Lin *et al. Carbon Research* (2023) 2:15
https://doi.org/10.1007/s44246-023-00046-4

Carbon Research

ORIGINAL ARTICLE

Open Access

Source contribution of PCBs and OC/EC at a background site in the Yangtze River Estuary: insight from inter-comparison by Positive Matrix Factorization (PMF)

Tian Lin[1*], Wanqing Zhou[1], Shizhen Zhao[2], Minqiao Li[3] and Zhigang Guo[4]

Abstract

Background air samples, including gas-phase components and suspended particulates, were collected over one year (2013-2014) for an investigation of polychlorinated biphenyls (PCBs) and OC/EC in the Yangtze River Estuary. PCB concentrations exhibited great seasonal variability and ranged between 43 and 720 pg·m^{-3}. They mainly were associated with the gas phase, and levels peaked in the summer time at 327 ± 177 pg·m^{-3}. By contrast, concentrations of particulate PCBs and OC/EC were higher in the cold seasons, which was due to high-temperature combustion emission and frequent haze events. According to the results of a positive matrix factorization, the combustion and non-combustion sources of PCBs accounted for 30% and 70% of total PCBs, respectively. Meanwhile, SOC/OC value was 54.7% ± 20.1%, which suggests gas-to-particle conversion process plays a significant role in contributing to atmospheric particles. To this end, the influence of OC/EC on both combustion and non-combustion PCBs in the long-range atmospheric transport deserves further research.

Highlights

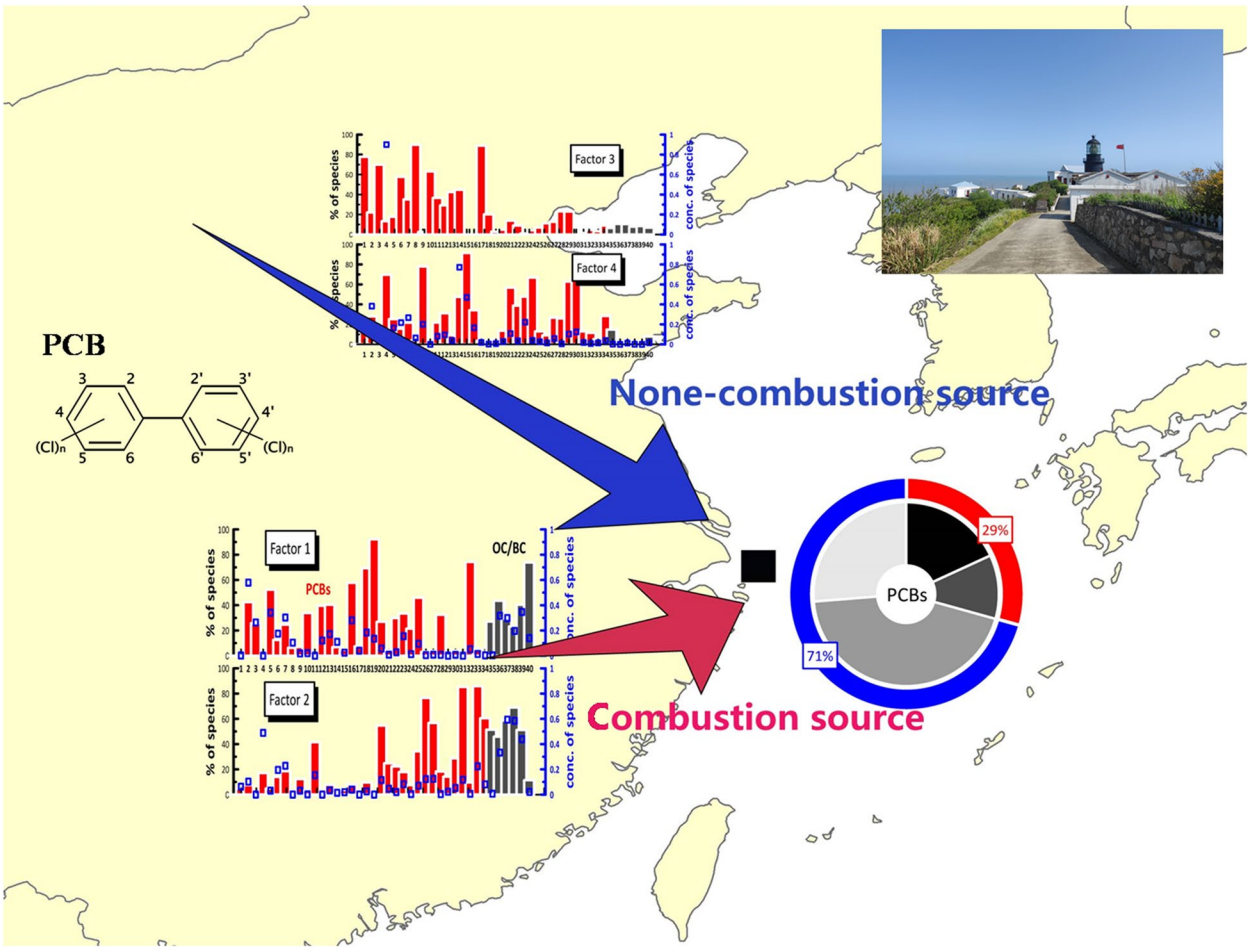
- Concentrations of particulate PCBs and OC/EC were higher in the cold seasons.
- An approach was proposed to estimate the source of PCB combustion with OC/EC.
- The combustion source of PCBs accounted for 30% of total PCBs.

Keywords Polychlorinated biphenyls, Organic carbon, Element carbon, Positive matrix factorization, Yangtze River Estuary

Handling Editor: Fengchang Wu.

*Correspondence:
Tian Lin
lintian@vip.gyig.ac.cn
Full list of author information is available at the end of the article

Graphical Abstract



1 Introduction

Polychlorinated biphenyls (PCBs) are a class of synthetic organic compounds with 209 isomers according to the number and location of chlorine atoms on the benzene ring (Zhao et al. 2019). PCBs have the advantage of chemical stability, insulation and nonflammability. Hence, PCBs are widely used in chemical industry and commerce, such as heat transfer fluids, plasticizers, rubber, plastic products and flame retardants (Erickson and Kaley 2011). Although PCBs have been restricted worldwide since the 1970s, they could still be transferred in different media through sedimentation, migration, and re-volatilization, especially in estuary and coastal zones where land–ocean interactions occur (Hu et al. 2011; Cabrerizo et al. 2013; Guo et al. 2020). In recent years, a growing number of researchers have suggested that atmospheric PCB levels might have increased, and unintentionally produced PCBs (UP-PCBs) have been shown to be the source of the high concentrations of PCBs in some Chinese urban areas (Wu et al. 2019). Zhao et al. (2017) have demonstrated that the UP-PCBs could become the main emission source of PCB pollution in China from 2035. Thus, the distinction and quantification of UP-PCBs separated from IP-PCBs (intentionally produced PCBs) are

particularly important. Major sources of UP-PCBs include industrial thermal processes (e.g., steel production, cement, waste incineration, etc.), burning of fossil and biomass fuels, as well as dye production emissions and e-waste dismantling (Liu et al. 2013). Combustion-associated emission has been considered as an important source of UP-PCBs.

Organic carbon (OC) and elemental carbon (EC) are important components of atmospheric particulate matter. EC mainly derives from incomplete combustion of biomass and fossil fuels. EC is a kind of black organic substance with graphite structure generated directly at high temperature (Fang et al. 2016), and a group of PCBs are generated simultaneously (Sajjadi et al. 2019). This means that the EC and PCBs can provide useful information to the pollutant source (Fang et al. 2016). In addition, the concentration of air PCBs will be largely influenced by gas-particle partitioning resulting from the long-range transport of contaminated air mass containing high levels of OC or EC in aerosols. Previous studies focused on identification and contribution of PCB sources by positive matrix factorization (PMF) (Saba and Su 2013; Capozzi et al. 2023). Since PCBs share similar sources with OC/EC, and the two pollutants have a strong association in atmospheric transport, new insights can be inferred from inter-comparison by PMF to distinguish the combustion sources of PCBs in atmosphere. Besides, it can provide in-depth results to understand the regional geochemical behavior between PCBs and OC/EC in the atmosphere.

The PMF model was originally used as a receptor model for source resolution and has been continuously improved and optimized in applications (Hopke 2016). Its advantage is that the relative contribution of one or more groups of compounds from the specific source categories can be quantitatively estimated. PMF model is widely used for source apportionment of pollutants, e.g. polycyclic aromatic hydrocarbons, PCBs. In this study, the concentrations of PCBs and OC/EC present in the Yangtze River Estuary (YRE), coastal East China Sea (ECS) atmosphere were determined. The outcomes of the PMF analyses enabled the identification of source and quantitative pollution contribution of combustion and non-combustion, which is helpful for our understanding of UP-PCBs and IP-PCBs.

2 Materials and methods

2.1 Sampling

The sampling site was located at Huaniao Island (HND, a point of 30.86°N, 122.67°E with an area of 3.28 km^2) in Zhoushan, Zhejiang Province (Fig. 1). The island is dominated by fishing, breeding and tourism, with little agricultural or industrial activity. The Yangtze River Delta, which is connected with the ECS, is a highly developed urban agglomeration of urbanization and industrialization in China. Within this context, the YRE and coastal ECS receive extensive hydrocarbon pollutant discharge and emission from their industrialized/urbanized surroundings, particularly through Yangtze River runoff and atmospheric transport (Qi et al. 2014; Wang et al. 2017a). A detailed analysis of polybrominated diphenyl ethers, polycyclic aromatic hydrocarbons, dichlorodiphenyltrichloroethane, hexachlorocyclohexane levels, temporal trends, and source/fate from the fixed-point sampling campaign was reported previously (Guo et al. 2020; Jiang et al. 2018; Li et al. 2015, 2017).

Samples were collected by the medium-volume atmospheric sampler from Guangzhou Mingye Technology Company, and the sampling velocity was 300 L/min. Quartz filters (issuquartz-2500QA-UP, Pall) was used to collect total suspended particulate components (TSP). The filter was wrapped in aluminum foil and burned in a muffle furnace at 450℃ for 4 h to remove volatile components. Polyurethane foam plugs (PUF, 8.0 cm length, 6.25 cm diameter, 0.035 $g{\cdot}cm^{-3}$ density) were used to collect gaseous components. During the sampling period, 94 pairs of air samples were collected, namely that is 20 Oct-12 Nov 2013 ($n=24$), 22 Dec 2013-14 Jan 2014 ($n=23$), 27 Mar-18 Apr 2014 ($n=23$), and 29 Jul-26 Aug 2014 ($n=24$), and the invalid samples caused by power outages and sampler failure were removed. The sample collection period lasted from 8:30 a.m. on the first day to 8:00 a.m. on the second day, with continuous sampling for about 23.5 h. Two blanks filters were obtained in every quarter. Field blank filters were collected by loading and unloading in the sampler without running. After sampling, the quartz filter was folded in half to prevent the particles drop. The quartz filter and PUF were wrapped in original aluminum at -4℃ until analysis.

2.2 Sample extraction of PCBs

The mixture of 13C-labled PCB-52 and PCB-209 was added as the recovery indicator of PCB and each sample was extracted with dichloromethane (DCM) for 48 h. The extraction was concentrated to 1 ~ 2 mL by the rotary evaporator, solvent exchange and then concentrated by n-hexane. The concentrate flowed through the chromatographic column (8 mm internal diameter). The column was filled with alumina (3 cm), silica gel (3 cm), 50% (by weight) sulfuric acid silica (2 cm), and anhydrous sodium sulfate (1 cm) from the bottom to the top. The PCB fraction was eluted with 50 mL DCM/ n-hexane (1:1 v/v) mixture solution and transferred to brown cell bottles, and then concentrated to 0.2 mL by N_2, finally transferred to 1.5 mL cell flask.

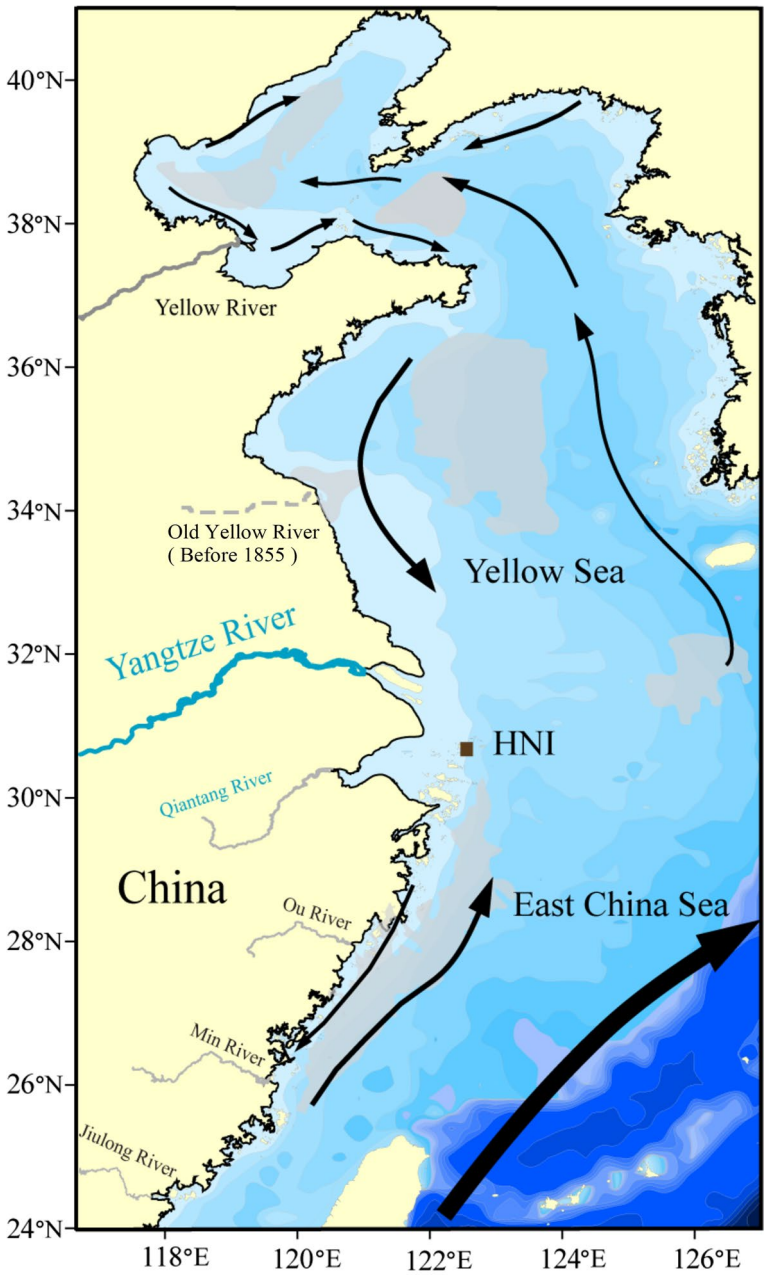


Fig. 1 Map of sampling area (black arrows represent ocean currents)

2.3 The analysis of PCBs

All concentration analyses of PCB were conducted using gas chromatograph (GC, HP-5890 Series II, Agilent, USA) with HP-5 capillary column (50 m × 0.25 mm × 0.25 μm). The initial temperature of GC heating procedure held at 100℃ for 1 min. The oven rose from 1.8℃/min to 170℃ and held for 3 min, rising to 226℃ at 2.5℃/min, and then was ramped to 280℃ at 4℃/min, and kept for 10 min.

In this study, the PCB target compounds determined included the following: PCB17, PCB18, PCB31/28, PCB52, PCB49, PCB44, PCB74, PCB70, PCB95, PCB101, PCB99, PCB87, PCB110, PCB82, PCB151, PCB149, PCB118, PCB153, PCB138/158, PCB187, PCB183, PCB128, PCB177, PCB171, PCB156, PCB180, PCB191, PCB170, PCB201, PCB208, PCB195, PCB194, PCB205 and PCB206.

2.4 The analysis of OC/EC

The Thermal/Optical Carbon Analyzer (DRI Model 2015) was used to detect OC and EC by thermal optical

reflectance (TOR). OC and EC require different oxidation reaction temperature and accordingly the 0.5026 cm^2 even granular sample membrane was heated by IMPROVE method. The specific setting procedure of the DRI Model 2015 was as follows. Firstly, the sample was heated to 120℃, 250℃, 450℃, 550℃ under pure helium and four OC fractions (OC1, OC2, OC3, OC4) were determined separately. Secondly, three EC fractions (EC1, EC2, EC3) were produced in the helium atmosphere with 2% oxygen at 550℃, 700℃ and 800℃, respectively. The 635 nm laser was used to illuminate the sample. The starting point of EC was set when the intensity of the reflected light returned to the initial state, thus accurately defining the pyrolyzed carbon (OP). According to the IMPORVE protocol, OC = OC1 + OC2 + OC3 + OC4 + OP, EC = EC1 + EC2 + EC3-OP (Fang et al. 2018; Han et al. 2007).

2.5 Quality assurance and quality control

The quality assurance and quality control were carried out with recoveries for PCB measurement. Field blanks, laboratory blanks and spiked blanks were analyzed with the same method as sample analysis. No target compounds were detected in field blanks and laboratory blanks. Spiked blanks were preformed after every ten samples. The recovery rates were 73 ± 28% and 88.0 ± 8.5% for the blank and spiked blank samples, respectively.

Before each analysis, the Bake procedure was executed until TC concentration of blank filters was below 0.2 μg C/cm^2. Thereafter, the AutoCalib method was run to test the stability of instrument. The relative standard deviation of OC3, EC1 and Calibration peak area was calculated and ensured to be less than 5%. After the instrument was stable, a certain amount of potassium hydrogen phthalate was injected to calculate the recovery rate to ensure that the value is above 95%. Two field blank filters were collected each quarter. The average blank filter was deducted as the background value when calculating the content of OC and EC in the sample. One replicate analysis was performed after every ten samples. The relative standard deviation of OC and EC was controlled within ± 5%.

2.6 PMF analysis

Source resolution of pollutants was performed using the version 5.0 PMF receptor model developed by the US Environmental Protection Agency, and source contribution rates were provided for the time series. In this study, a dataset consisting of 94 TSP samples × (34 PCBs + 4 OCs + 2 ECs) was prepared, excluding EC3 with concentration of most samples below the detection limit. Before the analysis, undetectable values (null values) were calculated using 1/2 minimal detectible level.

3 Results and discussion

3.1 Concentrations and profile of PCBs

HNI is a remote site in the YRE with a potential influence from urban centers, e.g., Shanghai and Hangzhou. Even the westerly and East Asian Monsoon can influence the atmospheric transport of air PCBs in YRE, especially in spring and winter (Zhang et al. 2011). Figure 2 shows the seasonal variation trend of PCB concentration in atmospheric gas and particle phase at the HNI site in the YRE. The air concentrations of 34 PCB congeners were 43 ~ 720 pg·m^{-3}, with the average of 180 pg·m^{-3} and the median of 140 pg·m^{-3}. The median level of PCBs in this study was significantly higher than that at the regional background CAWNET site located geographically near the center of China, where the air concentrations of the 32 PCBs ranged between 3.40 and 115 pg·m^{-3}, with an average of 39.4 ± 27.1 pg·m^{-3} (Zhan et al. 2017). Nevertheless, the median level concentration found in urban center zones in China was higher than that in this study, with the former concentration higher by magnitudes of up to several thousand (Cui et al. 2017a). The existence of point-source emissions (including e-waste recycling stations and industrial emissions) and secondary release from historical usage could lead to higher concentrations of total PCBs in urban areas of northern and eastern China (Cui et al. 2017a, b).

The levels of PCBs at HNI were compared with those measured at remote/background/rural sites around the globe between 2010 and 2019 (Table 1). PCB concentrations in the atmosphere at HNI were higher than those in Arctic regions and Antarctica (Hao et al. 2019; Li et al. 2012; Wang et al. 2017b). This might be attributable to higher concentrations of anthropogenic source pollutants from the adjacent areas. Polar areas are always deemed places much less disturbed by humans. The lowest concentration in our study (43 pg·m^{-3}) was also higher than the PCB concentrations in open oceans (Pegoraro et al. 2016; Wang et al. 2014) and the equatorial Indian Ocean (Huang et al. 2014). Longer distances from pollution sources could have resulted in enhanced washing, precipitation, and degradation of PCBs, resulting in the lower concentrations observed in the open ocean. However, the values of the present study were lower than the concentrations reported in the atmosphere at southwestern Lake Michigan, Sleeping Bear, and Sturgeon Point, USA, which are more similar to sites having some urban influence (Boesen et al. 2020).

According to the different number of chlorine atoms, the composition of PCBs homologue was analyzed, as shown in Figure S1. Tri- and tetra-CBs were the dominant homologues in the samples, in total comprising 72.7% and 73.4% of the samples, respectively. This congener composition profile was similar with those reported that the major

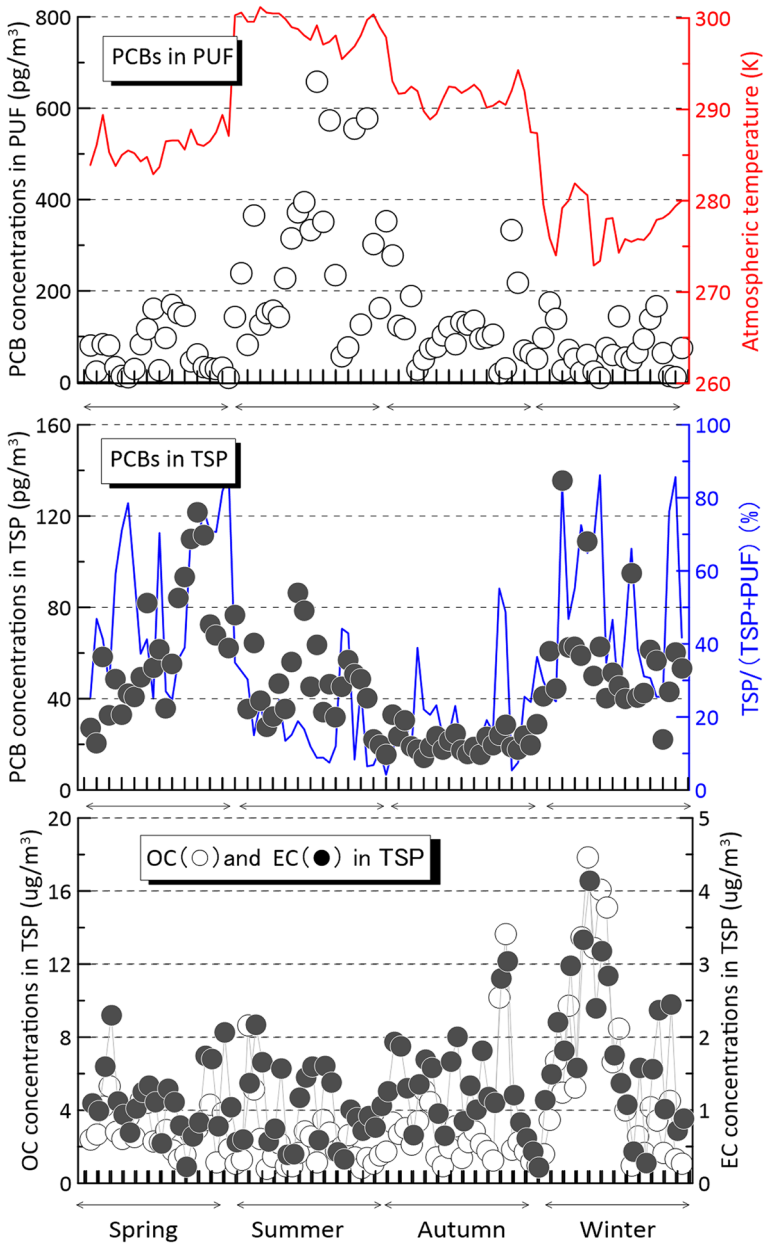


Fig. 2 Concentration variations of PCBs (pg·m^{-3}) and OC/EC (μg·m^{-3}) in the atmosphere of HNI

congeners were tri- and tetra-CBs from Chinese background site soils (Li et al. 2011; Ren et al. 2007). Tri-CB was used primarily in the production of electronic equipment. Higher-chlorinated PCBs can be released during the combustion of coal and wood (Lee et al. 2005).

3.2 Seasonal variation of PCBs and influence of temperature

The gas/particle partitioning of pollutants is affected by temperature, especially gaseous PCB concentrations. In this study, a significant increase in the proportion of gas-phase PCBs was observed in summer and autumn (up to ~ 90%). Studies have shown that higher temperatures will promote the re-volatilization of pollutants in water and soil, and promote the conversion of PCBs from granular to gaseous. When the proportion of particle phase to total concentration of PCBs is analyzed, it exhibits a greater shift from gas phase to particle phase under colder ambient temperatures.

Table 1 Total PCB (∑PCB) concentrations in the atmosphere ($pg \cdot m^{-3}$)

Location	Sampling period	N[a]	∑PCBs	Reference
King George Island, Antarctica	Summer, 2009–2010	20	1.66–6.50	(Li et al. 2012)
Chinese Great Wall Station, West Antarctica	Jan 2011–Jan 2014	20	5.87–72.7	(Wang et al. 2017b)
Arctic	2017–2018	19	10.8–26.9	(Hao et al. 2021)
North Pacific to Arctic Ocean	Jul–Sep 2012	26	7.97–67.66	(Wang et al. 2014)
Equatorial Indian Ocean	Apr–May 2011	21	2.0–29	(Huang et al. 2014)
Near-coast South Atlantic Ocean	Oct 2014	50	9.56–130	(Pegoraro et al. 2016)
Southwestern Lake Michigan, USA	Sep 2010	209	190–1,100	(Boesen et al. 2020)
Huaniao Island	Oct 2013–Aug 2014	36	43–720	This study

[a] Number of PCB congeners

To assess the influence of temperature-affected gas–particle phase partitioning quantitatively, the following model was used (Trinh et al. 2018):

$$log\ Kp = mp * logP_L^0 + bp, \quad (1)$$

where the slope (m_p) and intercept (b_p) are fitting constants obtained from formula (1).

Sub-cooled liquid vapor pressure ($P_L{}^0$) was determined by temperature (T) and calculated from a pair of estimated values of Ap and slopes Bp of log-linear relationships according to the following equation (Paasivirta et al. 1999):

$$log\ P_L^0 = A_p + B_p/T. \quad (2)$$

For total PCBs, a slope that deviates from −1 indicates a non-equilibrium state of gas–particle partitioning (mp = −0.4, R^2 = 0.1). This disequilibrium is considered to be independent of temperature, and is frequently observed in the urban environment. Possible explanations for this observation include fresh emissions from the coastal area. Or a sustained air–water exchange of PCBs from river runoff water due to the higher ambient temperature or wind speed affects the gas–particle partition at HNI (Guo et al. 2020). However, a slope that approaches −1 was observed for several PCBs (PCB, -99, -101, and -180). It suggests that these high chlorinated PCBs can reach equilibrium in a short period of time.

3.3 OC and EC concentrations and seasonal variability

The temporal variation of carbon aerosol concentration in TSP from autumn 2013 to summer 2014 is shown in Fig. 2. During the sampling period, the annual mean concentrations of OC and EC were 3.73 ± 3.45 and 1.22 ± 0.79 $\mu g \cdot m^{-3}$, respectively, and the concentration of OC was about three times that of EC. The concentrations of both showed the lowest in summer and the highest in winter. The concentrations of OC in summer and winter were 2.28 ± 1.64 and 6.48 ± 5.11 $\mu g \cdot m^{-3}$, and correspondingly EC were 0.87 ± 0.5 and 1.79 ± 1.01 $\mu g \cdot m^{-3}$. The increase in TSP concentration caused by frequent dust in spring did not significantly increase the concentration of OC and EC, which may be related to the fact that OC and EC mainly exists in fine particulate matter (Wang et al. 2006, 2014), which are concentrations of OC and EC in atmosphere at HNI were compared with those measured sea/mountain/urban sites across the globe (Table 2). Generally, OC/EC concentrations in the atmosphere of HNI were much lower than those in urban areas, which were more affected by human activities. That might be due to atmospheric pollutants emitted by the combustion of coal, automobile exhaust and resident's daily energy consumption which are concentrated in urban area. The carbonaceous aerosol pollution level of HNI is close to that of Bohai Sea, the Yellow Sea and other open sea areas, and slightly lower than that of the Arabian Sea. Sea/mountain sites, often used as a background point, are far away from urban and thus are less affected by direct emissions.

Pearson correlation coefficient (R) can be used to analyze the correlation between different components, and can preliminarily judge the stability and consistency of carbon components to some extent (Turpin and Lim 2001). In this study, the R value of OC and EC was 0.8377, which is between 0.8 and 1.0, indicating a strong correlation. The OC/EC value was 2.96, 2.61, 2.70 and 3.62 in spring, summer, autumn and winter, respectively, showing the lowest in summer and the highest in winter. OC/EC values of 1.0–4.2 suggests carbon components mainly derived from motor vehicle exhaust emission (Saarikoski et al. 2008; Schauer et al. 2022). Differently, the OC/EC ratios in some areas were extremely high, e.g., the OC/EC ratio reached 7.04 ± 2.30 in Qingdao, which was considered to be more contributed by biomass combustion and coal combustion sources. With the exception of urban pollution in the YRE, there are almost no indigenous sources of carbon aerosols on HNI each year. Abundant rain in summer can remove carbon particles with

Table 2 Carbonaceous species concentration in TSP (unit: μg·m^{-3})

Location	Sample quantity (period)	OC	EC	OC/EC	Reference
Lumbini, Nepal	156 (Apr 2013-Dec 2015)	32.1 ± 21.7	6.44 ± 3.17	4.82 ± 2.27	(Chen et al. 2019)
Lanzhou, China	40 (Dec 2015-Nov 2016)	25.4 ± 13.0	6.7 ± 3.52	3.79 ± 1.60	
Mardan, Pakistan	68 (Jun 2015-Dec 2016)	44.7 ± 32.1	11.7 ± 5.39	3.68 ± 1.47	
Delhi, India	47 (Jan-Feb 2019)	63 ± 38	16 ± 8	2.1–5.9	(Jangirh et al. 2022)
Kathmandu, Nepal	24 (Mar-May 2018)	57.15 ± 30.05	12.51 ± 5.77	4.64 ± 1.73	(Bhattarai et al. 2022)
Ioannina, Greece	60 (Jan-Feb 2013)	25.37 ± 27.06	4.14 ± 3.67	5.98 ± 2.58	(Kaskaoutis et al. 2020)
Heraklion, Greece		3.86 ± 2.99	2.29 ± 2.40	2.03 ± 1.05	
Karachi, Pakistan	113 (Feb 2015-Mar 2017)	37.2 ± 28.0	8.53 ± 6.97	4.20 ± 2.50	(Chen et al. 2020)
Dushanbe, Tajikistan	62 (Sep 2018-Aug 2019)	11.9 ± 3.0	5.13 ± 2.24	-	(Chen et al. 2021)
Bolu Mountain, Turkey	42 (24–29 Jun 2018)	31.3 ± 11.5	27.1 ± 13.1	-	(Karşı et al. 2020)
the Arabian Sea	17 (6–24 Dec 2018)	6.7 ± 3.3	1.4 ± 0.5	4.9 ± 1.8	(Bikkina et al. 2020)
Shanxi Province, China	28 (Sep 2015)	16 ± 8	2.2 ± 1.0	-	(Du et al. 2020)
Qingdao, China	27 (5 Oct 2015–25 Jun 2016)	19.28 ± 5.33	-	7.04 ± 2.30	(Ding et al. 2019)
the Yellow Sea, China	8 (29 Jun-20 Jul 2016)	1.84 ± 0.93	-	4.34 ± 3.05	
Bohai Sea, China	4 (29 Jun-20 Jul 2016)	2.50 ± 0.43	-	5.63 ± 1.99	
Huaniao Island	94 (October 2013–August 2014)	3.62 ± 3.50	1.33 ± 0.08	2.65 ± 1.36	This study

strong hydrophilicity from the atmosphere, thus reducing the OC/EC concentration in the atmosphere (Safai et al. 2014). Besides, the size of OC particles is larger than that of EC, and the rainwater removal efficiency is greater than that of EC. In winter, northern China is heated by the burning of fossil fuels, and air masses carried by northwest winds over HNI carry carbon pollutants. The contribution of low-temperature combustion of wood and coal in winter will increase the OC/EC ratio to a certain extent. As a result, the level of atmospheric particulate pollution is the lightest in summer and the worst in winter.

3.4 Source apportionment according to the PMF model

The plots of the temperature and gas-phase concentrations indicated that gaseous PCB concentrations increased with air temperature. However, as discussed above, the shallow slope and low R^2 ($R^2 = 0.1$ weak linear correlations between log Kp and log P_L^0) suggest that there were other influencing factors. Meanwhile, no correlation between PCBs and EC in TSP was found. Thus, we introduced PMF to investigate the sources and potential factors influencing seasonal variation. The factors generated by the PMF model are shown in Fig. 3, along with their time trends of percent contributions to total PCBs (Fig. 4).

Factor 1 was highly loaded with PCB-138, -153, -101, -28 and explained 18% of the total variance. As ones of seven indicator PCBs, they have been frequently studied in China since they are the main monitoring pollution associated with PCB-containing materials and predominant PCB products used in the past. Other higher loading of typical Aroclor congeners included PCB-17 and PCB-49. Zhao et al (2019) pointed out that very similar to the survey of e-waste sources sampling in Taizhou and Qingyuan, e-waste was the main contribution (~ 60%) of combustion PCBs in the atmosphere, especially for indicator PCBs. Referring to seasonal variation characteristics of factor contribution, the main contribution appeared in summer and autumn. This is consistent with the previous research results of polybrominated diphenyl ethers from HNI, which were largely input by the air masses coming from or passing over the Southeast coast of China (Li et al. 2015). More importantly, as an indicator of high-temperature combustion, EC2 accounted for up to 70% in the factor. Thus, Factor 1 was derived from combustion produced PCBs during the high temperature dismantling of e-waste.

Compared with the other factors, Factor 2 mostly consisted of moderately and higher-chlorinated PCBs. In addition, PCB187 and 205 are major indicators for coal combustion (Kim et al. 2004). In summer and winter, Factor 2 showed a higher contribution. In cold season, regional central heating mainly use coal combustion to provide energy. According to Lee et al. (Lee et al. 2005), high chloring substituted PCBs will be released during the combustion of coal and wood. In North and East China, man-made straw burning and natural deep forest fires often occur in summer (Cui et al. 2017a). Furthermore, the overall contribution of OC1-OC4 in this factor was as high as 45–68%, which also supported "Factor 2" as a low-temperature combustion source of PCBs and accounted for 11% of the total PCBs in the sample dataset in this study.

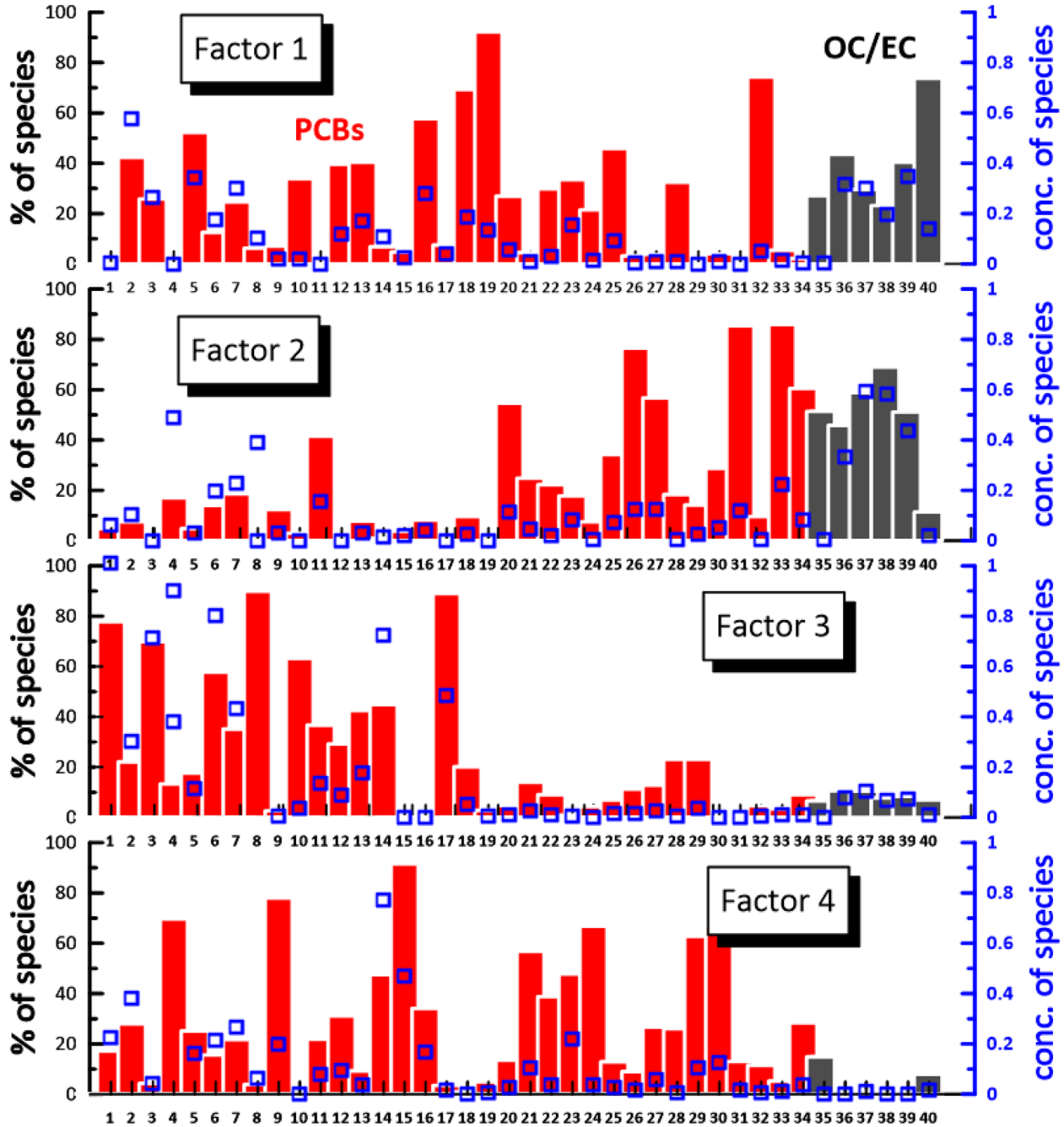


Fig. 3 Four-factor loadings by PMF analysis from PCBs and OC/EC data of 94 TSP samples from the HNI (No.1–40 refer to PCB-17, 18, 31/28, 52, 49, 44, 74, 70, 95, 101, 99, 87, 110, 82, 151, 149, 118, 153, 138/158, 187, 183, 128, 177, 171, 156, 180, 191, 170, 201, 208, 195, 194, 205, 206, unit: pg·m^{-3} and OC1, OC2, OC3, OC4, EC1, EC2, unit: μg·m.$^{-3}$)

Totally different with Factor 2, Factor 3 was highly loaded with lower-chlorinated congeners (tri- and tetra-homologue groups). Cui et al. (Cui et al. 2017a, b) pointed out the di- and tetra-CBs may be derived from UP-PCBs. Besides, contributions of Factor 3 exhibited lower frequency variations with clear pollution episodes, with the exception of springtime (accounting for 80% of whole year). The frequency of sand-dust event in spring is the highest, approximately accounting for 87% of the annual frequency, while 11% occurs in winter (Kim 2008). Obviously, the high atmospheric loading for lower-chlorinated PCBs was related to the long-range transport of desert

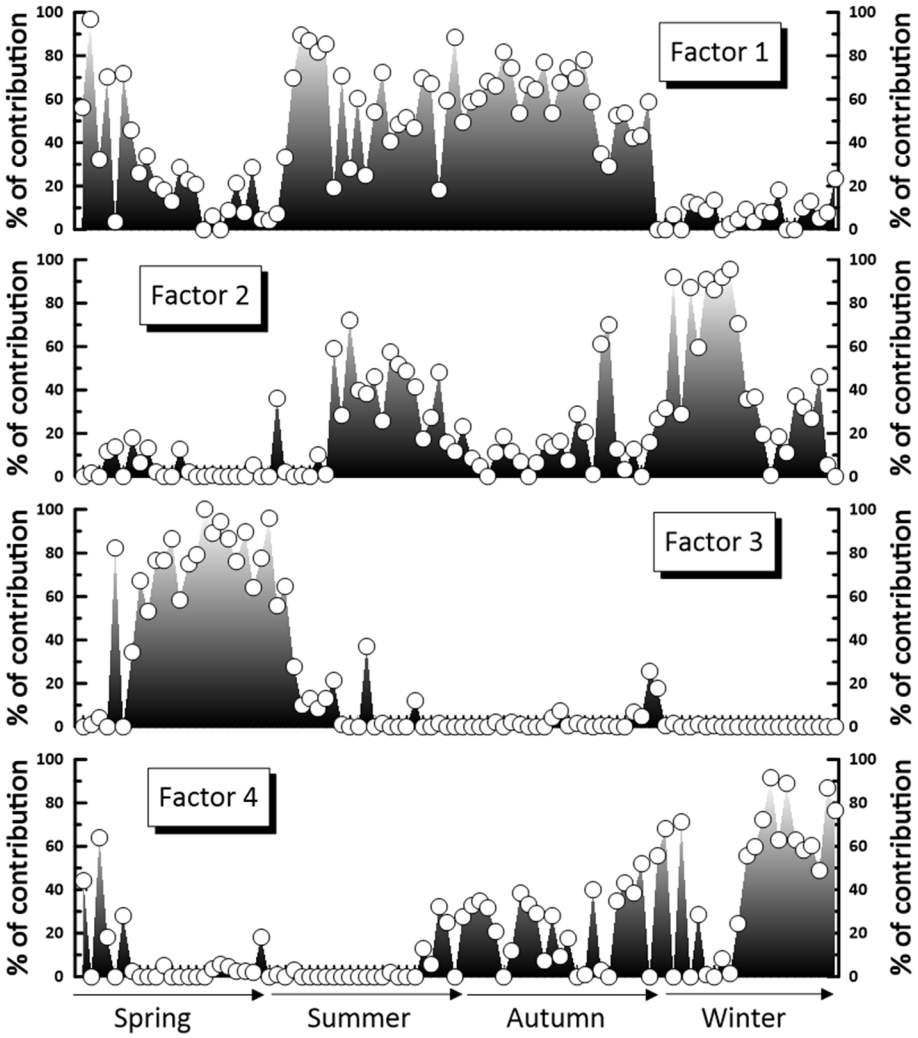


Fig. 4 Contribution of the four-factor to TSP samples from the HNI

dust to the open oceans in springtime. Actually, only such a dry, cold, and fast-moving dust can carry lower-chlorinated PCBs more effectively for long-range transport. The overall contribution of OC and EC included in this factor was less than 5%. It suggests that the role of dust particles was significantly higher than that of carbonaceous particles.

Factor 4 was highly loaded with PCB-52, -95, -82, -151, -183, -177, -171, -201, -208, and -206 and explained 44% of the total variance. These species are the main components of industrial PCB mixtures, such as Arochlor 1254, Arochlor 1260, and Kaneklor 600 (Praipipat et al. 2017). This pattern suggests the presence of a reemission or weathering source from historical applications of technical PCB mixtures. Autumn and winter are the seasons of high incidence of haze in our country. Typically caused by fine suspended particles, wind, relative humidity, precipitation, and sunshine duration play a more important role affecting the formation and dissipation of haze. Similar to Factor 3, the overall contribution of OC and EC included in Factor 4 was less than 5%. It can be considered conservatively that these two factors were not related with the combustion sources. Those PCBs can likely be transferred from gaseous phase to particulate one, or adsorbed onto atmospheric aerosols under such meteorological condition.

3.5 Contributions of combustion sources to particulate PCBs and implication for the role of OC/EC

According to PMF results along with OC/EC as an indicator of combustion source, Factor 1 and Factor 2 can be used as the combustion sources of PCBs in this study, and their total contribution was about 30% (Fig. 5). Different with Zhao et al. result, the UP-PCBs account for 65% of the total PCB emissions and have become the main emission source in China, including volatile emission from

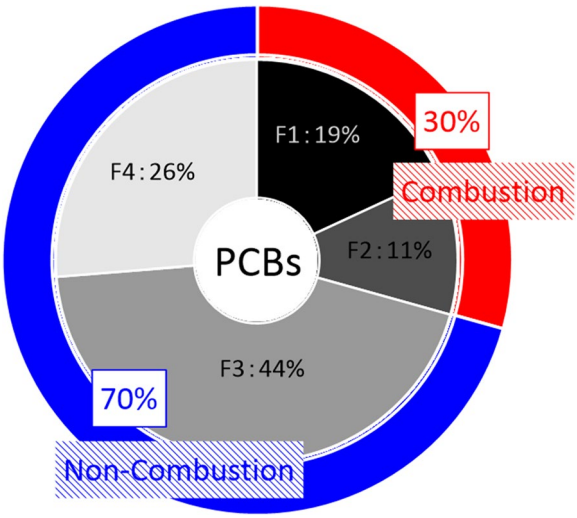


Fig. 5 Average combustion and non-combustion contributions to PCB mass concentration in TSP samples

pigment/painting and plastic industry (Zhao et al. 2019). Obviously, combustion source PCBs are not the same as UP-PCBs, the latter includes the non-combustion emission from pigment/painting and plastic industry.

Compared with the urban sampling sites near the emission source, it is challenging to differentiate remote or background sites far away from the emission source as either intentional or unintentional because of unknown pathway of PCB origins. The bigger challenge is the role of OC/EC in the distribution of PCBs during transport. 1) Although combustion source PCBs can be directly released into the atmosphere, the soil near the emission source is still an important sink for the deposition of those PCBs. It is not yet clear whether there is a so-called "non-exchangeable fraction" between PCBs and EC (that is PCBs with a common combustion source to EC) could be broken away in soil in a short period time. Theoretically, the PCBs that can volatilize away from the soil particles are more of the non-combustion source PCBs (or IP-PCBs) that remain in the soil without BC absorption. Thus, the contribution of the secondary source of IP-PCBs seemed to be significantly higher and was mainly associated with dust events in this study. 2) Production of secondary OC (SOC) also might be a tracer for the source (Day et al. 2015; Wu and Yu 2016). According to the minimum value of OC/EC, it can be estimated that the contribution of SOC to OC in HNI air TSP was 54.7% ± 20.1%. This shows that the conversion rate of SOC is high in the YRE. The relatively high SOC/OC ratio in remote areas such as HNI is probably due to the intensification of secondary organic aerosols under the combined effect of photochemical products and high humidity during long-range transport (Zhang et al 2018). Especially in hazy weather, PCB signals from the different sources might be disturbed by transformation between SOC and OC during the pathway under the monsoon system. To this end, the influence of combustion source OC/EC particles on IP-PCBs during the long-range atmospheric transport still deserves further research.

4 Conclusion

In general, temperature will affect the residence time, migration and transformation of semi volatile PCBs in different environment media. However, seasonal variations in PCB levels were affected by the atmospheric source or pathway under the monsoon system, especially in a remote area in this study.

PMF method is very effective for the identification of combustion PCBs that have a common combustion source with OC/EC. Although combustion PCBs are not the same as UP-PCBs, the quantitative contribution of combustion and non-combustion PCB is helpful for our understanding of source and pathway of UP-PCBs and IP-PCBs.

Abbreviations

PCBs	Polychlorinated biphenyls
OC	Organic carbon
EC	Element carbon
PMF	Positive Matrix Factorization
UP-PCBs	Unintentionally-produced PCBs
IP-PCBs	Intentionally-produced PCBs
YRE	Yangtze River Estuary
ECS	East China Sea
TSP	Total suspended particulates
PUF	Polyurethane foam

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1007/s44246-023-00046-4.

Additional file 1: Fig. S1. Seasonal variations of PCB compositions at gas and particle phases in HNI. **Fig. S2.** The plot of log Kp versus log PL0 for selected PCB.

Authors' contributions

Tian Lin: Resources, Conceptualization, Supervision, Writing–original draft. Wanqing Zhou: Writing, Data curation. Shizhen Zhao: Investigation, Data curation, Field surveys. Minqiao Li: Field surveys, Writing. Zhigang Guo: Resources, Conceptualization, Supervision. All authors read and approved the final manuscript.

Funding

This work was supported by National Natural Science Foundation of China (Grant numbers: 41573134, 42076205).

Availability of data and materials

The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author details

[1]College of Marine Ecology and Environment, Shanghai Ocean University, Shanghai 201306, China. [2]State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China. [3]State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China. [4]Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention, Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China.

Received: 4 October 2022 Revised: 7 March 2023 Accepted: 12 March 2023
Published online: 06 April 2023

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