

Polychlorinated Biphenyls in the Drinking Water Source of the Yangtze River: Characteristics and Risk Assessment

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Keywords: polychlorinated biphenyls (PCBs), Yangtze River, risk assessment, drinking water source

Posted Date: December 30th, 2019

DOI: <https://doi.org/10.21203/rs.2.19713/v1>

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Version of Record: A version of this preprint was published at Environmental Sciences Europe on February 26th, 2020. See the published version at <https://doi.org/10.1186/s12302-020-00309-6>.

Abstract

Background As the longest river in Asia, the Yangtze River flows through the most industrialized cities in China and provides critical ecological services for agriculture, industry, and transportation.

Polychlorinated biphenyls (PCBs) have been banned for many years, but trace amounts of PCBs still exist as persistent organic pollutant in drinking water and are an ecotoxicological problem. In this work, we collected water, sediment, and suspended particulate matter (SPM) samples along the Yangtze River, to study the distribution and transport of PCBs for the risk assessment of the Yangtze River as a drinking water source.

Results The Σ PCBs concentrations in water, sediment, and SPM ranged in 0.044–10.98 ng/L, 0.33–69.43 ng/g, and 0.72–152.66 ng/L, respectively. The main pollutants were PCB17, 18, 28, 47 and 118 in the Yangtze River. The ff SW value of PCB18 and PCB28 all exceeded 0.5, whereas the ff SW value of PCB47 and PCB118 (except for PCB118 at Y-4) were less than 0.5. The toxic equivalent quantity (TEQ) range of PCBs in water and in sediment was 0–5.55 pg-TEQ/L and 0–2.51 pg-TEQ/g, respectively.

Conclusions In both water and sediment, lower chlorinated PCBs are dominant in the middle reaches and higher chlorinated PCBs are dominant at downstream. In contrast, SPM is dominated by tetra-CBs. The ff SW values reveal a net redissolution of lower PCBs from sediments to water but a net sorption of higher PCBs from water to sediment. The PCBs in water and sediment at downstream pose potential ecological risk, but the drinking water does not have a non-carcinogenic risk for humans.

Background

As the longest river in Asia, the Yangtze River supports the livelihood of > 400 million inhabitants and flows through a basin of about 1.8×10^6 km² in size [1], covering regions with the highest level of urbanization and industrialization in eastern China that contribute to about 42% of China's Gross Domestic Product [2]. It is an important freshwater resource in China and provides numerous ecological services for agriculture, industry, and transportation. The lower reaches of the Yangtze River provide 80–100% of the drinking water for economically developed and densely populated regions like Jiangsu and Shanghai [3, 4], but water quality has been menaced in recent years by rapid industrialization and accelerated urbanization.

Polychlorinated biphenyls (PCBs) include 209 congeners whose physicochemical properties and toxicity depend on the number and position of chlorine atoms in the molecule [5]. Because of their chemical stability and heat resistance, PCBs were once widely used as additives in heat exchange fluids, components for transformers and large capacitors, carbon-free copy paper, as well as paints and plastics, before their persistence and toxicity were examined and revealed [6–8]. In general, PCB28, 52, 101, 118, 138, 153, and 180 are indicative polychlorinated biphenyls (ind-PCBs) and are often used as indicators of environmental pollution [9], whereas coplanar polychlorinated biphenyls (co-PCBs) including PCB77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189 are used to estimate the toxicity of PCBs [10, 11].

The PCBs ubiquitous in the environment are notorious for their bioaccumulation and ability to transport over long distance [12]. Upon entering the water environment, PCBs are adsorbed by particulate matter because they are insoluble in water and eventually precipitate in the sediment [13]. However, sediment can resuspend when environmental factors change (e.g., turbulence caused by ship motion or flooding), and adsorbed PCBs are then released back into the water to start another cycle of environmental contamination [14, 15] and eventually accumulate in organisms. In fact, the transport of PCBs along the food web poses a serious risk to ecological integrity and human health [16, 17]. Therefore, it is of great importance to understand the transport of PCBs in water, sediment, and suspended particulate matter (SPM) to evaluate their risks in drinking water source.

Some studies have reported on the occurrence and distribution of PCBs in the source water and sediment of the Yangtze River [18, 19, 20], but the transport of PCBs along the Yangtze River in water, sediment, and SPM remains overlooked. In this work, we calculated the fugacity fraction of PCBs in sediment based on the PCBs concentrations determined by isotope dilution HRGC/HRMS, to evaluate the sediment–water diffusion of PCBs and their risk in the drinking water source of the Yangtze River.

Materials And Methods

Study area

Samples were collected in November 2016 from drinking water sources along the Yangtze River at specified sites (Figure 1). Overlying water was firstly filled into 1 L brown glass bottles, stored in ice box, then brought back to the laboratory and passed through a 0.45 μm filter membrane to give the water sample and the SPM sample. Sediment was collected from the surface of the riverbed, wrapped in aluminum foil and sealed in polythene bag, then freeze-dried and frozen at $-20\text{ }^{\circ}\text{C}$ until chemical analysis. To reduce possible random variation during collection, at least three replicates were collected at each site for both overlying water and sediment, and the eventual sample of the site was a composite of all replicates after thorough mixing.

Chemicals and materials

All solvents were of pesticide residue grade and purchased from Fisher (Fair Lawn, NJ, USA) and Tedia (Fairfield, OH, USA). The standard solutions EC-4977 (method 1668A toxics/LOC/window defining) and EC-4979 (method 1668A injection internal standard solution) were supplied by Cambridge Isotope Laboratories (Andover, MA, USA).

Anhydrous sodium sulfate, silver nitrate and sodium hydroxidewere of analytical grade and purchased from Beijing Chemical Factory (Beijing, China). Silica gel (100–200 mesh) was purchased from Qingdao Haiyang Chemical Co., Ltd. (Shandong, China). Basic aluminum oxide (100–200 mesh) was purchased from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China.

Analytical method

Water samples were spiked with the EC-4977 standard (1 ng) and equilibrated for at least 2 h prior to solid phase extraction. Samples of sediment and suspended particle matter were pretreated according to the methods of Zhao et al. [21]. That is, the solid sample (about 2 g) were spiked with 1 ng of ^{13}C -labeled EC-4977 and homogenized with sodium sulfate (10 g) with mortar and pestle. The homogenized mixture was allowed to equilibrate for at least 12 h before accelerated solvent extraction (ASE) was carried out. The extracts was cleaned up, and passed through multi-layer silica gel and basic aluminum oxide, and the recovered compounds were finally identified and quantified by isotope dilution HRGC/HRMS.

Quality control and quality assurance (QA/QC)

The laboratories must show its ability to generate acceptable method blank, precision, and recovery. Initial precision and recovery tests were performed according Zhao et al. [21], including sample preparation, extraction, and clean-up. The limit of detection was defined as the level of S/N = 3:1 in the measurement of spiked samples. The limit of detection for PCBs ranged in 0.3–1.8 pg/L for water and 0.9–6.5 pg/g for sediment (dry weight). The recoveries of PCBs spiked in water and sediment samples ranged in 57.8%–79.8% and 69.6%–97.5% respectively, and the relative standard deviation (RSD) was less than 20%.

Data and statistical analyses

The SPM water partition coefficient K_d (kg SPM or sediment per L water) was calculated as follows [22]:

[Due to technical limitations, the formulas could not be displayed here. Please see the supplementary files section to access the formulas.] (1)

where C_s (mg PCBs/L SPM) and C_w (mg PCBs/L water) are the concentrations of PCBs in the aqueous and solid phases, respectively.

The normalized partition coefficient of organic carbon (K_{OC}) is an empirical value of the steady state environment simulated in the laboratory and can be predicted by K_{OW} (*n*-octanol/water partition coefficient) [23]:

[See supp. files] (2)

K_{OC}^* is the modified normalized partition coefficient of organic carbon found in the field and can be calculated as [24]:

[See supp. files] (3)

where f_{OC} is the mass fraction of the organic carbon in the sediment (based on the measured TOC).

The fugacity of PCBs in sediment and water was calculated as follows [20]:

[See supp. files] (4)

(5)

where f_S and f_W are the fugacity of PCBs in sediment and water, respectively, and ρ_S is the density of the solid sediment ($1.5 \times 10^3 \text{ kg/m}^3$ for all calculations).

The fraction of the total fugacity (ff) was calculated as [25]:

[See supp. files] (6)

where ff_{SW} represent the fugacity fraction of PCBs in sediment.

The carcinogenic risk and non-carcinogenic risk are calculated as follows [26]:

[See supp. files] (7)

(8)

where C_i is the concentration of PCBs in water (mg/L); IR is the daily drinking water volume (1.85 L); EF is the adult exposure frequency (365 d/a); SF is the carcinogenic slope factor (2 kg·d/mg); BW is the body weight of an adult (60 kg); AT is the average time of the onset of carcinogenic effect (25550 d); ED is the adult exposure period (70 a); and RfD is the exposure dose (0.02 mg/d·kg). All values were taken from the Chinese population exposure parameter manual [27].

Results And Discussion

PCB levels in the water, sediment, and SPM

Table S1 lists the total concentrations of PCBs (Σ PCBs) and predominant PCB congeners in the water, sediment, and SPM of the Yangtze River, which range in 0.044–10.98 ng/L, 0.33–69.43 ng/g, and 0.72–152.66 ng/L, respectively. The concentrations of ind-PCBs in water, sediment, and SPM are 0.0047–0.38 ng/L, 0.062–21.49 ng/g, and 0.25–4.04 ng/L, respectively. For the 12 types of co-PCBs, their concentration in the three phases range in 0–0.23 ng/L, 0–8.41 ng/g, 0–1.27 ng/L, with a median value of 0.03 ng/L, 0.03 ng/g, and 0.2 ng/L, respectively. The Σ PCBs level in water is thus comparable to that of the Pearl River Estuary (China) (0.02–14.8 ng/L) [28] and the Houston waterway (up to 12.49 ng/L) [29], but significantly lower than that of the Ebro River (up to 108.0 ng/L) [30]. The Σ PCBs level in sediment is comparable to that of the Pearl River Delta surface sediment (3.5–48.3 ng/g) but is at the low end of the global PCBs level (0.2–400 ng/g) [31]. The Σ PCBs level in SPM is higher than that of the Dalian River estuary (6.78–66.55 ng/L) and the Portuguese coast (4.2–30.1 ng/L) [32], but much lower than that of the Newark bay estuary (466–966 ng/L) [33]. It can be seen from the above comparisons with other systems

around the globe that the PCBs pollution at the Yangtze River is at a low to medium level [34]. Generally, the Σ PCBs level tends to be higher in water and SPM in the middle reaches than the downstream, but the reverse is true for sediment.

The PCBs concentration is the highest at Y-2, Y-5, Y-7, and Y-9 (all in the vicinity of Wuhan) for SPM and at Y-25 and Y-28 (both in the Jiangsu province) for sediment, respectively. Point sources are likely involved in these localized high PCBs level. Besides, various other factors including hydrodynamics, crystal size, *etc.*, may also drive up the PCBs concentration [35]. In fact, Y-25 is in the tributary of the Yangtze River, where sediment particle size and low hydrodynamics may contribute to high PCBs.

Characteristics of PCBs in the water, sediment, and SPM

Figures 2-4 present the distribution PCB homologues in water, sediment, and SPM. In water, di-CBs and tetra-CBs are the dominant congeners (>40% of the total) in the middle reaches, whereas di-CBs and penta-CBs are the dominant congeners in the lower reaches near Jiangsu (Figure 2). The findings are consistent with the results of Zhang et al. [19], in which tetra-CBs were found to be the predominant contaminants. The PCBs in the sediment are mainly tri-CBs and tetra-CBs, and the proportion of penta-CBs and hexa-CBs increases in the downstream (Figure 3). The PCBs distribution in the sediment of the Yangtze River resembles that of the coastal East China Sea [18]. The PCBs in the SPM are mainly tetra-CBs (>70%), although the proportion of di-CBs and tri-CBs increases in the downstream section (Figure 4).

In general, for both water and sediment, lower chlorinated biphenyls are the dominant congeners in the middle reaches and higher chlorinated biphenyls are the dominant congeners in the lower reaches. Tetra-CBs are the dominant congeners in SPM.

Two representative types of PCBs (ind-PCBs and co-PCBs) were analyzed as the indicators of environmental pollution and toxicity [9]. The proportion of seven ind-PCBs to Σ PCBs varied between 1.54% and 64.44%. The congener profile of the ind-PCBs is dominated by the tri-CB PCB28 and the penta-CB PCB118, and other congeners contribute much less. Besides, the concentration of co-PCBs is significantly lower in water than in SPM, likely due to the low polarity, poor water solubility, and low content of co-PCBs in water.

The fingerprint of the detected PCBs differs in the three phases. PCB11 is detected in all water samples. PCBs 47 and 41 are detected more frequently in SPM and in sediment than in water, whereas PCB 138 is detected more frequently in sediment than in water and in SPM. In contrast, Zhang et al. [19] found for the Yangtze River that the most abundant congeners are PCB28, 18, 66, 44, and 52 in water and PCB 138, 153, 156, 194, and 209 in SPM.

Principal component analysis (PCA) was carried out for PCBs that were detected with >55% frequency and representative in the three phases, including PCB11, 17, 18, 28, 41, 47, 51, 66, 70, 74, 110, 118, 138, 153, and 209. In the PCA results (Figures 5 and 6), PC1 accounts for 69.26% of the variation, and its representative congeners include PCB47, 41, 51, 11, 18, 17, 66, and 28. The PC2 accounts for 14.81% of the variation, and its representative congeners include PCB70, 110, 138, 118, 153, and 74. PC1 has higher loading for the lower chlorinated homologues like tri- and tetra-CBs, whereas PC2 has higher loading for the higher chlorinated homologues like penta- and hexa-CBs (Figure 5). The PCA results also indicate that at sites 25 and 28, the sediment is affected by PCB138, 110, 70 (Figure 6), water is mainly affected by PCB11, and the SPM is affected by PCB47, 41, 51. This is also consistent with the results of [36], where higher PCBs are more readily adsorbed by natural organic matter (NOM) with time.

Transport of PCBs among multi-matrix compartments

The exchange of pollutants among multi-matrix compartments is a very complicated process that involves many factors including van der Waals forces, particle size, covalent bonds, temperature, *etc.* Multiple parameters are used to represent the transport of pollutants between media[36].

In this study, predominant PCB congeners in water, sediment, and SPM (including PCB18, 28, 47, and 118) were selected to evaluate the transport of PCBs among multi-matrix compartments. The SPM-water partition coefficient (K_d) reflects whether the adsorption of chemical substances favors the SPM than the aqueous environment. The $\log K_d$ values of PCB18, 28, 47 and 118 were calculated from Equation (1) to be 3.50–4.16, 3.20–4.28, 3.17–4.90, and 3.03–4.07, respectively. Hence, the four PCBs have similar K_d . Organic matter content, temperature, mineral composition, and median particle size are all important factors affecting K_d value.

The diffusion of dissolved contaminant from sediment to water is one of the main transport processes. The K_{OC} value is compared with K_{OC}^* to predict whether PCBs have reached equilibrium between sediment and water in the field [25]. The $\log K_{OC}^*$ values (Figure 7) of PCB18, PCB28, PCB47, and PCB118 range in 2.99–4.90, 3.00–4.20, 2.97–4.71, and 2.91–4.18, respectively, all much less than the predicted equilibrium $\log K_{OC}$ value (5.16, 5.18, 5.87, and 6.19, respectively). That is, the transport of PCB18, PCB28, PCB47, and PCB118 in sediment and water did not reach equilibrium yet. Thus, just like organochlorine pesticides (OCPs), known as persistent organic pollutants, PCBs have not reached equilibrium in their exchange between sediment and water either [36].

The fugacity fraction is commonly used to demonstrate the equilibrium state because the two directions of the equilibrium line ($ff = 0.5$) represent the same level of the deviation from the balanced state. The value of the fugacity fraction (ff) can be used as one of the indicators to evaluate the transport of PCBs between two phases. Specifically, transport occurs from sediment to water when $ff_{SW} > 0.5$ and in the reverse when $ff_{SW} < 0.5$.

The ff_{SW} values (Table 1) indicate that PCB18 and 28 have net redissolution from sediment to water, except for that PCB18 at Y-5. In contrast, PCB47 and PCB118 in most cases (except for PCB118 at Y-4) have $ff_{SM} \leq 0.5$, corresponding to net sorption from water to sediment. As the number of chlorine atoms increases, PCBs tend to reach saturation in water and show migration from water to sediments. PCBs are different from OCPs according to Jin and Liu et al, whose ff_{SW} values show net sorption from water to sediment [27, 36].

Table 1. The ff_{SW} values of PCB18, 28, 47, and 118 in the Yangtze River.

Sampling site	ff_{SW}			
	PCB18	PCB28	PCB47	PCB118
Y-3	0.83	0.94	0.06	0.19
Y-4	0.73	0.77	0.05	0.90
Y-5	0.33	0.70	0.01	0.05
Y-7	0.79	0.96	0.01	0.15
Y-12	0.79	0.98	0.04	0.09
Y-13	0.92	0.98	0.06	0.10
Y-15	0.81	0.94	0.27	0.06
Y-16	0.99	0.99	0.29	0.39
Y-19	0.80	0.89	0.03	0.14
Y-20	0.70	0.60	0.11	0.17

A fugacity fraction in 0.2-0.8 may not represent a significant deviation from equilibrium, and there are other parameters that are difficult to include in the error. The sediment-water model provides a preliminary basis to describe the transport of pollutants in two media and needs to be further verified.

Ecotoxicological and health risk assessment

PCBs in sediment have certain effects on the environment, and benthic organisms are also affected by sediment-bound PCBs [37]. We use the sediment quality guidelines (SQG) to assess the potential ecotoxicological impacts of organic contaminants in the surface sediment of the Yangtze River. Specifically, the range-low (ER-L) and range-median (ER-M)

values refer to the lower 10 percentile and median values, respectively [38]. In this study, the Σ PCBs concentration is below the ER-L (22.7 ng/g) except at Y-25 and Y-28, and neither (69.43 ng/g for Y-25 and 59.44 ng/g for Y-28) is greater than the ER-M (180 ng/g). Because the Σ PCBs level is much lower than ER-L, the ecological risk of PCBs in the sediments is deemed reasonably low, but the PCBs pose potential ecological risk in 5.9% of the sampling sites.

According to the Canadian soil quality guidelines, the standard value of polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) is 4 pg/g [39]. In this study, the TEQ of PCBs in sediment falls in 0–2.51 pg-TEQ/g, with a median value of 0.0007 pg-TEQ/g. Therefore, the sediment has a low ecotoxicological risk.

The TEQ of co-PCBs in water ranges from 0 to 5.55 pg-TEQ/L. The TEQ is lower than the environmental quality standard (1 pg-TEQ/L) set by the Japanese government [15] at sites other than Y-26, Y-27, Y-28, Y-29, and Y-30. The sites in the Jiangsu province have higher TEQ of PCBs in water, and thus the ecotoxicological risk in the lower reaches of the Yangtze River.

The carcinogenic and non-carcinogenic health risk assessment models were also applied [40]. The carcinogenic and non-carcinogenic risk of PCBs in water varies from 3.87×10^{-11} to 9.67×10^{-9} and from 6.78×10^{-8} to 1.69×10^{-5} , respectively. The carcinogenic risk of PCBs is much less than the risk threshold (1×10^{-6}) defined by the US EPA and thus negligible. Because the PCBs are harmful to humans when the non-carcinogenic risk exceeds 1, the PCBs in water do not show a non-carcinogenic risk either.

Conclusions

In this study, the PCBs of water, sediment, and suspended particulate matter (SPM) in the drinking water source of the Yangtze River were determined with isotope dilution HRGC/HRMS. The Σ PCBs concentration ranges in 0.044–10.98 ng/L, 0.33–69.43 ng/g, and 0.72–152.66 ng/L in water, sediment, and SPM, respectively. Compared with other regions, the Yangtze River has a low to medium level of PCBs pollution.

In general, for both water and sediment, lower chlorinated biphenyls are the dominant congeners in the middle reaches, and higher chlorinated biphenyls dominate in the lower reaches. Among all congeners, PCB11, 17, 18, 28, and 118 have higher detection frequency in water, sediment, and SPM. The distribution of PCBs varies across the three phases. The SPM mainly contains PCB47, 41, and 51, whereas the sediment mainly contains PCB138, 110, and 70.

The value of ffSM indicates that the diffusion of PCB18, 28, 47, and 118 in sediment and water is away from equilibrium. The results show that PCB18 and 28 have net redissolution from sediment to water,

while PCB47 and PCB118 have net sorption from water to sediment. Risk assessment results indicate potential ecological risk from PCBs in the water and sediment of the Yangtze River, but neither carcinogenic nor non-carcinogenic risk was found from the PCBs.

Abbreviations

PCBs Polychlorinated biphenyls

SPM particulate matter

TEQ toxic equivalent quantity

ind-PCBs indicative polychlorinated biphenyls

co-PCBs coplanar polychlorinated biphenyls

QC quality control

QA quality assurance

RSD relative standard deviation

PCA Principal component analysis

NOM natural organic matter

OCPs organochlorine pesticides

SQG sediment quality guidelines

ER-L range-low

ER-M range-median

PCDD/Fs polychlorinated dibenzo-*p*-dioxins/dibenzofurans

Declarations

Acknowledgements

None applicable.

Authors' contributions

XZ , XC and JD were involved in the experiments and manuscript writing. ZH was responsible for the data analysis. CL, XQ and XW collected the samples. BZ and JS contributed to correction of the manuscript.

All authors read and approved the final manuscript.

Funding

This work was supported by the Water Pollution Control and Management (No. 2018ZX07502-001-005).

Competing interests

The authors declare that they have no competing interests.

Availability of data and materials

The datasets obtained and analyzed in the current study are available from the corresponding author on reasonable request.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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Figures



Figure 1

Sampling sites along the Yangtze River. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

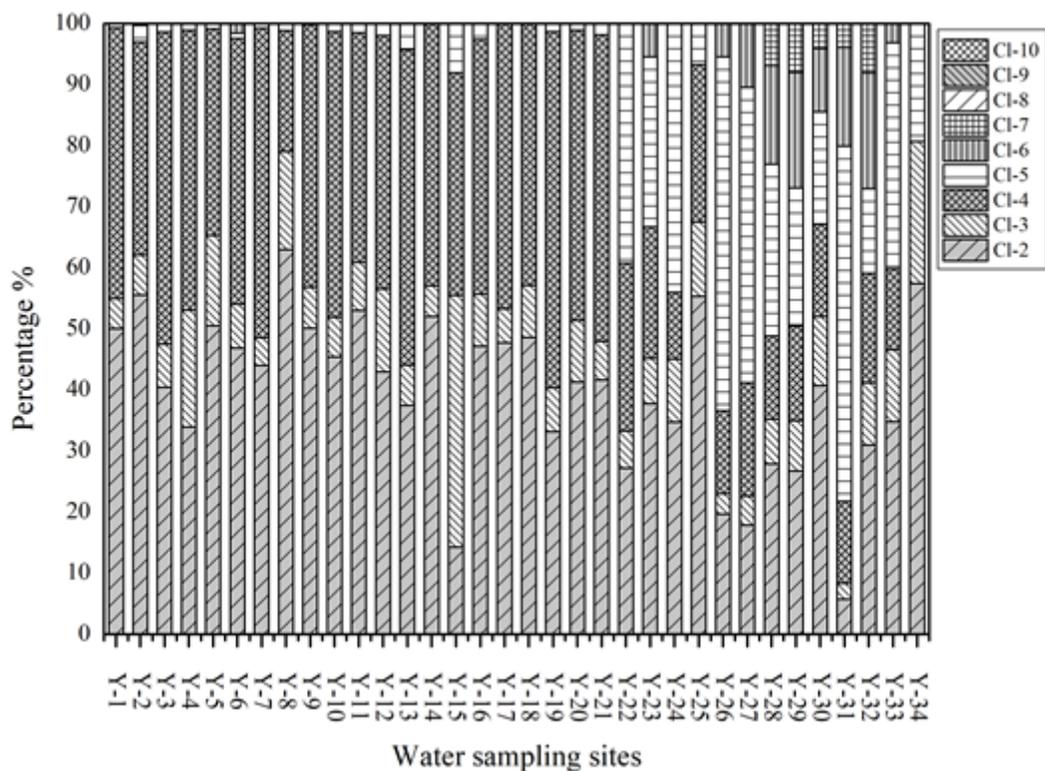


Figure 2

The percentage of PCB homologues in the water of the Yangtze River.

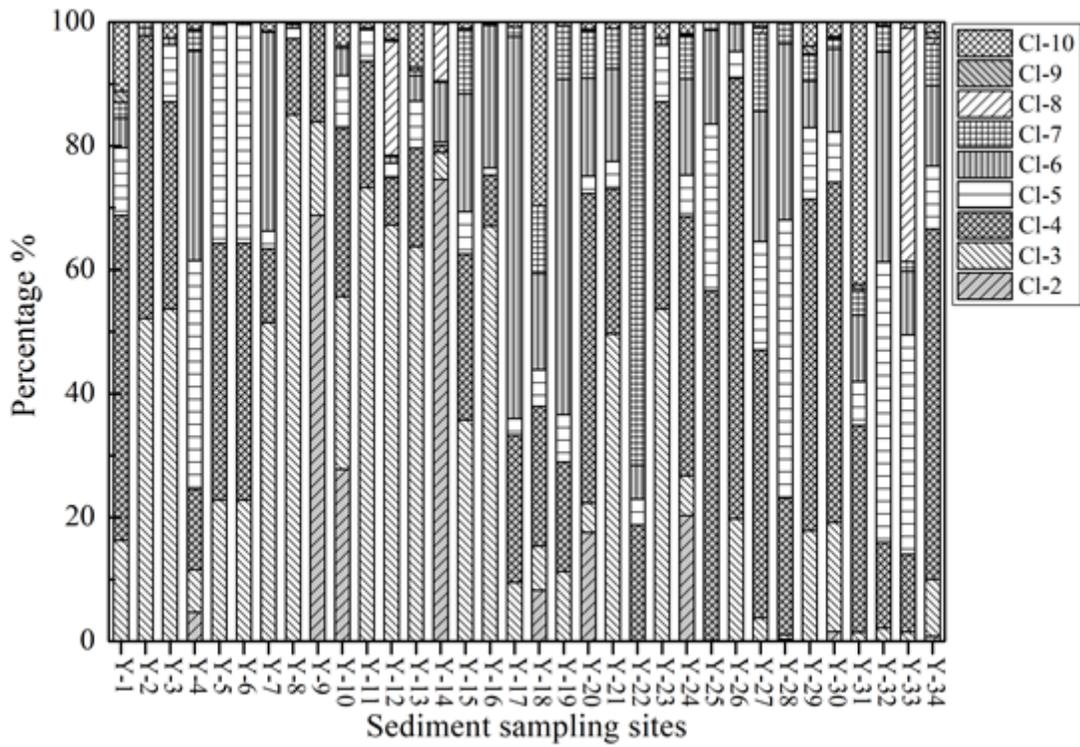


Figure 3

The percentage of PCB homologues in the sediment of the Yangtze River.

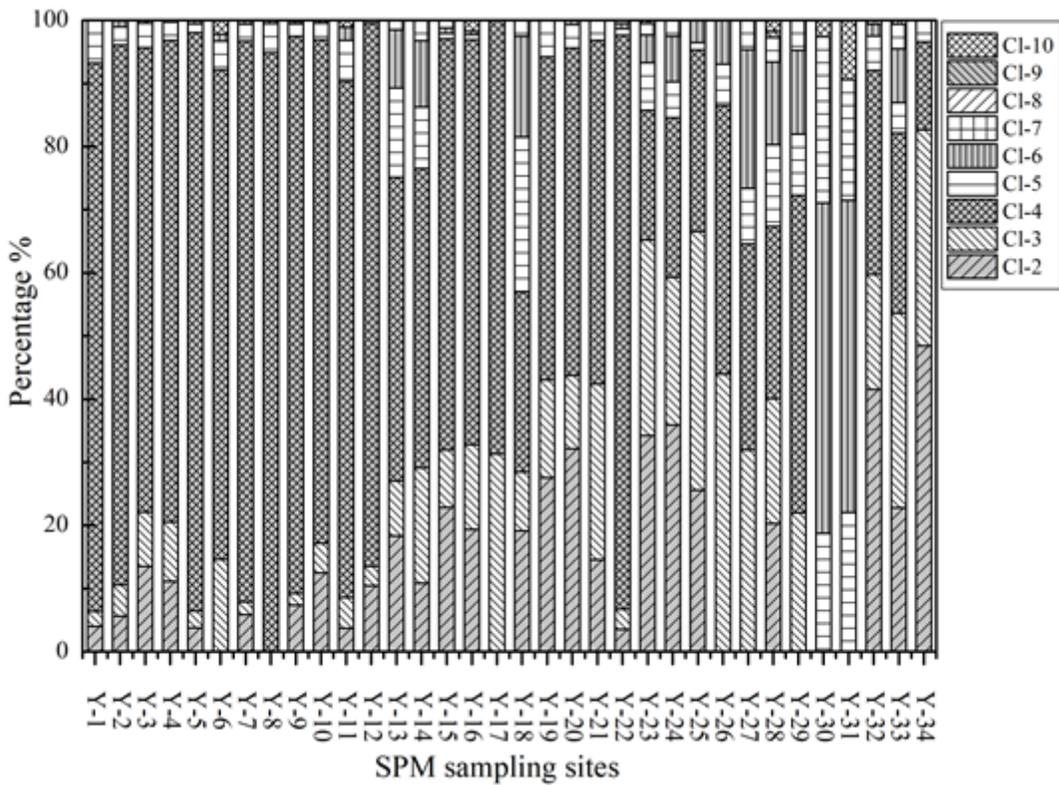


Figure 4

The percentage of PCB homologues in the SPM of the Yangtze River.

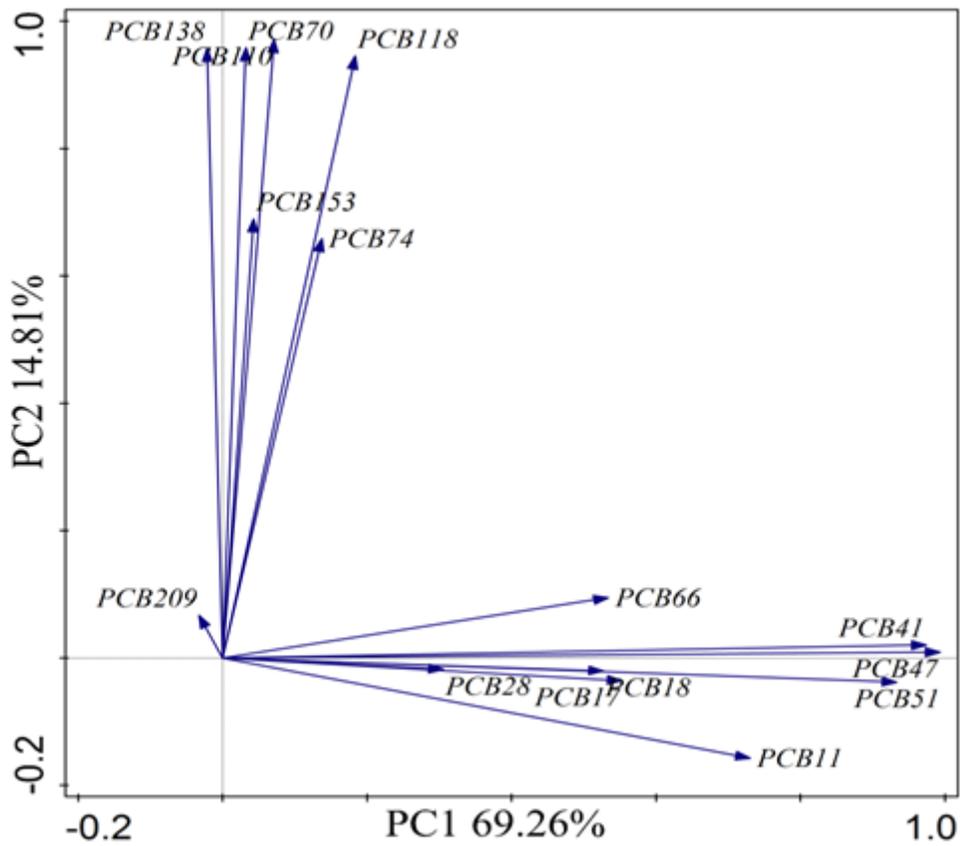


Figure 5

Principal component profile of PCBs in the water, sediment, and SPM of the Yangtze River.

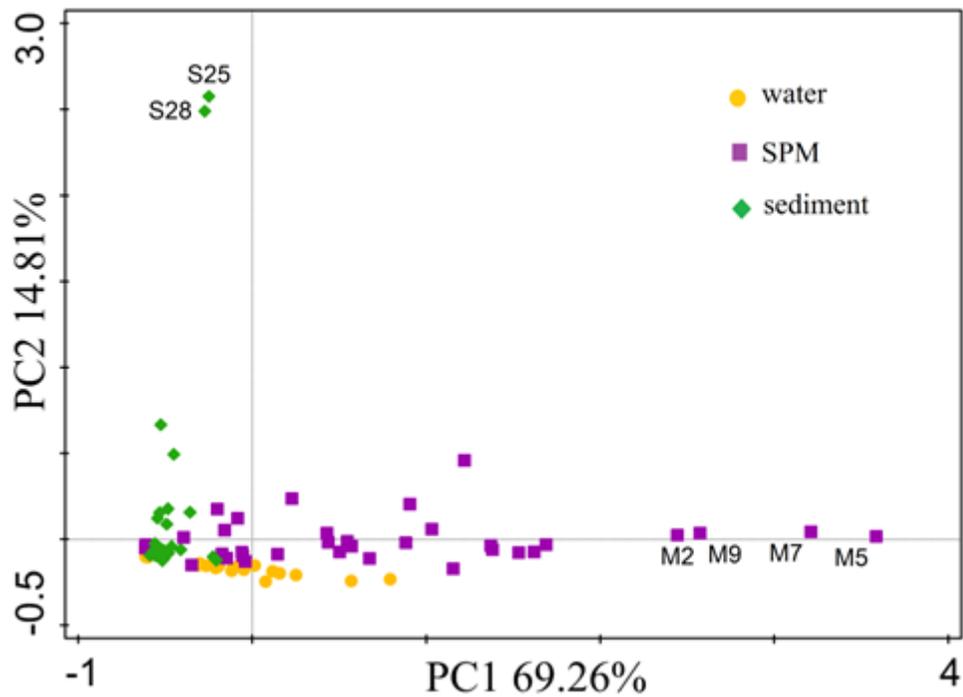


Figure 6

PCA score plot of PCBs in the water, sediment, and SPM of the Yangtze River.

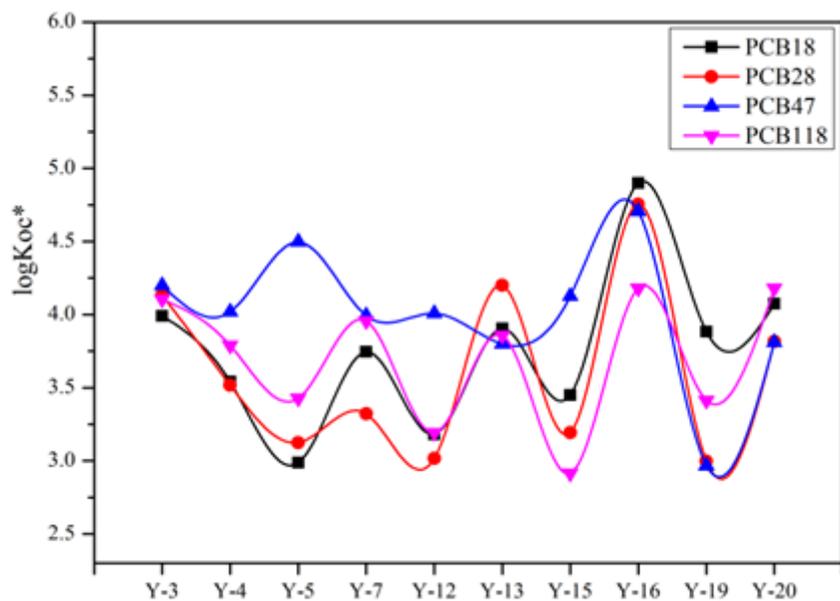


Figure 7

The logKOC* values of PCB18, 28, 47, and 118 in the Yangtze River.

Supplementary Files

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