Panel Data Analysis of Environmental PCB in Japan: National and Local Concentration Trends

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Introduction

PCB (Polychlorinated Biphenyl) is listed in Annex A "Elimination" and Annex C "Unintentional Production" of The Stockholm Convention on Persistent Organic Pollutants[1]. Therefore, it is essential to understand and develop countermeasures against PCB pollution conditions. In Japan, the Ministry of the Environment has been conducting a survey on the presence of chemicals including PCB in the general environment since 1974; the results of this survey are published in "Chemicals in the Environment" every year[2]. In addition, JESCO (Japan Environmental Storage & Safety Corporation) has been undertaking a project on PCB waste treatment since 2004[3].

Numerous studies have been conducted on the environmental fate of PCB on a global scale. Breivik et al. estimated the historical global PCB production, consumption, and emission by taking into consideration its mass balance[4]. Li et al. quantified the global PCB emissions using a dynamic substance flow model[5]. However, only a few studies focusing on the environmental fate of PCB or its concentration trends on a national scale have been conducted.

In addition, some studies have investigated the air pollution caused by PCB on a local scale. For example, Diefenbacher et al. showed that the air PCB concentration measured in areas within a radius of 150–200 m of a housing complex where joint sealants containing PCB were used was higher than that in the other areas of the same city[6]. In Japan, most of such contaminated buildings are likely to have been built on or before 1972, because both the production and the use of PCB were banned in 1972[7]. Therefore, it can be assumed that the air PCB concentration in the areas within a radius of several hundred m of the old buildings is likely to be higher than that in the areas close to new buildings. (In this study, the buildings that were built on or before 1972 are referred to as "old buildings," whereas the buildings that have been built after 1972 are referred to as "new buildings.")

The objective of this study was to understand the recent PCB pollution condition in Japan via panel data analysis of the results of a monitoring survey conducted by the Ministry of the Environment. First, the national PCB concentration trends in air, water, and sediment were estimated. Second, the PCB concentration trends on a local scale were estimated by taking into consideration the year in which the sampling sites were built.

Materials and Methods

Results of the air PCB concentration and temperature reported in "Chemicals in the Environment" were used for panel data analysis. Further, the PCB concentration in water and sediment has been measured once a year from 2002, and the PCB concentration in air has been measured twice a year from 2003 to 2013 (following which it has been measured once a year from 2014). The number of sampling sites for air, water, and sediment were 33–38 (51 in total), 45–49 (49 in total), and 62–64 (67 in total), respectively[2]. This study analyzed all the homologues (mono- to deca-CB). The half value of the detection limit concentration was substituted for the non-detection data.

The correlation between the PCB concentration and the sampling year in water or sediment was analyzed using Eq. 1. Further, in order to explain the PCB air concentration, the sampling year, the average temperature during the sampling time, and $dummy_{old}$ were included in Eq. 2. These two models were estimated for each homologue. Eq. 1 was estimated using a fixed effect model. On the other hand, Eq. 2 was estimated using a random effect model because it included a constant value ($dummy_{old}$). The PCB concentration of each homologue in any year and at any temperature was estimated by solving Eq. 2 for concentration (λ, $\varepsilon = 0$), and the total-PCB concentration was estimated as the sum of all the homologue concentrations.

$$\mathbf{lnC}_{\mathrm{media}} = \mathbf{a}_{\mathbf{i,media}} + \boldsymbol{\beta}_{\mathbf{year,media}}(\boldsymbol{year} - \mathbf{2000}) + \boldsymbol{\varepsilon}_{\mathbf{i,year,media}} \quad \textbf{(Eq.1)}$$

$$\mathbf{lnC_{air}} = \mathbf{b_0} + \boldsymbol{\delta}_{\mathbf{old}}\boldsymbol{dummy}_{\boldsymbol{old}} + \left(\boldsymbol{\beta}_{\mathbf{year,air}} + \boldsymbol{\delta}_{\mathbf{year}}\boldsymbol{dummy}_{\boldsymbol{old}}\right)(\boldsymbol{year} - \mathbf{2000}) + \left(\boldsymbol{\beta}_{\mathbf{temp,air}} + \boldsymbol{\delta}_{\mathbf{temp}}\boldsymbol{dummy}_{\boldsymbol{old}}\right)\frac{\mathbf{1}}{\mathbf{273.15} + \boldsymbol{T}} + \boldsymbol{\lambda}_{\mathbf{i}} + \boldsymbol{\varepsilon}_{\mathbf{i,year,T}} \quad \textbf{(Eq.2)}$$

$\mathbf{C_{air}}$: measured air PCB concentration [pg/m^3], $\mathbf{C_{sediment}}$: measured sediment PCB concentration [pg/g-dry],
$\mathbf{C_{water}}$: measured water PCB concentration [pg/L], $\mathbf{a_{i,media}}$: fixed effect of site i for each media,
$\mathbf{b_0}$: intercept, $\boldsymbol{\lambda_i}$: random effect for sampling sites, $\boldsymbol{\beta}$ and $\boldsymbol{\delta}$: coefficient, $\boldsymbol{\varepsilon}$: error term, **year**: sampling year,
T: average temperature during sampling time [°C],
$\mathbf{dummy_{old}}$: a dummy variable that equals to one if the building of the sampling site is built on or before 1972; otherwise, it equals to zero.

Results and Discussions

Yearly PCB concentration trends in each media on a national scale

In air, water, and sediment, $\beta_{year} < 0$ was statistically significant at a significance level of 0.1% for all the homologues (Fig. 1-a). Therefore, it is likely that the PCB concentration decreased in recent years in these three environmental media. For example, total-PCB concentration was estimated to decrease by approximately 40% in air and sediment, on the other hand, it was estimated to decrease by approximately 60% in water from 2006 to 2015 (Fig. 1-b).

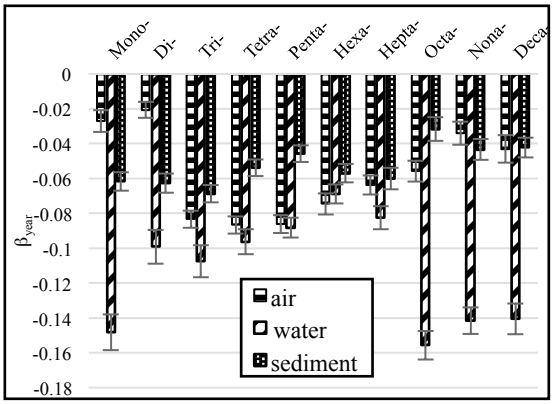


Fig. 1-a Estimation results of β_{year} (error bars indicate the standard errors)

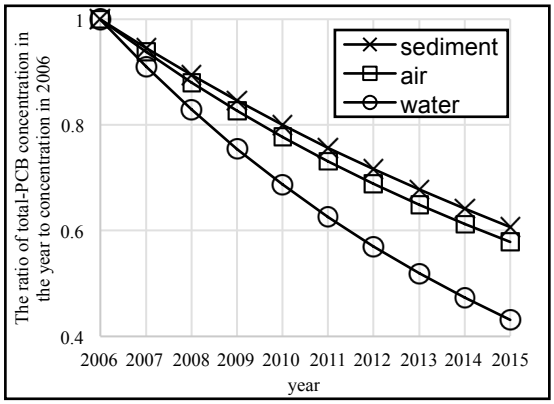


Fig. 1-b Estimation results of concentration trends in air, water, and sediment

It is assumed that the main causes for these decreasing trends are inflow decrease and degradation in the environmental media. The decreasing trend in air varies for each homologue, unlike that observed in sediment. Tri-, tetra-, and penta-CB, which were the main homologues of Kanechlor (PCB mixture mainly used in Japan[8]), showed the highest decreasing trends in air, whereas mono- and di-CB, which were the main homologues of unintentional emission sources[8], showed the lowest decreasing trends. This result suggests that emission from the PCB product waste may have been reduced by PCB waste management project; moreover, emission reduction could possibly be the main cause of decreasing trends in air. On the other hand, in the case of sediment, degradation of PCB accumulated during relatively high PCB emission could be the main cause for the decreasing trends because there should be no direct PCB emission for sediment. In the case of water, mono- and di-CB also showed high decreasing trends probably because of a decrease in the resuspension from the sediment following a decrease in the sediment PCB concentration. However, it is essential to conduct further studies such as those on the estimation of PCB emission and concentration using an environmental fate model on a national scale for more accurate consideration of the cause for these decreasing trends.

The correlation between the PCB concentration in air and the year in which the sampling sites were built

$\delta_{old} > 0$ was statistically significant at a significance level of 1% in the case of penta-, hexa-, and hepta-CB (Table. 1). Therefore, if the concentration of these homologues is measured in the same year and at the same temperatures, the concentration of the homologues in the air surrounding old buildings will be higher than that of the homologues surrounding new buildings. The concentration of the total-PCB in the air surrounding old buildings was estimated to be approximately 1.5–2 times higher than that of the total-PCB in the air surrounding new buildings (Fig. 2-a).

$\delta_{year} > 0$ was statistically significant at a significance level of 1% in the case of di- to hepta-CB (Table. 1). This result shows that the decreasing trend in the concentration of these homologues in the air surrounding old buildings is lower than that observed in the case of the concentration of the homologues in the air surrounding new buildings. The PCB concentration in the air surrounding old buildings was estimated to decrease by only approximately 25%, whereas the PCB concentration in the air surrounding new buildings was estimated to decrease by more than 40% from 2006 to 2015(Fig. 2-a).

$\delta_{temp} < 0$ was statistically significant at a significance level of 5% in the case of penta-, hexa-, and hepta-CB (Table. 1). Thus, the above results indicate that the temperature sensitivity of the PCB concentration in the air surrounding old buildings is higher than that in the air surrounding new buildings in the case of these homologues.

Further, the estimated proportion of the PCB homologues in the air surrounding old buildings was characteristic. The proportion of high-chlorinated homologues, particularly penta- and hexa-CB, were higher than those of high-chlorinated homologues in the air surrounding new buildings (Fig. 2-b).

These results suggest that in the case of some homologues, the local concentration trends in the air surrounding old buildings may differ from the national trends. However, the cause for these trends is unknown; therefore, it is necessary to carry out further investigations such as additional on-site sampling.

Table. 1 Results of estimation of Eq. 2

(Standard error) is indicated below **estimated value**.

	Mono-	Di-	Tri-	Tetra-	Penta-	Hexa-	Hepta-	Octa-	Nona-	Deca-
b_0	**2.83**** (0.870)	**20.4***** (0.632)	**22.2***** (0.684)	**23.7***** (0.677)	**22.8***** (0.697)	**21.0***** (0.830)	**17.1***** (0.754)	**12.1***** (0.827)	**-0.374** (0.913)	**-8.13***** (1.07)
β_{year}	**-0.027***** (0.00629)	**-0.0206***** (0.00456)	**-0.0833***** (0.00493)	**-0.0867***** (0.00489)	**-0.0862***** (0.00505)	**-0.0745***** (0.00606)	**-0.0636***** (0.00548)	**-0.0557***** (0.00602)	**-0.0340***** (0.00671)	**-0.0430***** (0.00790)
β_{temp}	**-112** (249)	**-4930***** (180)	**-5220***** (194)	**-5840***** (192)	**-5780***** (197)	**-5580***** (236)	**-4880***** (214)	**-4040***** (235)	**-853**** (262)	**1380***** (309)
δ_{old}	**-0.201** (1.66)	**-0.297** (1.21)	**1.18** (1.30)	**2.47** (1.28)	**3.93**** (1.31)	**6.17***** (1.56)	**6.04***** (1.42)	**2.61** (1.55)	**0.937** (1.72)	**1.29** (2.02)
δ_{year}	**0.0234** (0.0131)	**0.0311**** (0.00953)	**0.0373***** (0.0102)	**0.0299**** (0.0102)	**0.0415***** (0.0105)	**0.0395**** (0.0126)	**0.0295**** (0.0114)	**0.0136** (0.0125)	**-0.0200** (0.0139)	**-0.0195** (0.0163)
δ_{temp}	**8.36** (475)	**106** (344)	**-302** (370)	**-578** (364)	**-939*** (370)	**-1590***** (444)	**-1580***** (402)	**-583** (441)	**-150** (492)	**-252** (580)
R^2	0.0462	0.582	0.616	0.664	0.644	0.545	0.528	0.380	0.164	0.226
n	831	831	848	861	890	890	890	890	890	890
Statistically significant at 5(*), 1(**), 0.1(***)% level										

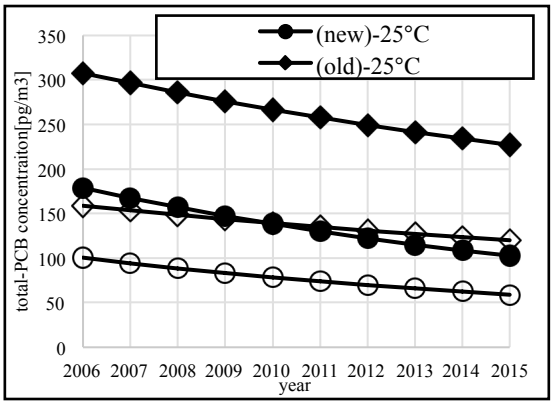
Conclusions

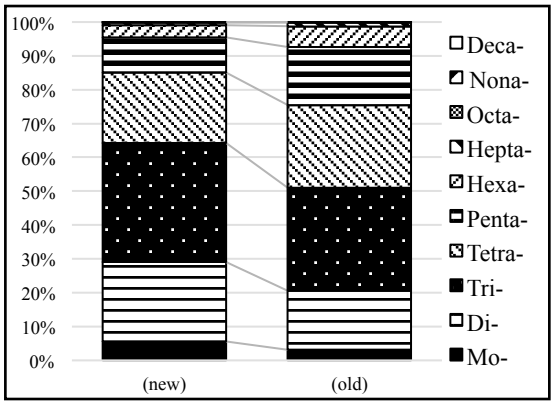
The results of panel data analysis carried out on a national level showed that in Japan, the PCB concentration in air, water, and sediment was estimated to have decreased at a significantly high confidence level. The yearly decreasing trend in each media differed from each other on the basis of a homologue pattern. The decreasing trend in air could most

likely be attributed to mainly emission reduction from PCB product waste. On the other hand, in the case of sediment, the PCB concentration is likely to have decreased because of degradation.

In addition, the decreasing trend observed in the air surrounding old buildings may differ from the trend observed in the air surrounding new buildings for some homologues on a local scale; for example, the air surrounding old buildings is likely to show higher PCB concentration, lower decreasing trend in PCB concentration, and higher temperature sensitivity than the air surrounding new buildings in the case of some homologues.

It is essential to conduct further investigations in order to understand the cause for these decreasing trends on both of a national and a local scale.





(new) indicates the area surrounding new buildings that have been built after 1972 and $dummy_{old}$ =0.
(old) indicates the area surrounding old buildings that were built on or before 1972 and $dummy_{old}$ =1.

Fig. 2-a Estimated concentration trends from 2006 to 2015 at 15 ℃ or 25 ℃

Fig. 2-b Estimated ratio of air PCB homologues in 2006 at 25 ℃

Acknowledgements

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