



Polybrominated Dibenzo-*p*-dioxins/Furans (PBDD/Fs) and Diphenyl Ethers (PBDEs) in the Indoor and Outdoor of Gymnasiums

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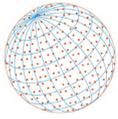
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ABSTRACT

It is still unknown whether the organobromine compounds in the indoor environment of the gyms would affect human health. This study tried to investigate PM_{2.5} and indoor-dust levels of polybrominated dibenzo-*p*-dioxins/furans (PBDD/Fs) and diphenyl ethers (PBDEs) in the gyms and then to further assess the health risks of the gym users and employees who were exposed to these organobromines through inhalation and ingestion. The gyms were to be examined for PBDD/DF/DEs on PM_{2.5} indoors and outdoors and in indoor dust. The results showed that indoor-dust, indoor PM_{2.5}, and outdoor PM_{2.5} levels of PBDD/Fs were 37.8 ± 13.7 pg WHO₂₀₀₅-TEQ g⁻¹ and 0.00650 ± 0.00340 and 0.00469 ± 0.00101 pg WHO₂₀₀₅-TEQ m⁻³, respectively, in the gyms, while for PBDEs, they were 2670 ± 2330 ng g⁻¹ and 9.54 ± 4.68 , and 8.27 ± 3.46 pg m⁻³. The PBDEs levels observed in this study were relatively lower when compared with the current global data of PM_{2.5} and indoor dust levels of PBDEs. The observed PM_{2.5}-bound PBDDFs and PBDEs in the indoor gyms were not related to those in the outdoor gyms, but certain PBDE level was associated



with PBDD/Fs in the indoor dust of the gyms. It suggested that the outdoor emission might not be correlated with the indoor emission, whereas indoor-dust PBDD/Fs might be converted from the pyrolysis of indoor-dust PBDEs. To assess the risks, the non-dietary daily intakes of PBDEs and PBDD/Fs were estimated as $1.61\text{--}2.11 \times 10^{-7}$ and $4.15\text{--}5.42 \times 10^{-13}$ mg kg b.w.⁻¹ day⁻¹, respectively, in the employees and $5.04\text{--}6.58 \times 10^{-8}$ and $1.29\text{--}1.69 \times 10^{-13}$ mg kg b.w.⁻¹ day⁻¹, respectively, in the gym users. The values didn't exceed the critical levels when the gym employees and users were assessed by non-cancer and cancer risks of organobromines. In conclusion, although the gym employees and users inhaled more PM_{2.5} during the exercise, the risks of PBDD/DE/DFs were still under the acceptable levels.

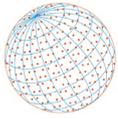
Keywords: PM_{2.5}, Indoor dust, PBDEs and PBDD/Fs, Gym, Health risks

1 INTRODUCTION

Due to the high prevalence of hypertension, hyperglycemia, and hyperlipidemia in Taiwan (Hsu *et al.*, 2021), these chronic diseases are related to metabolic syndromes and are also associated with people with insufficiently physical exercise (Booth *et al.*, 2012). For the improvement of the health of the Taiwanese citizens, the Health Promotion Administration, and the Ministry of Health and Welfare, Taiwan encourage the citizens for sufficient activities which in turn accelerated the exponentially increased numbers of gymnasiums or gyms in Taiwan (Chou and Huang, 2021). The gyms were provided for sports and physical activity in the indoor environment. The indoor air quality (IAQ) in the microenvironments of the gyms was related to human health, especially for physical activities. The exponentially increased numbers of indoor sports facilities and workers were observed due to the dramatical increase of sports enthusiasts in the last decade in Taiwan (Chou and Huang, 2021). Thus, the concerns of both public and government (Environmental Protection Administration, Taiwan (TEPA)) for IAQ of the gyms have been raised. It was observed that the concentration levels of carbon dioxide (CO₂) and particulate matters (PMs) were significantly affected by the time of elevated human occupation, frequent human activity, and poor ventilation in fitness centres or gyms (Castro *et al.*, 2015; Andrade and Dominski, 2018; Slezakova *et al.*, 2018; Salonen *et al.*, 2020). A gym located in a university was known to contain higher levels of PM_{2.5}, PM₁₀, and carbonyl compounds including acrolein and formaldehyde indoors than that outdoors (Alves *et al.*, 2013; Castro *et al.*, 2015). PM_{2.5} levels in gyms or fitness centres were also notably associated with human occupation and frequent activities (Castro *et al.*, 2015). This indicated that the short-term physical activity like cardio activities might have significant effects on the daily inhalation dose and it was also demonstrated that the exercising females possibly inhaled the ultrafine particulate (UFP) 1.2 times higher when compared with the exercising males in the fitness centres (Slezakova *et al.*, 2018).

PM_{2.5} is ubiquitously existed in the indoor and outdoor environment (Bai *et al.*, 2021; Navinya *et al.*, 2020; Yu *et al.*, 2021). Several types of environmental pollutants, including metals, organic compounds, and microbes, are contaminated on PM_{2.5} (Chen *et al.*, 2021; Shen *et al.*, 2020; Siudek and Ruczyńska, 2021; Vega *et al.*, 2021). To raise the public concerns, PM_{2.5} has been demonstrated to cause the adverse effects on animal models and human bodies (Chao *et al.*, 2018; Chung *et al.*, 2020; Zhang *et al.*, 2020). In our previous study (Chao *et al.*, 2016), respirable PBDEs were detectable in the indoor (i.e., library, rail station, and supermarket) and outdoor (i.e., urban, rural, and industrial areas) environments.

Like the other indoor environments in workplaces, a lot of consumer products or electronic equipment are now available in the gyms also. It is still unknown whether the brominated flame retardants (BFRs), such as polybrominated diphenyl ethers (PBDEs) and the dibrominated by-product of BFRs, including polybrominated dibenzo-*p*-dioxins/furans (PBDD/Fs), are widespread in the gym environments. However, it has been demonstrated to have a negative impact on human exposure to PBDEs or PBDD/Fs leading to human health effects such as decreased semen quality, alteration of thyroid and growth hormone homeostasis, delay of neurological behaviour and neurodevelopment, and disruption of menstrual cycles and pregnancy periods (Chao *et al.*, 2014a). Our previous studies indicated that PBDEs or PBDD/Fs were found in several indoor environments including residential



houses, classrooms in elementary schools, offices, medical and dental clinics, and workplaces in electronic and vehicle dismantler factories (Chao *et al.*, 2014c; Shy *et al.*, 2015; Chao *et al.*, 2016; Chou *et al.*, 2016; Gou *et al.*, 2016a, 2016b). PM_{2.5}-bound PBDEs were also observed in public indoor environments like libraries, rail stations, department stores, offices, hospitals, and supermarkets in Taiwan (Chao *et al.*, 2016). Among PBDEs, BDE-209 was the predominated PBDE congener in the Taiwanese microenvironments (Chao *et al.*, 2014c; Shy *et al.*, 2015; Chao *et al.*, 2016; Chou *et al.*, 2016; Gou *et al.*, 2016a, 2016b). Our previous findings (Shy *et al.*, 2015) showed that for the non-dietary daily intake of PBDEs in the indoor environment of the residential houses, indoor air inhalation intake was 1.88 and 1.29% of the total in male and female adults, respectively, and the house dust was the main exposure pathway of the non-dietary PBDEs intake.

To the best of our knowledge, this was the first study addressed the investigation of the POPs in the indoor environment of the gyms especially for PM_{2.5}-bound PBDD/DF/DEs and these organobromines in indoor dust. It was studied that the increment of respiratory ventilation and the inhalation of air through the mouth rather than the nasal, which has particle-filtering apparatus, during the exercise was accelerated in the case of gym users with exposure to the environmental contaminants via inhalation or ingestion. Finally, this study also assessed the health risks of the gym users and employees after they were exposed to PBDD/DF/DEs.

2 METHODS

2.1 Chemicals and Reagents

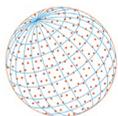
The native mixtures of standards of twelve congeners of PBDD/Fs (2,3,7,8-TeBDF, 1,2,3,7,8-PeBDF, 2,3,4,7,8-PeBDF, 1,2,3,4,7,8-HxBDF, 1,2,3,4,6,7,8-HpBDF, OctBDF, 2,3,7,8-TeBDD, 1,2,3,7,8-PeBDD, 1,2,3,7,8,9-HxBDD, 1,2,3,4,6,7,8-HxBDD, 1,2,3,4,6,7,8-HpBDD, and OctBDD) and fourteen congeners of PBDEs (BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, and 209) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). The ¹³C₁₂-labeled internal standards of organobromine compounds, such as nine ¹³C₁₂-labeled PBDEs (BDE-28, 47, 99, 153, 154, 183, 197, 207, and 209) and PBDD/Fs (2,3,7,8-TeBDF, 1,2,3,7,8-PeBDF, 2,3,4,7,8-PeBDF, 1,2,3,4,7,8-HxBDF, 1,2,3,4,6,7,8-HpBDF, OctBDF, 2,3,7,8-TeBDD, 1,2,3,7,8-PeBDD, 1,2,3,7,8,9-HxBDD, 1,2,3,4,6,7,8-HxBDD, 1,2,3,4,6,7,8-HpBDD and OctBDD) were obtained from Wellington Laboratories (Guelph, Canada). The HPLC grade solvents including n-hexane, toluene, were HPLC purchased from Tedia (Fairfield, USA), Sigma-Aldrich (St. Louis, USA), and Merck (Darmstadt, Germany). Sodium sulfate, alumina oxide, potassium oxalate, and silica gel of the highest-grade purity were obtained from Merck.

2.2 Sampling Areas and Sample Collection

The gyms, which were the commercial gyms such as Les Mills or Equinox with various exercise devices (e.g., treadmill, exercise bike, and fitness equipment), including 4 gyms in Kaohsiung and 2 gyms in Pingtung were in southern Taiwan and the indoor dust, indoor PM_{2.5}, and outdoor PM_{2.5} was collected between November 2018 and October 2019. All the gyms were nearby the road to gather the indoor dust and the proximities of 4 gyms (3 gyms in Kaohsiung and 1 gym in Pingtung) to the main traffic roads were chosen for the collection of samples for indoor dust and indoor PM_{2.5} and outdoor PM_{2.5}.

The samples for indoor dust, indoor PM_{2.5}, and indoor PM_{2.5} were simultaneously collected for two weeks in a gym. Indoor dust on the floor was collected by using a vacuum cleaner (Nilfisk Advance Euroclean UZ934 HEPA canister vacuum cleaner) equipped with an independent filter bag as described previously (Gou *et al.*, 2016a). The indoor dust sample was immediately transported to the laboratory at the National Pingtung University of Science and Technology (NPUST) after the daily dust (5–10 g) was gathered in the gym during the sampling period of two weeks. Prior to the chemical analysis, each dust sample was sieved through 100 mesh (< 0.149 mm) by shaking for 10 min, with the removal of hair, trash, leaf, and possibly interfered materials before all the dusts were collected from a gym was pooled together.

The indoor and outdoor PM_{2.5} was followed by TEPA standard methods (NIEA A205.11C). PM_{2.5} samples were gathered on quartz fibre filters under the pre-heated condition at 600°C for 2 h prior to sampling. The PM_{2.5} filter paper was conditioned in an electronic desiccator at the



temperature of 20–23°C and humidity of 30–40% and weighed by a balance with an accuracy of 0.1 mg before and after the collection of PM_{2.5} samples. The indoor PM sampler, such as Model 200-Personal Environmental Monitor™ (PEM™) for PM_{2.5} (2.5 μm) (MSP Corporation, Minnesota, USA) was used to collect PM_{2.5} indoors in the gyms based on our previous report with minor modification (Chao *et al.*, 2016). After two-week indoor air sampling with the flow rate of 5.00 L min⁻¹ (7.2 m³ day⁻¹ and 100.8 m³ for two weeks), the PM_{2.5} samples were pooled together from the four continuous 3-day sampling samples by considering the collection of sufficient PM_{2.5}-bound PBDD/DF/DEs for further chemical analysis. Collection of the outdoor PM_{2.5} referenced by our previous study was done by using a high-volume air sampler, such as SIBATA HV-1000R (Saitama, Japan) following U.S. EPA Reference Method TO9A (Chung *et al.*, 2019). The outdoor PM_{2.5} in the gyms was gathered for 48 h with a flow rate of 800 L min⁻¹ for a triplicate repeat during the two-week sampling and three outdoor PM_{2.5} samplers were pooled for the further chemical analysis. The PM_{2.5} sample to be conditioned in an electronic desiccator after the sampling was immediately transferred into the lab at NPUST.

2.3 Extraction and Clean-up

Soxhlet extraction and multi-clean-up processes, including a multi-layered silica column, alumina column and activated carbon column, were followed based on the previous study (Wu *et al.*, 2014) and our previous report (Chao *et al.*, 2016; Gou *et al.*, 2016a) with minor modification. In brief, the isotopes and surrogate standards were spiked in the PM_{2.5} filter paper for estimating the organobromines loss during the aerosol sampling process before PM_{2.5} sampling indoors and outdoors. To understand the PBDD/DFs and PBDEs loss in the extraction and clean-up procedures, the isotope internal standards were spiked into dust and PM_{2.5} samples were mixed well with toluene before extraction. PM_{2.5} filter samples and 20 g of indoor dust were extracted with 250 mL of toluene by the Soxhlet extractor for 24 h before evaporation. The elute was concentrated to 2 mL for the following multi-clean-up procedures to be eluted by 15 mL hexane, 25 mL dichloromethane/hexane (1/24, v/v), 5 mL toluene/methanol/ethyl acetate/hexane (1/1/2/16, v/v/v/v), and 40 mL toluene. The final elute was concentrated to approximately 1 mL in a vial. The elute was evaporated to near dryness by a gentle stream of gaseous nitrogen to quantity the final volume of 0.2 mL.

2.4 Chemical Analysis of PBDEs and PBDD/DFs

The analytical method used in this study was previously described (Chao *et al.*, 2014b; Gou *et al.*, 2016a). The congeners of 14 PBDEs and 12 PBDD/DFs were analyzed by a high-resolution gas chromatograph (Hewlett–Packard 6970 Series gas, CA) equipped with a high-resolution mass spectrometer (Micromass Autospec Ultima, Manchester, UK) (HRGC/HRMS) installed with a DB-5HT silica capillary column (L = 60, i.d. = 0.25 mm, film thickness = 0.1 μm) (J&W Scientific, CA) and a splitless injector.

The analytical assurance and quality control (QA/QC) of the analytical method used in this study were referenced by U.S. EPA method 1614 and TEPA NIEA M803.00B to be briefly shown as follows. The blanks including field and lab blanks (i.e., solvent or glassware blanks) were regularly tested in each batch of the analyses. Recoveries of spiked blanks, prelabelled internal standards, surrogate, and precision and recovery (PAR) of all the relevant standards were within the acceptable criteria (70–130%). The limits of detection (LODs) were defined as the ratio of signal to noise (S/N) over 3 and method detection limits (MDLs) were obtained from LODs to be calculated as 2.5–5 folds of the estimated LODs. The limit of quantification (LOQ) was defined as the ratio of S/N over 10. MDLs values of PBDEs and PBDD/Fs ranged from 4.33–290 pg g⁻¹ and 0.0310–25.6 pg g⁻¹, respectively.

2.5 Risk Assessment and Statistical Analysis

The risk assessments of the gym users and professional staff through a non-dietary pathway were referenced by our previous report with minor modifications (Chou *et al.*, 2016; Gou *et al.*, 2016a; Xu *et al.*, 2016; Zhang *et al.*, 2020). Non-dietary daily intakes of PBDEs and PBDD/Fs in the gyms covered the pathways of inhalation from PM_{2.5} and ingestion from indoor dust. The calculation is done using the following equation; DI (mg kg b.w.⁻¹ day⁻¹) = (C × IEF × IR × AB)/(BW) when DI, C,

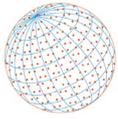


Table 1. The parameters of risk assessments in this study^{a,b}.

	Age ^c (years)	Body Weight ^c (kg)	IR for inhalation ^c		IR for ingestion ^d	AT ^e
			Inactive (m ³ day ⁻¹)	Active (m ³ day ⁻¹)	Indoor dust (mg)	(years)
Male					30	78.1
	20–24	68.57	9.74	18.42		
	25–34	71.38	9.56	20.01		
	35–54	71.72	9.58	16.57		
	54–64	67.19	9.29	16.81		
Female					30	84.7
	20–24	52.46	7.14	11.86		
	25–34	58.61	7.47	11.79		
	35–54	59.38	7.50	13.72		
	54–64	61.07	7.58	11.54		

^a $DI_{dust} (ng\ kg\ b.w.^{-1}\ day^{-1}) = (C_{dust} \times IEF \times IR_{indoor\ dust} \times AB_{intestine\ tract}) / (BW)$ whereas $AB_{intestine\ tract}$: triBDEs to nonaBDEs = 0.508, BDE-209 = 0.630, and PBDD/Fs = 0.950 and $DI_{PM_{2.5}} (ng\ kg\ b.w.^{-1}\ day^{-1}) = (C_{PM_{2.5}} \times IEF \times IR_{PM_{2.5}} \times AB_{alveoli}) / (BW)$ whereas $AB_{alveoli} = 1.00$.

^b The IEF was determined as 0.104 (2.5 hours) for the gym users and 0.333 (8 hours) for the gym staff, respectively, based on “The report from surveillance of gym industry in 2021 Trends & Insights” written by [Chou and Huang \(2021\)](#).

^c The risk factors were referenced by the report of “Compilation of Exposure Factors” by [Taiwanese DOH \(2008\)](#).

^d The indoor dust daily intake was followed by the report of “Child-specific Exposure Factors Handbook” by [U.S. EPA \(2008\)](#).

^e AT was from Ministry of The Interior, Taiwan for 2021 ([Ministry of The Interior, 2022](#)).

IEF, IR, AB, BW were defined as daily intake, concentration (PM_{2.5} or dust), indoor exposure time (gym user: 0.104 for 2.5 hr day⁻¹ and staff: 0.333 for 8 hr day⁻¹), intake rate (inhalation of PM_{2.5} or ingestion of indoor dust), absorption rate of PBDEs (ingestion: triBDEs to nonaBDEs = 0.508 and BDE-209 = 0.630 and inhalation: PBDEs = 1.000) or PBDD/Fs (ingestion = 0.950 and inhalation = 1.000), body weight (Table 1). DI was a summary of DI_{PM_{2.5}} and DI_{dust}. The non-cancer and cancer risks were assessed based on the calculation of chronic daily intake (CDI) ($CDI = (DI \times EF \times ED) / (AT \times 365)$) as EF = exposure frequency per year of 200 days/year, ED = exposure duration of 44 years at the age of 20–64, and AT = average lifespan. The non-cancer risk (Hazard quotient, HQ) was assessed as the equation of $HQ = CDI$ (chronic daily intake)/RfD (reference dose) and cancer risk (R) was evaluated as the equation of $R = CDI \times SF$ (slope factor). RfD was obtained from the U.S. EPA Integrated Risk Information System (IRIS) to express BDE-47, -99, -153, and -209 as 0.0001, 0.0001, 0.0002, and 0.007 mg kg⁻¹ day⁻¹ ([U.S. EPA, 2014](#)), respectively, and 2,3,7,8-TBDD as 0.7 pg kg⁻¹ day⁻¹ ([U.S. EPA, 2012](#)). SF was from for oral 2,3,7,8-TBDD ([Simon et al., 2009](#)) and BDE-209 was 0.0001 per pg kg⁻¹ day⁻¹ and 0.0007 per mg kg⁻¹ day⁻¹ ([U.S. EPA, 2014](#)), respectively.

Measurements of PBDEs and PBDD/Fs below MDLs were recognized as zero for further statistical analysis. Descriptive analysis was used to determine the means and standard deviations (SD) of indoor dust, indoor PM_{2.5}, and outdoor PM_{2.5}. Mann-Whitney *U* tests were used to determine the differences in levels of PBDD/Fs and PBDEs on PM_{2.5} indoors and outdoors because PM_{2.5}-bound PBDEs and PBDD/Fs were not normally distributed and the sample size was small. The correlations or sources of PBDEs and PBDD/Fs in the gyms were examined by the principal component analysis (PCA). For the assessment of risk in the gym users and the staff in the gyms, the Monte Carlo method was used to simulate the non-dietary intakes of PBDEs and PBDD/Fs via ingestion or inhalation. The critical values of HQs and Rs were set as 1.00 and 1.00×10^{-6} . All statistical analyses were performed using Statistical Product and Service Solutions, version 12.0.

3 RESULTS AND DISCUSSION

3.1 Levels of PBDD/Fs in the Gyms' Environment

Table 2 shows PBDD/F levels in outdoor PM_{2.5}, indoor PM_{2.5}, and indoor dust in the gyms located in southern Taiwan. It was found that $\sum_{12}PBDD/F$ concentrations (mean ± SD: 0.202 ± 0.200 pg m⁻³) and PBDD/Fs-TEQ levels (0.00650 ± 0.00340 pg WHO₂₀₀₅-TEQ m⁻³) in indoor PM_{2.5} were higher than that of those in outdoor PM_{2.5} (PBDD/Fs: 0.163 ± 0.0951 pg m⁻³ vs. PBDD/F-TEQs: 0.00469

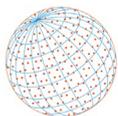


Table 2. PBDD/F concentrations on PM_{2.5} and indoor dust collected from the gyms.

	TEF	PM _{2.5} (pg m ⁻³)			Indoor Dust (pg g ⁻¹)
		Outdoor PM _{2.5} Mean ± SD (n = 4)	Indoor PM _{2.5} Mean ± SD (n = 4)	p value ^a	Mean ± SD (n = 6)
Congeners					
2,3,7,8-TeBDF	0.1000	< LOD	< LOD	1.000	2.98 ± 2.58
1,2,3,7,8-PeBDF	0.0300	< LOD	0.000561 ± 0.00112	0.686	3.54 ± 3.35
2,3,4,7,8-PeBDF	0.3000	0.00413 ± 0.00165	0.00388 ± 0.00460	0.042*	8.45 ± 3.90
1,2,3,4,7,8-HxBDF	0.1000	0.0176 ± 0.00800	0.0236 ± 0.0219	0.083	107 ± 49.6
1,2,3,4,6,7,8-HpBDF	0.0100	0.0938 ± 0.0478	0.136 ± 0.108	0.083	2270 ± 942
OctBDF	0.0003	0.0463 ± 0.0370	0.0378 ± 0.0755	0.166	4660 ± 3920
2,3,7,8-TeBDD	1.0000	< LOD	< LOD	1.000	< LOD
1,2,3,7,8-PeBDD	1.0000	< LOD	< LOD	1.000	< LOD
1,2,3,4,7,8-HxBDD	0.1000	< LOD	< LOD	1.000	< LOD
1,2,3,7,8,9-HxBDD	0.1000	< LOD	< LOD	1.000	< LOD
1,2,3,4,6,7,8-HpBDD	0.0100	0.000726 ± 0.00145	< LOD	0.317	6.55 ± 5.57
OctBDD	0.0003	< LOD	< LOD	1.000	27.3 ± 38.0
∑ ₆ PBDFs		0.162 ± 0.0939	0.202 ± 0.200	1.000	7050 ± 4690
∑ ₆ PBDDs		0.000726 ± 0.00145	< LOD	0.317	33.8 ± 38.1
∑ ₁₂ PBDD/Fs		0.163 ± 0.0951	0.202 ± 0.200	1.000	7090 ± 4680
∑ ₆ PBDFs WHO ₂₀₀₅ -TEQs ^b		0.00395 ± 0.00176	0.00492 ± 0.00465	0.773	37.8 ± 13.7
∑ ₆ PBDDs WHO ₂₀₀₅ -TEQs ^b		0.000007 ± 0.0000145	< LOD	0.317	0.0737 ± 0.0562
∑ ₁₂ PBDD/Fs WHO ₂₀₀₅ -TEQs ^b		0.00469 ± 0.00101	0.00650 ± 0.00340	0.248	37.8 ± 13.7

^a The significant levels were between indoor PM_{2.5} and outdoor PM_{2.5}; * $p < 0.05$.

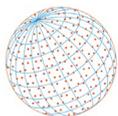
^b The WHO₂₀₀₅-TEQs were expressed as pg-WHO₂₀₀₅-TEQ m⁻³ and pg-WHO₂₀₀₅-TEQ g⁻¹ for PM_{2.5} and dust, respectively.

± 0.00101 pg WHO₂₀₀₅-TEQ m⁻³) with statistically insignificant differences (n = 4). There were a few articles reported on PM_{2.5}-bound PBDD/Fs indoors and outdoors.

The indoor dust level of ∑₁₂ PBDD/Fs was 7090 ± 4680 pg g⁻¹ and its PBDD/Fs-TEQ level was 37.8 ± 13.7 pg WHO₂₀₀₅-TEQ g⁻¹ in the gyms (n = 6). Several studies investigated and reported on PBDD/Fs in indoor dust (Ma *et al.*, 2009; Takigami *et al.*, 2009; Suzuki *et al.*, 2010; Tue *et al.*, 2010, 2013; Gou *et al.*, 2016a). In our previous study (Gou *et al.*, 2016a), the dust PBDD/F levels in the normal and computer classrooms of the elementary schools in southern Taiwan were reported as 4.72–7.51 and 3.60–6.28 ng g⁻¹ respectively, while the dust levels of PBDD/F-TEQs were 0.0401–0.0636 ng WHO₂₀₀₅-TEQ g⁻¹ in the normal classrooms and 0.0281–0.0474 ng WHO₂₀₀₅-TEQ g⁻¹ in the computer classrooms. A comparison between the present work and the previous work reported by us (Gou *et al.*, 2016a), indicated that the indoor dust levels of PBDD/F WHO₂₀₀₅-TEQs (0.0378 pg WHO₂₀₀₅-TEQ g⁻¹) in the gyms were lower when compared with those in the elementary schools from our previous study. Based on the current global data concerned with PBDD/Fs in indoor dust, it was suggested that the PBDD/F concentrations (7.09 ng g⁻¹) in the gyms had a distinctly lower magnitude when compared with those from the floor dust in the e-waste plant of China (89.3–143 ng g⁻¹) (Ma *et al.*, 2009) and house dust in the e-waste sites of Vietnam (7.70–63.0 ng g⁻¹) (Tue *et al.*, 2010) as well as the comparable magnitude with those from the indoor dust in the classrooms (1.46–6.39 ng g⁻¹) (Gou *et al.*, 2016a), house dust in the US (0.33–150 ng g⁻¹) and Japan (1.10–12.1 ng g⁻¹) (Suzuki *et al.*, 2010; Tue *et al.*, 2013), indoor dust in the Japanese offices (1.62–31.3 ng g⁻¹) (Suzuki *et al.*, 2010), and indoor dust in a Japanese hotel (0.740–15.4 ng g⁻¹) (Takigami *et al.*, 2009). Based on the distribution of PBDD/F profiles in PM_{2.5} and in indoor dust, it was observed that PM_{2.5}-bound PBDD/Fs were also similarly distributed in the gyms indoors and outdoors, but the indoor dust did not have the similarity of PBDD/F profiles with indoor PM_{2.5} (Fig. S1).

3.2 Levels of PBDEs in the Gyms' Environment

PBDE levels on indoor and outdoor PM_{2.5} and indoor dust in the Taiwanese gyms are presented in Table 3. ∑₁₄PBDEs in outdoor PM_{2.5}, indoor PM_{2.5}, and indoor dust in the gyms were expressed as

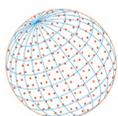
**Table 3.** Levels of PBDEs on PM_{2.5} and indoor dust collected from the gyms.

Congeners	PM _{2.5} (pg m ⁻³)			Indoor Dust (pg g ⁻¹)
	Outdoor PM _{2.5} Mean ± SD (n = 4)	Indoor PM _{2.5} Mean ± SD (n = 4)	p value ^a	Mean ± SD (n = 6)
BDE-28	0.00266	0.00845	0.008*	519 ± 923
BDE-47	0.0448 ± 0.0323	0.130 ± 0.0713	0.081	128000 ± 285000
BDE-100	0.0189 ± 0.0133	0.0497 ± 0.0728	1.000	46600 ± 98200
BDE-99	0.122 ± 0.0950	0.264 ± 0.366	0.554	177000 ± 334000
BDE-154	0.0742 ± 0.0551	0.0661 ± 0.0337	0.773	42500 ± 92000
BDE-153	0.211 ± 0.214	0.170 ± 0.0849	0.773	51400 ± 104000
BDE-183	0.349 ± 0.220	0.680 ± 0.470	0.248	13200 ± 11300
BDE-197	0.223 ± 0.105	0.477 ± 0.408	0.564	10300 ± 7700
BDE-203	0.152 ± 0.0711	0.379 ± 0.324	0.564	5180 ± 2960
BDE-196	0.212 ± 0.0941	0.428 ± 0.343	0.773	12700 ± 9270
BDE-208	0.332 ± 0.144	0.361 ± 0.245	0.773	26500 ± 12700
BDE-207	0.920 ± 0.392	0.982 ± 0.696	1.000	106000 ± 52600
BDE-206	0.801 ± 0.351	0.698 ± 0.483	0.564	169000 ± 85100
BDE-209	4.80 ± 1.90	4.85 ± 2.23	0.773	1880000 ± 251000
Σ ₁₄ PBDEs	8.27 ± 3.46	9.54 ± 4.68	0.564	2670000 ± 2330000

mean ± SD, such as 8.27 ± 3.46 pg m⁻³, 9.54 ± 4.68 pg m⁻³, and 2670000 ± 2330000 pg g⁻¹ (2670 ± 2330 ng g⁻¹), respectively. The results indicated that PM_{2.5}-bound PBDEs and BDE-209 (4.85 pg m⁻³) in indoor PM_{2.5} was higher than that of those in outdoor PM_{2.5} (BDE-209: 4.80 pg m⁻³), however their difference was found to be not significant. Based on the obtained results, it was suggested that BDE-209 was the predominated congener consisting of 58.0% and 50.8% of the total PBDEs in outdoor and indoor PM_{2.5}, respectively. The study conveyed that for indoor dust in the gyms, BDE-209 was also consisting of 70.4% of the total PBDEs. According to the homologue distribution of PBDEs in the gyms including outdoor and indoor PM_{2.5} and indoor dust as shown in Fig. S2, the distribution of PM_{2.5}-bound PBDEs in the gyms indoors and outdoors was very similar, but PBDE homologues in indoor dust were different from those in PM_{2.5}.

Based on the current global data in Table 4, it was claimed that not many studies were addressed PM_{2.5}-bound PBDEs in the outdoor and indoor environment (Beser *et al.*, 2014; Wang *et al.*, 2014; Li *et al.*, 2015; Xu *et al.*, 2015; Chao *et al.*, 2016; Deng *et al.*, 2016; Liu *et al.*, 2016; Xu *et al.*, 2016; Guo *et al.*, 2020; Tames *et al.*, 2020). According to the current data, it is very difficult to conclude whether indoor levels of PM_{2.5}-bound PBDEs are higher or lower than outdoor levels of PM_{2.5}-bound PBDEs or indoor and outdoor PM_{2.5}-bound PBDEs are highly correlated. The results of this present study conveyed that the data of PM_{2.5}-bound PBDEs indoors and outdoors in the gyms were not lower when compared with those from the certain outdoor and indoor environments, such as residential houses, rural areas, and urban areas.

A large variation of indoor PBDE concentrations in the gyms was found in this study. Based on the current global data it was emphasized that indeed this was the first report of indoor PBDEs in the gyms. Indoor dust BDE-209 as shown in Fig. S2 was the most abundant congener among PBDEs in the gyms which was very similar to the other microenvironments, such as offices, day-care centres, houses, cars, classrooms, and the public places (Gou *et al.*, 2016a; Brits *et al.*, 2019; Jin *et al.*, 2019; Lee *et al.*, 2020; Al-Harbi *et al.*, 2021; Klinčić *et al.*, 2021). Although the results obtained in the present study were in the same order of magnitude when compared with the other microenvironments (offices, medical and dental clinics, the classrooms of the elementary schools, and residential houses), the pattern of PBDE homologue in indoor dust from the gyms was not similar to the other Taiwanese microenvironments according to our previous studies (Chao *et al.*, 2014c; Shy *et al.*, 2015; Chou *et al.*, 2016; Gou *et al.*, 2016a). It was demonstrated that the unique PBDE homologue pattern in indoor dust of the gyms might be due to three reasons: (1) the electronic equipment used in the gyms was quite different from the one used in other indoor environments; (2) the gyms we chose were established and opened within these 5 years whereas the microenvironments we used for our previous studies were opened before 2015 (Chao *et al.*, 2014c; Shy *et al.*, 2015; Chou *et al.*, 2016; Gou *et al.*, 2016a). (3) Based on the

**Table 4.** Summary of the current data on PM_{2.5}-bound PBDEs.

Region (country)	Source	Reference	∑PBDEs ^a	Mean or mean ± SD
Outdoor				
Region of Valencia Government (Spain)	Households	Beser <i>et al.</i> , 2014	∑ ₁₂ PBDEs	0.213 pg m ⁻³
Beijing (China)	Industrial	Deng <i>et al.</i> , 2016	∑ ₄₂ PBDEs	146 pg m ⁻³
East China Sea (China)	Ocean	Li <i>et al.</i> , 2015	∑ ₁₁ PBDEs	0.97 ± 0.52 pg m ⁻³
Metropolis areas (China) ^b	Urban	Liu <i>et al.</i> , 2016	∑ ₉ PBDEs	35 ± 150 pg m ⁻³
Central and southern Taiwan (Taiwan)	Urban	Chao <i>et al.</i> , 2016	∑ ₁₄ PBDEs	67.9 ± 12.9 pg m ⁻³
Central and southern Taiwan (Taiwan)	Industrial	Chao <i>et al.</i> , 2016	∑ ₁₄ PBDEs	112 pg m ⁻³
Central and southern Taiwan (Taiwan)	Rural	Chao <i>et al.</i> , 2016	∑ ₁₄ PBDEs	62.7 pg m ⁻³
Kaohsiung and Pingtung (Taiwan)	Urban	The present study	∑ ₁₄ PBDEs	8.27 ± 3.46 pg m ⁻³
Indoor				
Guangzhou (China)	Households	Wang <i>et al.</i> , 2014	∑ ₂₆ PBDEs	239 pg m ⁻³
Hong Kong (China)	Households	Wang <i>et al.</i> , 2014	∑ ₂₆ PBDEs	43.8 pg m ⁻³
Shanghai (China) ^c	Households	Xu <i>et al.</i> , 2016	∑ ₁₈ PBDEs	48.5 ± 17.2 pg m ⁻³
Shanghai (China) ^c	Workplaces	Xu <i>et al.</i> , 2016	∑ ₁₈ PBDEs	105 ± 45.5 pg m ⁻³
Hong Kong (China)	Kindergartens	Deng <i>et al.</i> , 2016	∑ ₁₃ PBDEs	453 ± 345 pg m ⁻³
Central and southern Taiwan (Taiwan) ^d	Workplaces	Chao <i>et al.</i> , 2016	∑ ₁₄ PBDEs	116 ± 112 pg m ⁻³
Cordoba (Argentina)	Rural	Tames <i>et al.</i> , 2020	∑ ₇ PBDEs	18.28 pg m ⁻³
Harbin (China)	Houses	Guo <i>et al.</i> , 2020	∑ ₁₀ PBDEs	4.19 ± 3.17 pg m ⁻³
Kaohsiung and Pingtung (Taiwan)	Gyms	The present study	∑ ₁₄ PBDEs	9.54 ± 4.68 pg m ⁻³

^a The congeners of ∑PBDEs were shown in Table S1 and S2 in the supplementary materials.

^b The metropolis areas in China including Beijing, Shanghai, Guangzhou, Nanjing, Wuhan, Taiyuan, Chengdu, Lanzhou, Guiyang, and Xinxiang.

^c Xu *et al.* (2016) did not show the types of workplaces in Shanghai City.

^d The workplaces in the present study were library, rail station, department store, office, hospital, and supermarket.

latest announcement of persistent organic pollutants on Stockholm Convention in 2019 to list BDE-209 or commercial decaBDE mixture as Annexure A for elimination, the new building materials, electronic equipment, or decoration might be largely used for PBDE-replacement formulation of fire retardants in the gyms.

3.3 Correlations of PBDEs and PBDD/Fs in the Gyms' Environment

Fig. 1 shows the correlations of PBDD/Fs or PBDEs in the gyms indoors and outdoors after examination of PCA tests. Firstly, PM_{2.5}-bound PBDD/Fs indoors and outdoors were examined by PCA to classify two groups as follows: ∑₁₂PBDD/Fs, PBDDs, PBDFs, and certain PBDD/Fs in outdoor PM_{2.5} in a group and indoor PM_{2.5}-bound PBDD/Fs including PBDDs, PBDFs, and certain PBDD/Fs in the other group (Fig. 1(A)) (percentage of variance (σ) = 94.9%). It was stated that outdoor PM_{2.5}-bound PBDD/Fs including PBDDs, PBDFs, and certain PBDD/Fs were independent of those in indoor PM_{2.5}, indicating that indoor PM_{2.5}-bound PBDD/Fs were not associated with outdoor PM_{2.5}-bound PBDD/Fs and it could also be explained as the different emission sources of PBDD/Fs between indoors and outdoors. Fig. 1(B) shows the statistically independent correlation of PM_{2.5}-bound PBDEs between indoors and outdoors and this result could thus explain the 94.9% of the data (σ = 94.9%). It clearly showed that the outdoor PM_{2.5}-bound PBDEs were not related to indoor PM_{2.5}-bound PBDEs. According to our previous study (Gou *et al.*, 2016b), airborne PBDEs from Di-BDEs to Nona-BDEs including gas and particle-phase were significantly independent both indoors and outdoors of the vehicle dismantling factories, but the outdoor BDE-209 was significantly correlated with indoor PBDEs from Hexa-BDEs to Deca-BDE.

For the indoor dust of PBDD/Fs and PBDEs, three rotated principal components (RPCs) were displayed (Fig. 2(A)). RPC1 (σ = 61.1%) was composed of BDE-28, 1,2,3,7,8-PeBDF, BDE-47, BDE-154, BDE-206, BDE-100, BDE-153, BDE-208, BDE-207, and BDE-99. RPC 2 (σ = 27.1%) was composed of 1,2,3,4,6,7,8-HpBDD, PBDDs, BDE-203, PBDEs, and BDE-209 and RPC 3 (σ = 11.8%) was composed of PBDD/Fs, PBDFs, BDE-183, 1,2,3,4,7,8-HxBDF, 1,2,3,4,6,7,8-HpBDF, 2,3,4,7,8-PeBDF, and OctBDF. Liang *et al.* (2020) indicated that certain PBDD/Fs were released from the pyrolysis of PBDEs,

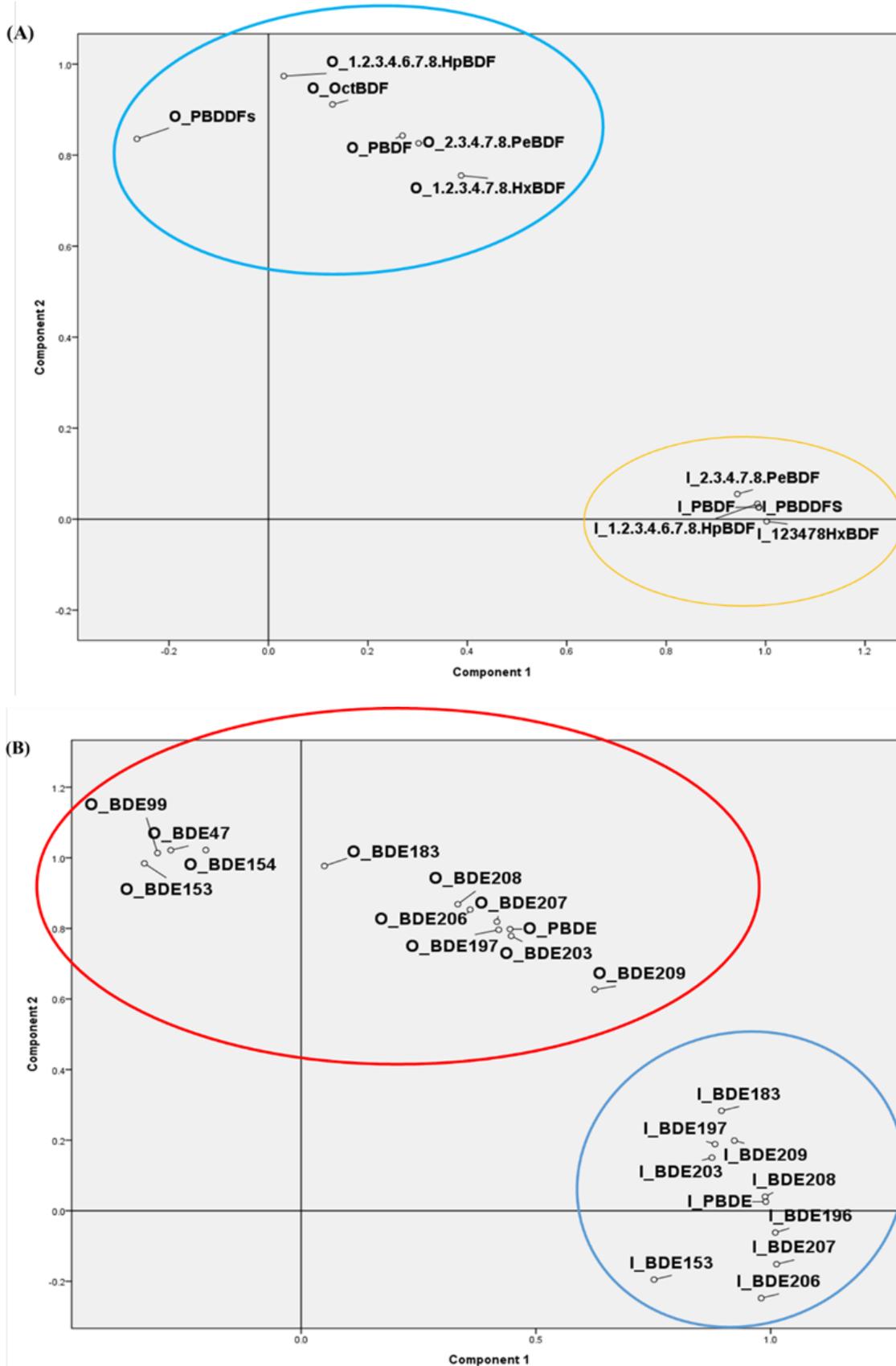
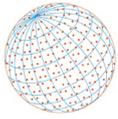
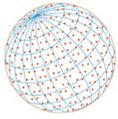
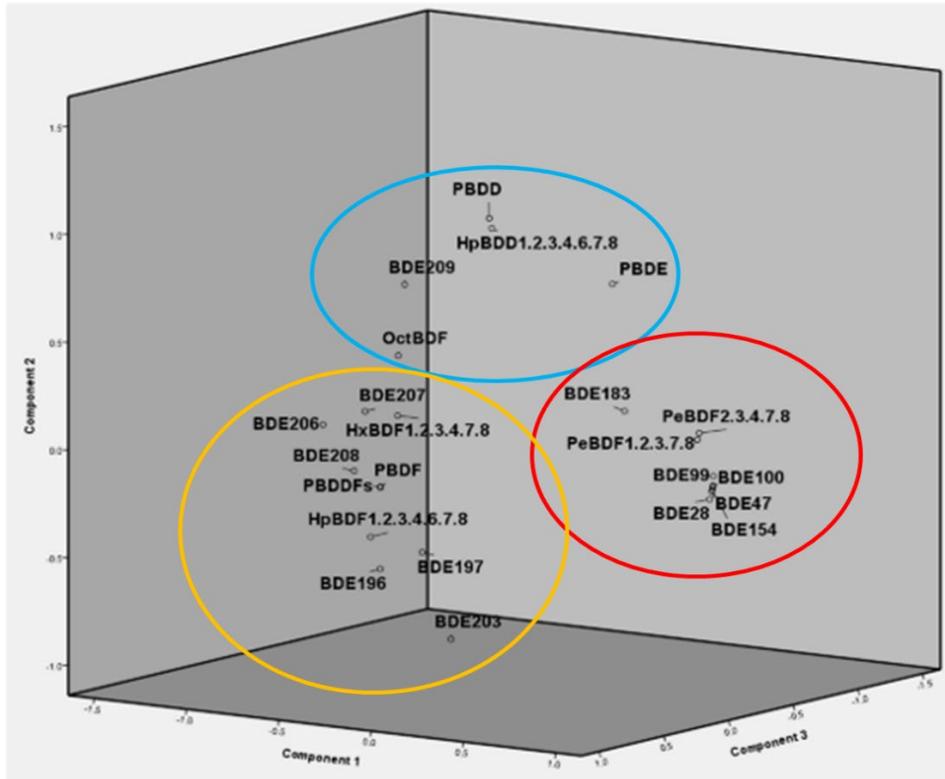


Fig. 1. The relationship between PBDEs and PBDD/Fs in the gyms examined by principal component analysis (PCA). For an example, indoor PBDEs and outdoor PBDD/Fs were expressed as I_PBDEs and O_PBDD/Fs, respectively. The RPCs were shown as follows: (A) PM_{2.5}-bound PBDD/Fs indoors and outdoors and (B) PM_{2.5}-bound PBDEs indoors and outdoors



(A)



(B)

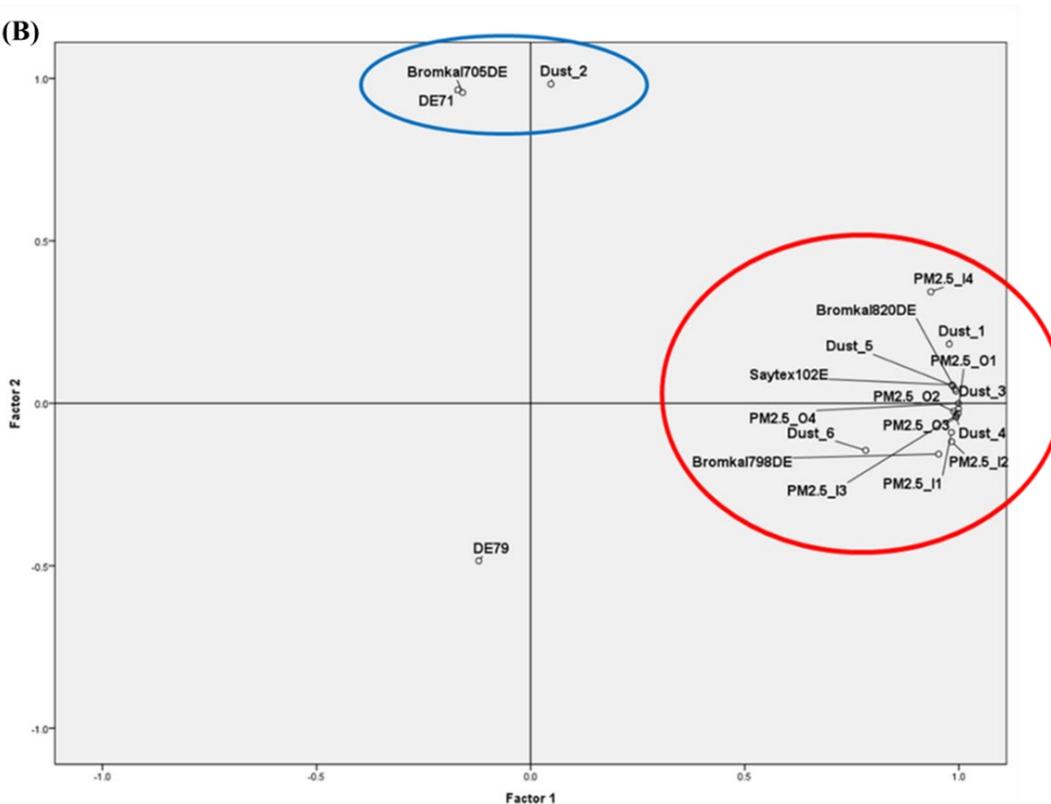
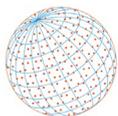


Fig. 2. Principal component analysis was used to examine (A) the relationship between PBDEs and PBDD/Fs in indoor dust and (B) the correlations of PBDEs between indoor PM_{2.5}, outdoor PM_{2.5}, indoor dust, and commercial formulation of brominated flame retardants (BFRs). PM_{2.5}_I, PM_{2.5}_O, and Dust were expressed as indoor PM_{2.5}, outdoor PM_{2.5}, and indoor dust, respectively. Commercial formulations of BFRs are DE-71 (Penta-BDE mixture), Bromkal 70-5DE (Penta), DE-79 (Octa), Bromkal 79-8DE (Octa), Saytex 102E (Deca), and Bromkal 82-0D (Deca). The commercial formula of Bromkal 79-8DE was contained 49.6% of BDE-209.



such as the disruption of the ether bond of the lower brominated numbers of PBDEs would be generated as PBDD/Fs (i.e., BDE-153 converting to 2,3,7,8-tetraBDD); while the higher brominated numbers of PBDEs favored the formation of polybromobenzene by cleavage of ether bond. The previous studies showed a significantly positive correlation between dust PBDD/Fs and dust PBDEs (Ma *et al.*, 2009; Gou *et al.*, 2016a). Based on our findings both in the present and the previous studies (Ma *et al.*, 2009; Gou *et al.*, 2016a; Liang *et al.*, 2020), certain PBDEs and PBDD/Fs were assigned into the same group after PCA tests in the indoor dust of the gyms which suggested that PBDD/Fs and PBDEs might come from the same emission sources. Fig. 2(B) shows the commercial formulation of Saytex 102E, Bromkal 82-0, and Bromkal 79-8DE in relation to indoor and outdoor PM_{2.5}-bound PBDEs and indoor dust PBDEs except for an indoor dust sample in a gym located at Kaohsiung to be classified as the groups of Penta-BDE mixtures (DE71 and Bromkal705DE). It is meant that the indoor and outdoor environment of the gyms were mainly contaminated from the Deca-BDE commercial formulations which are released from the surface of consumer or electronic products.

3.4 Risk Assessments in the Gyms' Environment

The risk factors and the exposure levels were used to assess the risk assessments, such as daily intakes, non-cancer risks (HQs), and cancer risks (Rs), via non-dietary routes based on two exposure scenarios including the gym users and employees (Table 1 and Table 5). According to Table 1, the active inhalation rates were 1.69–2.10 times higher than the inactive inhalation rates in Taiwanese. The values of daily intakes, HQs, and Rs for the gym users and staff through ingestion of indoor dust were found to be notably higher than that of those through inhalation of PM_{2.5} as shown in Table 5. The Korean study also announced that the contribution of dust ingestion was still main after the estimation of multiple-exposure assessment of PBDEs (Lee *et al.*, 2020). The summary and individual calculation of HQs and Rs from certain PBDEs and \sum PBDD/Fs for the gym users and the employees were still under the critical values of HQ = 1.00 and R = 1.00×10^{-6} . Thus, it was concluded that no non-cancer and cancer risks were found in the gym users and employees through indoor PM_{2.5} and indoor dust in the gyms. A comparison with our previous report revealed that the school-age children at the age of 7–9 consumed between 9.40×10^{-12} and 1.34×10^{-11} mg kg b.w.⁻¹ day⁻¹ from the classroom dust (Gou *et al.*, 2016a). However, in the present study, the estimated daily intakes of \sum PBDD/Fs (1.29×10^{-13} – 5.42×10^{-13} mg kg b.w.⁻¹ day⁻¹) for the adults through the indoor dust of the gyms were found to be low. The gym-dust daily intakes of PBDEs for the male and female employees were found to be 1.61×10^{-7} and 2.11×10^{-7} mg kg b.w.⁻¹ day⁻¹ (0.161 and 0.211 ng kg b.w.⁻¹ day⁻¹), respectively. Several studies reported the estimated daily intakes via indoor dust for adults as BDE-209 of 0.2 ng kg b.w.⁻¹ day⁻¹ in the South African dust (Brits *et al.*, 2019), PBDEs of 0.56–0.57 ng kg b.w.⁻¹ day⁻¹ in the Korean dust (Lee *et al.*, 2020), PBDEs of 0.557–0.727 ng kg b.w.⁻¹ day⁻¹ in the Taiwanese dust (Shy *et al.*, 2015), and PBDEs of 0.283–0.311 ng kg b.w.⁻¹ day⁻¹ (Xu *et al.*, 2015). The results represented that the data of HQs and Rs in the present study was consistent with our previous study (Shy *et al.*, 2015) and also the Korean report (Lee *et al.*, 2020) suggesting that the non-cancer and cancer risks have not occurred.

4 CONCLUSIONS

This study was suggested to be the first report to announce the levels of PBDEs and PBDD/Fs on indoor and outdoor PM_{2.5} and indoor dust in the gyms. PM_{2.5}-bound brominated compounds, such as PBDD/Fs and PBDEs were not correlated in the gyms indoors and outdoors, but the profiles of PBDEs or PBDD/Fs on PM_{2.5} indoors and outdoors are similarly distributed. Certain PBDD/F and PBDE congeners in indoor dust of the gyms were potentially correlated. In the indoor environment, the profiles of PBDEs and PBDD/Fs were not similarly distributed between PM_{2.5} and indoor dust. The assessment of human health risks including the gym users and staff indicated that the daily intakes, non-cancer risks, and cancer risks of PBDEs and PBDD/Fs were still found to be in the acceptable values.

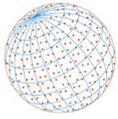


Table 5. Risk assessment of PBDEs and PBDD/Fs in the indoor environment of the gyms.

	Male gym user	Female gym user	Male Staff	Female Staff
Indoor dust^a				
Daily intake (mg kg b.w. ⁻¹ day ⁻¹)				
BDE-47	1.18×10^{-9}	1.54×10^{-9}	9.41×10^{-9}	1.23×10^{-8}
BDE-99	1.62×10^{-9}	2.12×10^{-9}	1.32×10^{-8}	1.73×10^{-8}
BDE-154	3.91×10^{-10}	5.11×10^{-10}	3.14×10^{-9}	4.10×10^{-9}
BDE-209	1.74×10^{-8}	2.27×10^{-8}	1.39×10^{-7}	1.83×10^{-7}
∑PBDEs	5.04×10^{-8}	6.58×10^{-8}	1.61×10^{-7}	2.11×10^{-7}
∑PBDD/Fs-WHO ₂₀₀₅ TEQs	1.29×10^{-13}	1.69×10^{-13}	4.15×10^{-13}	5.42×10^{-13}
Non-cancer risk (HQs) ^b				
BDE-47	6.34×10^{-6}	7.64×10^{-6}	5.06×10^{-5}	6.10×10^{-5}
BDE-99	8.73×10^{-6}	1.05×10^{-5}	7.10×10^{-5}	8.55×10^{-5}
BDE-154	1.05×10^{-6}	1.27×10^{-6}	8.43×10^{-6}	1.02×10^{-5}
BDE-209	1.34×10^{-6}	1.61×10^{-6}	1.07×10^{-5}	1.29×10^{-5}
∑PBDD/Fs-WHO ₂₀₀₅ TEQs	9.93×10^{-6}	1.14×10^{-5}	3.19×10^{-5}	3.84×10^{-5}
Cancer risk (Rs) ^b				
BDE-209	6.55×10^{-12}	7.88×10^{-12}	5.26×10^{-11}	6.34×10^{-11}
∑PBDD/Fs-WHO ₂₀₀₅ TEQ	6.96×10^{-18}	8.38×10^{-18}	2.23×10^{-17}	2.70×10^{-17}
Indoor PM_{2.5}^c				
Daily intake (mg kg b.w. ⁻¹ day ⁻¹)				
BDE-47	1.45×10^{-12}	1.23×10^{-12}	6.15×10^{-12}	5.91×10^{-12}
BDE-99	2.96×10^{-12}	2.49×10^{-12}	1.25×10^{-11}	1.20×10^{-11}
BDE-154	7.41×10^{-13}	6.23×10^{-13}	3.15×10^{-12}	3.01×10^{-12}
BDE-209	5.42×10^{-11}	4.56×10^{-11}	2.29×10^{-10}	2.19×10^{-10}
∑PBDEs	1.67×10^{-10}	8.98×10^{-11}	4.51×10^{-10}	4.32×10^{-10}
∑PBDD/Fs-WHO ₂₀₀₅ TEQ	2.96×10^{-13}	3.87×10^{-13}	3.08×10^{-12}	2.95×10^{-12}
Non-cancer risk (HQs)				
BDE-47	7.84×10^{-9}	6.08×10^{-9}	3.32×10^{-8}	2.93×10^{-8}
BDE-99	1.59×10^{-8}	1.24×10^{-8}	6.71×10^{-8}	5.93×10^{-8}
BDE-154	1.99×10^{-9}	1.55×10^{-9}	8.46×10^{-9}	7.47×10^{-9}
BDE-209	4.17×10^{-9}	3.23×10^{-9}	1.76×10^{-8}	1.56×10^{-8}
∑PBDD/Fs-WHO ₂₀₀₅ TEQ	2.27×10^{-8}	2.74×10^{-8}	2.37×10^{-8}	2.09×10^{-8}
Cancer risk (Rs)				
BDE-209	2.04×10^{-14}	1.58×10^{-14}	8.64×10^{-14}	7.63×10^{-14}
∑PBDD/Fs-WHO ₂₀₀₅ TEQ	1.59×10^{-17}	1.92×10^{-17}	1.66×10^{-17}	1.46×10^{-17}

^a Oral ingestion.

^b The dimensionless unit.

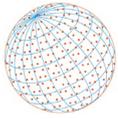
^c Inhalation.

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DISCLAIMER

The authors declare no conflicts of interest in this study.

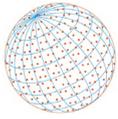


SUPPLEMENTARY MATERIAL

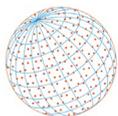
Supplementary material for this article can be found in the online version at <https://doi.org/10.4209/aaqr.220264>

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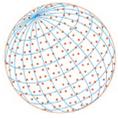
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