 pg I-TEQ g^{-1}). In 2009, PCDD/Fs level is 66.82 – 1155.45 pg g^{-1} (0.69 – 3.54 pg I-TEQ g^{-1}) with an average of 277.73 pg g^{-1} (1.70 pg I-TEQ g^{-1}). In 2010, PCDD/Fs level ranges from 85.01 to 1279.39 pg g^{-1} (0.65 – 6.07 pg I-TEQ g^{-1}) with an average of 395.24 pg g^{-1} (2.83 pg I-TEQ g^{-1}). In the 2010 survey, the extraordinary sample is S5, and the increase compared to the value in 2009 is up to 460.15 pg g^{-1} (1.50 pg I-TEQ g^{-1}). So it is re-analyzed, and there is almost no difference between two measurements. In the on-site place of S5, there is no obvious specific pollution source. S5 is located in a hillside without herb or plants, and rain wash up is noticeable there. The possible explanation is that pollutants on soil surface were washed by rain and enriched in S5. Certainly, the persistent pollutant concentration in soil is the multi-result of pollution, distribution, deposition and bio-degradation.

The overall variation of PCDD/Fs level in soil is shown in Fig.4 and Fig.5. Figure 4 is the box plot of PCDD/Fs concentration each year, and Fig.5 is the comparison of PCDD/Fs baseline and the average of PCDD/Fs level after MWI operation (2008 to 2010) in every sites. In Fig.4, the PCDD/Fs variation is clear. PCDD/Fs level after operation is always higher than the baseline, and there is a little drop in 2009 compared to 2008. As analyzed in the previous paper (Li et al., 2010), the dioxin emission from this factory was largely reduced because medical waste combustion decreased and a series of improvements according to best available technique and best environment practice (BAT/BEP) were implemented in August 2008 (Lu et al., 2008). After the improvement, PCDD/Fs concentration in the stack gas and fly ash reduced by 96.7% and 83.15 %, respectively. This is the major reason of the PCDD/Fs decrease in the 2009 survey. In Domingo's research

(2002), a similar result was observed around a MSWI, 40% reduction in soil after technical alteration in the MSWI. Lee et al. (2007) found PCDD/Fs concentration in air around MSWI decreased approx. 50% after the introduction of a new flue gas treatment, as well as, 99.98% reduction of PCDD/Fs in stack gas samples. However, the PCDD/Fs level continues to increase in 2010 survey. The PCDD/Fs distribution in different sites and the relation of PCDD/Fs variation with distance from MWI is present in Fig.5. In the baseline, all of the sites almost stay in the same level of PCDD/Fs, and there is no specific trend with distance. After operation, the level curve (AO) goes up, particularly in the close sites (S1 to S4). With the amount comparison, the largest increase of PCDD/Fs (629.31 pg g^{-1}) is in S2, which is the closest point from MWI. Furthermore, S1 is the same distance away the stack as S2, and its increase (271.47 pg g^{-1}) is much lower than S2' increase. The main reason is the different characteristic surface in these two sites, as the thick grass covers in S1. Grass can reduce the adsorption of PCDD/Fs in soil, even absorb and degrade these toxic substances. And the curve (AO) of TEQ after operation displays a slight decline with distance. Meanwhile, the variation of PCDD/Fs is not significant in the farther sites than S5. So approx. 500 m radius is thought as the influence area in this case, which is consistent with another study (Kim et al., 2008). In this possible influenced area, there are no inhabitants except the staff of this plant, so the workers had better take strict protection to avoid health risk.
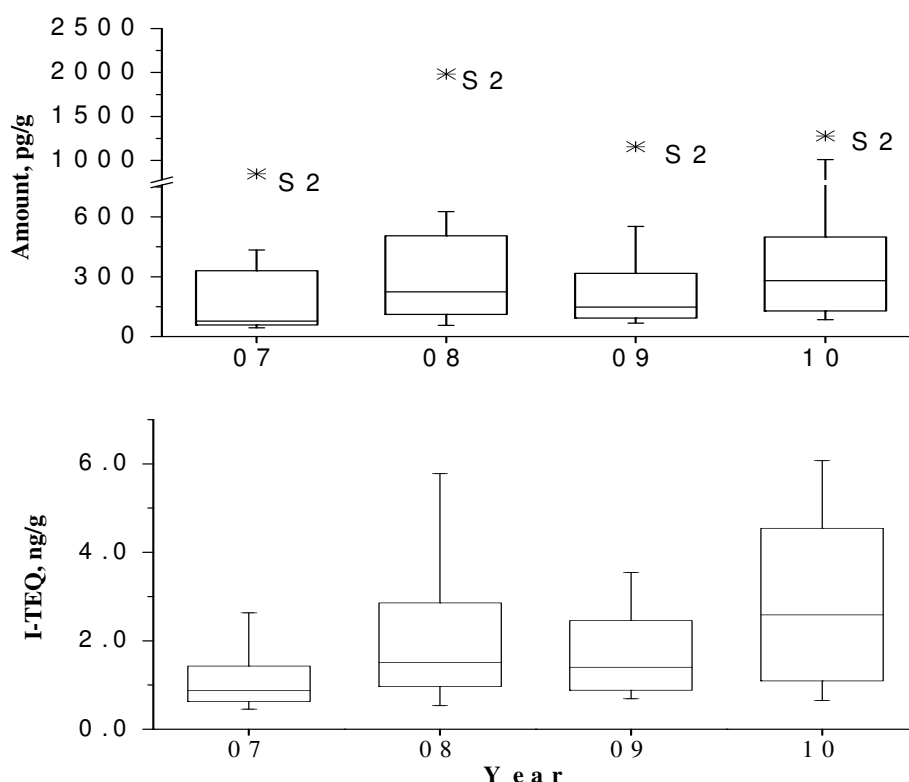

Fig. 4. Box plot of PCDD/Fs concentration in soils.

Figure 6 summarizes the average PCDD/Fs level in soil samples in the 2010 year survey and the comparison with different sites from Spain (Jiménez et al., 1996; Domingo et al., 2000), Taiwan (Cheng et al., 2003), Italy (Caserini et al., 2004; Capuano et al., 2005), Switzerland (Schmid et al., 2005), Norway (Andersson & Ottesen, 2008), South Korea (Kim et al., 2008), China (Yan et al., 2008), USA (Lorber et al., 1998) and Japan (Takei et al., 2000). The present PCDD/Fs level in this studied region is in the normal level as shown in Fig.6.
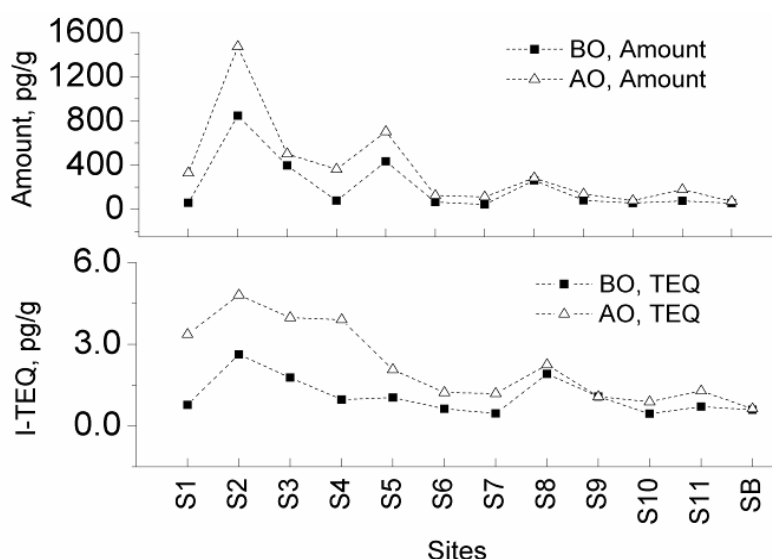
Fig. 5. Comparison of PCDD/Fs in soils collected before operation (BO, 2007) and after operation (AO, average of 2008 to 2010).
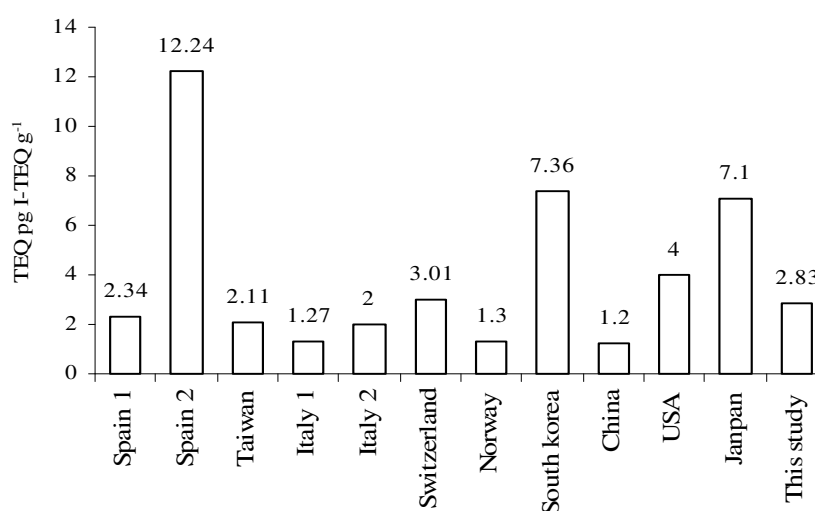

Fig. 6. The average of PCDD/Fs level in soil around worldwide.

3.3 Analysis of PCDD/Fs homologue pattern

Jiménez et al. (1996) found a slight PCDD/Fs contamination in soil near a medical waste incinerator in Madrid Spain, but did not clarify whether this plant was the only PCDD/Fs source responsible for the contamination. Homologue pattern or specific congener/isomer is defined as the fingerprint of PCDD/Fs. PCDD/Fs homologue distribution in soil, fly ash and stack gas are present in Table 2 to 6. The average PCDD/Fs homologue pattern in different surveys is present in Fig.7. Different PCDD/Fs sources have different fingerprint (Alcock et al., 1999; Domingo et al., 2001). In generally, the ratio of PCDFs to PCDDs from combustion processes is larger than 1, and a maximum weight distribution is PeCDF or HxCDF (Huang & Buekens, 1995). OCDD predominates PCDD/Fs homologue in the soil samples, which is consistent with other surveys. The deposition of OCDD on soil is easier and OCDD has longer degradation half-life time (Sinkkonen & Paasivirta, 2000). In the stack gas and fly ash, the dominant compound is HxCDF and PeCDF, and OCDD proportion is

less than 5%. In 2007 survey, percentage of OCDD is in the range of 40.81 to 90.97 with an average of 58.51, and the average ratio of PCDFs to PCDDs is 0.40. In 2010, the average percentage of OCDD distribution is 43.51 and the mean ratio is 0.72. That means the proportion of OCDD decreases and the ratio of PCDFs to PCDDs increases, and this change might be caused by PCDD/Fs source from combustion or other thermal processes.

2007	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
TeCDD	7.40	0.24	2.15	2.08	0.86	5.18	2.82	4.81	2.12	1.44	1.61	3.00
PeCDD	3.19	0.12	0.77	1.00	0.18	2.58	2.59	2.41	0.64	ND	2.49	3.61
HxCDD	2.02	0.39	1.23	3.42	0.72	3.82	3.80	2.15	2.46	2.51	1.23	3.86
HpCDD	7.49	2.14	5.07	7.60	3.55	7.07	7.74	5.44	4.93	5.25	7.41	6.38
OCDD	41.0	91.0	79.3	48.7	88.3	42.2	41.3	58.0	59.4	59.2	56.6	40.8
TeCDF	17.2	1.76	4.30	9.80	2.82	16.2	9.51	14.5	6.02	8.09	5.87	13.8
PeCDF	6.23	0.79	1.55	8.12	1.00	6.13	7.41	3.34	7.43	6.11	6.57	4.69
HxCDF	7.37	1.31	3.01	9.13	1.28	7.85	9.35	4.26	7.22	8.94	7.28	10.2
HpCDF	6.95	1.16	1.39	8.04	0.89	5.99	10.24	2.99	6.31	5.21	6.31	7.52
OCDF	1.15	1.13	1.24	2.10	0.44	2.98	5.26	2.17	3.46	3.21	4.68	6.14

Table 2. PCDD/Fs homologue distribution in soil of 2007, %.

2008	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
TeCDD	0.80	0.26	1.76	0.83	0.20	2.31	1.97	2.59	1.92	3.10	1.26	2.02
PeCDD	0.74	0.29	0.78	0.85	0.31	2.94	1.60	2.64	2.31	2.45	1.51	2.24
HxCDD	1.14	0.27	2.63	2.09	0.67	4.58	2.55	2.81	3.43	4.44	1.69	6.78
HpCDD	1.17	1.55	5.43	3.49	3.44	5.16	3.08	4.60	4.17	5.59	3.22	4.42
OCDD	58.4	73.8	56.3	36.1	91.6	32.6	18.5	43.6	40.8	28.9	26.8	43.3
TeCDF	6.70	1.87	10.2	4.94	1.37	13.0	15.7	11.4	8.92	14.2	6.19	17.7
PeCDF	6.28	1.33	8.40	5.89	0.77	13.8	14.8	12.3	6.45	7.42	4.68	11.4
HxCDF	5.78	1.47	8.00	5.27	0.66	12.11	14.3	8.16	7.68	12.3	4.64	5.48
HpCDF	2.87	1.43	4.33	5.22	0.60	7.21	7.77	4.56	7.27	11.4	6.05	3.96
OCDF	16.1	17.7	2.22	35.4	0.35	6.33	19.8	7.38	17.1	10.3	44.0	2.72

Table 3. PCDD/Fs homologue distribution in soil of 2008, %.

2009	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
TeCDD	3.06	0.36	1.14	3.40	0.68	3.09	3.10	3.49	1.77	2.82	3.05	2.57
PeCDD	3.41	0.45	1.42	3.21	0.68	5.00	4.42	3.37	1.86	3.62	3.40	2.70
HxCDD	4.51	0.63	2.37	5.81	1.04	4.30	5.17	2.92	3.68	3.74	4.02	3.90
HpCDD	4.42	2.02	5.24	5.23	3.69	5.17	5.67	5.10	4.35	6.16	5.63	4.66
OCDD	41.9	88.2	70.0	25.8	84.2	33.1	37.7	55.8	52.9	40.5	39.1	44.4
TeCDF	14.4	2.53	5.53	20.7	3.29	13.1	11.0	9.50	11.7	10.6	19.5	11.8
PeCDF	9.36	1.71	5.14	10.5	2.16	8.18	9.65	9.91	8.89	7.28	7.65	11.6
HxCDF	10.3	1.68	4.37	11.9	1.87	9.67	11.6	4.71	6.88	11.0	8.08	8.00
HpCDF	6.50	1.15	3.13	8.84	1.55	8.52	7.09	3.27	4.76	9.22	6.09	5.93
OCDF	2.23	1.23	1.71	4.52	0.83	9.86	4.57	1.98	3.16	5.15	3.46	4.40

Table 4. PCDD/Fs homologue distribution in soil of 2009, %.

2010	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
TeCDD	5.85	0.53	1.79	3.61	0.46	2.21	4.17	2.26	1.65	3.05	1.72	2.79
PeCDD	5.53	0.66	2.06	5.73	0.51	4.33	4.96	3.06	2.61	6.24	2.71	1.75
HxCDD	7.45	1.15	3.73	6.40	0.94	4.89	4.30	4.00	3.64	5.71	2.19	6.02
HpCDD	4.60	2.52	5.30	5.05	3.19	6.16	5.47	5.28	4.45	5.41	2.92	5.14
OCDD	14.2	83.4	56.8	21.7	78.8	49.7	34.9	47.7	52.8	32.1	20.8	29.4
TeCDF	16.9	3.06	9.50	17.9	11.3	9.35	14.6	7.86	13.1	15.9	10.8	20.3
PeCDF	14.1	2.35	6.26	12.8	1.37	6.45	8.24	6.06	6.99	8.01	2.71	12.5
HxCDF	15.5	2.37	6.92	13.5	1.40	7.20	9.25	8.17	5.75	8.88	5.65	9.26
HpCDF	11.4	1.86	5.33	9.01	1.20	6.37	8.57	11.7	5.54	9.20	13.6	8.68
OCDF	4.49	2.16	2.26	4.25	0.82	3.38	5.62	3.84	3.44	5.58	36.9	4.21

Table 5. PCDD/Fs homologue distribution in soil of 2010, %.

	TeCDD	PeCDD	HxCDD	HpCDD	OCDD	TeCDF	PeCDF	HxCDF	HpCDF	OCDF
Fly ash	3.57	6.76	10.76	7.19	3.39	18.48	11.39	20.51	14.71	3.24
Stack gas	3.02	6.99	5.44	3.93	2.31	20.33	17.20	23.64	13.40	3.73

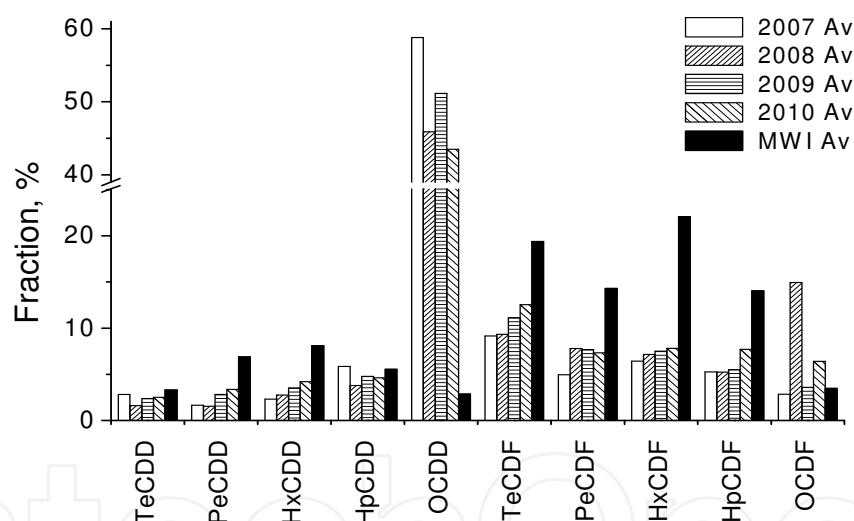Table 6. PCDD/Fs homologue distribution of fly ash and stack gas, %.

Fig. 7. PCDD/Fs Homologue pattern of soil and MWI samples (Av, Average).

Principal component analysis (PCA) is used to estimate the similarity and difference of homologue pattern between soil and the presumed source (MWI), as shown in Fig.8. Accumulation information of component 1 and component 2 is up to 77.98%, means these two components can well represent the total information of all samples. Component 1 mainly depends on OCDD, HxCDF and HxDD, as well as component 2 is related to OCDF and HpCDD. The sites of fly ash and stack gas locate on the right of the PCA score plot, separates from soil samples, which indicates a clear difference between MWI emission and soils in the homologue distribution. Overall, 2007 survey soils are mainly located top left, 2008 soils are mainly in bottom, 2009 and 2010 year soils are mainly in the centre. The groups of each year illuminate homologue patterns in soil change with time, and show a close relation in the soils collected 2009 and 2010. Considering the average distance between

each year soil group and fly ash (stack gas), soils points move closer to fly ash and stack gas with the time, especially S1 and S4 of 2010 year. It demonstrates there is a possible influence of the MWI in neighboring soil that accumulates with year's past. By the way, the fly ash and stack gas samples can not completely display MWI characteristic emission because PCDD/Fs emissions change with different operation parameters. And other combustion process like open burning, firewood usage, and vehicle might release similar PCDD/Fs. In addition, since fly ash is a major output of PCDD/Fs in incinerators (over 50%) (UNEP Chemicals, 2005; Huang & Buekens, 1995), a good and scientific collection and storage of fly ash must be conducted, to avoid leaking and diffusing into the surrounding environment.
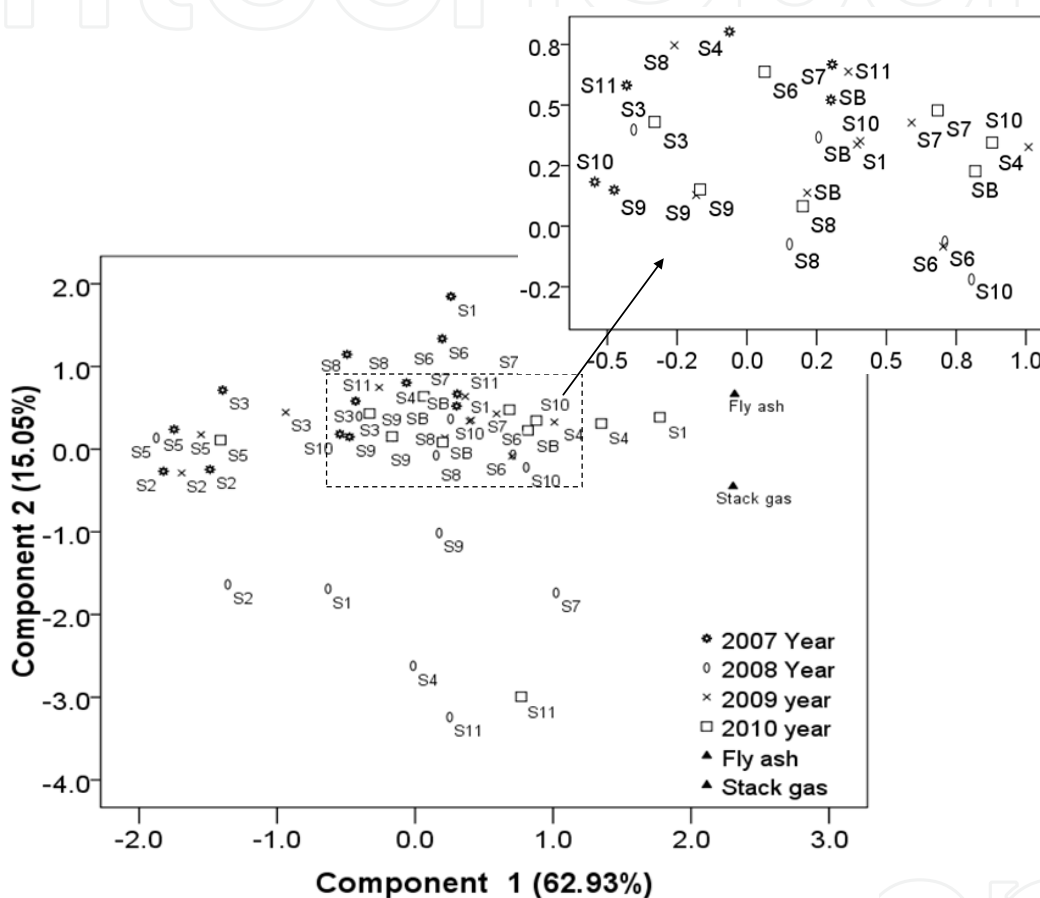
Fig. 8. PCA plot of PCDD/Fs homologue.

3.4 Analysis of HxCDFs isomer profile

PCDD/Fs from Tetra- to Octa-chlorination have ten homologues with different molecular structure and different substituted chlorines, and these compounds have different chemical and biological properties. PCDD/Fs are emitted from source, deposited on earth surface, distributed and decomposed in soil and organism, lot different activities would happen in this process, which deteriorate the relation of soil and source in PCDD/Fs homologue pattern. In order to minimize these possible changes, further analysis focuses on isomer profile of the same homologue. The isomer pattern is expressed as the relative percentage of an isomer with each homologue, which is useful for source identification to compensate for homologue-dependent difference (Ogura et al., 2001; Xu et al., 2008). HxCDF is the

dominant homologue in MWI samples (Table 6), so HxCDF is chose to investigate the isomer profile. Table 7 to 10 are HxCDFs isomer distribution in soil samples, stack gas and fly ash, respectively. There are 16 isomers of HxCDF besides 4 toxic species whose 2,3,7,8 position are occupied by chlorine atom. 124678-HxCDF is the same peak with 134678-HXCDF in gas-chromatographic elution, 123679-HxCDF is also the same peak with 123469-HxCDF, so these two isomers are not assigned; meanwhile, 123489-HxCDF is difficultly separated from 123789-HCDF, so 123489-HxCDF is not assigned too. Fig.9 shows the average of HxCDF isomer pattern in different surveys, the dominated species is 134678-HxCDF, as well as, 123467-HxCDF, 123478-HxCDF and 123678-HxCDF. The average isomer profile among soil and MWI emission (Fig.9) is more similar than the average homologue pattern (Fig.7).

Position	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
123468	6.06	7.43	7.97	7.75	9.64	11.71	9.60	10.7	5.95	9.14	11.08	8.02
134678	44.0	21.62	33.2	32.43	37.0	18.8	28.1	33.3	21.9	32.8	9.79	34.4
134679	ND	ND	1.98	3.91	1.23	0.90	ND	ND	ND	ND	7.75	ND
124679	7.88	2.16	ND	ND	7.04	6.50	5.83	1.26	2.22	6.04	ND	5.09
124689	0.79	1.35	ND	ND	ND	1.67	ND	1.93	ND	ND	ND	0.16
123467	7.37	7.92	ND	9.56	6.22	17.6	12.9	10.2	8.58	11.0	15.3	16.2
123478	4.28	32.5	13.0	15.7	11.8	ND	13.4	9.48	30.2	10.9	19.5	15.6
123678	5.64	13.3	14.1	15.1	10.5	10.5	9.89	5.30	11.6	10.2	9.43	ND
123479	ND	ND	ND	ND	ND	8.37	ND	ND	ND	ND	ND	ND
123469	ND	ND	ND	ND	ND	ND	ND	5.92	ND	ND	ND	ND
123689	8.33	5.98	ND	2.12	5.90	7.31	6.27	5.92	5.59	ND	9.32	5.59
234678	9.54	ND	9.36	11.1	8.21	5.06	8.67	8.15	10.6	9.08	17.3	7.91
123789	6.09	7.67	20.3	2.38	2.48	11.6	5.34	7.89	3.38	10.9	ND	7.07

Table 7. HxCDF isomer distribution of 2007 year soil, %.

Position	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
123468	11.8	7.30	10.5	13.6	11.0	11.0	11.5	10.9	8.66	9.94	10.4	13.1
134678	29.5	20.8	26.6	33.3	35.3	26.9	29.1	28.9	27.7	24.9	26.3	30.3
134679	ND	0.96	1.73	6.54	2.10	1.87	1.97	1.66	1.08	1.46	2.20	2.79
124679	6.53	3.37	4.38	6.27	ND	5.48	4.06	4.07	4.18	2.19	4.23	3.42
124689	1.56	1.74	2.03	ND	2.73	1.61	0.39	0.46	2.05	2.03	3.51	1.18
123467	12.1	9.33	12.8	13.8	4.99	12.6	11.9	13.2	9.66	14.8	9.44	11.8
123478	11.2	22.4	7.16	ND	8.75	10.7	13.4	13.0	4.23	13.1	9.48	13.5
123678	11.0	11.2	10.5	11.3	9.89	10.8	10.9	11.5	8.63	9.42	10.3	11.0
123479	2.89	2.97	3.56	ND	6.10	1.98	3.27	3.34	5.97	4.75	3.58	2.78
123469	1.23	1.55	1.44	2.61	ND	1.51	1.19	1.53	2.55	0.85	0.61	1.42
123689	2.97	4.17	5.00	ND	ND	4.31	3.48	1.06	7.71	4.42	4.35	1.95
234678	6.53	9.52	7.32	12.6	12.7	6.74	5.67	6.61	12.0	9.20	10.4	4.41
123789	2.78	4.68	6.98	ND	6.50	4.58	3.29	3.83	5.58	2.97	5.21	2.47

Table 8. HxCDF isomer distribution of 2008 year soil, %.

Position	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB
123468	10.9	9.80	9.35	9.70	7.05	9.68	10.1	8.67	8.07	8.76	9.40	8.73
134678	27.2	24.6	26.1	28.0	27.4	24.3	26.1	22.8	26.8	26.3	27.3	24.5
134679	2.14	1.39	1.82	1.29	ND	1.16	1.61	ND	1.12	1.60	0.77	1.54
124679	4.48	3.89	3.94	3.82	5.47	5.39	5.53	7.98	4.61	3.11	5.45	4.08
124689	2.00	1.83	0.41	1.25	1.54	3.13	1.86	5.71	1.70	1.63	2.19	1.36
123467	12.1	11.3	11.6	11.8	12.2	13.8	13.2	13.5	13.5	13.2	14.7	12.5
123478	9.74	15.7	8.44	9.90	9.50	8.97	8.32	10.0	8.47	9.91	7.29	18.7
123678	9.78	10.6	9.86	10.3	9.49	8.35	10.6	10.1	9.49	10.7	8.40	10.6
123479	1.99	2.50	2.78	2.94	2.85	2.68	2.24	4.31	5.46	4.90	3.90	ND
123469	2.12	1.38	1.23	1.58	1.57	2.65	1.39	1.14	1.10	1.77	1.67	0.96
123689	3.13	4.25	4.68	5.36	7.52	4.61	5.54	4.89	8.34	5.44	6.11	4.72
234678	10.6	8.56	9.08	10.2	10.4	10.2	10.1	7.20	7.92	8.84	8.65	7.96
123789	3.74	4.19	10.7	3.85	5.00	5.07	3.49	3.65	3.35	3.89	4.11	4.40

Table 9. HxCDF isomer distribution of 2009 year soil, %.

Position	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	SB	Ash	Gas
123468	11.4	9.34	9.76	10.7	8.78	8.95	10.1	7.61	8.17	9.36	5.99	10.3	8.62	9.83
134678	26.5	25.1	26.1	28.7	25.4	25.5	27.1	23.6	24.5	25.7	18.6	28.3	20.2	29.9
134679	1.66	1.45	1.03	1.90	1.56	1.63	ND	1.06	1.65	1.80	ND	2.14	1.59	1.88
124679	3.39	3.50	2.96	3.88	4.07	2.23	3.12	2.74	5.82	4.00	2.94	4.45	3.28	3.32
124689	1.83	1.87	1.30	1.85	1.70	0.67	1.22	1.55	ND	ND	1.34	ND	2.12	1.56
123467	13.4	11.1	13.3	10.2	11.8	11.8	13.7	13.5	10.8	12.4	33.8	18.2	13.4	10.4
123478	10.4	14.9	9.72	9.70	12.3	11.8	9.95	16.3	11.9	12.0	8.78	ND	14.0	9.66
123678	10.5	11.2	11.1	10.2	10.3	10.8	11.5	11.4	11.2	10.5	14.9	12.5	13.2	11.2
123479	2.14	2.22	2.37	2.73	2.44	2.70	2.96	2.33	4.20	3.88	2.60	2.74	1.52	1.27
123469	2.02	1.74	1.88	2.04	1.44	1.45	ND	1.14	1.01	1.75	1.58	1.27	2.73	1.94
123689	2.85	3.74	3.66	4.23	5.65	6.28	5.01	3.53	7.68	5.36	3.87	2.67	2.49	3.07
234678	11.3	9.86	10.8	10.5	10.5	9.77	12.0	10.6	9.84	9.19	5.67	14.5	13.5	13.0
123789	2.62	3.96	6.03	3.28	4.08	6.46	3.34	4.60	3.16	4.01	ND	3.09	3.33	2.97

Table 10. HxCDF isomer distribution of 2010 year soil, fly ash and stack gas of MWI, %.
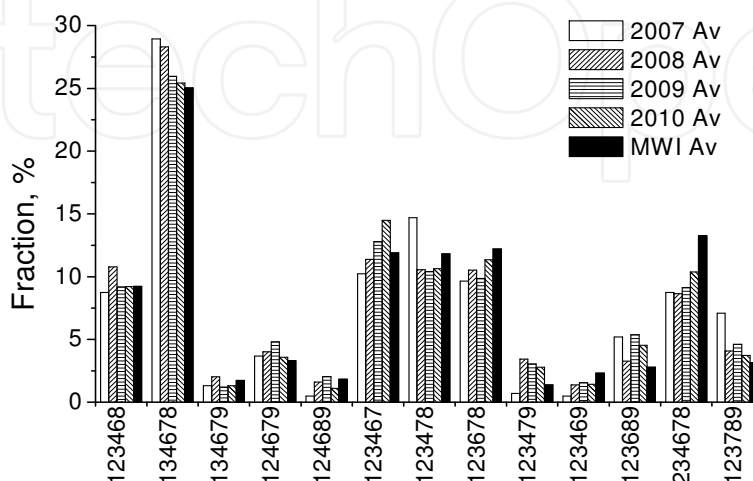

Fig. 9. HxCDF isomer pattern of soil and MWI samples (Av, Average).

The PCA result of HxCDFs isomer profile is shown in Fig.10. Two principal components are extracted from the analyzed 13 isomers. Although component 1 and component 2 can only explain 32.72% of samples' information, while it still can give some useful information for investigating the relation among soils and MWI emission by PCA of isomer profile. In Fig.10, it is observed that 2007 soil spots locate in a large scale, apart from each other, and far away fly ash and stack gas, which means significant difference between 2007 soils and MWI emission. Other year soils have slight trend of assemblage together, meanwhile, become closer to the location of fly ash and stack gas. The points of 2008 and 2010 survey soils are closer to MWI than the sites of 2009 soils, and the group of 2010 survey soils has comparatively closest relation with MWI emission in the PCA plot. This is in line with the variation of PCDD/Fs concentration, and the HxCDF isomer profile also become more likely with MWI emission with year's past.
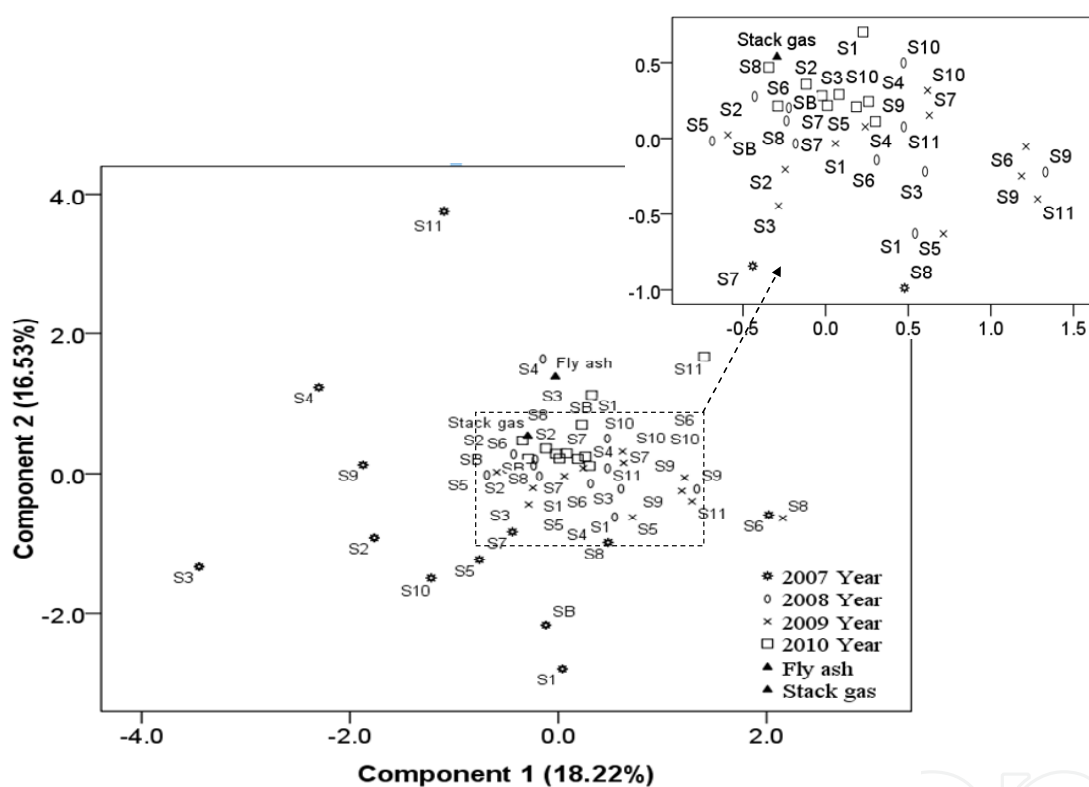
Fig. 10. PCA plot of HxCDF isomers distribution.

4. Conclusion and future work

In the present study, it was observed that dioxin level varied in the analyzed four years, generally, the average level increased after this MWI operation started, as long as just a slight increase in the background samples. The most significant variation is detected in the sites close to this plant, and accounting for the relation of variation and distance away the stack, a limited region near MWI (approx. 500 m) is assumed to be under the influence of PCDD/Fs emission from this MWI. By the PCA of PCDD/Fs homologue pattern and HxCDF isomer profile, PCDD/Fs characteristic distribution in soil became more and more similar with the character of MWI emission. The present PCDD/Fs concentration in this region is in the normal level by the comparison with other studies over the worldwide. In

China, the monitor of dioxin emission for every plant by the environment protection bureau is just one time per year (three samples for a time), so that the information of daily emission is unknown. Some incinerators would release higher concentration of pollutants, which would cause the pollution in the vicinity of plants. Thus, a comprehensive supervised system and more stringent emission limit standard should be established. This tracking monitoring study will be continued in the future years, and the air samples also would be collected and determined to better clarify the environmental impact of waste incinerators. At present, the baseline survey of PCDD/Fs in vicinity soil must be done and noted before the operation of new incinerators according to latest Chinese regulations. The baseline will be used to the comparison with the vale of PCDD/Fs in soil years later, which is basic method to assess the environmental impact of plant.

5. Acknowledgement

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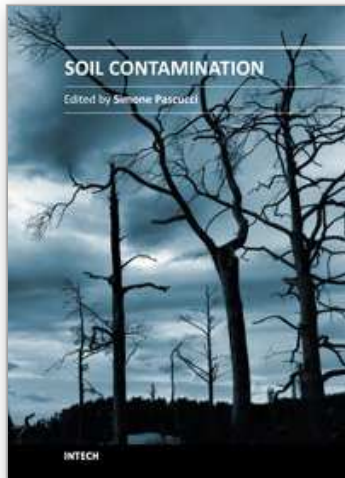
Soil Contamination

Edited by MSc Simone Pascucci

Soil contamination has severely increased over the last decades, mainly due to petroleum hydrocarbons, solvents, pesticides, lead and other heavy metals from industrial wastes and human activities. The critical point regarding contaminated soil monitoring is the intrinsic difficulty in defining fixed monitoring variables and indicators as the establishment of any a priori criterion and threshold for soil quality can be still considered subjective. This book is organized into eight chapters and presents the state-of-the art and new research highlights in the context of contaminated soil monitoring and remediation strategies, including examples from South America, Europe and Asia. The chapters deal with the following topics: - monitoring of dioxin, furan, hydrocarbons and heavy metals level in soils - bioindicators and biomarkers for the assessment of soil toxicity - use of reflectance spectroscopy for soil contaminants and waste material detection - remediation technologies and strategies.

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