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Comparison of Historical Record of PCDD/Fs, Dioxin-Like PCBs, and PCNs in Sediment Cores from Jiaozhou Bay and Coastal Yellow Sea: Implication of Different Sources

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Abstract The concentrations of polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs), dioxin-like polychlorinated biphenyls (PCBs), and polychlorinated naphthalenes (PCNs) were measured in two sediment cores collected from Jiaozhou Bay. The concentrations of PCDD/Fs, dioxin-like PCBs, and PCNs in the cores were in the range of 2.8–26.3, 7.1–82.4 and 3.9–56.4 pg/g dw, respectively. The depth profiles of total concentrations PCDD/Fs and dioxin-like PCBs were similar in the sediment core J37 inside Jiaozhou Bay, but different from those in the sediment core J94 outside the bay, suggesting the different sources. In both cores Tri-CNs and Tetra-CNs were dominant, similar to the PCNs composition of some Halowax technical products. The maximal PCNs contamination occurred in the mid-1970s (outside the Bay) and early-1990s (inside the Bay). An increase of the indicator CN congeners characteristic for thermal source in the top layers of the sediment core inside the bay indicated that the contribution from the municipal solid waste incineration has been more important in recent years.

Keywords Polychlorinated dibenzo-*p*-dioxins/dibenzofurans · Dioxin-like polychlorinated biphenyls · Polychlorinated naphthalenes · Sediment core

Because of low vapor pressure and degradation-resistance properties, polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs), dioxin-like polychlorinated biphenyls (PCBs), and polychlorinated naphthalenes (PCNs) may enter marine ecosystem through the river discharge, atmospheric dry and wet depositions (Dalla Valle et al. 2005; Falandysz 1998; Zheng et al. 1997). Sediment cores can keep good records of pollution history, and providing information on pathways and sources for POPs pollutants. The behavior and fate of POPs in coastal sea areas is an important link for the environmental pollution control in the coastal zones, especially those of marginal seas (Mirza et al. 2012). Studies on PCDD/Fs, dioxin-like PCBs, and PCNs in sediment cores from estuaries and bays have been reported (Yamashita et al. 2000; Kim et al. 2008; Okumura et al. 2004; Dalla Valle et al. 2005), while little research on sediment cores has been conducted in China.

This study presents the analytical results of PCDD/Fs, dioxin-like PCBs, and PCNs determined in two dated sediment cores collected from Jiaozhou Bay which is surrounded by the industrial city Qingdao, Shandong Peninsular and the western part of the Yellow Sea. In the past, more than 500 factories discharging waste waters into Jiaozhou Bay. The Yellow Sea coastal current (YSCC) flows southwestwards approximately along the direction of the 40–50 m isobar outside of the bay. The objectives of this work are to preliminarily evaluate the historical

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records and toxic equivalent quotients (TEQ) of PCDD/Fs, dioxin-like PCBs and PCNs, and to identify their possible sources in the sediments linking with the marine current circulation system in the Yellow Sea.

Materials and Methods

Two sediment cores were collected using a box core sampler. The cores J37 (36°09.34′N, 120°14.16′E; water depth: 35 m) and J94 (35°58.620′N 120°27.130′E; water depth: 25 m) were 84 and 82 cm in length, respectively, both were gray mud in structure throughout the cores. The sampling locations of the two sediment cores were shown in Fig. 1. Immediately after sampling, the samples were frozen during transportation and storage. The sediment cores were sectioned for every 2 cm segment. Each sediment segment was freeze-dried and stored at −20°C until analysis.

All chemicals used were of analytical reagent grade. The anhydrous sodium sulfate, silica gel, and alumina were oven baked at 500°C for 12 h. The method of Hanari et al. (2004) was used for analysis of PCDD/Fs, dioxin-like PCBs and PCNs. Around 10 g of freeze-dried samples were spiked with internal standard materials consisting of ^{13}C-labeled PCBs and ^{13}C-labeled PCDD/Fs and then Soxhlet-extracted with 50 % DCM in *n*-hexane for 16 h. After evaporation the extract was passed through a multi-layer silica gel cleanup column (from top to bottom: 0.8 g silica gel, 3 g 2 % KOH-silica gel, 0.8 g silica gel, 4 g 44 % H_2SO_4-silica gel, 4 g 22 % H_2SO_4-silica gel, 0.8 g silica gel, 8 g 10 % $AgNO_3$-silica gel, 5 g anhydrous sodium sulfate). The extract was eluted with 200 mL *n*-hexane and concentrated to 4 mL, then passed through an activated basic alumina column packed with 10 g alumina and 2 g anhydrous sodium sulfate on the top. The first fraction which was eluted with 40 mL of 0.5 % DCM in *n*-hexane contains most of PCNs. Then the second fraction containing PCDD/Fs and dioxin-like PCBs was eluted with 100 mL of 50 % DCM in *n*-hexane. All fractions were micro-concentrated to 200 μL under a gentle stream of nitrogen. PCNs were further fractionated by HPLC using a porous graphitic carbon column. Prior to extract fraction the column was pre-washed with 50 % DCM in *n*-hexane and toluene. The 150 μL aliquot of the fraction was injected. The remaining part of the extract was reserved and kept sealed under deep-freezing condition. The Hypercarb-HPLC column was forward eluted using 50 % DCM in *n*-hexane and back flushed using toluene. Each fraction was micro-concentrated to 100 μL, ready for HRGC-HRMS analysis.

HP 6890 GC coupled with JEOL JMS-700D HRMS (Japan) was used. Tetra- to Hexa-CDD/Fs were separated and quantified using a SP-2331 capillary column

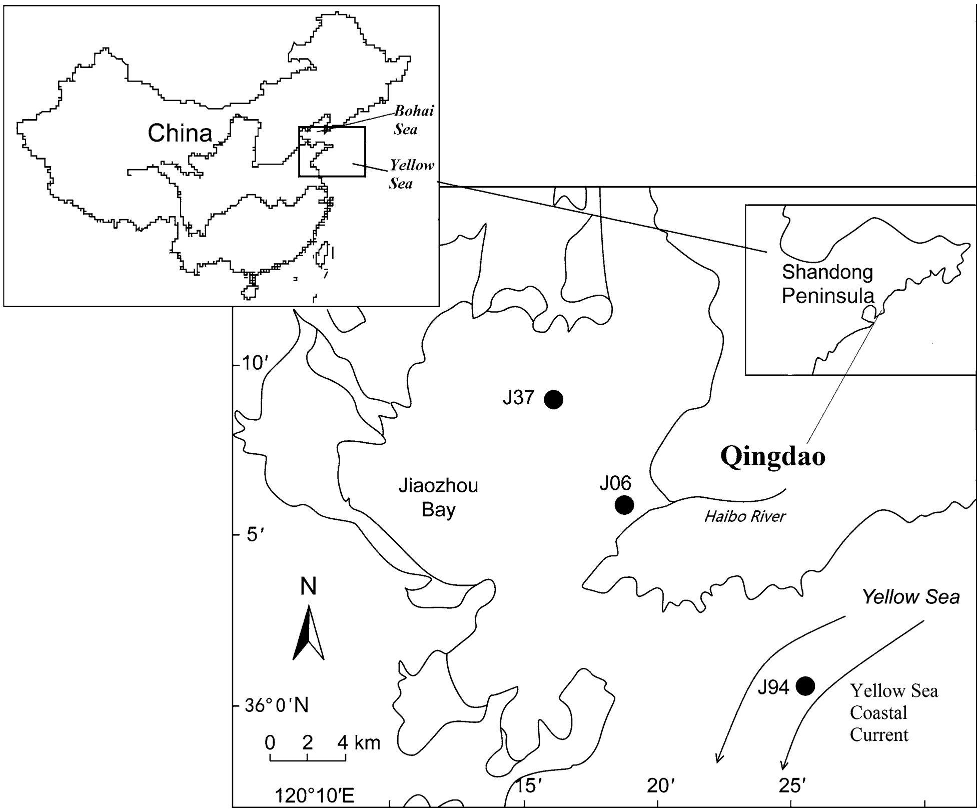


Fig. 1 Sampling sites of the sediment cores J37 and J94 (The *arrows* denote the Yellow Sea Coastal Current)

(60 m × 0.25 mm × 0.20 μm; Supelco, USA); Hepta- and Octa-CDD/Fs, dioxin-like PCBs and PCNs were separated and quantified using a DB-17 column (30 m × 0.25 mm × 0.25 μm; J&W Scientific); The GC column temperature program for Tetra- to Hexa-CDD/Fs measurement was 100°C (1 min) to 200°C at a rate of 20°C/min and then to 260°C at 2°C/min, with a final hold time of 35 min. For dioxin-like PCBs and Hepta-, Octa-CDD/Fs were 70°C (1 min) to 200°C at a rate of 15°C/min and then to 270°C at 4°C/min, with a final hold time of 15 min. For PCNs was 70°C (1 min) to 180°C at a rate of 15°C/min and then to 270°C at 2°C/min, with a final hold time of 10 min.

Blank spikes and blank spike duplicates, matrix spikes and matrix spike duplicates, replicate samples and procedural blanks were performed in QA/QC protocols. All samples were spiked with ^{13}C-labeled PCBs and ^{13}C-labeled PCDD/Fs internal standard before the Soxhlet extraction. Any sample with recovery below 40 % was not quantified. All the results were corrected for blanks. The recoveries of PCDD/Fs, PCBs and PCNs were in the range of 75.6 %–121.8 %, 77.2 %–126.8 % and 69.5 %–107.2 %, respectively and the detection limits were 0.01–1.00 pg/g dw.

The chronological data for cores J37 and J94 were obtained using ^{210}Pb (half-life: 22.3 a) dating method. The sub-samples collected at different depths were added ^{209}Po spike and a silver disc in 0.5 mol/L HCl at 75–80°C. The α-radioactivity of ^{210}Po was measured with a multi-channel α-energy spectrometer. The excess ^{210}Pb was calculated by subtracting the ^{210}Pb supported by its present ^{226}Ra from the total measured ^{210}Pb. The sedimentation rate was obtained by plotting the excess ^{210}Pb against depth in semi-logarithm coordinates.

Results and Discussion

Total concentrations of PCDD/Fs (ΣPCDD/Fs) in J37 were in the range of 3.1–26.3 pg/g dw, with an average of 14.1 pg/g dw. The range of ΣPCDD/Fs in J94 was 2.8–11.5 pg/g dw, with an average of 6.5 pg/g dw, one-half lower than that of J37. These concentrations are much lower than the surface sediment in Bohai Bay, China (500–35,000 pg/g dw) (Hu et al. 2005), Masan Bay, Korea (1,300 pg/g dw) (Hong et al. 2009), Tokyo Bay (34,900 pg/g dw) and Ariake Bay, Japan (95–17,000 pg/g dw) (Yamashita et al. 2000; Kim et al. 2008). Total dioxin-like PCB concentrations of J37 (ΣPCBs) were 7.1–82.4 pg/g dw, with an average at 49.8 pg/g dw. The ΣPCBs of J94 were 23.1–61.2 pg/g dw, with an average at 33.8 pg/g dw, significantly lower than that of J37. Total PCN concentrations (ΣPCNs) in core J37 were 3.9–56.4 pg/g dw, with the mean of 29.8 pg/g dw. ΣPCNs in core J94 ranged from 9.9 to 29.5 pg/g dw, with the mean of 18.9 pg/g dw, significantly lower than those in J37 core. Most of the Hepta-CNs and Octa-CN are below the detection limit. The concentrations of total PCNs in J37 and J94 cores were lower than those in surface sediment from the Haibo River estuary (1.2 ng/g dw) (Pan et al. 2007) and Tokyo Bay (4.4 ng/g dw) (Yamashita et al. 2000).

Depth profiles of PCDD/Fs, dioxin-like PCBs and PCNs in core J37 and J94 were showed in Fig. 2. The ΣPCDD/Fs in the cores inside and outside Jiaozhou Bay showed a similar trend that ΣPCDD/Fs gradually increased from the bottom of cores (1940s–1950s) and reached the maximum values in the 10–12 cm depth layer (the early 1990s), and then showed a slight decrease in the top (0–10 cm) layers (Fig. 2a, b). The ΣPCDD/Fs in the top layer of J37 were 9 times higher than in the bottom layers. In comparison, PCDD/Fs emission maximum occurred in 1970s in European countries (Gevao et al. 2000). The PCDD/Fs in sediment cores in Tokyo Bay, Sentai Bay and Ariake Bay, Japan (Yamashita et al. 2000; Kim et al. 2008) began to increase in 1950s and 1960s and reached maximum in early 1980s. The maxima in the sediment cores inside and outside Jiaozhou Bay showed a time lag behind those in the Japan sediment cores for about 10 years.

The PCDD/Fs homologue compositions of the two cores showed similar patterns to a certain degree (Fig. 3). OCDD were dominated in both cores (J37: 27.5 %–48.7 %, J94: 21.2 %–56.1 %) with more OCDD outside of the bay. The next abundant homologue in both cores was OCDF, but more OCDF was found inside the bay (J37 average 18.7 %; J94 average 10.5 %) (Fig. 3). The historical change in the homologue concentrations was also different. In both cores, OCDD showed rapid growth in 1960s and 1970s and reached the maximum in early 1990s, then gradually decreased to the surface values in both cores. The OCDD/OCDF ratios range from 1.39 to 4.27 in J37, while the ratios are from 3.04 to 8.88 in J94. The ratios in both cores show a decreasing trend since 1950s. Low PCDD/PCDF ratio (about 0.25) was found in sediments from the Venice Lagoon and PCDFs, especially OCDF, appear to provide a marker for sediments in the industrial district (Dalla Valle et al. 2003). J37 in Jiaozhou Bay may receive pollution discharge from the river input around the bay. The influence of the Haibo River on PCDD/Fs concentration was evidenced from an estuary site J06 (Pan et al. 2007) exhibiting high OCDF, indicating that there was a distinctive PCDD/F source which could be the discharge from the Haibo River. On the other hand, J94 could be less affected by the local industrial discharge.

The more input of OCDD in the sediment outside the bay may suggest a different source for PCDD/Fs. Alcock and Jones (1997) found that the pentachlorophenol PCP-based textile preservatives, and chloranil-based dyes and pigments contain large amount of OCDD. PCP and PCP-Na have been manufactured for nearly half a century in the

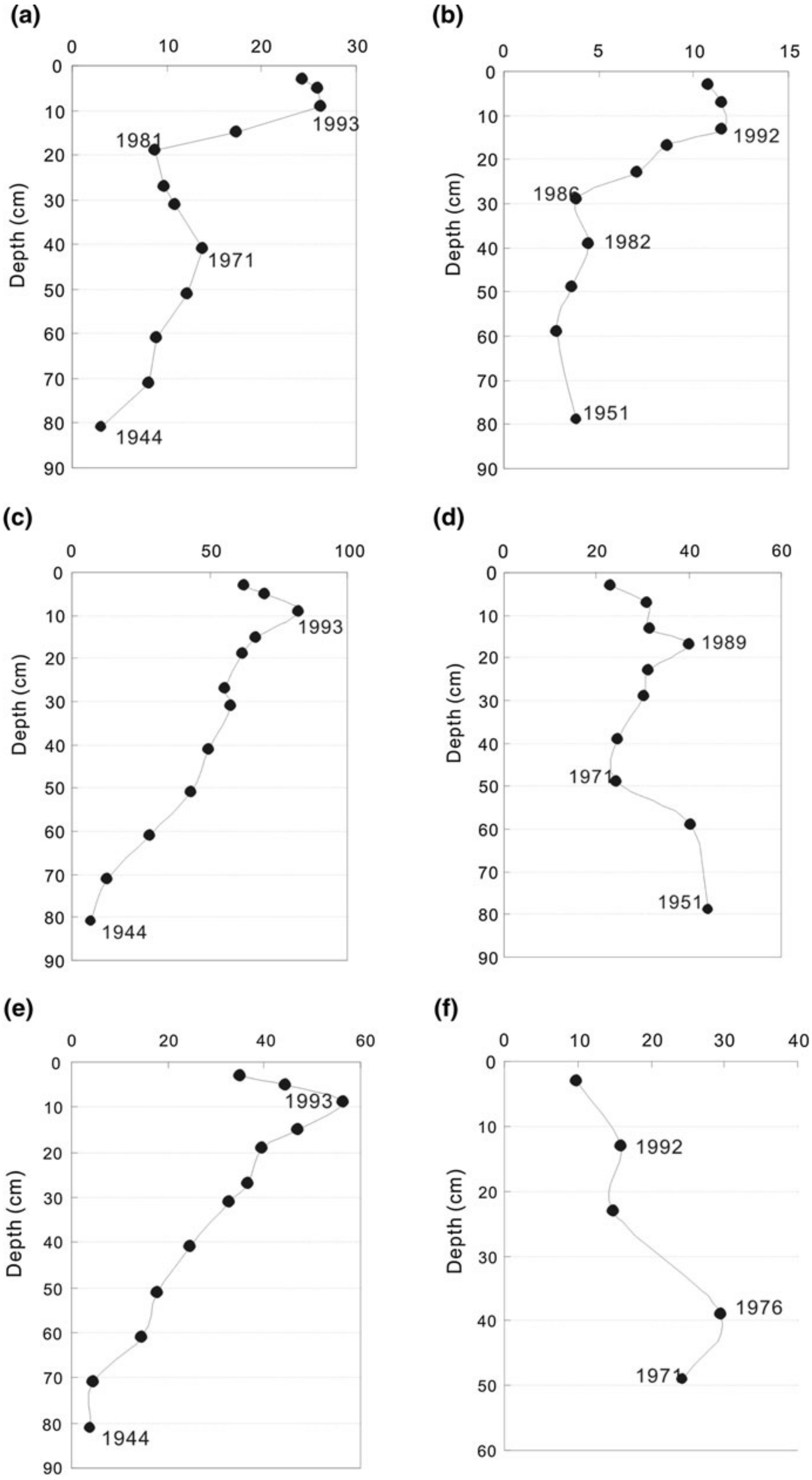
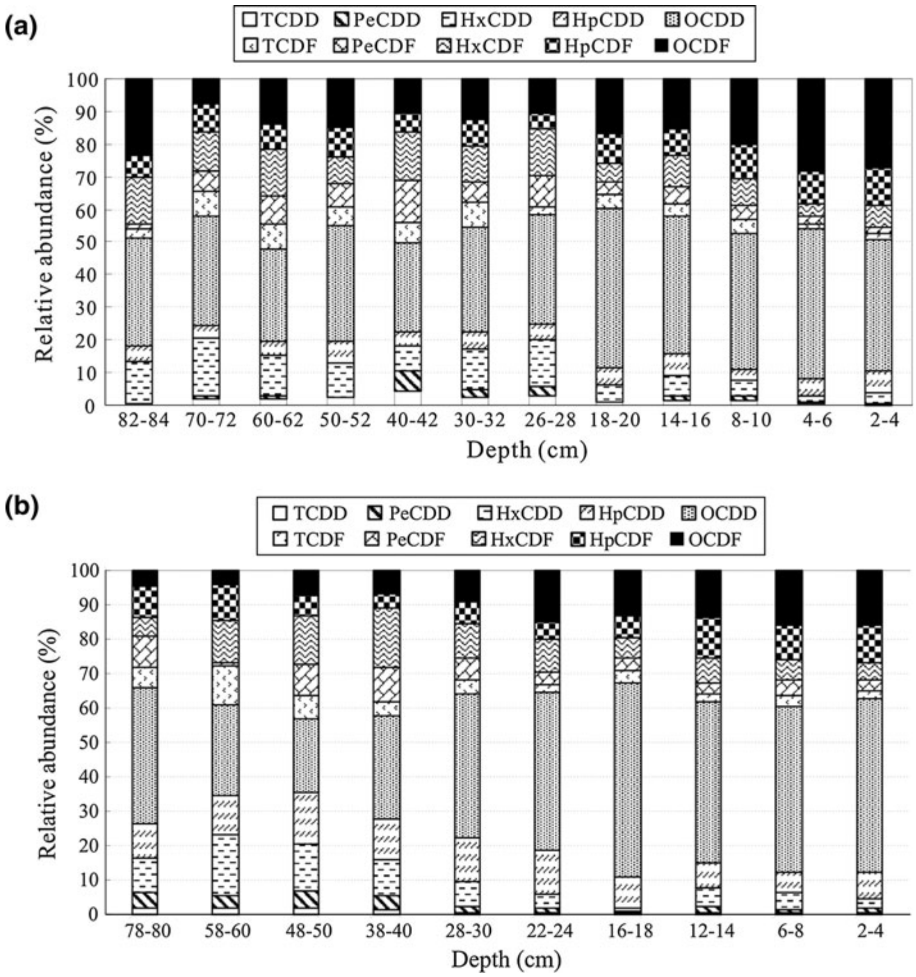


Fig. 2 Depth profiles of PCDD/Fs (**a**, **b**), dioxin-like PCBs (**c**, **d**), and PCNs (**e**, **f**) in core J37 and J94

industrial city Tianjin in western coast of the Bohai Sea and similar congener profiles of 2,3,7,8-PCDD/Fs for the river sediments in Tianjin and sediment cores from Bohai Bay to those in PCP and PCP-Na were reported (Hu et al. 2005; Liu et al. 2007). Our results suggest that PCP and PCP-Na might be possible sources for J94.

Fig. 3 Down-core variations in proportion of PCDD/F homologues for J37 (**a**) and J94 (**b**)



PCDD/Fs were measured in sediments from the Bohai Sea and the Yellow Sea coastal areas of South Korean by Naile et al. (2011). Spatial distribution indicated some point sources in Korea while Chinese sources were more widespread and diffuse. Because of the Yellow Sea Warm Current and the coastal current formed a counter-clockwise vortex in the Yellow Sea (Fig. 1), part of PCDD/Fs in Qingdao coastal sediments, especially J94 located in the open sea, was likely transported by sea current from the Bohai Sea, the eastern Yellow Sea, and probably as far as Korean coastal area. The relatively higher OCDD in J94 compared with that in J37 was possibly affected by PCDD/Fs brought from the coast areas of the Bohai Sea and Yellow Sea.

The ΣPCBs in the two cores showed different trend (Fig. 2c, d). PCBs in J37 increased nearly straight forward from the bottom of the core (1940s) and reached the maximum in the 8–10 cm layer (early 1990s), and then decreased in the top sediment (the end of 1990s), similar to the trend of PCDD/Fs. The ΣPCBs maximum in J37 was 11 times higher than that in the bottom layer of the core. On the other hand, the concentrations of PCBs in J94 at the bottom of the core (the early 1950s) were relatively high, then decreased till early 1970s, and increased again with the maximum in the 16–18 cm layer (the end of 1980s) and then decreased in the top sediment (the end of 1990s). China began to produce PCBs in 1965 and reached the highest production in 1971. PCBs production in China was reduced at the end of 1974 and stopped in 1980s. The higher ΣPCBs level at the bottom of the core (before 1960s) may have been caused by sea current transportation from the neighboring regions of the Yellow Sea. In comparison, PCB industrial products were imported and used in Japan, before the 1950s and began to produce PCBs in early 1950s, and were banned in 1972 (Takasuga et al. 2006). The PCBs maxima appeared at the subsurface of sediment core J37 (early 1990s) and J94 (late 1980s) after China and neighboring countries had banned production of PCBs, suggesting a time lag between the time of banning and the maximum sedimentation of PCBs in the cores.

The dominant homologue of dioxin-like PCBs in the sediment cores both inside and outside Jiaozhou Bay was Penta-CBs, accounting for 73 %–84 % in J37 and 62 %–89 % in J94, respectively. The most abundant PCB 118 in the

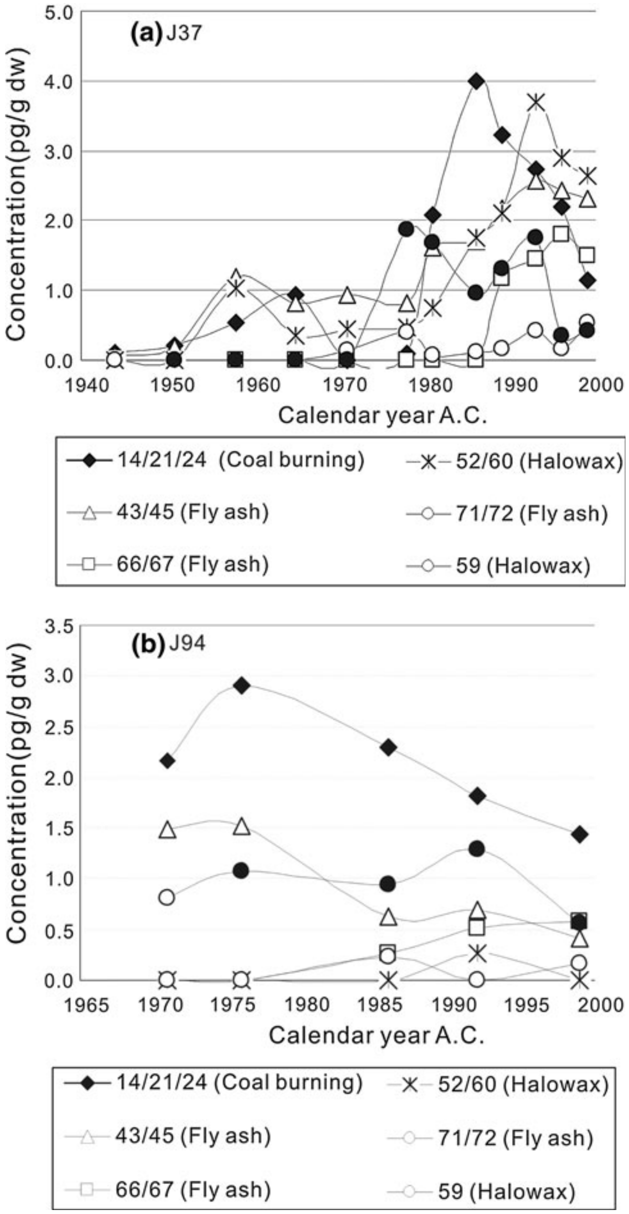


Fig. 4 Time trends of different source indicators for PCNs in cores **a** J37 and **b** J94

sediments inside and outside the bay accounted for 25 %–56 % and 29 %–57 % of the total dioxin-like PCB, respectively, followed by PCB 105 and 77. These isomers are the main component of PCB industrial products such as Japanese Kanechlor 400. PCB 169 which may have come from combustion processes (Yao et al. 2002) was relatively low in our samples, suggesting that the PCB mainly came from PCB products.

ΣPCNs in core J37 is characterized by the lowest concentration of 3.86 pg/g dw at the bottom layer, then increased to the maximum at the subsurface layer (8–10 cm) dated as the early 1990s (56.4 pg/g dw), and gradually decreased to the current surface layer (34.9 pg/g dw) around year 2000 (Fig. 2e). The ΣPCNs in the bottom segment were 18 times lower than that of the observed maximum value in subsurface layer. The PCN concentrations in J94 core reached the maximum in the mid-1970s, and then gradually decreased to the surface layer in the late 1990s (Fig. 2f). This decreasing trend in the top layers of sediment cores typically reflects the chemicals being banned for production and usage in recent decades. The occurrence of the PCN maximum in core J37 was in accordance with that of PCDD/Fs and dioxin-like PCBs (i.e. in the early 1990s). On the other hand, the maximum PCN concentration in core J94 occurred in the layer corresponding to mid-1970s, 10-year earlier than those for PCBs, which was in consistent to the fact that the global usage of PCNs was earlier than those for PCBs (Gevao et al. 2000).

In core J37, PCN 14, 22/23 and 25 were the predominant congeners for Tri-CNs, PCN 27 and 46 for Tetra-CNs, PCN 52/60 and 62 for Penta-CNs, PCN 66/67, 69 and 71/72 for Hexa-CNs. In core J94, Tri-CNs and Tetra-CNs are predominant homologues, accounting for 43 % and 36 %, respectively. Some indicator congeners can be used to identify the sources of PCNs. PCN 54 can be produced as a by-product in combustion processes (Falandysz 1998). According to a previous report, PCN 39, 44, 45/36, 66/67 and 73 can be found in fly-ash (Falandysz 1998; Meijer et al. 2001). Some chloronaphthalene (CN) congeners are negligible in technical PCN mixtures and also as by-side impurity in PCB commercial products. Consequently, these PCNs are considered as the suitable markers to identify thermal sources of PCNs (Orlikowska et al. 2009).

Temporal variations of some source indicator PCN congeners in cores J37 and J94 are presented in Fig. 4, which shows that the trend of different source contributions are different for the cores inside and outside of the bay. PCN 14, which is representative of the contribution from coal burning process (Meijer et al. 2001), showed an increasing trend in 1980s and reached a maximum in core J37 between 1980s and 1990s. In core J94 PCN 14 reached a maximum between 1970s and 1980s, and then decreased, reflecting the changes in the fuel structure in China and the regions surrounding the Yellow Sea. The congeners PCN 71/72 and PCN 59, which are representatives of the technical product Halowax 1014 (Yamashita et al. 2000), began to increase since the early 1950s in core J37. This may reflect the reconstruction and industrialization processes in the regions surrounding the Yellow Sea after the World War II with the PCN pollution transported through the Yellow Sea current system to Jiaozhou Bay area. This PCN source reached its maximum in the late 1990s in both J37 and J94 cores, with a slight time-lag in core J37, and diminished since then, reflecting the ban of PCN production and usage. There were rapid increases of indicator congeners PCN 66/67 and PCN 52/60, representatives of the PCNs in fly ash formed by municipal solid waste

(MSW) incineration, since the late 1970s in core J37. The results may indicate that the contribution from MSW incineration becomes more important. Enormous solid wastes were produced since the fast economic growth in China in recent decades, and this trend of contribution from the MSW incinerations is now still showing no sign of slowing down.

The PCDD/F-TEQ in the sediments of J37 and J94 were 0.12–2.26 pg TEQ/g dw and 0.24–0.59 pg TEQ/g dw according to WHO-TEF values. The dioxin-like PCB-TEQ in the sediments of J37 and J94 were in a range of 0.01–0.14 pg TEQ/g dw and 0.02–0.06 pg TEQ/g dw, respectively. The contributions of PCDD/Fs to the ΣTEQ (PCDD/Fs + dioxin-like PCBs) were in the range of 81 %–97 % and 88 %–95 % for J37 and J94, respectively. PCN-TEQ was calculated based on the relative potencies (REPs) following the Blankenship and co-workers' method (Blankenship et al. 2000). PCN-TEQ in all samples was less than 0.1 pg-TEQ/g dw.

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