



Monitoring of PCBs at facilities related with PCB-containing products and wastes in South Korea

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ABSTRACT

Polychlorinated biphenyl (PCB) contents were analyzed in samples collected from facilities related to PCB-containing products or wastes in South Korea. Average concentrations of the atmospheric Σ_{209} PCBs were 7420 (37.0–104,048) pg m^{-3} and 16.8 (ND–34.2) fg WHO-TEQ m^{-3} in indoor air samples; and 1670 (106–13,382) pg m^{-3} and 5.64 (ND–36.0) fg WHO-TEQ m^{-3} in outdoor air samples. The highest levels were observed in indoor air samples from disposal facilities (7336–104,048 pg m^{-3}), followed by production (330–25,057 pg m^{-3}), recycling, and storage facilities, indicating that PCB emissions from PCB-containing products and wastes remains very high and the facilities related with those may be an important source to atmospheric PCBs. Principal component analysis of PCB profiles showed that the homologue patterns of PCBs in outdoor and indoor air samples collected from the facilities were similar to those of boundary air samples and PCB commercial products, e.g. Aroclor 1016, 1221, 1232 and 1242. Evaluation of the PCB mass balance in a facility, dismantling and solvent-washing PCB-contaminated transformers, showed that of the total PCBs treated in this facility, approximately 0.0022% was emitted to the atmosphere, and most was transferred to waste oil for disposal by incineration or chemical methods.

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1. Introduction

Polychlorinated biphenyls (PCBs) are classified and regulated as one of the 12 persistent organic pollutants (POPs) under the Stockholm Convention on POPs [1]. The sources of PCBs can be divided into two major categories: intentional chemicals produced in the chemical industries, and unintentionally *de novo* synthesized by-products during thermal processes [2,3]. The production and consumption of global PCBs for industrial purposes are relatively well established. PCBs were mostly produced commercially from 1929 to the early 1970s. During this period, total global production of PCBs was estimated approximately 1.3 million tons [4]. The commercial PCBs are known by a variety of trade names, such as Aroclor (USA, UK), Kanechlor (Japan), Sovol (Russia), Chlophen (Germany, Poland), and Phenoclor (France) [5,6].

In South Korea, industrial PCB mixtures have never been produced and their use in electronic equipments has been banned since 1979, and their import and use was completely banned in 1996. Kim et al. [7] reported that the ambient air in Korea was more influenced by combustion processes than that in Japan and also the contribution of PCB commercial products was relatively small. PCB levels in iron and steel complexes in South Korea have been reported to be higher than those in residential areas, indicating that iron and steel complexes are probably an important source of PCBs [8]. However, the emission of PCBs caused by *de novo* synthesis is not believed to contribute significantly to the global historical PCB mass balance [9]. The relative importance of atmospheric emissions from various source categories is not well known with considerable uncertainty [10]. Jamshidi et al. [11] reported that the principal contemporary source of PCBs in UK conurbation was ventilation of indoor air and not volatilization from soil.

According to the Korean Law, wastes that contain PCBs ($>0.0001 \text{ mg kg}^{-1}$ in solids or $>0.01 \text{ mg kg}^{-1}$ in liquids) are considered as “PCB-containing wastes” which must be treated by specialized methods [12]. Recycling of PCB-containing wastes only limits for wastes which contain less than 2 mg kg^{-1} PCBs. In 2007, the amount of PCB-containing wastes generated in South Korea that were contaminated with $>2 \text{ mg kg}^{-1}$ PCBs was 2543 tons [13]. Therefore, the emission of PCBs from PCB-containing products and

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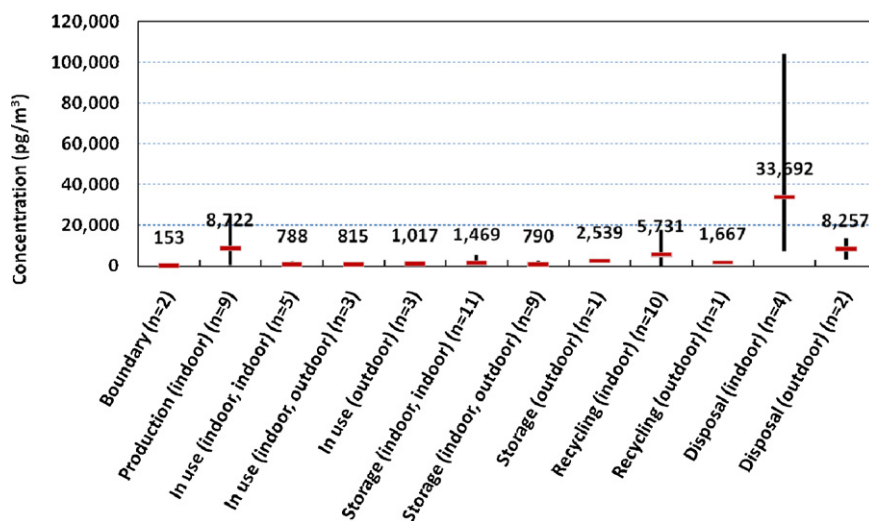


Fig. 1. PCB levels in air samples collected using HVAS from facilities related to PCB-containing products or wastes. Cross bars are mean values and vertical represent maximum and minimum concentrations.

wastes remains very high, even 30 years after PCB production ceased. Emission inventories are essential for identifying, evaluating, and prioritizing sensible control strategies on a regional or a global scale [1,14]. Also, to understand and predict the long range transport features and environmental fates of these substances, quantitative information on their atmospheric releases is deemed essential [15,16]. Hosomi et al. have estimated volatilization of PCBs from PCB-containing ballast in a fluorescent lamp [17]. However, few studies have investigated PCB contamination in facilities related to PCB-containing products and wastes rather than unintentional sources such as incinerators; no such evaluation has been performed in South Korea.

In this study, we investigated atmospheric levels and distribution of PCBs in facilities related to PCB-containing products and wastes. These facilities include production, in-use, recycling, storage and disposal facilities across South Korea. We also evaluated PCB emission factors and the mass balance in a PCB disposal facility. The emission of PCBs caused by *de novo* synthesis was not considered in this study. This is the first study to investigate PCB emissions from facilities related to PCB-containing products and wastes in South Korea, providing valuable data in planning for comprehensive management and final elimination of PCB-containing products and wastes.

2. Experiment and method

2.1. Sampling

Air samples were collected from 44 sites (9 production, 8 in-use, 14 storage, 10 recycling, 1 disposal, and 2 boundary) related to PCB-containing products or wastes across South Korea from October 2007 to July 2008 (Fig. S1). The specific information of sampling was shown in the Table S1. Samples were collected using high volume air sampling (HVAS, DHA-1000S, SIBATA). A glass fiber filter (GFF) and two consecutive polyurethane foam plugs (PUF) were used to collect airborne particles and vapor-phase PCBs, respectively. Before sampling, the GFFs were baked at 450 °C for 12 h, and the PUF disks were Soxhlet extracted for 16 h with acetone, then for 16 h with dichloromethane, then dried in a desiccator under vacuum for 24 h. A total of 20 outdoor air samples and 39 indoor air samples were collected for 24 h and at a flow rate of 700 L/min. It is important to note that

room sizes of a few in-use facilities were smaller than collected air volumes (1000 m³). Therefore, PCB concentrations could be underestimated by dilution effects in those small facilities. Outdoor air samples were collected within 5 m from the facilities (or rooms). Boundary PCB concentrations were measured at sites situated at the boundaries (500–800 m) of facilities. An additional 89 bottom samples were collected at the 37 facilities by wiping floor dust with hexane rinsed glass wool. At each site, 1–3 bottom samples were collected according to its facility size. For mass balance case study, several final product samples, such as copper, silicon steel plate, waste paper, waste oil etc., were collected from a dismantling and cleaning facility of PCB-containing wastes.

2.2. Analytical methods

In the laboratory, samples were treated, extracted and analyzed according to the methods established at the USEPA's method 1668A [18]. Briefly, the samples were spiked with the internal standard containing 27 ¹³C-labeled PCB congeners (1, 3, 4, 15, 19, 37, 54, 77, 81, 104, 105, 114, 118, 123, 126, 155, 156, 157, 167, 169, 188, 189, 202, 205, 206, 208, and 209) (Wellington, 1668-LCS), then Soxhlet-extracted for 24 h using toluene. The extracts were then washed with concentrated H₂SO₄ followed by hexane-saturated H₂O. Sample cleanup was performed using multi-layer silica and florisil columns. The eluent was reduced to 0.5 mL by rotary evaporation and a gentle stream of N₂ gas. Finally, the extracts were transferred to GC vials, and ¹³C-labeled PCBs (9, 28, 52, 101, 111, 138, 178, and 194) were added as recovery standards. PCB contents were analyzed using an Agilent Hewlett-Packard 6890 gas chromatograph/Jeol JMS-700T high resolution mass spectrometer (GC/HRMS) with a DB-5MS column (J&W Scientific, 60 m length, 0.25 mm ID, 0.25 μm film thickness). The instrument was operated using He as the carrier gas with a constant flow of 1 mL min⁻¹. The temperature program of the GC oven was as follows: the temperature was held at the initial value of 110 °C for 2 min, then raised at 40–200 °C min⁻¹ and held for 3 min, then raised at 2–230 °C min⁻¹, then raised at 7–300 °C min⁻¹ and held for 7 min. 1 μL sample was injected at a temperature of between 280 and 300 °C for the analysis of PCB contents. The GC/HRMS was operated under positive EI conditions (38 eV) with a resolution of 10,000. Data were obtained in the selected ion monitoring (SIM) mode.

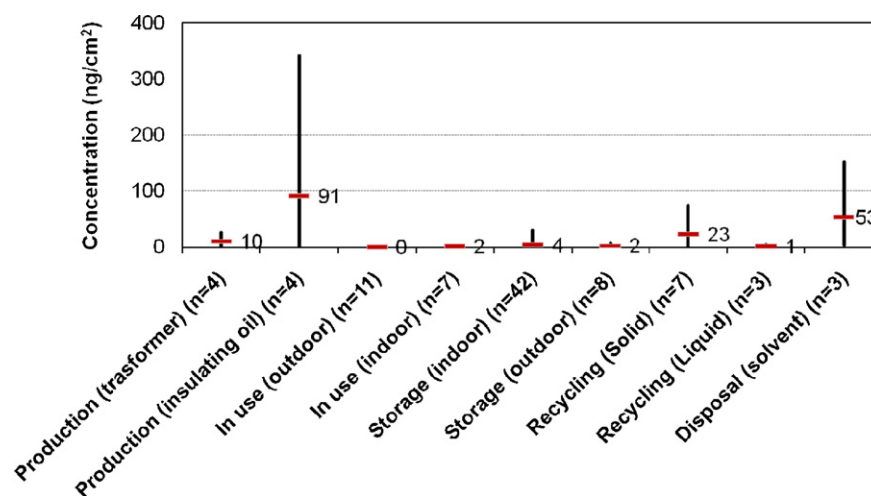


Fig. 2. PCB levels in bottom samples. Cross bars are mean values and vertical represent maximum and minimum concentrations.

Peak assignment was conducted to quantify 209 PCB congeners, but typically only 120 PCB congeners were detected. Bottom samples were analyzed using a GC with an electron capture detector (HP 6890, Agilent) following the Korean waste official method [19].

2.3. Quality assurance/quality control

Several steps were taken to obtain data that would allow an assessment of the accuracy and reliability of the data. Analytical blanks were included at a rate of one per 10 samples. All data have been blank corrected. The average recoveries of 27 ^{13}C -labeled PCB congeners ranged from 25 to 93% (Table S2), which satisfied the criteria (25–150%) recommended by US EPA method 1668A. Recovery statistics are given in Table S6. The method detection limit was calculated as 3 times the standard deviation of seven blank replicate samples (Table S3). The criteria for the quantification of analytes were as follows: retention time within 2 s of that of the standard, isotope ratio within 20% of that of the standard, and signal-to-noise ratio ≥ 3 .

3. Results and discussion

3.1. PCB levels

Detected levels of PCBs were generally lower in outdoor air samples than in indoor air samples, although the range was very large (Table 1, Fig. 1). The mean PCB concentration (Σ_{209} PCBs) in outdoor air samples was 1670 pg m^{-3} ($5.64 \text{ fg WHO-TEQ m}^{-3}$) and ranged from 106 pg m^{-3} at a PCB-containing waste storage site to $13,400 \text{ pg m}^{-3}$ at a PCB disposal (dismantling and cleaning) facility. These PCB concentrations in outdoor air samples were consistent with the PCB levels in the ambient air of South Korea in a previous study [7] and comparable with those in global urban sites (mean: 1700 pg m^{-3}) [20]. The mean PCB concentrations of indoor air samples were 7420 pg m^{-3} ($16.8 \text{ fg WHO-TEQ m}^{-3}$) and ranged from 37 pg m^{-3} at an indoor transformer site (containing 46.6 tons of transformer oil contaminated with 0.15 mg kg^{-1} PCBs) to $104,048 \text{ pg m}^{-3}$ at a PCB disposal facility. PCB levels in both indoor and outdoor air samples were highest at the disposal facility followed by the production facility, the recycling facility, the storage site and the in-use site. The high concentration at the PCB disposal facility might be due to the volatilization of

Table 1
PCB levels (Σ_{209} PCBs) in indoor and outdoor air samples from different sites using HVAS.

Site type	Sample type	Mean PCB concentration		References
		(pg m^{-3})	(fg WHO-TEQ m^{-3})	
Boundary	Outdoor (n=2)	153 (146–161)	1.44 (0.029–2.85)	This study
Production	Indoor (n=9)	8722 (330–25,057)	11.50 (0.073–31.0)	This study
In use (indoor)	Indoor (n=5)	788 (37–2273)	4.79 (0.157–12.3)	This study
In use (indoor)	Outdoor (n=3)	815 (199–1159)	8.20 (ND–24.6)	This study
In use (outdoor)	Outdoor (n=3)	1017 (671–1561)	8.76 (ND–14.9)	This study
Storage (indoor)	Indoor (n=11)	1469 (353–5404)	0.72 (ND–3.74)	This study
Storage (indoor)	Outdoor (n=9)	790 (106–2527)	6.81 (ND–36.0)	This study
Storage (outdoor)	Outdoor (n=1)	2539 (2539–2539)	0.033	This study
Recycling	Indoor (n=10)	5731 (160–17,710)	6.05 (0.018–41.3)	This study
Recycling	Outdoor (n=1)	1667 (1667–1667)	ND	This study
Disposal	Indoor (n=4)	33,692 (7336–104,048)	114 (ND–342)	This study
Disposal	Outdoor (n=2)	8257 (3131–13,382)	2.31 (1.70–2.91)	This study
Background	Outdoor	180–280		Kim et al. [7]
Industrial		2080–5820		
Residential		240–28,000		
In use condenser (containing PCBs)	Indoor	26,000–110,000		Hosomi [17]
Industrial area	Outdoor (n=3)		21–27	Martínez et al. [29]
Urban area	Outdoor (n=3)		19–46	
Background	Outdoor		0.6 (1–1.9)	Menichini et al. [30]

more-volatile PCB congeners during transformer dismantling and solvent washing. PCB levels were also investigated in 89 bottom samples collected from 36 facilities (Fig. 2). PCBs were detected in about 80% of bottom samples. PCB concentrations ranged from ND to 342 ng cm⁻² and the highest level was found in samples from a transformer oil production site (ND–342 ng cm⁻²), followed by a disposal facility (4–152 ng cm⁻²).

PCB concentrations of 26,000–110,000 pg m⁻³ have been observed in the indoor air of an office where fluorescent lamps with PCB-containing ballast had been used [17]; the PCB volatilization rates from this ballast were temperature-dependent and the PCB composition of the emission gas was similar to that observed in the ballast samples collected. Our samples were not collected to quantify the effect of temperature on PCB levels in indoor air samples; however, the highest PCB concentrations were observed in samples collected in a disposal facility during summer (104,000 pg m⁻³ in July vs. 7340 pg m⁻³ in October), followed by a production facility (25,100 pg m⁻³). In a major UK conurbation, the principal contemporary source of PCBs has been reported to be not the volatilization from soil but the ventilation of indoor air; existing structures, especially older buildings in which PCBs had been used in the past, were the major source of PCBs in outdoor air [11]. Generally, urban areas are more polluted by PCBs than rural areas [21]. Based on our data and previous studies, it seems that PCBs volatilized from the PCB-containing products or wastes are important sources of PCBs in the ambient air in South Korea. Future reductions in PCB concentrations in the outdoor air and ultimately in human exposure may be best achieved by actions to these remaining sources of PCBs from PCB-containing products and wastes.

In our air samples, the average contribution of gas phase PCBs to total PCBs was about 96%, which was consistent with the previous study of 24 PCB congeners in South Korea [22]. Gas-particle partitioning of PCBs in air samples from each type of facilities related to PCB-containing products or wastes showed similar patterns (Fig. S5). Gas phase contribution to total PCBs decreased from 89 (mono-CBs) to 24% (deca-CB) in indoor air samples, and from 92 (mono-CBs) to 22% (deca-CB) in outdoor air samples (Fig. S6). These results suggest that PCBs in the air exist predominantly in the gas phase and that the contribution of PCB congeners to the gas phase decreases as congeners become more-highly chlorinated (i.e., less volatile).

3.2. Homologue patterns

The homologue patterns of PCBs in air samples were similar at all sampling sites (Fig. S2). In all air samples, the dominant PCB homologues found were low chlorinated PCBs such as mono-, di- and tri-PCBs which accounted for about 14%, 35%, and 33% of total PCBs in indoor air samples and about 15%, 41%, and 30% of total PCBs in outdoor air samples, on average, respectively (Fig. S3).

Many sources of PCBs can influence atmospheric PCB levels, including incinerators, industrial thermal processes, and PCB-containing products and wastes [23]; homologue patterns can provide clues to where and how these substances originated [24]. PCBs in the outdoor air in this study were apparently influenced by the indoor air PCBs due to the higher levels of PCBs in the indoor environment. For further source identification, principal component analysis (PCA) of the data was conducted using SPSS 12.0 software (SPSS, Inc.) and homologue patterns of air samples from this study were compared to those of commercial mixtures of Aroclor 1016, 1221, 1232, 1242, 1248, 1254, 1260, 1262 and 1268 from other studies [6,25]. Total concentrations of each homologue PCBs (i.e. 1 Cl, 2 Cls, . . . , 10 Cls) were used for PCA analysis. PCB data were normalized by dividing by the total PCB concentrations for each sample, producing data ranging from 0 to 1. Finally, these normalized PCB compositions were used as input data for PCA. As a result,

PC1 and PC2 accounted for 60% of the total variance (Fig. 3). In the loading plot, the variables are well grouped by the number of chlorine. The homologue patterns of PCBs in air samples from various sites in this study were similar to commercial mixtures such as Aroclor 1016, 1221, 1232, and 1242, suggesting that the homologue patterns of many air samples were simultaneously influenced by these commercial mixtures. It is consistent with another previous study that the homologue patterns of PCBs found in sediments in South Korea indicated that their sources were commercial mixtures such as Aroclor 1016, 1242, 1254, and 1260 or corresponding Kanechlor products [26].

PCA was also used to compare homologue patterns of air samples in this study to those of ambient soil [27], incineration flue gas, and cement plant flue gas samples from other researches [6,28]. As a result, PC1 and PC2 accounted for 90% of the total variance (Fig. 4). The homologue patterns of PCBs in our air samples from various sites in South Korea were different from those of ambient soil, incineration flue gas, and cement plant flue gas samples, suggesting that there are other significant sources. However, all our air samples including boundary air samples had similar homologue patterns with the general ambient air samples (Korea, $n = 15$; Japan, $n = 11$) [7], indoor air samples from a disposal facility (Japan, $n = 5$), and indoor air samples of a room where PCB-containing sealant was used (Japan, $n = 3$) [6] (Fig. S4). Kim et al. [7] reported that the PCB levels in the ambient air of South Korea were more influenced by combustion processes than that in Japan, and also that the contribution of commercial PCB products was relatively small. However, our results strongly suggest that the ambient air in South Korea is contaminated by mixtures of commercial Aroclor products with various chlorine contents, particularly lowly chlorinated mixtures.

3.3. A case study of PCB mass balance

To evaluate the PCB mass balance, a PCB disposal facility was selected, where PCBs in the indoor air showed the highest level. A series of air, bottom, and final product samples were collected from each process of dismantling and cleaning. This facility mainly treats waste transformers which contain PCB-contaminated transformer oil (>2 mg kg⁻¹). The main processes are removal of transformer oil, dismantling of outer transformer cases, first extraction with toluene, dismantling of transformer inner assemblies, and second extraction with toluene. The final products are recycled (metals) or passed on for further disposal (waste oil). The mean PCB concentrations were measured as 14,560 pg m⁻³ in indoor air samples and 8130 pg m⁻³ in outdoor air samples (Fig. 5). Since all PCB sampling was conducted only in autumn season, no seasonal variation of PCBs in outdoor air was reflected. This is one of the limitations in the present study. Limited field monitoring data need further study in the future.

If the concentrations of PCBs in the indoor air are relatively constant over the operation period, air PCB equilibrium between indoor and outdoor air can be described by the following Eq. (1):

$$V \frac{dC_1}{dt} = E_a Q - VR(C_1 - C_0) = 0 \quad (1)$$

and the PCB air emission factor E_a in this facility can be calculated using Eq. (2),

$$E_a = \frac{(C_1 - C_0)VR}{Q} \quad (2)$$

where V is the volume of the room (m³), C_1 is the concentration of PCBs in the indoor air (ng m⁻³), C_0 is the concentration of PCBs in the outdoor air (ng m⁻³), t is time, Q is the quantity of PCBs treated in the facility, and R is the natural air exchange rate at which outdoor air replaces the whole indoor air. Generally, natural air exchange rate of the concrete building is 7–24 day⁻¹ [17],

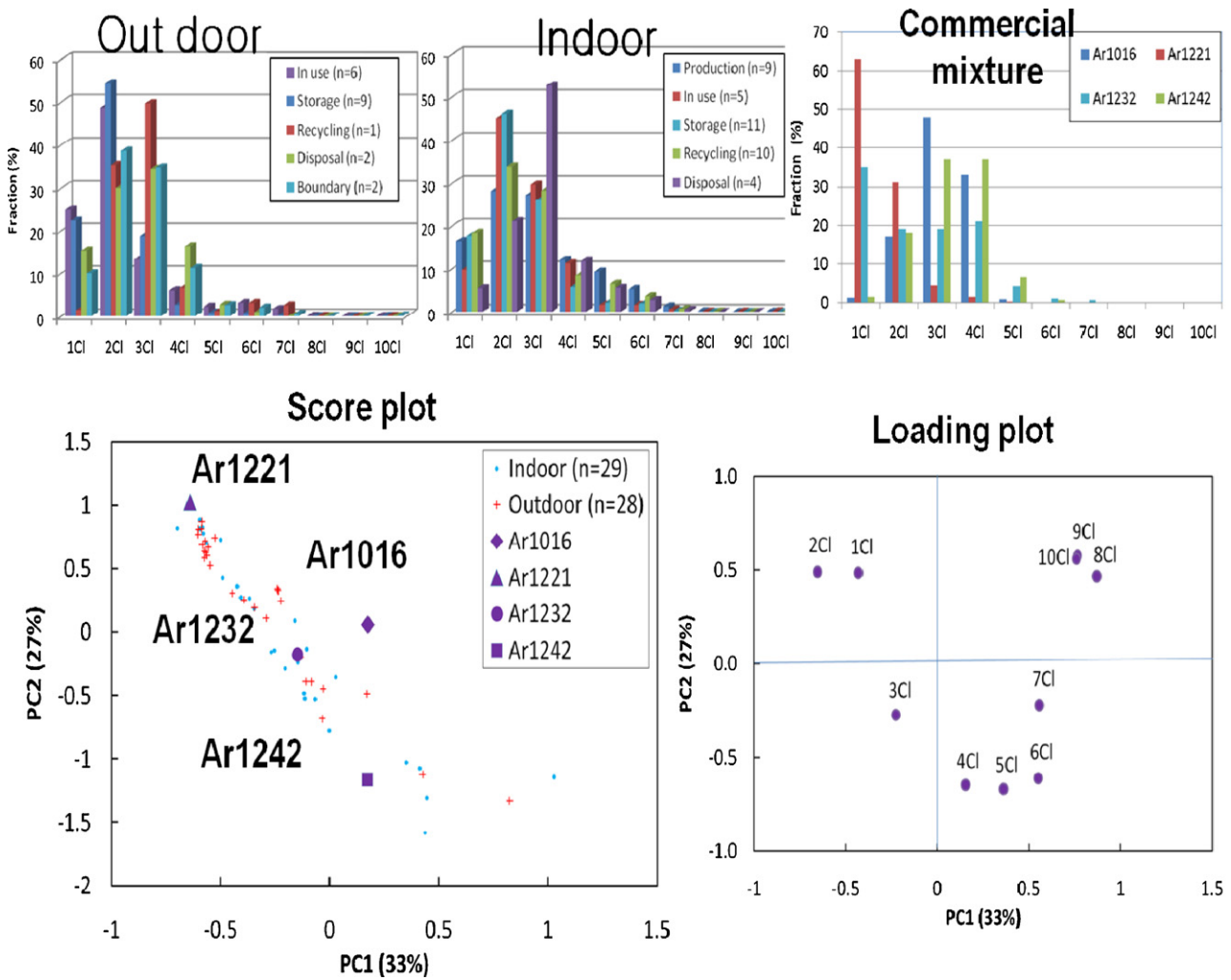


Fig. 3. Comparison of homologue patterns of air samples from this study with commercial mixtures of PCBs.

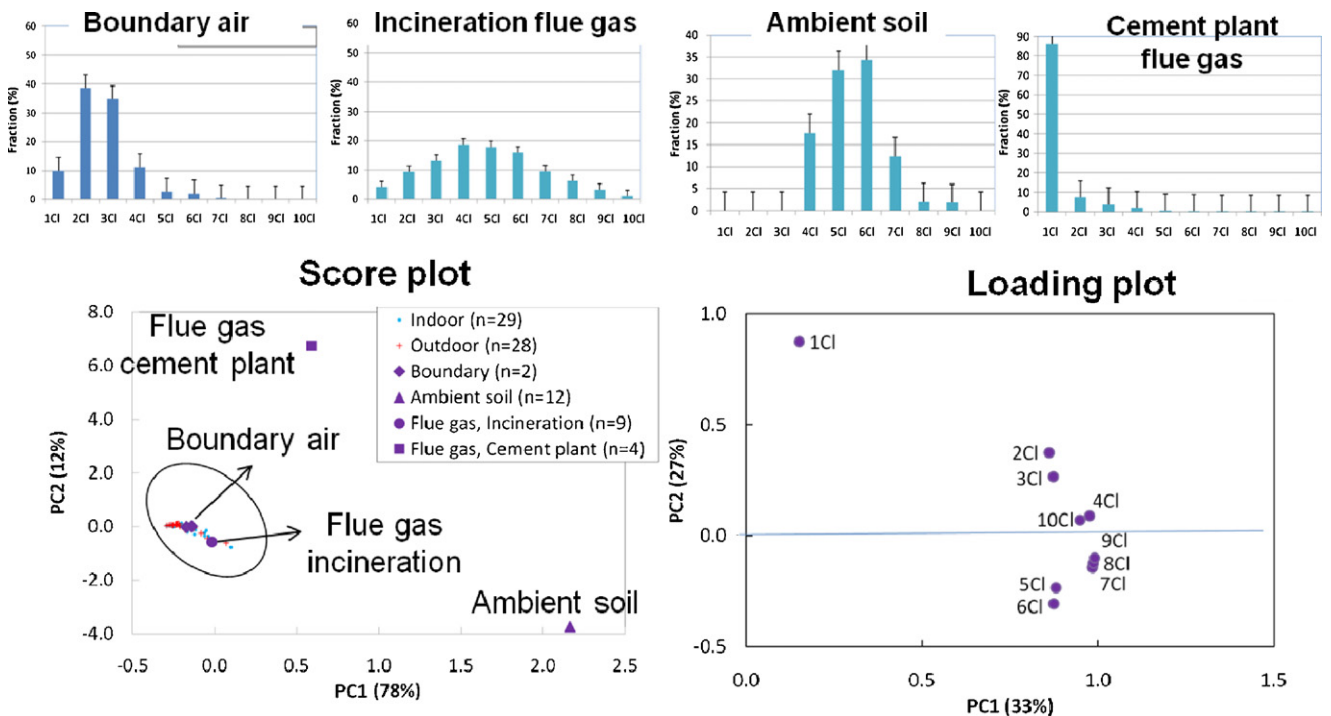


Fig. 4. Comparison of homologue patterns of air samples from this study with samples from other sources.

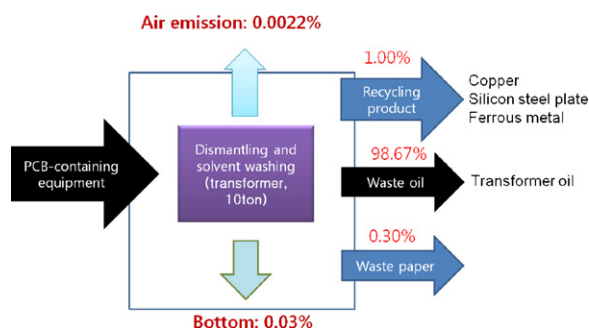


Fig. 5. PCB mass balance in a PCB disposal facility.

which was used in this study. Using the data observed in this facility (Table S4), E_a was estimated to be 9.8×10^{-4} to 3.4×10^{-3} g-PCB g-PCB $^{-1}$ yr $^{-1}$. This value is comparable or slightly higher than that reported in a previous study [4]; E_a ranged from 1.58×10^{-5} to 2.56×10^{-2} g-PCB g-PCB $^{-1}$ yr $^{-1}$ for open in-use and storage sites, and from 3.38×10^{-9} to 5.22×10^{-4} g-PCB g-PCB $^{-1}$ yr $^{-1}$ for closed in-use and storage sites [9]. In the previous study, Breivik et al. [4] has reported emission factors of only 22 PCB congeners, which have high uncertainties. The specific amounts of 22 congeners in the Aroclor mixtures in South Korea are not available. However, direct comparison of PCB air emission factors reported in this study with those above might be reasonable, because the 22 congeners were dominant congeners of PCBs and both air emission factors had the same unit. The uncertainty of air emission factor calculation in this study mainly comes from natural air exchange rate, variation of PCB concentrations in indoor and outdoor air samples, temperature etc.

PCB bottom emission factor E_b in this facility can be calculated using Eq. (3):

$$E_b = \frac{C_b A}{Q} \quad (3)$$

where C_b is the average concentration of PCBs in bottom samples, and A is the area of the facility. Using the data measured in this facility, E_b was estimated for bottom samples to be 3.0×10^{-4} g-PCB g-PCB $^{-1}$ yr $^{-1}$.

The PCB mass balance in this disposal facility was calculated (Fig. 5). There are major uncertainties, like Breivik et al. [9], which mainly come from natural air exchange rate, variation of PCB concentrations in air samples and temperature etc. Of the total PCBs disposed in this facility, approximately 0.0022% was emitted to the atmosphere and 0.03% was deposited to the indoor bottom as dust particles or transformer oil leakage. Meanwhile, most PCBs (98.7%) were transferred as waste transformer oil for later disposal by incineration or chemical treatment. If this facility were to operate at its maximum capacity of 100 tons/week, the estimated maximum Σ_{209} PCB emission to the air would be 13 g/yr. This is much smaller than the previous estimation, where estimated PCB emissions to air in South Korea was 199 kg (for 22 PCB congeners, mid scenario, maximum is hundreds-fold of minimum scenario) in the reference year 2008 [4]. Although the production, import and use of PCBs have been banned in South Korea since 1999, however, the amounts of in-use PCB-containing products are still huge. In South Korea, the amount of PCB-containing waste, which is contaminated with >2 mg kg $^{-1}$ PCBs, was 2543 tons in 2007 [13]. Therefore, atmospheric emission of PCBs from PCB-containing products and wastes still can be a significant source for some period by the time of their complete elimination.

4. Conclusion

In this study, we investigated PCBs from various types of facilities and calculated the PCB mass balance in a facility related to

PCB-containing products and wastes in South Korea. The total PCB concentrations ranged from 37.0 to 104,048 pg m $^{-3}$ in indoor air samples and from 106 to 13,382 pg m $^{-3}$ in outdoor air samples. The homologue patterns of PCBs in outdoor and indoor air samples collected from various facilities were similar to those of boundary air samples and the PCB commercial mixtures of Aroclor 1016, 1221, 1232 and 1242. These results suggest that PCB emissions during the production, recycling, in-use and disposal of PCB-containing products and wastes can be an important source of atmospheric PCBs. Therefore, it provides valuable data in planning for comprehensive management and final elimination of PCB-containing products and wastes.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2011.09.030.

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