



Reduction of Toxic Pollutants Emitted from Heavy-duty Diesel Vehicles by Deploying Diesel Particulate Filters

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ABSTRACT

In this study, the emission characteristics of PCDD/Fs, PCBs and PBDEs from heavy-duty diesel vehicles, and the reduction of these above toxic pollutants by deploying three kinds of diesel particulate filters (DPFs), that is, mobile metal filter plus catalyzed diesel particulate filter (MMF + CDPF), diesel oxidation catalysts (DOC) + CDPF and partial diesel particulate filter (PDPF) were investigated. The tested vehicles were maintained at 60 km/h for 10 minutes on a standard chassis dynamometer. The results show that vehicles with greater mileage had higher PCDD/Fs, PCB and PBDE concentrations in the exhaust. After the tested vehicles were equipped with DPFs, some trials showed the formation of PCDD/Fs and PCBs occurring inside the DPF. This could be due to the de novo and precursor formation resulting from the combined effects of the accumulated particulate in the DPFs, favorable temperature and longer retention time for the exhaust. The PDPF exhibited the largest reduction in these toxic pollutants emitted from HDDVs, which reached 83.9%–95.3% on mass basis, and 54.2%–71.9% on toxicity basis. However, significant differences existed among the trials of these three DPFs, revealing that the influential factors for the reduction of these toxic pollutants could be more complex than those for reduction of particulate matter.

Keywords: Diesel particulate filters; PBDEs; PCDD/Fs; PCBs; Heavy-duty diesel vehicles.

INTRODUCTION

Ambient particulate matter (PM) has an adverse impact on human health and environmental quality. Recent toxicological and epidemiologic studies have shown the adverse effects of PM are strongly associated with particle diameter, and smaller particles, such as PM_{2.5} and ultrafine

particles, are more toxic than larger ones. The exhausts of heavy-duty diesel vehicles have long been recognized as the largest mobile source of ambient PM_{2.5}, and because vehicles are often used in densely populated areas, they have a significant influence on human health.

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) are semi-volatile organic compounds, which are similar to polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs), and polybrominated diphenyl ethers (PBDEs), and these exist in both gas and particle phases in the ambient air and stack flue gases (Lee *et al.*, 1996a; Lee *et al.*, 1996b; Sheu *et al.*, 1996; Lai *et al.*, 2007; Wang and Chang-Chien, 2007; Wang *et al.*, 2008b; Wang *et al.*, 2010f). PCDD/F emissions

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are serious issues in many countries. In the past, many studies focused on the PCDD/F emissions from various sources, such as waste incinerators (Wang *et al.*, 2003b; Wang *et al.*, 2007; Wang and Chang-Chien, 2007; Lin *et al.*, 2010a; Wang *et al.*, 2010d), iron ore sintering (Wang *et al.*, 2003c; Kuo *et al.*, 2011), power plants (Lin *et al.*, 2007; Lin *et al.*, 2010b; Wang *et al.*, 2010e; Wu *et al.*, 2010), boilers (Chen *et al.*, 2011), electric arc furnaces (EAFs) (Lee *et al.*, 2004; Lee *et al.*, 2005; Li *et al.*, 2008; Wang *et al.*, 2010c; Chiu *et al.*, 2011), secondary aluminum smelters (ALS) (Lee *et al.*, 2005; Li *et al.*, 2007), crematories (Wang *et al.*, 2003a), and the open burning of rice straw (Lin *et al.*, 2007) and wood (McNamara *et al.*, 2011). However, mobile sources are almost as important as stationary sources in the contribution to PCDD/F emissions (Chuang *et al.*, 2010a; Chuang *et al.*, 2010b), and the relative contribution fraction of PCDD/Fs from mobile sources has gradually risen in many countries since the execution of more stringent PCDD/F emission standards for stationary sources.

We recently found that PBDEs can form during the combustion processes through similar formation conditions as PCDD/Fs (Wang *et al.*, 2010a; Wang *et al.*, 2010b; Wang *et al.*, 2010c; Artha *et al.*, 2011). The GM PBDE concentrations measured from the exhaust of UGFVs and DFVs were 46.7 ng/Nm³, and 29.1 ng/Nm³, respectively (Wang *et al.*, 2010b), and the highly brominated PBDEs in urban ambient air were contributed by combustion sources, such as vehicles (Wang *et al.*, 2010b, 2011).

Air pollution control devices (APCDs), such as bag filters (Lin *et al.*, 2008; Li *et al.*, 2010; Yamada *et al.*, 2011), electrostatic precipitators (EP) (Ruttanachot *et al.*, 2011), activated carbon injection (Li *et al.*, 2010) and other measures (Lai *et al.*, 2007; Wang *et al.*, 2008a), are used to remove pollutants from stack flue gases of stationary emission sources. Diesel particulate matter has been identified as a toxic air contaminant (Wenger *et al.*, 2008), and diesel particulate filters (DPFs) are widely-used devices which can decrease the PM from diesel engine exhausts by up to 90%. Diesel oxidation catalysts (DOC), which are commonly used in combination with a DPF, have been used to oxidize total hydrocarbon and carbon monoxide to water and carbon dioxide.

Heavy-duty diesel vehicles emit potentially harmful and unregulated toxic pollutants to the atmosphere, such as PAHs, PCDD/Fs, PCBs and PBDEs (Wenger *et al.*, 2008;

Wang *et al.*, 2010b, 2011;). In this study, an analytical method was developed to simultaneously measure several pollutants, including PAHs, PCDD/Fs, PCBs and PBDEs, from one exhaust sample. The emission characteristics of the above toxic pollutants from heavy-duty diesel vehicles were investigated in this study, except for PAHs, which will be discussed in another work. Furthermore, the reduction of toxic pollutants by deploying three kinds of DPF as pollution control devices was evaluated to clarify their removal.

MATERIALS AND METHODS

Vehicles and DPF

The engine volume, mileage, manufacturing year, and the types of DPF of the tested vehicles are listed in Table 1. The manufacturing years of the vehicles ranged from 1997 to 2003, and the mileage were from 73000 to 145000 km. Three types of DPF were deployed in these vehicles, including a mobile metal filter plus catalyzed diesel particulate filter (MMF+CDPF) for vehicles A and B, DOC + CDPF for vehicles C – F, and a partial diesel particulate filter (PDPF) for vehicle G.

Sampling Procedures

The tested vehicles were maintained at 60 km/h for 10 minutes on a standard chassis dynamometer. Exhausts of the heavy-duty diesel vehicles (vehicular tailpipes) were directly sampled with constant flow rate at 30 L/min using U.S. EPA Modified Method 5 sampling trains before and after deploying the DPF as a pollution control device. Gaseous and particulate phase toxic pollutants were collected by XAD-2 resins and quartz fiber filters, respectively. The sampled flue gas volumes were normalized to the dry condition of 760 mmHg and 273 K, and denoted as Nm³.

Analytical Procedures

The quartz fiber filter and XAD-2 resin of each exhaust sample were combined for analysis to represent a whole exhaust sample. Samples were analyzed for sixteen PAHs, seventeen 2,3,7,8-substituted PCDD/F, twelve dioxin-like PCBs and thirty PBDE congeners. PBDE, PCB and PCDD/F analyses were performed following the U.S. EPA Method 1614, Modified Method 1668A, and Modified Method 23, respectively. PAHs, PCBs, PCDD/Fs and

Table 1. Basic information concerning the tested vehicles and DPFs.

Vehicles	Volume (cm ³)	Manufacturing date (year, month)	Mileage (km)	DPF	Methods for filtering particulate
A	7545	2003. 11.	72900	MMF + CDPF	Wall flow
B	7545	2002. 06.	141000		
C	6925	1997. 07.	142000	DOC + CDPF	Wall flow
D	6925	2001. 01.	122000		
E	3907	2001. 04.	96600		
F	7639	1999. 11.	110000		
G	6925	2001. 01.	145000	PDPF	Partial flow

MMF: mobile metal filter; CDPF: catalyzed diesel particulate filter; PDPF: partial diesel particulate filter.

PBDEs internal standards were spiked to the samples before Soxhlet extraction with *n*-hexane, and were used to monitor the extraction and cleanup procedures. After extraction, the extract was then concentrated, silica column cleaned-up and re-concentrated to exactly 1.0 mL for gas chromatography/mass spectrometry (GC/MS) analyses. The retained solution was treated with concentrated sulfuric acid, and this was followed by a series of sample cleanup and fractionation procedures, including a multi-layered silica column, alumina column and activated carbon column. The activated carbon column was used for well-separating the PBDEs and PCDD/Fs due to differences in their planarity and absorption properties, as is the case with separations of mono-ortho PCBs from PCDD/Fs (Choi *et al.*, 2003). The column was sequentially eluted with 25 mL dichloromethane/hexane (40/60, v/v) for PBDEs, followed by 35 mL of toluene for PCDD/Fs. The eluate was concentrated to approximately 1 mL and transferred to a vial. The concentrate was further concentrated to near dryness using a stream of nitrogen. Immediately prior to analysis, 10 μ L of the standard solution for recovery checking was added to the sample extract to minimize the possibility of loss. The detailed analytical procedures for PBDEs and PCDD/Fs are given in our previous works (Wang *et al.*, 2003a; Wang *et al.*, 2010c)

Instrumental Analysis

A high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PBDE and PCDD/F analyses. For PBDEs, the HRGC (Hewlett-Packard 6970 Series gas chromatograph, CA) was equipped with a DB-5HT capillary column (L = 15 m, i.d. = 0.25 mm, film thickness = 0.1 μ m) (J&W Scientific, CA). For PCDD/Fs, the HRGC was equipped with a DB-5MS fused silica capillary column (L = 60 m, i.d. = 0.25 mm, film thickness = 0.25 μ m) (J&W Scientific, CA). The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) was used with a resolving power of 10000. The electron

energy and source temperature were specified at 35 eV and 250°C, respectively. The detailed instrumental analysis parameters of PBDEs and PCDD/Fs are given in our previous works (Wang *et al.*, 2003a; Wang *et al.*, 2010a).

RESULTS AND DISCUSSION

PCDD/F, PCB and PBDE Concentrations in the Vehicular Exhaust before Installing DPF

PCDD/F, PCB and PBDE concentrations in the exhaust of these tested heavy-duty diesel vehicles before and after deploying DPFs as pollution control devices are listed in Tables 2 to 4, respectively. The samples of vehicles D and E are combined into a composite sample in order to prevent the analyst below the detection limit after deploying DPF. Before DPF were installed in these vehicles, the PCDD/F concentrations in the exhausts ranged from 0.983 to 22.3 pg I-TEQ/Nm³. Among these HDDVs, vehicles B, C and G had greater mileage, and their emitted PCDD/F concentrations were also the highest, at 7.93, 7.40 and 22.3 pg I-TEQ/Nm³, respectively. The PCDF/PCDD ratios were all less than 1, indicating that PCDDs dominated the total PCDD/Fs in the exhaust of HDDVs. Huang and Buekens (1995) reviewed research regarding the formation mechanisms of PCDD/Fs, and concluded that de novo synthesis can produce PCDD/Fs with the characteristic of a PCDFs/PCDDs ratio > 1, while precursor formation produces PCDD/Fs with a PCDFs/PCDDs ratio << 1. Consequently, the PCDFs/PCDDs ratio obtained in this work reveals that the PCDD/Fs from HDDVs are produced via precursor formation.

Chuang *et al.* (2010b) found that the mean PCDD/F I-TEQ concentrations in exhaust gases of the six sport utility vehicles (SUVs), six diesel passenger vehicles (DPVs), and three HDDVs, which were tested in accordance with regulated driving cycles, were 54.4 (RSD = 70.3%), 33.7 (88.6%), and 72.4 pg I-TEQ/Nm³ (56.7%), respectively. These reported concentrations were all higher than our observations in this study, revealing that the vehicle testing procedure is an influential factor affecting the PCDD/F I-TEQ concentrations in exhaust gases.

Table 2. PCDD/F concentrations in the exhaust of the tested heavy-duty diesel vehicles before and after deploying DPF as pollution control devices.

PCDD/Fs	A		B		C		D + E		F		G	
	before	after	before	after	before	after	before	after	before	after	before	after
PCDDs	151	146	260	1030	141	43.8	43.0	18.8	19.1	16.7	1720	91.9
PCDFs	83.1	77.3	84.7	315	87.8	35.8	20.9	9.27	9.43	6.72	274	43.9
PCDFs/PCDDs ratio	0.56	0.53	0.33	0.31	0.62	0.82	0.49	0.49	0.49	0.40	0.16	0.48
Total PCDD/Fs (pg/Nm ³)	233	224	345	1340	229	79.7	63.9	28.0	28.5	23.5	1990	136
Total I-TEQ (pg I-TEQ/Nm ³)	6.22	2.95	7.93	34.1	7.40	4.65	2.99	0.680	0.983	0.905	22.3	6.27

Table 3. PCB concentrations in the exhaust of the tested heavy-duty diesel vehicles before and after deploying DPF as pollution control devices.

PCBs	A		B		C		D + E		F		G	
	before	after	before	after	before	after	before	after	before	after	before	after
Total PCBs (pg/Nm ³)	588	802	1310	2400	1810	801	295	69.0	41.2	31.0	7500	1200
Total TEQ (pg WHO-TEQ/Nm ³)	0.909	1.83	1.29	5.35	1.66	0.791	0.249	0.0894	0.0727	0.0821	2.86	1.31

Table 4. PBDE concentrations in the exhaust of the tested heavy-duty diesel vehicles before and after deploying DPF as pollution control devices.

PBDEs	A		B		C		D + E		F		G	
	before	after	before	after	before	after	before	after	before	after	before	after
\sum_{2-8} BDE	6.76	4.13	11.1	7.94	7.49	3.99	1.49	0.750	0.840	0.470	24.2	5.39
\sum_{9-10} BDE	29.8	12.4	181	32.3	23.5	22.0	39.4	7.42	11.2	1.53	361	11.3
Total (ng/Nm ³)	36.6	16.5	192	40.3	31.0	26.0	40.9	8.17	12.0	2.00	385	16.7

The PCB concentrations in the exhausts of the tested heavy-duty diesel vehicles before installing DPF ranged from 0.0727 to 2.86 pg WHO-TEQ/Nm³, while for PBDEs they were from 12.0 to 385 ng/Nm³. Similar to PCDD/Fs, vehicles with greater mileage had higher PCB and PBDE concentrations in their exhausts. The PCB concentrations emitted from vehicles B, C and G were 1.29, 1.66 and 2.86 pg WHO-TEQ/Nm³, while for PBDEs, their concentrations were 192, 31.0 and 385 ng/Nm³.

PCDD/F, PCB and PBDE Congener Profiles in the Vehicular Exhaust before Installing DPF

The PCDD/F, PCB and PBDE congener profiles in the vehicular exhaust before installing DPFs are illustrated in Fig. 1. The exhaust of these tested vehicles contained an abundance of highly chlorinated-substituted congeners, like OCDD, 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF and OCDF, as seen in the results of our previous work (Wang et al., 2010b), and for unleaded gas-fueled vehicles and diesel-fueled vehicles (DFLs) in the USEPA's database for mobile sources (U.S. EPA, 2006). The identical distribution of PCDD/F congeners for gasoline and diesel passenger cars suggests that the difference in fuels and engine types did not result in significant variations in the PCDD/F congener profile.

For the PCB congener profiles in the exhaust, PCB-118, -156, -105 and -167 are the major congeners. For PBDEs, the dominant PBDE congeners are highly brominated-substituted ones, including BDE-209, -208, -207, and -206. Among the low to medium brominated congeners, BDE-47 and BDE-99 are more dominant. Unlike the EAFs and secondary ALSs, there are no raw materials containing PBDEs fed into the combustion systems of HDDVs. Consequently, the relatively higher fractions of highly brominated substituents compared to EAFs and secondary ALSs reveal that the PBDE formation during the combustion process prefers highly brominated congeners.

Reduction of the Toxic Pollutants by DPFs

Reductions of PCDD/Fs, PCBs and PBDEs emitted from HDDVs by deploying three kinds of DPFs as pollution control devices are listed in Table 5. The estimated reductions of PCDD/Fs, PCBs and PBDEs by DPFs are calculated as follows.

$$\text{Reduction efficiency (\%)} = (A - B)/A \times 100\%, \quad (1)$$

where A is the concentration of the toxic pollution emitted from HDDVs before deploying a DPF, and B is the concentration of the toxic pollution after deploying one.

For MMF + CDPF, the reduction in PBDEs (67.9%–80.5%) is higher than that of PCDD/Fs and PCBs, which actually showed a negative reduction, that is, there was more formation of PCDD/Fs and PCBs. This could be due to the de novo and precursor formations of PCDD/Fs and PCBs resulting from the combined effects of the accumulated particulate in the DPFs, favorable temperature and longer retention time for the exhaust.

Among these three types of DPFs, PDPF exhibited the largest reduction in these toxic pollutants emitted from HDDVs, which reached 83.9%–95.3% on mass basis, and 54.2%–71.9% on toxicity basis. Generally speaking, the reduction of these toxic pollutants by DOC + CDPF is higher than by MMF + CDPF. In the trial of vehicles D + E, the reduction of these toxic pollutants ranged from 56.1% (mass of PCDD/Fs) to 80.1% (mass of PBDEs). Wenger et al. (2008) investigated the AhR mediated activity of the exhaust generated by a heavy-duty diesel engine. AhR agonists were quantified using the DR-CALUX reporter gene assay. They found 54–60 ng 2,3,7,8-tetrachlorodibenzo-*p*-dioxin CALUX equivalents (TCDD-CEQs) per m³ of exhaust in unfiltered samples, and 6–16 ng TCDD-CEQ/m³ in DPF treated samples. DPF thus decreased the TCDD-CEQ concentrations by almost 90%.

Significant differences existed between the trials, revealing that the influential factors on the reduction of these toxic pollutants could be more complex than those for the reduction of particulate matter. We speculate that the most influential factors that need to be considered concerning the reduction of these toxic pollutants by DPF are the existence of these in both particulate and gaseous phases, as well as the formation of these pollutants inside the DPF.

The reductions in PBDEs by these three types of DPFs are generally higher than those of PCDD/Fs and PCBs. This could be attributed to the weaker binding energy of C-Br compared to C-Cl (Weber and Kuch, 2003), and the different formation mechanisms of these pollutants.

CONCLUSIONS

Before DPF were installed in these vehicles, the PCDD/F and PCB concentrations in the exhaust of these tested heavy-duty diesel vehicles ranged from 0.983 to 22.3 pg I-TEQ/Nm³, and from 0.0727 to 2.86 pg WHO-TEQ/Nm³, respectively. Vehicles with greater mileage had higher PCDD/Fs, PCB and PBDE concentrations in the exhaust. After the tested vehicles were equipped with DPFs, some trials showed the formation of PCDD/Fs and PCBs

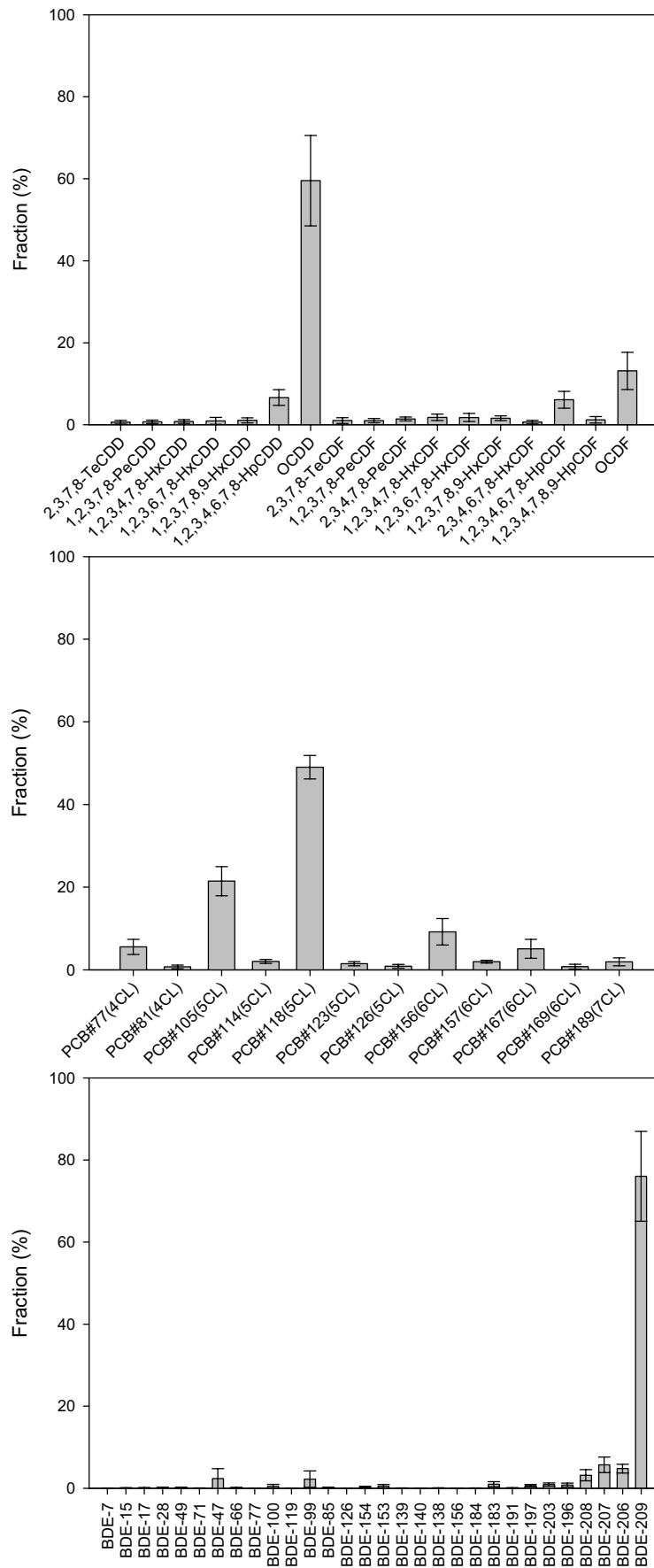


Fig. 1. PCDD/F, PCB and PBDE congener profiles in the vehicular exhaust before installing DPFs.

Table 5. Reductions of PCDD/Fs, PCBs and PBDEs emitted from HDDVs by deploying three kinds of DPFs.

		MMF + CDPF		DOC + CDPF		PDPF	
		A	B	C	D + E	F	G
PCDD/Fs	mass	3.8	−290	65.1	56.1	17.6	93.2
	I-TEQ	52.6	−330	37.2	77.3	7.9	71.9
PCBs	mass	−36.4	−83.9	55.7	76.6	24.9	83.9
	WHO-TEQ	−101	−313	52.4	64.1	−13.0	54.2
PBDEs	mass	67.9	80.5	22.7	80.1	84.3	95.3

occurring inside the DPF, resulting from the de novo and precursor formations. PDPF exhibited the largest reduction in these toxic pollutants emitted from HDDVs among the three DPFs, and the reduction reached 83.9%–95.3% on mass basis, and 54.2%–71.9% on toxicity basis. However, significant differences existed between the trials of the three DPFs. We speculate that the most influential factors that need to be considered concerning the reduction of these toxic pollutants by DPF are their existence in both particulate and gaseous phases, as well as their formation inside the DPF. The reductions of PBDEs by these three types of DPFs are generally higher than those of PCDD/Fs and PCBs. This phenomena could be attributed to the weaker binding energy of C-Br compared to C-Cl, and the different formation mechanisms among these pollutants.

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