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Characterization of dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the atmosphere of different workplaces of a sinter plant

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Abstract

This study investigated concentrations, contents and congener profiles of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the workplaces of a sintering plant. Air samples were collected from the charging zone, the sintering zones of the sintering grate and the rough roll shredder, and the control room. The charging zone was found to have a higher total suspended particulate (TSP) but lower PCDD/F concentrations ($=4969 \mu\text{g nm}^{-3}$ and 7.61 pg nm^{-3} , respectively) than the sintering zone ($=1422\text{--}1448 \mu\text{g nm}^{-3}$ and $19.16\text{--}23.17 \text{ pg nm}^{-3}$, respectively). The TSP and PCDD/F concentrations were lowest in the control room ($=98 \mu\text{g nm}^{-3}$ and 1.75 pg nm^{-3} , respectively). However, the above concentrations were within the range of a typical urban-industrial area. Quite similar PCDD/F contents were found in particles in the sintering zones and control room ($=11.72\text{--}14.30$ and 15.85 ng g^{-1} , respectively) suggesting that the sintering zone and the control room are very similar. In both charging and sintering zones, 2,3,4,7,8-PeCDF contributed 40–60% to the total I-TEQ in both the gas phase and particle phase. However, in control room, both congeners 2,3,4,7,8-PeCDF and 2,3,7,8-TCDD were the main contributors; the later contributed 30% of the total I-TEQ in gas phase. In control room, the contribution of 2,3,7,8-TCDD to the total PCDD/F concentration in the gas phase greatly exceeded that in particle phase ($=5.5\%$ and 0.8% , respectively). Therefore, solutions must be sought to avoid the transfer of not only the particle-phase but also the gas-phase PCDD/Fs from the sintering zone to the control room in the future.

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Keywords: PCDD/Fs; Sinter plant; Content; Congener profile

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1. Introduction

Iron and steel are important in construction and other industries. The manufacture of iron and steel involves a series of closely linked steps, including the preparation of raw materials (pelletizing and sintering), iron-making, steel-making and finishing processes. During sintering, iron ore fines, iron-bearing recovery materials (such as iron-bearing dusts and slag) and fluxes (lime or dolomite) are thoroughly mixed with ~5% of a finely divided fuel, such as coke breeze or anthracite, and are deposited on a traveling grate. The traveling grate resembles the endless loop of a conveyor belt, forming a shallow trough with small holes in the bottom. The bed of materials deposited on the grate is ignited after an ignition burner fired with natural gas and air. Next, air is pulled down through the bed as the grate moves slowly toward the discharge end. As the coke fines burn in the bed, the generated heat sinters/or fuses the fine particles. The temperature of the bed reaches approximately 1300 to 1500 °C. Average production rates of 20–40 metric tons $\text{m}^{-2} \text{d}^{-1}$ of the grate area are expected, depending on the characteristics of the ore materials and the sintering conditions (U.S. EPA, 2001). Typical operating conditions for the sintering of iron ore have been described in more detail elsewhere (Cieplik et al., 2003; Wang et al., 2003a).

Iron ore sintering is known to be major sources of airborne emissions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) (Lahl, 1993, 1994). Numerous studies have demonstrated that PCDD/F formation in the sintering process was most likely to occur through a *de novo* synthesis from carbon sources and inorganic chloride (Huang and Buekens, 1996; Fisher et al., 1998; Buekens et al., 2001). Emissions of PCDD/Fs to the atmosphere from the sintering of iron ore have been studied in several countries. Sinter plants in Japan accounted for 3.2% of total dioxin emissions (Japan Ministry of the Environment, 2002); those in Europe accounted for 19.6% of such emissions (Quaß et al., 2000). In the UK, the percentage contribution of sinter plants to the total dioxin inventory raised from 3–7% to 11–34% between 1995 and 1998 (Alcock and Jones, 1996; Douben, 1997; Anderson and Fisher, 2002), because other sources such as incineration have declined over this period. In Taiwan, Wang (2003) reported that

sinter plants became the most important PCDD/F emission sources (59%) in the area in 2002, because stricter regulations ($0.1 \text{ ng I-TEQ nm}^{-3}$) have applied to municipal waste incinerators constructed after 2001.

The most recent study in Taiwan focused on the PCDD/F emissions from the stack flue gas of various sinter plants and their impact on the surrounding environment (Wang et al., 2003b). Notably, however, PCDD/F fugitives can also be released into the atmosphere of the workplace during the preheating of feed materials, sinter cooling, and sinter discharging. To the best of the authors' knowledge, this issue has never been studied. In two works on municipal waste incinerators, although no air sample was collected, significant increases in the PCDD/F concentrations in workers' serum samples were found (Kumagai et al., 2000, 2002). Given the high PCDD/F emissions from the sintering process, these data reveal the need to evaluate PCDD/F concentrations in various workplaces in sinter plants.

In this work, static air was sampled from three workplaces during sintering process. Workers spend most of their time in a control room during the sintering, so PCDD/F concentrations in the control room were also determined for comparison. The data can help to evaluate the effect of PCDD/F emissions from the sintering process on the workplace atmosphere, and can serve as a basis for further studies of workers' exposure to dioxins.

2. Materials and methods

One sinter plant in southern Taiwan, with an electrostatic precipitator (ESP) and a selective catalytic reduction (SCR) device for reducing air pollution, was the subject of this study. Three sampling sites located approximately 2 m from the raw material inlet (site A), from the sintering grate (site B) and from the rough roll shredder (site C) were selected, to characterize the PCDD/F concentrations in the workplaces of the sinter plant. Site A represents fugitives from raw materials before sintering, whereas sites B and C represent fugitives from the sintering process. In the selected sinter plant, the air that was introduced into the control room (located at the end of the sintering grate) was directly drawn from the workplace of the

sinter plant but was filtered by an air conditioner. Based on this, air samples were also collected inside the control room (site D) to characterize fugitives transferred from the sintering zone to the control room. Field samples were also taken from the outside ambient environment (~ 1 km) near the selected sinter plant to compare with the results obtained from the various workplace environments.

All air samples were collected using a high-volume PS-1 sampler (Greaseby Anderson, GA). This sampler was equipped with a quartz-fiber filter to collect PCDD/Fs in the particle phase, and then a XAD cartridge was used to collect PCDD/Fs in the gas phase. The above sampling method follows the revised EPA Reference Method T09A. The sampling flow rate was specified at $\sim 0.225 \text{ m}^3 \text{ min}^{-1}$. Each sample was collected continuously on four consecutive days (sampling volume $\sim 1300 \text{ m}^3$). Samples were taken from the control room; the effluent gas from PS-1 sampler was discharged to the outside of the control room to prevent the dilution of the TSP or PCDD/F concentration collected from the control room.

A known quantity of surrogate standard was spiked in the glass cartridge in the laboratory before field sampling was conducted. Analyses of the collected air samples strictly followed the US EPA Reference Method T09A and were performed in the Super Micro Mass Research and Technology Center of the Cheng Shiu University. This center has been certified by the Taiwan EPA to analyze PCDD/Fs and passed the international inter-calibration of PCDD/Fs in fly ash, sediment, mothers' milk, human blood and cod liver. More detailed information regarding this center can be found in the authors' previous work

(Wang et al., 2003b). A high-resolution gas chromatograph (HRGC) coupled with a high-resolution mass spectrometer (HRMS) was used to quantify PCDD/F contents of the samples. The HRGC was a Hewlett Packard 6970 Series gas chromatograph, equipped with a DB-5 (J&W Scientific, CA, USA) fused silica capillary column (60 m, 0.25 mm ID, 0.25 μm film thickness), and splitless injection. The initial oven temperature was 150 $^{\circ}\text{C}$. The temperature was programmed as follows: 150 $^{\circ}\text{C}$, held for 1 min; increased by 30 $^{\circ}\text{C min}^{-1}$ to 220 $^{\circ}\text{C}$, held for 12 min; increased at 1.5 $^{\circ}\text{C min}^{-1}$ to 240 $^{\circ}\text{C}$, held for 5 min; increased at 1.5 $^{\circ}\text{C min}^{-1}$ to 310 $^{\circ}\text{C}$, and held for 20 min. Helium was the carrier gas. The HRMS was a Micromass Autospec Ultima (UK) mass spectrometer with a positive electron impact (EI+) source. The analyzer mode was the selected ion monitoring (SIM) with a resolving power of 10,000. The electron energy was set to 35 eV, and the source temperature was set to 250 $^{\circ}\text{C}$.

3. Results and discussion

3.1. TSP and PCDD/F concentrations in the workplace atmosphere and the ambient environment of the sinter plant

Table 1 shows the average total suspended particle (TSP) and total PCDD/F concentrations (gas phase + particle phase) collected from the atmospheres of the four selected workplaces and the outside ambient environments of the selected sinter plant. In workplace environments, the TSP was highest at site A ($=4969 \mu\text{g nm}^{-3}$), mainly because of the emissions

Table 1

Average TSP and total PCDD/F concentrations collected from the atmospheres of the four selected workplaces and outside the selected sinter plant

Concentration	Workplace atmosphere				Outside ambient environment ($n=3$)
	Site A ($n=2$)	Site B ($n=2$)	Site C ($n=2$)	Site D ($n=2$)	
TSP ($\mu\text{g nm}^{-3}$)	4969	1448	1422	98	40.4
Total PCDD/Fs (pg nm^{-3})	7.61	19.16	23.17	1.75	1.60
PCDDs (pg nm^{-3})	2.50	4.22	4.83	0.54	0.63
PCDFs (pg nm^{-3})	5.11	14.94	18.34	1.21	0.97
Total I-TEQ (pg I-TEQ nm^{-3})	0.55	1.76	2.14	0.16	0.07
PCDDs I-TEQ (pg I-TEQ nm^{-3})	0.08	0.19	0.24	0.04	0.01
PCDFs I-TEQ (pg I-TEQ nm^{-3})	0.47	1.56	1.90	0.12	0.06

from the raw material charging process. The TSP was second highest at sites B and C ($=1448$ and $1422 \mu\text{g nm}^{-3}$, respectively) but was much lower than at site A, perhaps because the strong airflow was pulled down through the bed, reducing TSP emission in the sintering zone. The TSP was lowest at site D ($=98 \mu\text{g nm}^{-3}$). This might be explained either by the location's far away from the site of the process or by the effect of filtration by the air conditioning device installed inside the control room. However, the above value still exceeded that in the ambient environment outside the sinter plant ($=40.4 \mu\text{g nm}^{-3}$). These data reveal that fugitive aerosols in the sinter plant were much more significant than those found in the outside ambient environment.

The average PCDD/F concentrations in the sintering zone (sites B and C $=19.16$ and 23.17 pg nm^{-3} , respectively) were approximately three times higher than that in the discharging area (site A $=7.61 \text{ pg nm}^{-3}$) and ten times higher than that in control room (site D $=1.75 \text{ pg nm}^{-3}$). The relatively low PCDD/F concentration in the control room can again be explained by its being far away from the sintering zone, by the partial isolation of the area by the walls, or by filtration effect by the air conditioning device. Meanwhile, the considerable difference between the high PCDD/F concentrations measured in the sintering zones and the other workplace atmospheres revealed that most of the PCDD/Fs generated by sintering. Additionally, total PCDD/F concentration measured in the control room was similar to that measured in the outside ambient environment ($=1.60 \text{ pg nm}^{-3}$). Wang et al. (2003b) found that total PCDD/F concentrations in ambient air samples collected from a rural area, a residential area, an urban area, an industrial area, and the vicinity of a crematory

were 0.32 , 0.849 , 1.43 , 2.71 and 6.57 pg nm^{-3} , respectively. Therefore, the PCDD/F concentration in the control room of the selected sinter plant is within the range of a typical urban-industrial area, but still much lower than that near a crematory. The average total I-TEQ for samples collected from the workplace atmosphere ($=0.55$, 1.76 , 2.14 , and $0.16 \text{ pg I-TEQ nm}^{-3}$ for site A, B, C, and D, respectively) and from the outside ambient environment ($=0.07 \text{ pg I-TEQ nm}^{-3}$) follow a similar trend.

3.2. PCDD/F contents in gas and particle phases

Table 2 shows the gas-phase and particle-phase PCDD/F concentrations as fractions of total PCDD/F and total I-TEQ concentrations in samples collected from the workplace atmospheres and outside the selected sinter plant. The values show that particle-phase PCDD/Fs (range $=88$ – 93%) were consistently higher than those of gas-phase PCDD/Fs (range $=6.9$ – 12%) in the workplaces. Meanwhile, sites B, C and D exhibited very similar proportions of PCDD/Fs in the gas phase and in the particle phase suggest that the particles in the control room may be similar to those in the sintering zones. Interestingly, the proportion of particle-phase PCDD/Fs in the surrounding ambient environment exceeded those in the workplaces, perhaps because condensation and coagulation were dominant when transporting particles away from the sinter plant and mixing with other pollution.

Table 3 presents the average PCDD/F contents in particles from the workplaces in the sinter plant. The PCDD/F content is obtained by dividing the particle-phase PCDD/F concentration by the particle concentration. In this work, the value in the charging zone ($=1.43 \text{ ng g}^{-1}$ and $0.10 \text{ ng I-TEQ g}^{-1}$) was much

Table 2

Gas-phase and particle-phase PCDD/F concentrations as fractions of total PCDD/F and total I-TEQ concentrations in samples collected from the workplace atmospheres and outside the selected sinter plant

	Site A		Site B		Site C		Site D		Outside environment	
	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)
Total PCDD/Fs	6.9	93.1	11.4	88.6	11.5	88.5	12	88	6.3	93.7
PCDDs	4.2	95.8	4.2	95.8	4.1	95.9	6.6	93.4	0.9	99.1
PCDFs	8.3	91.7	13.5	86.5	13.5	86.5	14.8	85.2	9.7	90.3
Total I-TEQ	12.6	87.4	25.2	74.8	25.7	74.3	29.1	70.9	19.1	80.9
PCDDs I-TEQ	11.5	88.5	23.8	76.2	24.4	75.6	35.6	64.4	12.5	87.5
PCDFs I-TEQ	12.8	87.2	25.4	74.6	25.8	74.2	26.6	73.4	20.1	79.9

Table 3

Average PCDD/F contents in particles of the workplace atmospheres and outside the selected sinter plant

Contents (ng g ⁻¹)	Site A	Site B	Site C	Site D	Outside environment
Total PCDD/Fs	1.43	11.72	14.30	15.85	37.10
PCDDs	0.48	2.79	3.25	5.19	15.58
PCDFs	0.95	8.92	11.04	10.66	21.68
Total I-TEQ	0.10	0.90	1.07	1.19	1.40
PCDDs I-TEQ	0.01	0.10	0.12	0.29	0.22
PCDFs I-TEQ	0.08	0.80	0.95	0.90	1.19

lower than that in the sintering zone (site B=11.72 ng g⁻¹ and 0.9 ng I-TEQ g⁻¹, site C=14.30 ng g⁻¹ and 1.07 ng I-TEQ g⁻¹) and the control room (=15.85 ng g⁻¹ and 1.19 ng I-TEQ g⁻¹). The values were lowest in the charging zone, indicating that the charging materials contained relatively little PCDD/Fs. However, the particles in the sintering zones and the control room have similar PCDD/F contents; therefore, the sintering zone and the control room are probably similar. These results also reveal that, despite the fact that the control room is far from the sintering zones (and certainly partially isolated in an area enclosed with walls), a significant transfer of toxic particles occurs from the sintering area into the control room. Risks to human health are associated with these particles so solutions that involve no transfer of particle-bound PCDD/Fs from the sintering zone to the control room are being sought. However, the values were highest in the surrounding ambient environment (=37.10 ng g⁻¹ and 1.40 ng I-TEQ g⁻¹). In workplace atmospheres, after the raw material has been sintered, the particle-phase PCDD/Fs in the sintering zones declined from 93% to 88% (Table 2). However, in outside surrounding ambient, 94% of the PCDD/Fs were found to be in the particle phase after mixing with other pollution sources in this industrial area. Aerosols are generally classified into three size groups as ultrafine, mid-sized and coarse particles. Ultrafine particles are produced by gas-to-particle conversions during combustion, when hot vapor condenses to produce primary particles, which coagulate. Following rapid condensation and coagulation, mid-sized particles dominate. Coarse particles are typically produced by mechanical attrition, soil erosion and sea spray (Lohmann and Jones, 1998). In this work, gas/particle-phase distributions are considered to be freely

exchangeable during transport away from the source and is considered to be strongly held by the particles in outside ambient.

Additionally, as stated above, when the bed of materials was fired with natural gas and air, air was pulled down through the bed as the burden moved slowly toward the discharge end. This process reveals that the generated coarse particles may not be able to be released into the workplace atmosphere, indicating that the workplace atmosphere contains only fine particles. At this stage, the sizes of particle associated with those released to the workplace are still unknown, and so warrant further investigation.

3.3. PCDD/F congener profiles of sinter plant fugitive emissions

Fig. 1 shows congener profiles of the gas phase, particle phase, and gas+particle phase of the seventeen 2,3,7,8-chlorinated PCDD/Fs detected in the four selected workplace atmospheres and in the outside ambient environment. Fig. 2 presents the congener profiles of I-TEQ in gas, particle, and gas+particle phases. The congener profiles of the particle phase reveal that the four selected workplace atmospheres were quite similar to each other, with more high-chlorinated congeners than low-chlorinated congeners. The congener profiles of the gas+particle phase indicate a similar result, perhaps because particle-phase PCDD/Fs (88–93%) contributed much more than gas-phase PCDD/Fs (7–12%) to the total PCDD/Fs in the four selected workplace atmospheres.

The congener profiles of gas+particle phase detected in the workplace differed considerably from those obtained from the stack flue gas of the sinter plants by Anderson and Fisher (2002) and Wang et al. (2003a). (These were dominated by low-chlorinated PCDFs.) Herein, the congener profiles of gas-phase PCDD/Fs in the workplace atmospheres were similar to those from the stack flue gas obtained by Wang et al. (2003a). This difference may be explained by the fact that most of the particulates that were originally contained in the offgas were removed by the air pollution control devices before they were discharged from the stack. Congener profiles of the gas+particle phase in the outside ambient environment were similar to those obtained by Wang et al. (2003a). How-

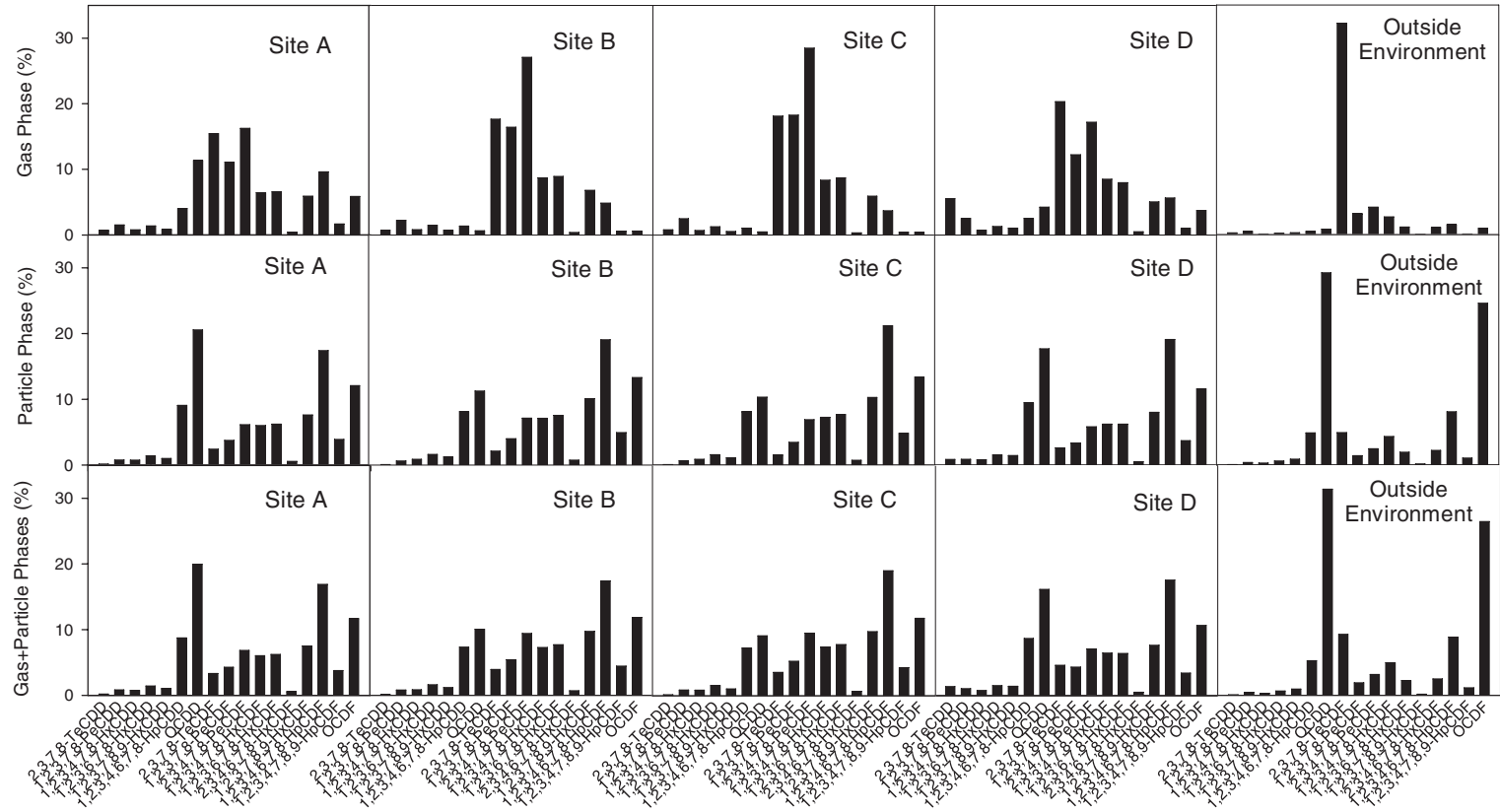


Fig. 1. Congener profiles of the gas phase, particle phase, and gas+particle phase of the seventeen 2,3,7,8-chlorinated PCDD/Fs detected in the four selected workplace atmospheres and in the outside ambient environment.

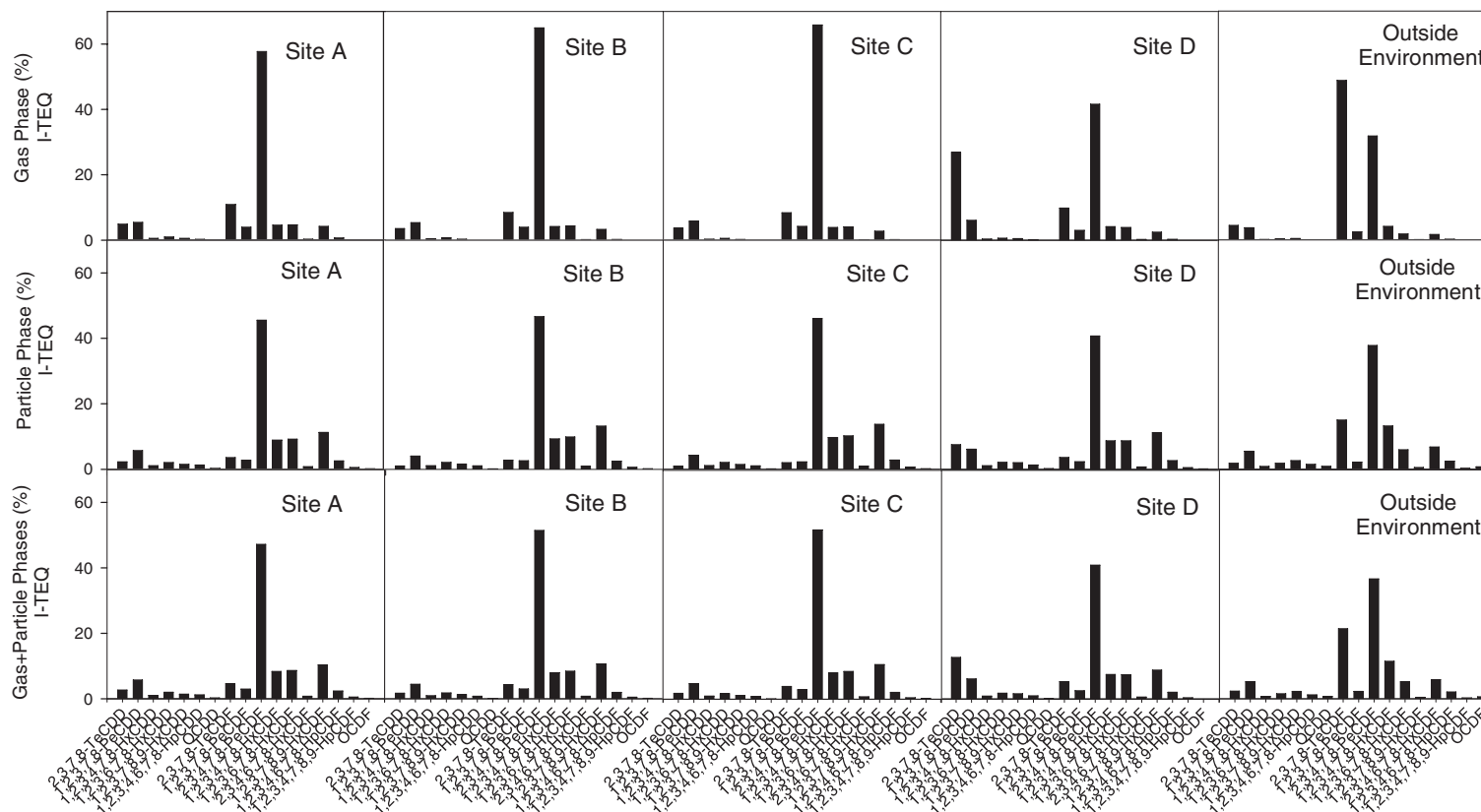


Fig. 2. Congener profiles of the gas phase, particle phase, and gas+particle phase of the seventeen 2,3,7,8-chlorinated I-TEQ detected in the four selected workplace atmospheres and in the outside ambient environment.

ever, these profiles differed from those in the workplaces, perhaps of the transportation away from the sinter plant and the mixing with other pollution in this industrial area. High-chlorinated congeners such as OCDD and OCDF were found to be the dominant congeners.

In this work, the contributions of individual congeners to the total I-TEQ of the particle phase were similar among the four selected workplace atmospheres. In these areas, 2,3,4,7,8-PeCDF contributed approximately 50% of the total I-TEQ in the particle phase. The congener profiles of gas phase in the charging and sintering zones were similar. However, in control room, both 2,3,7,8-TCDD and 2,3,4,7,8-PeCDF were the dominant congeners; in particular, the contribution of 2,3,7,8-TCDD to the total I-TEQ of the gas phase was found to be as high as 30%. In this work, in the control room, the contribution of 2,3,7,8-TCDD to the total PCDD/F concentrations of the gas phase and particle phase were merely 5.5% and 0.8%, respectively. Its contribution to the total I-TEQ was high clearly because of its high toxic equivalent factor (TEF). Since the contribution of 2,3,7,8-TCDD to total PCDD/F concentrations in the gas phase was much higher than that in the particle phase, means of preventing transfer of PCDD/F from the sintering zone to the control room, not only in the particle phase but also in the gas phase, are required. In the outside ambient environment, the contribution of individual congeners to the total I-TEQ in the particle phase were similar to those in the four selected workplace atmospheres, except 2,3,7,8-TCDF. The congener profiles of gas phase I-TEQ in the charging and sintering zones, as well as in the outside ambient environment were similar to each other, except 2,3,7,8-TCDF. 2,3,7,8-TCDF was found to be the dominant congener in both gas and particle phases in the outside ambient environment.

4. Conclusions

The charging zone had higher TSP but much lower PCDD/F concentrations than the sintering zone, indicating that charging materials contained relatively low PCDD/Fs and that most PCDD/Fs were generated by the sintering process. The TSP and PCDD/F concentrations were lowest in the control room, but its

PCDD/F concentration appears to be within the range of a typical urban-industrial area. PCDD/F contents in particles in the sintering zones and the control room are similar, indicating that the sintering zone and the control room are probably of the same nature. The congener profiles of the gas phase, rather than gas+particle phase, detected in the workplace were similar to that obtained from the stack flue gas of sinter plants. In the charging and sintering zones, 2,3,4,7,8-PeCDF was congener that contributes most to the total I-TEQ in both the gas and particle phases. However, in control room, both 2,3,4,7,8-PeCDF and 2,3,7,8-TCDD were the dominant congeners, and the latter contributed 30% of the total I-TEQ of the gas phase. The contribution of 2,3,7,8-TCDD to the total PCDD/F concentrations in gas phase was much higher than that in particle phase of samples collected from the control room. Accordingly, means of preventing the transfer of PCDD/F from the sintering zone to the control room, not only in the particle phase but also the gas phase.

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