

Sixty-Year Sedimentary Record of DDTs, HCHs, CHLs and Endosulfan from Emerging Development Gulfs: A Case Study in the Beibu Gulf, South China Sea

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Abstract The temporal distribution of organochlorine pesticides (OCPs) was examined in the ^{210}Pb dated sediment core from the Beibu Gulf, South China Sea. The total OCPs concentrations were in the range of 0.93–26.6 ng g⁻¹ dry weight. Dichlorodiphenyltrichloroethanes (DDTs) (0.17–24.8 ng g⁻¹), Hexachlorocyclohexanes (HCHs) (0.04–0.51 ng g⁻¹), Chlordane related compounds (CHLs) (0.22–1.72 ng g⁻¹) and endosulfan (n.d.–0.91 ng g⁻¹) were the predominant compounds. Similar to most Chinese coastal areas, the levels of DDTs in the Beibu Gulf became elevated since the early 1990s, especially since 2000 despite the ban in 1983 in China. This suggests that the concentrations of DDTs were controlled by several processes, such as land reclamation and soil runoff. The isomer ratios of (*p,p'*-DDE + *p,p'*-DDD)/*p,p'*-DDT, *p,p'*-DDT/DDTs along with construction land expansion indicated that economic activities, land reclamation, soil runoff and the use of DDT-containing antifouling paints might be responsible for the input of DDT. The ratios of α -HCH/ γ -HCH (and γ -HCH/HCHs) and *trans*-chlordane/*cis*-chlordane (TC/CC) indicated fresh inputs of lindane and chlordane, respectively. In addition, CC was found to be degraded faster than TC under anaerobic conditions in sediments from the Beibu Gulf.

Keywords Sediment core · OCPs · DDTs · HCHs · Endosulfan · Beibu Gulf

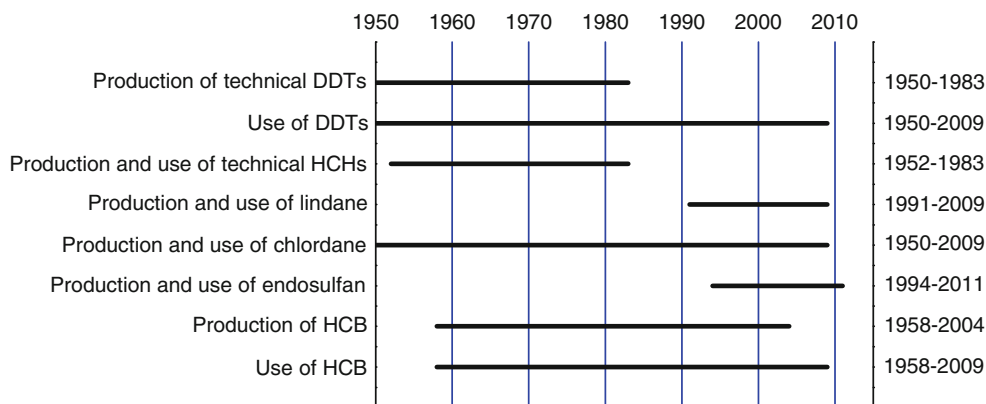
Organochlorine pesticides (OCPs) are chemical synthetic pesticides and environmental contaminants of great concern on a global scale due to their persistence, long-range

transport properties, bioaccumulation and health risks as potential carcinogens and mutagens. They are particle-reactive in aquatic ecosystems and tend to accumulate in sediments due to their hydrophobic character (Binelli et al. 2008). Thus, sediments were a significant sink of OCPs and reverted to becoming a secondary source following point source reduction. DDTs and HCHs, as the most extensively concerned OCPs compounds, were heavily produced and used in the world after the 1940s. China is the second largest pesticides producer in the world. The total production of DDTs and technical HCHs were 0.4 and 4.9 million tons, accounting for 1/5 and 1/3 of the total worldwide output, respectively (Zhang et al. 2002). The production and application history of individual OCPs is shown in Fig. 1.

The Beibu Gulf, located in the northwest of South China Sea, is a semi-enclosed gulf shared between China and Vietnam, covering an area of approximately 128,000 km² and reaching a maximum width of 333 km. Abundant nutrient and pollutants are discharged from the land to the gulf by the adjacent rivers (Chen et al. 2009). Pollutants in the gulf tend to deposit because of the lower water flow rate (Zhang et al. 2011a). The Beibu Gulf has traditionally played a significant role in the economies of China and the Association of Southeast Asian Nations (ASEAN) by providing highly productive and diverse marine products. As a new highlight of China-ASEAN cooperation, the Beibu Gulf economic rim emerged in 2006. This implied that economic activities, land reclamation and soil runoff have growing impact on the gulf. Besides, there has been a sharp increase in demand for seafood products due to the rapid population growth during the twentieth century. As a result, commercial fishing has also undergone fast growth with the introduction of engine boats in the gulf in the 1960s. The power of fishing boats in coastal areas of the

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Fig. 1 Production and application history of individual OCPs in China (MEP 2009; UNEP 2011)



gulf was twice as high as those of optimum yield in the early 1990s (Chen et al. 2008). This implied that a large number of fishing vessels were needed in the Beibu Gulf. DDTs can be released from fishing boats due to the presence of DDT-containing antifouling paint (GEF 2007). Thus, it is significant to evaluate the pollution history and transfer of OCPs in the Beibu Gulf, a typical emerging development gulf. To our knowledge, this is the first report of OCPs in a sediment core in the Beibu Gulf. The results will be of particular value for understanding the historical trend and sources of OCPs in a typical emerging development gulf.

Materials and Methods

A sediment core (Fig. 2) was collected from the Beibu Gulf, South China Sea (21°35.113'N, 108°45.479'E) in December 2010. A stainless steel static gravity corer (8 cm i.d.) was employed to minimize the disturbance of surface sediment layers. The core samples were sliced at 1 cm intervals to the depth of 50 cm aboard ship. The sliced sediment samples were sealed in polyethylene bags and stored at -20°C until analysis.

The procedures for extraction and purification of OCPs in sediment samples were described in detail elsewhere (Zhang et al. 2002). Briefly, sediment samples were freeze-dried, ground, homogenized at the first what? About 50 g of sediment samples, spiked with surrogate standards (TCmX and PCB 209), were Soxhlet extracted with 250 mL dichloromethane for 48 h. Activated copper granules were added to the collection flask before the extraction to remove elemental sulfur. The sample extracts were then concentrated on a rotary evaporator and solvent-exchanged to hexane, which were further reduced to 0.5 ml. The concentrated extracts were separated into two fractions. The first fraction was purified with a glass column, which was loaded with alumina, silica gel, sulfuric acid silica gel and anhydrous sodium sulfate from bottom to top in turn. The column was

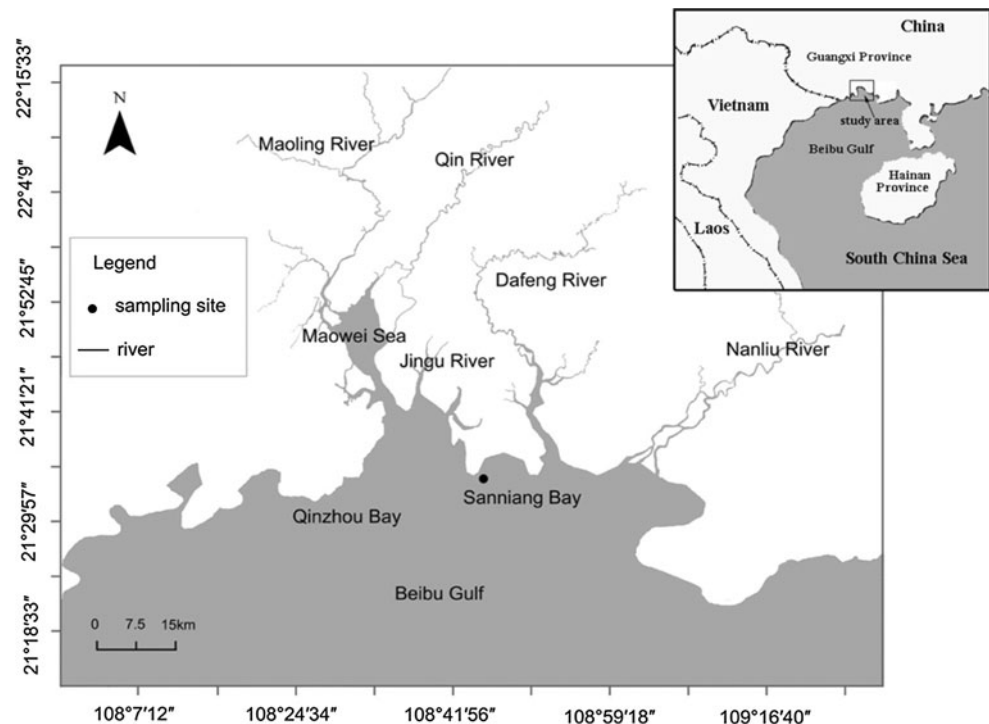
eluted with 15 ml of hexane/dichloromethane (1:1 v/v) for the collection of OCPs. The eluate was finally reduced to 0.5 ml under a gentle nitrogen stream. A known quantity of internal standard, PCNB (pentachloronitrobenzene), was added prior to gas chromatography-electron capture detector (GC-ECD) analysis.

Organochlorine pesticides in the sediment samples were measured by a Hewlett–Packard 5890 series II gas chromatograph (GC) equipped with a ^{63}Ni electron capture detector (μECD) and a HP-5 MS fused silica capillary column (30 m length, 0.25 mm diameter, 0.25 μm film thickness). As the carrier gas, nitrogen at 99.99 % purity was at a flow rate of 2.5 ml min^{-1} under the constant flow mode. Nitrogen was filtered with moisture, hydrocarbon and oxygen filters before entering the GC system. The oven temperature was initially isothermal at 60°C (1 min hold time), immediately rose to 290°C at $4^{\circ}\text{C min}^{-1}$ (25 min hold time). Splitless injector of a $1 \mu\text{L}$ sample was performed with 1 min solvent delay time. Data were acquired and processed using Agilent Chemstation software. Data acquisition and processing were controlled by HP-3365 Chemstation software.

In order to calculate the sedimentation rate and date the sediment core, a ^{210}Pb dating method was applied. In brief, the ^{210}Pb activities in sediment subsamples were determined by analysis of α -radioactivity of its decay product ^{210}Po , with the assumption that the two were in equilibrium. The Po was extracted, purified, and self-plated onto silver disks at $75\text{--}80^{\circ}\text{C}$ in 0.5 M HCl, with ^{209}Po used as a yield monitor and tracer in quantification. Counting was conducted by computerized multichannel α -spectrometry with gold–silicon surface barrier detectors. Supported ^{210}Po was obtained by indirectly determining the α -activity of the supporting parent ^{226}Ra , which was carried by coprecipitated BaSO_4 . A constant initial ^{210}Pb concentration (CIC) model was applied to date the sediment core to give an average sedimentation rate.

In the QC/QA program, the instruments were calibrated daily with calibration standards. For each batch of 10 field

Fig. 2 Sampling location of the sediment core in the Beibu Gulf



samples, a method blank (solvent), a spiked blank (standards spiked into the solvent), a sample duplicate and a US National Institute of Standards and Technology (NIST) standard reference material (SRM 1941) were processed. The recoveries of target OCPs in the SRM 1941 were $91\% \pm 11\%$ of the certified values. The method detection limits (MDLs) of OCPs were described as 3:1 signal versus noise value (S/N). MDLs of OCPs were in the range of $0.01\text{--}0.05\text{ ng g}^{-1}$. Recoveries for TCmX and PCB 209 were $60.66\% \pm 21.63\%$ and $89.79\% \pm 20.63\%$, respectively.

Results and Discussion

The sediments of the core were mainly silt. The lower water flow rate indicates a stable sedimentary dynamic environment at the sampling site. Based on this, the average sedimentation rate of 0.86 cm year^{-1} could be calculated using a constant initial ^{210}Pb concentration (CIC) model (Fig. 3). Vertical profiles of the excessive ^{210}Pb activity in the sediment core exhibited approximately a monotonic decline with depth (Fig. 3). A strong correlation ($R^2 = 0.75$, $p < 0.001$) was found between the values of $\ln(^{210}\text{Pb}_{\text{ex}})$ and the sample depths, which indicates that the slices of the core can reflect historic change. The deposition depth of 50 cm was between the computed date of 1951 and the core-top date of 2010. The sedimentary rate in the sampling site is 0.86 cm year^{-1} , which was between the estuary ($>0.9\text{ cm year}^{-1}$) and the marine areas ($<0.5\text{ cm year}^{-1}$) in the Beibu Gulf (Gan et al. 2013; Xu

et al. 2012). A similar declining trend of sedimentation rate from estuary to ocean was observed in Quanzhou Bay (Gong et al. 2007).

The total OCPs concentration varied from 0.93 to 26.6 ng g^{-1} (average 3.48 ng g^{-1}). The maximum concentration (26.6 ng g^{-1}) was found at the core-top corresponding to 2010 (Fig. 4a). DDTs ($0.17\text{--}24.8\text{ ng g}^{-1}$, mean 1.90 ng g^{-1}), HCHs ($0.04\text{--}0.51\text{ ng g}^{-1}$, mean 0.18 ng g^{-1}), CHLs ($0.22\text{--}1.72\text{ ng g}^{-1}$, mean 0.69 ng g^{-1}) and endosulfan (n.d.– 0.91 ng g^{-1} , mean 0.27 ng g^{-1}) were the predominant compounds. Among them, DDTs were found to be the most abundant compounds, accounting for 7–93% (mean 40%) of the total OCPs.

The down-core variations of DDTs (summation of *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDT and *p,p'*-DDT) concentrations are shown in Fig. 4b. Concentrations of DDTs began to increase slowly in 1980. An obvious peak concentration (5.24 ng g^{-1}) occurred in 1997, followed by a dramatical decrease to 2000 (0.89 ng g^{-1}). Then a sharp increase was observed to the maximum concentration (24.8 ng g^{-1}) in 2010.

DDTs started to be produced and used in the 1950s in China. They were heavily used in the 1970s to the 1980s, which were exhibited in the sediment cores from Quanzhou Bay (Gong et al. 2007) and the Pearl River Delta (Zhang et al. 2002), but not in those from Xinghua Bay (Zhang et al. 2011b), Daya Bay (Wang et al. 2008) and the Beibu Gulf. This depicts the regional differences in the use of DDTs in China. Although no technical DDT had ever been used in the Beibu Gulf before 1980 (Qiu et al. 2005), DDTs

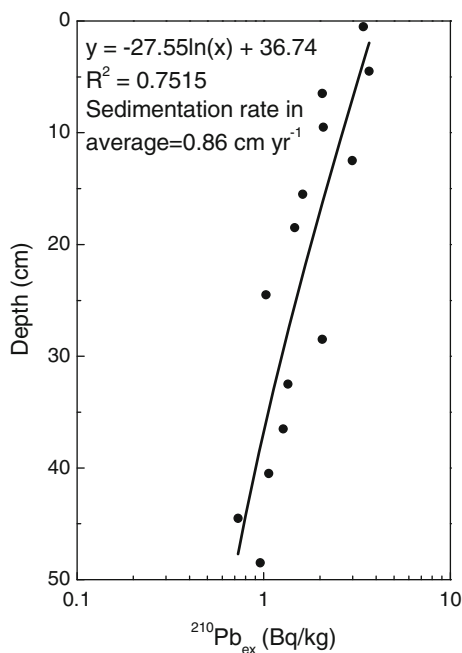


Fig. 3 Vertical distribution of $^{210}\text{Pb}_{\text{ex}}$

can still be detected from long-range atmospheric transport. The increase of DDTs concentrations from 1980 implies the start of use of DDTs in the Beibu Gulf (Wang et al. 2005). DDTs were banned from being used in China in 1983. Instead of declines in the 1980s, the levels of DDTs in the Beibu Gulf elevated from 1990. A recent increasing trend was also observed in the sediment cores from most Chinese coastal zones, such as the Pearl River Delta (Zhang et al. 2002), Daya Bay (Wang et al. 2008), Quanzhou Bay (Gong et al. 2007) and Xinghua Bay (Zhang et al. 2011b), as well as Mississippi River Delta and Galveston Bay in the United States (Santschi et al. 2001). The fact that DDTs concentrations didn't decrease after the ban suggests their concentrations are controlled by several processes, such as land reclamation and soil runoff.

1,1,1-trichloro-2,2-bis-(*p*-chlorophenyl)ethane (*p,p'*-DDT) is likely to be dehydrochlorinated to 1,1-dichloro-2,2-bis-(*p*-chlorophenyl)ethylene (*p,p'*-DDE) under aerobic conditions prior to sedimentation, but it can be reductively transformed to 1,1-dichloro-2,2-bis-(*p*-chlorophenyl)ethane (*p,p'*-DDD) in sediment (after sedimentation) under anaerobic conditions (Zhang et al. 2002). The ratio of (*p,p'*-DDE + *p,p'*-DDD)/*p,p'*-DDT can be used as an indicator of whether *p,p'*-DDT is "aged (degraded)" or "new (input recently)" (Qiu et al. 2004). A ratio >1 is observed generally for aged mixtures, and if the ratio <1, it indicates new application. The ratios of (*p,p'*-DDE + *p,p'*-DDD) to *p,p'*-DDT ranged from 0.40 to 10.2, higher than 1 for 90 % samples before 2000 in the Beibu Gulf (Fig. 4c), indicating an aged contamination of DDTs. After 2000, the ratios of (*p,p'*-DDE + *p,p'*-DDD) to *p,p'*-DDT (<1 for about 2/3 of

the samples) exhibited an obvious decline tendency with the increase of *p,p'*-DDT levels, indicating prevailing fresh inputs. In addition, the high ratios of *p,p'*-DDT to $\sum\text{DDTs}$ indicated recent DDT inputs (Lee et al. 2001). Residual *p,p'*-DDT in sediments ranged from 21 to 57 % (mean 40 %) of $\sum\text{DDTs}$ in the 2000s, indicating the same deduction in the Beibu Gulf. The input of DDTs after 2000 was also supported by the relatively high values of the organic pollution and nutrient salts of the Beibu Gulf (Lan 2011). A peak level of DDTs in 1997 and the increase from 2000 in the sediment core in the Beibu Gulf might be attributed to several factors as follows. It can't be ignored that the economic "overheat" period occurred in Southern China (Zhang et al. 2002) since the early 1990s. Characteristic of rapid regional urbanization, industrialization, and intensifying population, cities around the Beibu Gulf experienced the fast marine economic development and land reclamation. Remote-sensing studies of 1990, 2000 and 2010 on the structure of regional land use in the Beibu Gulf by mapping data (Yao and Yang 2012) also implied a large-scale land transformation taking place in the past 20 years. 32,979 and 43,740 hectares of land were transformed to construction land from 1990 to 2000 and from 2000 to 2010, respectively. Construction land expansion rate of the second decade (2.16 % year⁻¹) was faster than that of the first decade (1.94 % year⁻¹). Greater development happened when the Beibu Gulf Economic Development Zone was established in 2006. Additionally, erosion of the most severe flood in 1998 in the Pearl River System enhanced surface runoff. Thus, the economic activities, land reclamation and flood erosion since the early 1990s, especially since the 2000s, enhanced soil runoff to the gulf sedimentary system for the sloping from the northwest to the southeast. More DDTs were transported into the Beibu Gulf from soils and rivers (Wang et al. 2008). Furthermore, the Beibu Gulf is an important fishing harbor for a large number of fishery resources. With the rapid development of marine fisheries and marine transportation, fishing boats existed extensively in bays in China including the Beibu Gulf. DDT-containing antifouling paints used in fisherboats might be another possible DDT source since the 1990s (Liu et al. 2009). There are at least two Coating Manufacturing Company located near the Beibu Gulf (surveyed in 2005) that produced 160 and 180 tons DDT-based antifouling paint using 2.4 and 9 tons DDT per year, respectively (GEF 2007). The low value of DDTs concentration in 2000 may be attributed to the enlargement of comprehensive remediation of watershed pollution (Lan 2011).

The vertical profile (Fig. 4d) of HCHs (sum of α -, β -, γ -, and δ -HCH) is characterized with alternately high-low fluctuations. The peak value of HCHs (0.51 ng g⁻¹) was observed in the mid-1980s (1984), during which the technical HCHs usage reached 1,000–10,000 tons year⁻¹ in this area (Li et al. 1998). This was followed by a decrease for the production ban of HCHs in 1983 in China.

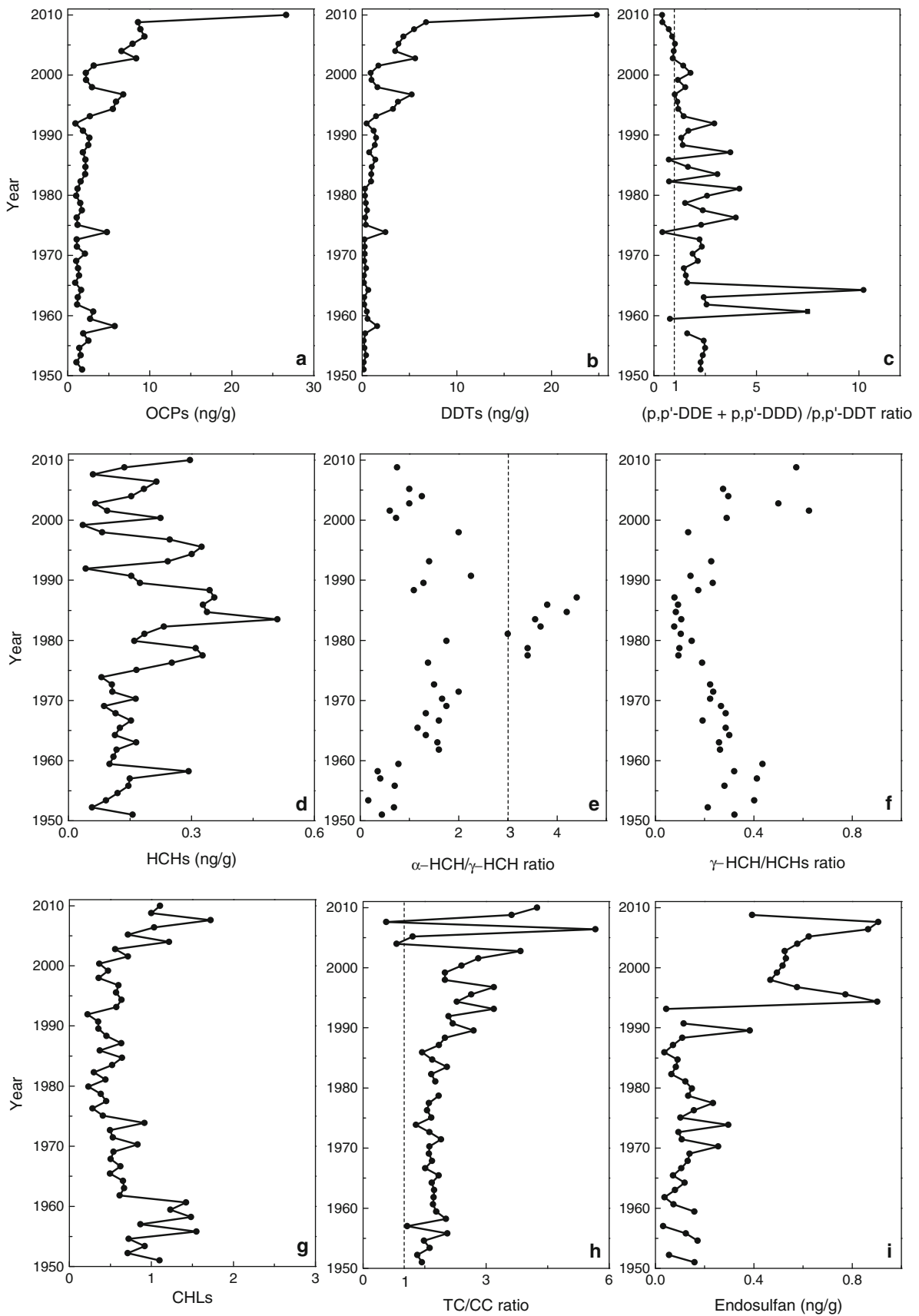


Fig. 4 Concentrations variations of OCPs and compositional ratios of $(p,p'$ -DDE + p,p' -DDD)/ p,p' -DDT, α -HCH/ γ -HCH, γ -HCH/HCHs and TC/CC

Hexachlorocyclohexane (HCH) could be available in two technical formulations: technical HCH (containing about 60–70 % α -HCH, 5–12 % β -HCH, 10–12 % γ -HCH, 6–10 % δ -HCH and minor proportions of other isomers) and lindane (γ -HCH > 99 %). The sources and fate of HCHs in the environment can be reflected by the isomer composition. Four isomers of HCHs were all detected in sediments and the mean percentages of α -, β -, γ -, and δ -isomers were 30, 33, 25, and 12 %, respectively. The percentage of α -isomer declined, while β - and γ -isomer increased when compared with those of technical HCH. One possible explanation was that α -HCH has a higher vapor pressure and Henry's law constant. The Henry's law constant of α -HCH is approximately twice as high as that of γ -HCH (Walker et al. 1999) and 20 times higher than β -HCH (Li et al. 2003), indicating that α -HCH is more likely to partition to the air than sediments. β -HCH, the most persistent HCH isomer, has a lower vapor pressure and a higher $\log K_{ow}$. α - and γ -HCH could be transformed into β -HCH after a long time (Wang et al. 2008). Based on the composition of technical HCHs, a high α -HCH/ γ -HCH ratio indicates historical evidence of HCHs and a high percentage of γ -HCH indicates a contamination of lindane (Zhang et al. 2011b). The ratios of α -HCH/ γ -HCH (Fig. 4e) were nearly equal with those of technical HCH (3–7) (Walker et al. 1999) during the 1970s and the early 1980s, when China was one of the most polluted countries in the world for the extensive application of technical HCH (Li et al. 1998). Then, the ratios of α -HCH/ γ -HCH were lower than 3, and showed a declining trend since the end of 1980s with the upraised γ -HCH/HCHs ratios (Fig. 4f), suggesting fresh inputs of lindane in this area in recent two decades.

Chlordane production began in the 1950s in China (NIP 2007), which was extensively used in China against termites until 2009 (MEP 2009). The temporal trend of CHLs (sum of *cis*-chlordane (CC), *trans*-chlordane (TC), heptachlor, heptachlor epoxide, and oxy-chlordane) concentrations in the sediment core is shown in Fig. 4g. A peak-value ($1.31 \pm 0.27 \text{ ng g}^{-1}$) period of CHLs was observed in the late 1950s, when chlordane was first heavily used in Guangdong, Guangxi and Hainan province in China as these areas were highly impacted by termites. This was followed by a progressive decrease to a relative constant concentration between the mid-1970s and mid-1980s ($0.38 \pm 0.01 \text{ ng g}^{-1}$). Some enterprises had been gradually stopping production of chlordane (NIP 2007) at that time. Next, a relatively higher constant concentration was shown between the mid-1980s and 2000 ($0.47 \pm 0.14 \text{ ng g}^{-1}$), during which some production facilities were established in succession because termites were very severe in the South China, and there was a lack of high efficiency and low cost alternatives for prevention and termite control (NIP 2007).

After 2000, an obvious increasing trend was noted for chlordane until 2009 (from 0.37 to 1.72 ng g^{-1}).

The ratios of TC/CC (>1 for nearly 96 % samples) exhibited an increasing trend in recent 20 years (Fig. 4h), suggesting fresh inputs of chlordane. While they were lower than 1.0 in several samples in the upper layer of the sediment core, indicating a “weathered” chlordane source from soil residues. The ratios of TC/CC >1 were found in most samples, corresponding to the ratios in technical mixture, indicating more TC occurred than CC in sediments. A similar predominance of TC in sediments was observed in the Kyeonggi Bay, Korea (Lee et al. 2001). Previous studies suggested that the levels of TC were less than CC in most soil samples (Martinez et al. 2012). A significant distinction between sediment and soil is that the former is anaerobic while the latter is aerobic. This implies that TC degrades faster under aerobic conditions than under anaerobic conditions. Moreover, TC can be oxidized to oxy-chlordane by biotic and abiotic processes (Offenberg et al. 2004). The concentrations of oxy-chlordane were below detection limit in most sediment samples in the Beibu Gulf, indicating the lack of biodegradation of TC in sediments.

Endosulfan is an ingested insecticide extensively used throughout the world for over 40 years on a variety of crops (Sutherland et al. 2000). It started to be produced and used in 1994 in China. The total output of endosulfan in China was estimated to be approximately 25,700 tons between 1994 and 2004 (Jia et al. 2009). It has been widely used until 2011 when the Parties of the Stockholm Convention on Persistent Organic Pollutants (POPs) agreed to stop the production and application of endosulfan on a global scale (UNEP 2011).

The historical trend of endosulfan (sum of α - and β -endosulfan) concentrations in the sediment core is shown in Fig. 4i, which agrees with the application history of endosulfan in China. It can still be detected though no endosulfan had ever been used in China including the Beibu Gulf before 1994. This suggests that endosulfan was transferred by long-range atmospheric transport from other countries, such as India, the world's largest consumer of endosulfan. The second peak value (0.90 ng g^{-1}) was shown in 1994 (Fig. 4i) when endosulfan started to be produced for the control of pests in cotton in China and two endosulfan formulator companies were set up near the Beibu Gulf (Jia et al. 2009). Next, a sharp decline occurs between 1994 (0.90 ng g^{-1}) and 1998 (0.47 ng g^{-1}), which agrees with the reducing use of endosulfan in China (Jia et al. 2009). This was followed by a progressive increase since 1998 (from 0.47 to 0.91 ng g^{-1}) when endosulfan was allowed to expand its application on wheat, tea, tobacco, apples, and other fruits for the first time. Subsequently, its application increased continuously in China.

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