Science of the Total Environment 539 (2016) 388–394

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

DDTs and HCHs in sediment cores from the coastal East China Sea

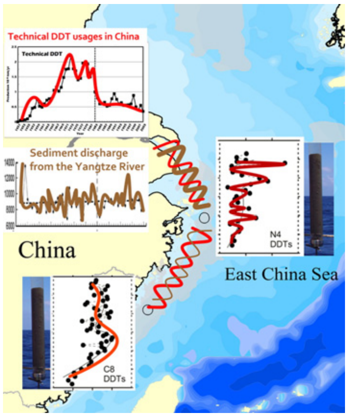
Tian Lin [a,b,*], Luca Nizzetto [c,d], Zhigang Guo [e], Yuanyuan Li [e], Jun Li [b], Gan Zhang [b]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[b] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[c] Research Centre for Toxic Compounds in the Environment, Masaryk University, Brno 62500, Czech Republic
[d] Norwegian Institute for Water Research, Gaustadalleen 21, Oslo 0349, Norway
[e] Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China

HIGHLIGHTS

- Sedimentary records, inventories and burdens of DDTs and HCHs were studied.
- The records in estuarine environment displayed remobilization of aged OCPs from soil.
- The records in open sea reflected historical trends of OCP application.
- The Yangtze outflow had a larger influence on the OCP exposure in the region.
- The Yangtze still delivers relatively high inputs of OCPs after the official ban.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 30 July 2015
Received in revised form 3 September 2015
Accepted 3 September 2015
Available online 12 September 2015

Editor: Adrian Covaci

Keywords:
HCHs and DDTs
Sedimentary record
Yangtze River
East China Sea

ABSTRACT

Four sediment cores were collected along the Yangtze-derived sediment transport pathway in the inner shelf of the East China Sea (ECS) for OCP analysis. The sediment records of HCHs and DDTs in estuarine environment reflected remobilization of chemicals from enhanced soil erosion associated to extreme flood events or large scale land use transformation. The sediment records in the open sea, instead, reflected long-term historical trends of OCP application in the source region. Unlike the so-called mud wedge distribution of sediment, inventories of HCHs and DDTs slightly increased from the mouth of Yangtze River alongshore toward south, suggesting the sediment deposition rate was one of factors on the exposure of chemicals within the inner shelf of the ECS. Re-suspension and transport of the Yangtze-derived sediment and consequent fractionation in grain size and TOC were also responsible for the spatial variation of inventories of catchment derived OCPs in a major repository area of the Yangtze suspended sediment. The total burdens of HCHs and DDTs in the inner shelf of the ECS were 35 tons and 110 tons, respectively. After 1983 (year of the official ban in China), those values were 13 tons and 50 tons, respectively. It appears that the Yangtze still delivers relatively high inputs of DDTs more than 30 years after the official ban. High proportions of DDD + DDE and β-HCH suggested those OCPs mainly originated from historical usage in the catchment recent years.

---

[*] Corresponding author at: State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China.
E-mail address: lintian@vip.gyig.ac.cn (T. Lin).

http://dx.doi.org/10.1016/j.scitotenv.2015.09.010
0048-9697/

1. Introduction

The river-derived material (water, sediments and pollutants) entering the seas has a tremendous ecological and toxicological influence on the estuaries, coastal zones, and even continental shelf (McKee et al. 2004; Chu et al. 2009). One of the best examples of the river-dominated ocean margins is the inner shelf of the East China Sea (ECS). The modern Yangtze River's annual sediment load has been about 400 million tons during 2000–2007. This river discharges ~87% of its annual sediments in the June–October flood season, when the northward Zhejiang–Fujian coastal current (ZFCC) is weak and the Taiwan Warm Current (TWWC) is strong due to the prevailing southeast monsoon. This leads to the accumulation of the Yangtze-derived sediments in the estuarine system, forming the Yangtze River Estuary mud area (YREMA) (Shen and Pan 2001; Su and Huh 2002). Short-term monthly deposition rates near the river mouth during this period are about 4.4 cm, in contrast to yearly integrated accumulation rates, which are an order of magnitude lower, ~1–5 cm/yr. (DeMaster et al. 1985). In winter, strong wind-induced waves readily re-suspend fine particles of the newly deposited sediments in YREMA which are then transported southward along the inner shelf by the southward ZFCC. This material is then constrained by the northward flowing TWWC to the inner shelf of the ECS. Such complex dynamics underpin the formation of the Min-Zhe coastal mud area (MZCMA) (Liu et al. 2006, 2007). Overall, a large amount of terrigenous sediment from the Yangtze River forms a “mud wedge” extending ~1000 km from the Yangtze mouth to the Taiwan Strait (Liu et al. 2006, 2007). Other sediment inputs from small rivers south of the Yangtze River, such as the Qiantang River, the Ou River and the Min River, and aerosol/dust, only make a relatively small contribution (<5%) to the mud wedge compared to those originating from the Yangtze River (Lim et al. 2007).

Until now, a large number of studies have been made in the ECS to document the fate of the Yangtze-derived sediment and various geological and geochemical materials (Lin et al. 2002; Liu et al. 2010a,b; Xing et al. 2011; Youn and Kim 2011; Xu et al. 2012; Duan et al. 2013a), however, little is known concerning the fate of Yangtze River-derived persistent organic pollutants (POPs) in this distinct depositional environment. A few studies were focusing on assessing regional distribution of surface sediments or vertical distribution of sediment cores only from the Yangtze Delta (Chen et al. 2002; Li et al. 2012; Duan et al. 2013b). Available information is however insufficient for understanding redistribution and fate of POPs in such a complex system.

Due to hydrophobic nature, the transport of POPs is largely linked to suspended sediment dynamics in the coastal environment (Latimer et al. 1999; Tsapakis et al. 2006). Analysis of sediment cores systematically collected along the sediment transport pathway is necessary to build up a more precise picture of historical pollutant inputs and their fate in the inner shelf of the ECS. The present work aimed at assessing levels, time trends and inventories of HCHs and DDTs in four sediment cores in a major repository area of the Yangtze suspended sediment plume from the ECS.

2. Materials and methods

2.1 Sample collection

In this study three sediment cores, namely N4 (123°00'E and 30°47'N, 148 cm long core), N6 (122°44'E and 30°30'N, 132 cm long core) and S5 (122°30'E and 29°00'N, 130 cm long core), were collected from June to September 2003, and one core (C8, 121°39'E and 27°38'N, 120 cm long core) was collected in April, 2009. The sediment cores (N4, N6, S5 and C8) located along the mud wedge area in the south–north direction (Fig. 1). All sediment cores were collected using a gravity corer deployed from the R/V Dong Fang Hong 2 of the Ocean University of China. The cores were cut into 1 cm (for core C8) or 2 cm (for cores N4, N6 and S5) sections along the length using a stainless steel cutter in the laboratory. The subsamples were packed in aluminum foil and stored at −20 °C until analysis.

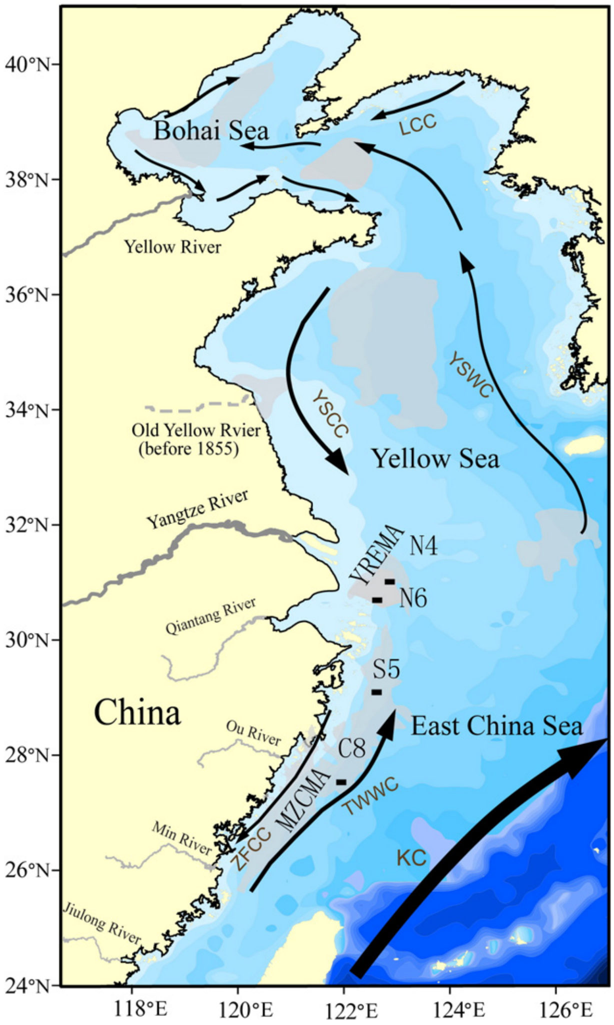


Fig. 1. Map of East China Sea showing the locations where sample cores were collected.

2.2 Sample treatment and analytical procedure

The sediment treatment and analytical procedures (including sediment grain size, sediment dry density, TOC and ^{210}Pb activities) are described in detail in the Supporting Information and previous papers (Guo et al. 2006, 2007).

2.3 Organic analysis of HCHs and DDTs

The organic analysis was completed for N4, N6 and S5 in 2006, and for C8 in 2011. As recorded in Table S1, there were 29 for core N4, 18 for core N6, 16 for core S5 and 48 for core C8 subsamples which had been selected and performed for OCPs analysis. In the laboratory, sediment samples were freeze-dried for 24 h, pulverized, and sieved through stainless steel sievers (80-mesh). About 5 g of the treated samples were extracted in a Soxhlet extractor for 48 h with dichloromethane (DCM) at a rate of 2–3 cycles/h. A mixture of 2,4,5,6-tetrachloro-m-xylene (TcmX) and decachlorobiphenyl (PCB209) was added to each of the samples as surrogate standards prior to extraction. Activated copper granules were added to the collection flask to remove elemental sulfur. The extract was concentrated and solvent-exchanged to hexane and purified using a 8 mm i.d. alumina/silica column packed (from the bottom to top) with neutral alumina (3 cm, 3% deactivated), neutral silica gel (3 cm, 3% deactivated), 50% (on a weight basis) sulfuric acid silica (2 cm), and anhydrous sodium sulfate. Alumina, silica gel, and anhydrous sodium sulfate were Soxhlet extracted for 48 h with DCM, and then baked for 12 h at 250, 180, and 450 °C before use, respectively. The column was eluted with 50 mL of dichloromethane/hexane (1:1) to yield the organochlorine pesticide (OCP) fraction. The fraction was solvent-exchanged to hexane and concentrated to 0.5 mL under a gentle nitrogen stream. A known quantity of pentachloronitrobenzene (PCNB) was added as internal standard prior to gas chromatography–electronic capture detector (GC–ECD) analysis.

OCPs were measured by GC–ECD (Agilent-6890 GC system, Hewlett-Packard, USA). Target analytes included 8 compounds: α-HCH, β-HCH, γ-HCH, δ-HCH, p,p′-DDT, o,p′-DDT, p,p′-DDE, p,p′-DDD in this study. A CP-Sil 8 CB capillary column (50 m, 0.25 mm, 0.25 μm; DB-5MS, Agilent, USA) was used, with helium as carrier gas at 1.2 mL $\min^{-1}$ under constant flow mode. Helium was filtered with moisture, hydrocarbon, and oxygen traps before entering the GC–ECD system. The oven temperature began at 60 °C for 1 min and increased to 290 °C (10 min hold time) at a rate of 4 °C $\min^{-1}$. Splitless injection of a 1 μL sample was performed with a 5 min solvent delay time. Injector temperature was 250 °C.

2.4 Quality control

All data were subject to strict quality control procedures, including the analysis of blanks and spiked blank samples. Seven method blanks, seven duplicate samples, and seven spiked blanks were analyzed (one every ten real samples). The average surrogate recoveries were 73.2 ± 10.1% and 94.9 ± 10.3% for TcmX and PCB209, respectively. The reported concentrations of HCHs and DDTs were not corrected for surrogate recoveries.

3. Results and discussion

3.1 Grain size distribution and sedimentation rates

Sediments near the Yangtze River Mouth (Cores N4 and N6) were dominated by clay (26%) and silt (62–70%), with a smaller fraction of sand (7%) (Figure S1). Xu et al. (2009) found that on average clay, silt and sand from the lower reaches of the Yangtze River were 18%, 72% and 9%. Concerning the vertical profile of grain composition, some anomalous values in the sand fraction were observed in several sediment samples in the cores N4 and N6 (Figure S1). The variation of the percentage of sand was explained by the inter-annual variation of water and sediment discharge, as well as the Yangtze River flood or ocean storm yearly (Wang et al. 2008). Sand fractions as low as 1% were detected in the sediments in the cores S5 and C8 collected from the MZCMA. These results confirm expectation regarding lower potential for re-suspension and long-range transport of coarse particles. In addition, constant clay/silt ratio in Cores S5 and C8 indicates a stable dynamic sedimentary environment in this area (Figure S1).

In general, sedimentation rates calculated from gradients of excess ^{210}Pb are valid only if the sediment column is not disturbed and the distribution of ^{210}Pb is at steady state. However, this ideal condition is not common, especially in a dynamic environment like the inner shelf of the ECS. Because of this it is more conservative to refer to the ^{210}Pb-based sedimentation rates determined here as apparent values. Cores N4 and N6 located in the YREMA displayed the highest apparent sedimentation rates, averaging 2.47 cm/yr. and 1.89 cm/yr., respectively (Figure S2). The average sedimentation rates for Core S5 and C8 (northern and middle part of the MZCMA) were about 1.0 cm/yr. (Figure S2). Depth distribution of excess ^{210}Pb in Core S5 and C8 ($R^2 > 0.8$) suggested the source of the sediments and sedimentation condition were stable over time (Figure S2).

Sedimentation rates and grain size distribution patterns obtained in this study are in agreement with previous reports (DeMaster et al. 1985; Huh and Su 1999). The highest sedimentation rates (>2.0 cm/yr) are found in the estuary of the Yangtze River from which is the largest source of sediments in the ECS. There exists a general decrease of sedimentation rates from the mouth of Yangtze River alongshore toward south, meanwhile partial fine-grained sediments from the Yangtze River were transported and later re-deposited southward along the inner shelf (Liu et al. 2006; Wang et al. 2007b).

The concentrations of HCHs and DDTs in the surface sediments generally increased southward along the mud wedge area (Fig. 2), possibly reflecting the decrease of sediment deposition rates, increase proportion of fine-grain sediments and increase TOC (Zhu et al. 2011; Lin et al. 2013). These findings are in good agreement with the gradual increase of the PAH concentrations in surface sediment reported in earlier work (Lin et al. 2013). Although the YRE and its adjacent YREMA serve as main sink for the fluvial-derived sediments, sediment re-suspension and transport play an effective role in remobilizing and consequently “diluting” the concentration levels over a broader area.

3.2 Vertical variation of HCHs and DDTs in sediment cores in the ECS

In Core S5 and C8 collected from the MZCMA, eight targeted compounds were detected in all samples. Differently, δ-HCH had detection frequencies of 83% (24/29) and 44% (8/18) in core N4 and N6, respectively (Table S1). The vertical variations of HCH and DDT concentrations in sediment cores are shown in Fig. 2. Although the Yangtze outflow is expected to be the predominant HCH and DDT source in the mud wedge area, with expectedly no significant contribution from other sources, sedimentary records varied considerably site-by-site along the monitored transect (Fig. 2). Substantial site-by-site differences of deposition rate along the sediment distribution pathway, and accompanying sediment re-suspension and transport have clearly played a crucial role in altering source signal from Yangtze River, and are the most likely behaviors responsible for such a spatial variability in time trend in the major repository area.

A considerable fluctuation of HCH and DDT concentrations along with core depth was the most obvious feature of Core N4 in proximity to the estuary. In this core, there is no identifiable general trend in distribution, presumably due to mixed effects of varied sedimentation rates, shallow water depth (<20 m) and strong re-suspension. In spite of this, the periods of the higher Yangtze sediment loading (associated with extraordinary flooding) coincided with those of the higher concentrations of HCHs or DDTs (e.g. 1971, 1991 and 1998, especially of DDTs) (Dai and Lu 2010). The accumulation of these pollutants in the sediment bed was sensitive to enhanced outflow due to the fact that sediment and associated pollutants were deposited rapidly and well-preserved in the estuary mud area during flooding. A sudden increase of pesticide concentrations was observed during the late 1980s and the 1990s (being this behavior more visible in Core N6). HCHs and DDTs were widely used in China between 1950s and 1980s, and they were officially banned in 1983. Since these contaminants are effectively retained in soils, the contaminated agricultural areas can behave as secondary source for many years after the ban. The sedimentary records of samples N4 and N6 at the estuary of the Yangtze confirms that soil erosion is a significant process affecting remobilization of OCPs as previously hypothesized by Zhang et al. (2002). Furthermore, the enhanced soil erosion associated with large scale land use change and other forms of disturbance can promote such a remobilization. The lower Yangtze region experienced a dramatic transformation from an agricultural economy to an industrial one after the middle 1980s and until year 2000, with an intense conversion of lands from agriculture to residential or industrial which may have caused a significant increase in soil erosion. Such a large scale transformation could have resulted in significant remobilization of persistent contaminants from agricultural soils (Figure S3). After year 2000 the rate of land use change significantly declined in this area (Figure S3). Notably this trend is reflected by a concomitant decline of OCP concentrations in the sediments (Fig. 2).

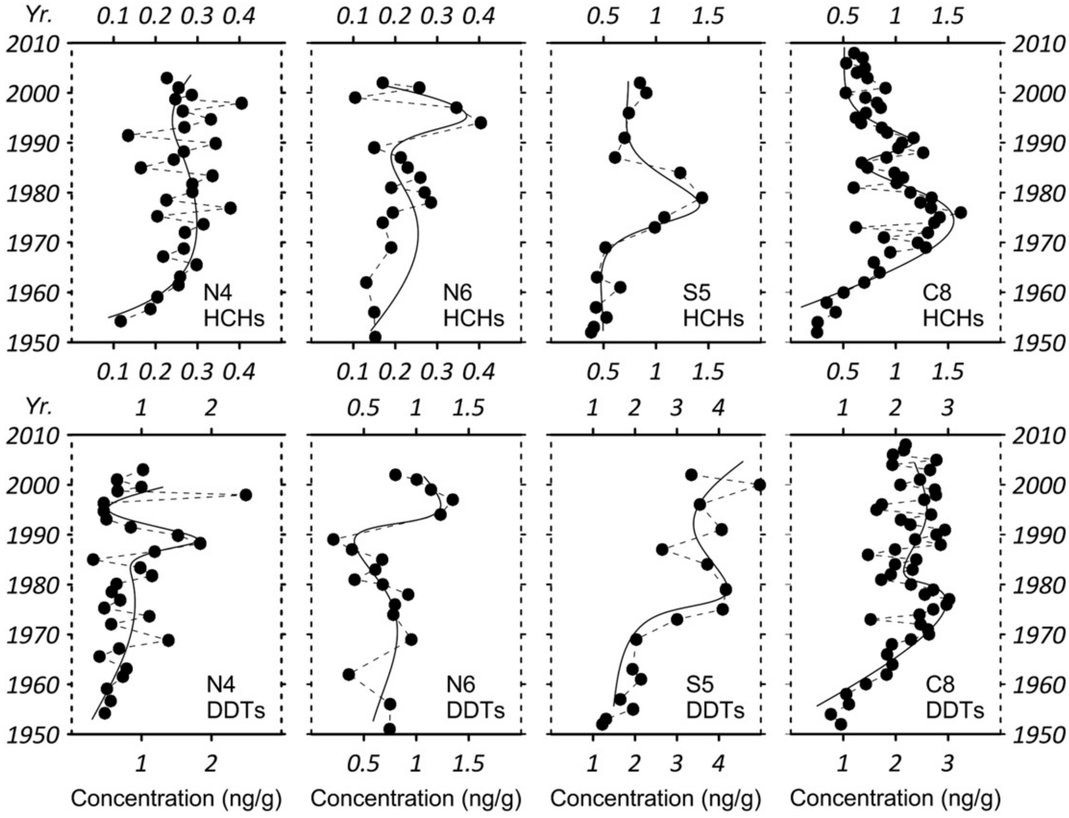


Fig. 2. Time trends of HCH and DDT concentrations in sediment cores N4, N6, S5 and C8.

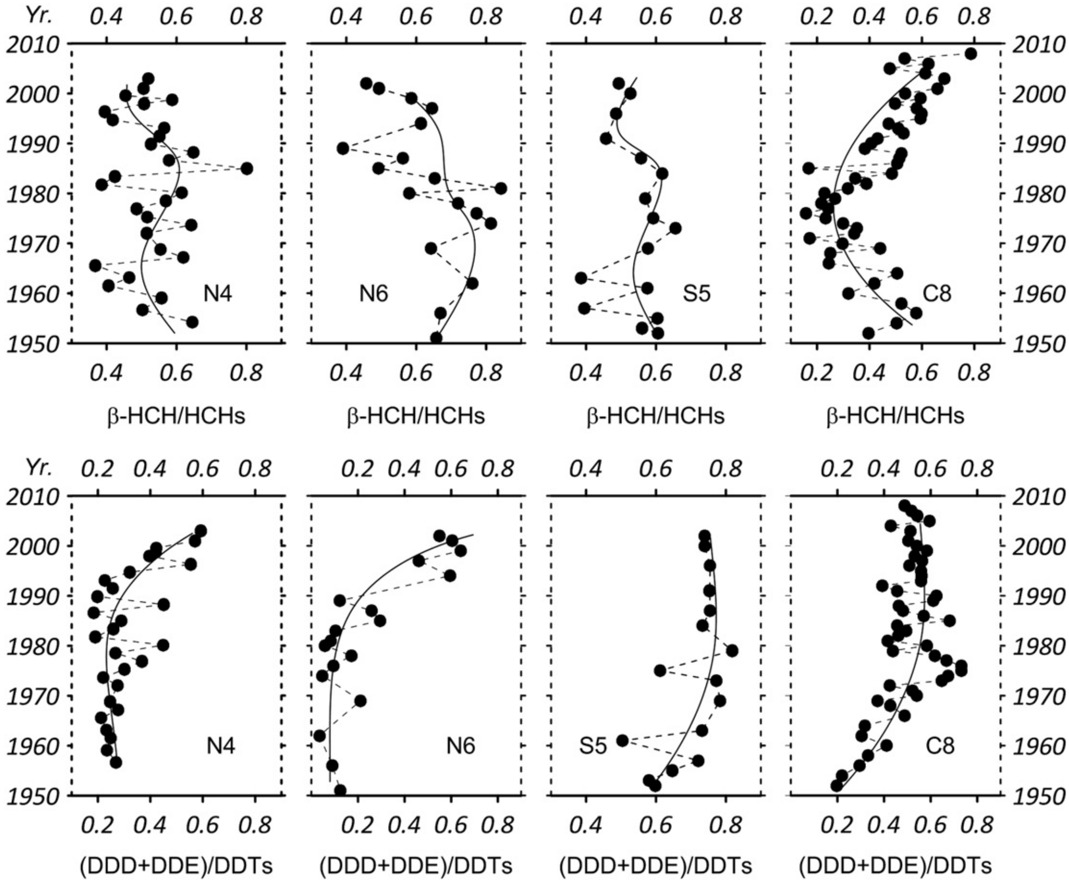


Fig. 3. Down-core variations of β-HCH/HCHs and (DDE + DDD)/DDTs ratios in the sediment cores in the ECS.

Present result represents an evidence of the link between intense land use change and increased remobilization of OCPs from soils over a vast geographical area. The vertical profiles of DDT metabolite signature in core N4 and N6, appear to confirm this link. DDE and DDD proportionally increased (relatively to DDTs) upward in the cores, suggesting catchment-derived aged DDT previously stored in soils was the principal source of sediment exposure (Li et al. 2006) (Fig. 3).

In Core S5 and C8 collected from the MZCMA, an initial increase in HCHs and DDTs levels was observed during the period 1960–1980. These corresponded to the period of most intensive use of HCHs and DDTs in China (Wang et al. 2005, Figure S4). The dramatic decline of HCHs immediately observed after the 1980s in these cores can be attributed to regulatory restrictions and ban in the agricultural usage (Lin et al. 2012a, Figure S4). The ratio of β-HCH/HCHs consistently increased after 1980 in core C8, also suggesting there was no fresh HCH input after the ban (Fig. 3). Similarly, a constant and higher ratio of (DDD + DDE)/DDTs (>0.6) can be observed following the 1980s, suggesting DDTs were mainly derived from secondary source. However, in the same cores there is no evidence of declining trend in DDTs following its ban in China in 1983. DDTs had lower volatilities compared with HCHs, and the DDT residues in agricultural soil should be more persistent than HCHs after the ban. Besides, although the general trend of DDTs in the sediment cores suggest aged DDTs stored in soils as the main source, on-going inputs of fresh DDTs from the Yangtze catchment to the inner shelf of the ECS cannot be excluded. After technical DDT was banned in agriculture, technical DDT was used for more than additional twenty years for other purposes, (e.g. DDT-activated antifouling paint and public health). This may help to partially explain the observed high p,p'-DDT levels in the sedimentary record of the inner shelf of the ECS after the ban (Lin et al. 2009, 2012b).

Since the Yangtze-derived HCHs and DDTs are deposited quickly with high sediment deposition rates in YREMA, the signal of soil erosion controlled by intra-annual variation of water and sediment discharge by the Yangtze can be well preserved as shown in sedimentary records of HCHs and DDTs in the estuarine environment. As a consequence, sediment deposition rate was key factor in determining sediment records of HCHs and DDTs in the estuarine environment. In contrast, expected diminishing signal of short term extreme events in sedimentary record was observed with sediment re-suspension and transport as it appears from the cores collected in the southern inner shelf of the ECS. A number of processes can play a role in determining observed variability during the sediment re-suspension and transport. These possibly include physical dispersion of sediments carrying different contaminant signatures and subsequent mixing in the water column, re-equilibration of contaminants between suspended sediments and the water phase, and contaminant degradation. Considering the time lag for the transport of sediment re-suspended from the estuarine environment by the ZFCC and TWWC toward their stable depositional environment, the redistribution between sediment and water tended to be in approximate balance in water column. As a result, sediment cores (e.g. C8) collected in the distal mud area in the southern inner shelf of the ECS, reflected the overall trend of OCP input into the ECS, which show sedimentary records fitting more with available data of historical application before the ban as well as significant rebound driven by enhanced soil erosion from the late 80s.

3.3 Mass inventories

The fluxes (F, ng/cm^2 yr) and inventories (I, ng/cm^2, 1950–2003) of HCHs and DDTs were calculated for each core according to the following equations (Eqs. (1)–(2)):

$$\mathrm{Fi} = \mathrm{CipiRi} \tag{1}$$

$$\mathrm{I} = \sum \mathrm{Fi} \tag{2}$$

where Ci (ng/g) is the concentration of HCHs or DDTs in sediment, pi (g/cm^3) is the dry sediment density, and Ri is sedimentation rate cm/yr. Since pesticide concentration was not measured in all the core sections representing individual years, linear interpolation was used to estimate flux data of the non-analyzed sections (Fig. 4, Table S2).

Although the OCP concentrations within the inner shelf of the ECS varied considerably across locations (factor of 3–5) (Fig. 3), inventories only ranged between 32 and 47 ng/cm^2 for HCHs and 86–144 ng/cm^2 for DDTs (Fig. 5). Unlike concentrations, the HCH and DDT fluxes and inventories remained relative constant in the sediment transport direction as a result of different sedimentation rates, re-distribution/release of OCPs across and from suspended sediments along the transport path.

Inventories estimated here are lower than those previously reported for estuaries or coastal environments of heavily polluted areas such as the Pearl River (36.6–1109.5 ng/cm^2 for DDTs and 11.2–226.3 ng/cm^2 for HCHs with a span of 30–50 years (Zhang et al. 2002)). However, the Yangtze OCP outflow was estimated to be higher (of a factor of ~6) compared to that of the Pearl River (Wang et al. 2007a; Chen et al. 2006). This result suggests that a large amount of the Yangtze River-derived OCP is deposited over a much larger areas resulting in lower local inventories in individual point.

3.4 Total burdens in the ECS

The calculated inventories of HCHs and DDTs in the sediment cores were used to provide a first rough estimate of the total burdens of HCHs and DDTs in the inner shelf of the ECS (Fig. 5). The depositional area covers a surface of approximately 100,000 km^2 within the inner shelf of the ECS. Burdens were calculated in each area (eight areas divided every 0.5° latitude from 27°N to 31°N) by assuming the mean value of inventory correlated with TOC and sediment rate across all the area (Fig. 5). This resolution is clearly insufficient for providing a detailed picture of burdens for such a large surface. Its result however can provide useful information on the magnitude of the sediment compartment as storage and potential secondary source for the aquatic environment. The total sediment burdens of HCHs and DDTs in the inner shelf of the ECS were estimated to be 35 tons and 110 tons, respectively. After 1983, the total sediment burdens of HCHs and DDTs in the inner shelf of the ECS were 13 tons and 50 tons, equivalent to one third and one half of the total, respectively. It appears therefore that the Yangtze still delivers relatively high inputs of DDTs more than 30 years after the official ban. Besides, this is important since this shallow water area routinely experiences extreme meteorological events associated to the monsoon regime, which can lead to sediment disturbance, resuspension and continuous upward re-mobilization of old OCPs.

Historical data on OCP production in China (Wei et al. 2007) report a total production of technical HCH (4.9 million tons) much higher than technical DDT (0.4 million tons). In contrast, the observed mean ratio of sedimentary burdens of HCHs and DDTs was 1:3 in the inner shelf of the ECS. From the source-to-sink point, the relative inventory/burden of HCHs compared to that of DDTs consistently declined. This is clearly due to differences in physicochemical and biochemical properties, as the HCHs have higher water solubility, vapor pressure and biodegradability, and consequently lower affinity to sediments and lower persistence compared to the DDTs (Shen and Wania 2005). Thus, HCHs can transport and disperse more widely than DDTs, while DDTs is more restrained in sediment/soil near the source regions. In addition, compared with its original components (both Technical HCH and Lindane), β-HCH was the most abundant one among the four isomers of HCHs (with a mean percentage of over 40%, Fig. 3) in all sediment cores, suggesting sediment is still the major sink of β-HCHs in the environment.

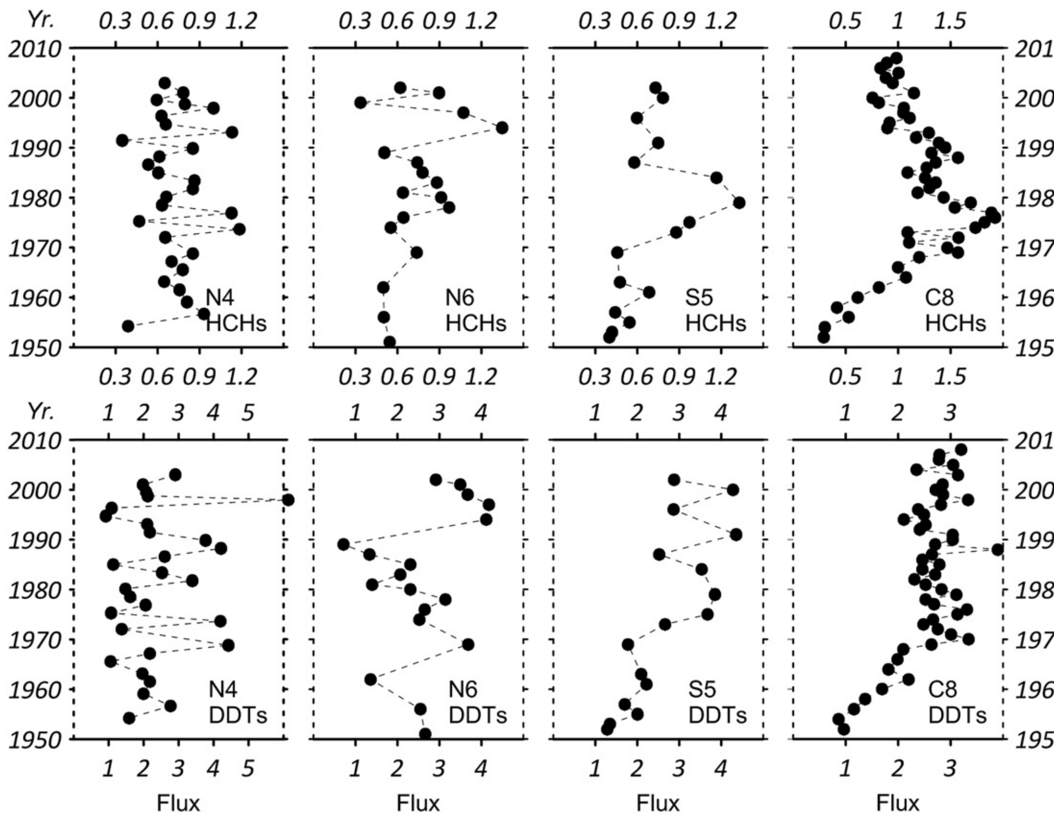


Fig. 4. Down-core variations of HCH and DDT fluxes in sediment cores in the ECS.

4. Conclusions

This study documented the influence of riverine input from catchment in defining sedimentary records over a very large receptor area in the continental shelf. The aged OCP remobilization associated to enhanced soil erosion in the Yangtze catchment are very well represented in the sedimentary records in the estuarine environment. Considering the time lag for the transport of sediment re-suspended from the Yangtze estuarine environment, direct influence of the Yangtze River sediment discharge was decreasing along the mud area. The sediment records from the distal mud area along the coast gave expected time trend of their regional application history.

Unlike the so-called mud wedge distribution of sediment, the HCH and DDT fluxes and inventories remained relative constant in the sediment transport direction, suggesting Yangtze River outflow had a large influence on the exposure over the ECS associated with re-suspension and transport of the sediment in the estuary. After OCPs official ban in China (1983), the total sediment burdens of DDTs still represented a large fraction of the total sediment burden, showing an on-going high input from the catchment associated with the enhanced soil erosion and new usage.

Acknowledgments

Data supporting are available in Supporting Information. This work was supported by the Natural Science Foundation of China (NSFC) (No. 41103046, 41076015), partially by the State Key Laboratory of Environmental Geochemistry, Institute Of Geochemistry, CAS. This work has also been partially supported by the RECETOX research infrastructure (project of the Ministry of Education of the Czech Republic LM2011028),

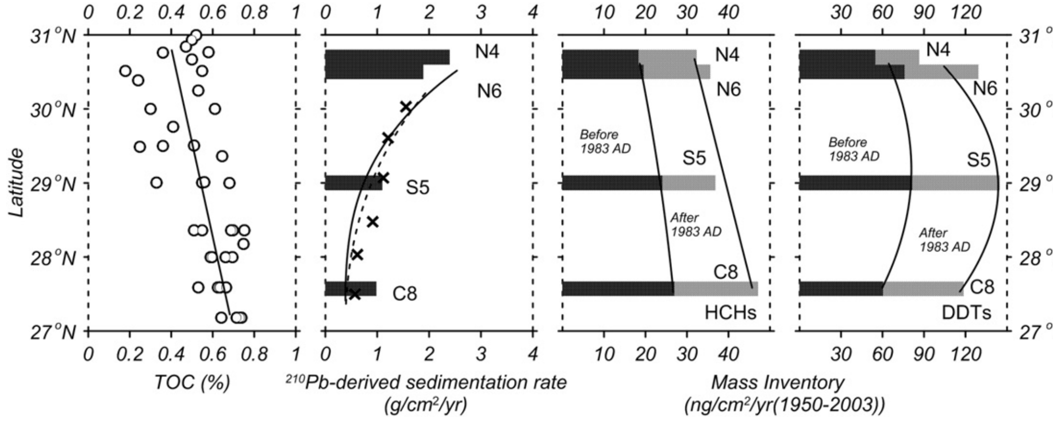


Fig. 5. The mass inventories of HCHs and DDTs estimated on the data from the Core N4, N6, S5 and C8. (Data of TOC ( ) and ^{210}Pb-derived sedimentation rate (×) was from published sources (Lin et al. 2013)).

and the CETOCOEN project of the European Structural Funds (CZ.1.05/2.1.00/01.001).

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.09.010.

References

Chen, J.F., Xia, X.M., Ye, X.R., Jin, H.Y., 2002. Marine organic pollution history in the Changjiang Estuary and Zhejiang coastal area—HCHs and DDTs stratigraphical records. Mar. Pollut. Bull. 45, 391–396.

Chen, S.J., Luo, X.J., Mai, B.X., Sheng, G.Y., Fu, J.M., Zeng, E.Y., 2006. Distribution and mass inventories of polycyclic aromatic hydrocarbons and organochlorine pesticides in sediments of the Pearl River Estuary and the northern South China Sea. Environ. Sci. Technol. 40 (3), 709–714.

Chu, Z.X., Zhai, S.K., Lu, X.X., Liu, J.P., Xu, J.X., Xu, K.H., 2009. A quantitative assessment of human impacts on decrease in sediment flux from major Chinese rivers entering the western Pacific Ocean. Geophys. Res. Lett. 36, L19603.

Dai, S.B., Lu, X.X., 2010. Sediment deposition and erosion during the extreme flood events in the middle and lower reaches of the Yangtze River. Quat. Int. 226, 4–11.

DeMaster, D.J., McKee, B.A., Nittrouer, C.A., Jiangchu, Q., Guodong, C., 1985. Rates of sediment accumulation and particle reworking based on radiochemical measurements from continental shelf deposits in the East China Sea. Cont. Shelf Res. 4, 143–158.

Duan, L., Song, J., Yuan, H., Li, X., Li, N., 2013a. Spatio-temporal distribution and environmental risk of arsenic in sediments of the East China Sea. Chem. Geol. 340, 21–31.

Duan, X., Li, Y., Li, X., Li, M., Zhang, D., 2013b. Distributions and sources of polychlorinated biphenyls in the coastal East China Sea sediments. Sci. Total Environ. 463–464, 894–903.

Guo, Z., Lin, T., Zhang, G., Yang, Z., Fang, M., 2006. High-resolution depositional records of polycyclic aromatic hydrocarbons in the central continental shelf mud of the East China Sea. Environ. Sci. Technol. 40, 5304–5311.

Guo, Z., Lin, T., Zhang, G., Zheng, M., Zhang, Z., Hao, Y., Fang, M., 2007. The sedimentary fluxes of polycyclic aromatic hydrocarbons in the Yangtze river estuary coastal sea for the past century. Sci. Total Environ. 386, 33–41.

Huh, C.A., Su, C.C., 1999. Sedimentation dynamics in the East China Sea elucidated from ^{210}Pb, ^{137}Cs and ^{239,240}Pu. Mar. Geol. 160, 183–196.

Latimer, J.S., Davis, W.R., Keith, D.J., 1999. Mobilization of PAHs and PCBs from in-place contaminated marine sediments during simulated resuspension events. Estuar. Coast. Shelf Sci. 49, 577–595.

Li, J., Zhang, G., Qi, S.H., Li, X.D., Peng, X.Z., 2006. Concentrations, enantiomeric compositions, and sources of HCH, DDT and chlordane in soils from the Pearl River Delta, South China. Sci. Total Environ. 372, 215–224.

Li, Y., Lin, T., Chen, Y., Hu, L., Guo, Z., Zhang, G., 2012. Polybrominated diphenyl ethers (PBDEs) in sediments of the coastal East China Sea: occurrence, distribution and mass inventory. Environ. Pollut. 171, 155–161.

Lim, D.I., Choi, J.Y., Jung, H.S., Rho, K.C., Ahn, K.S., 2007. Recent sediment accumulation and origin of shelf mud deposits in the Yellow and East China Seas. Prog. Oceanogr. 73, 145–159.

Lin, S., Hsieh, I.J., Huang, K.M., Wang, C.H., 2002. Influence of the Yangtze River and grain size on the spatial variations of heavy metals and organic carbon in the East China Sea continental shelf sediments. Chem. Geol. 182, 377–394.

Lin, T., Li, J., Xu, Y., Liu, X., Luo, C., Cheng, H., Chen, Y., Zhang, G., 2012a. Organochlorine pesticides in seawater and the surrounding atmosphere of the marginal seas of China: spatial distribution, sources and air–water exchange. Sci. Total Environ. 435–436, 244–252.

Lin, T., Hu, L., Shi, X., Li, Y., Guo, Z., Zhang, G., 2012b. Distribution and sources of organochlorine pesticides in sediments of the coastal East China Sea. Mar. Pollut. Bull. 64, 1549–1555.

Lin, T., Hu, L., Guo, Z., Zhang, G., Yang, Z., 2013. Deposition fluxes and fate of polycyclic aromatic hydrocarbons in the Yangtze River estuarine-inner shelf in the East China Sea. Glob. Biogeochem. Cycles 27, 77–87.

Lin, T., Hu, Z.H., Zhang, G., Li, X.D., Xu, W.H., Tang, J.H., Li, J., 2009. Levels and mass burden of DDTs in sediments from fishing harbors: the importance of DDT-containing antifouling paint to the coastal environment of China. Environ. Sci. Technol. 43, 8033–8038.

Liu, H., He, Q., Wang, Z., Weltje, G.J., Zhang, J., 2010a. Dynamics and spatial variability of near-bottom sediment exchange in the Yangtze Estuary, China. Estuar. Coast. Shelf Sci. 86, 322–330.

Liu, J.P., Li, A.C., Xu, K.H., Velozzi, D.M., Yang, Z.S., Milliman, J.D., DeMaster, D.J., 2006. Sedimentary features of the Yangtze River-derived along-shelf clinoform deposit in the East China Sea. Cont. Shelf Res. 26, 2141–2156.

Liu, J.P., Xu, K.H., Li, A.C., Milliman, J.D., Velozzi, D.M., Xiao, S.B., Yang, Z.S., 2007. Flux and fate of Yangtze River sediment delivered to the East China Sea. Geomorphology 85, 208–224.

Liu, S., Zhang, W., He, Q., Li, D., Liu, H., Yu, L., 2010b. Magnetic properties of East China Sea shelf sediments off the Yangtze Estuary: influence of provenance and particle size. Geomorphology 119, 212–220.

McKee, B.A., Aller, R.C., Allison, M.A., Bianchi, T.S., Kineke, G.C., 2004. Transport and transformation of dissolved and particulate materials on continental margins influenced by major rivers: benthic boundary layer and seabed processes. Cont. Shelf Res. 24, 899–926.

Shen, H.T., Pan, D., 2001. Turbidity maximum in the Changjiang estuary. China Ocean Press, Beijing, pp. 28–29 (in Chinese).

Shen, L., Wania, F., 2005. Compilation, evalution and selection of physical–chemical property data for organochlorine pesticides. J. Chem. Eng. Data 50 (3), 742–768.

Su, C.C., Huh, C.A., 2002. ^{210}Pb, ^{137}Cs and ^{239,240}Pu in East China Sea sediments: sources, pathways and budgets of sediments and radionuclides. Mar. Geol. 183, 163–178.

Tsapakis, M., Apostolaki, M., Eisenreich, S., Stephanou, E.G., 2006. Atmospheric deposition and marine sedimentation fluxes of polycyclic aromatic hydrocarbons in the eastern Mediterranean Basin. Environ. Sci. Technol. 40, 4922–4927.

Wang, H., Yang, Z., Wang, Y., Saito, Y., Liu, J.P., 2008. Reconstruction of sediment flux from the changjiang (Yangtze River) to the sea since the 1860s. J. Hydrol. 349 (3–4), 318–332.

Wang, J.Z., Guan, Y.F., Ni, H.G., Luo, X.L., Zeng, E.Y., 2007a. Polycyclic aromatic hydrocarbons in riverine runoff of the Pearl River Delta (China): concentrations, fluxes, and fate. Environ. Sci. Technol. 41, 5614–5619.

Wang, T.Y., Lu, Y.L., Zhang, H., Shi, Y.J., 2005. Contamination of persistent organic pollutants (POPs) and relevant management in China. Environ. Int. 31, 813–821.

Wang, Z.H., Li, L.Q., Chen, D.C., Xu, K.Q., Wei, T.Y., Gao, J.H., Zhao, Y.W., Chen, Z.Y., Masabate, W., 2007b. Plume front and suspended sediment dispersal off the Yangtze (Changjiang) River mouth, China during non-flood season. Estuar. Coast. Shelf Sci. 71, 60–67.

Wei, D.B., Kameya, T., Urano, K., 2007. Environmental management of pesticidal POPs in China: past, present and future. Environ. Int. 33, 894–902.

Xing, L., Zhang, H., Yuan, Z., Sun, Y., Zhao, M., 2011. Terrestrial and marine biomarker estimates of organic matter sources and distributions in surface sediments from the East China Sea shelf. Cont. Shelf Res. 31, 1106–1115.

Xu, K., Li, A., Liu, J.P., Milliman, J.D., Yang, Z., Liu, C.S., Kao, S.J., Wan, S., Xu, F., 2012. Provenance, structure, and formation of the mud wedge along inner continental shelf of the East China Sea: a synthesis of the Yangtze dispersal system. Mar. Geol. 291–294, 176–191.

Xu, K., Milliman, J.D., Li, A., Paul Liu, J., Kao, S.J., Wan, S., 2009. Yangtze- and Taiwan-derived sediments on the inner shelf of East China Sea. Cont. Shelf Res. 29, 2240–2256.

Youn, J., Kim, T.J., 2011. Geochemical composition and provenance of muddy shelf deposits in the East China ea. Quat. Int. 230, 3–12.

Zhang, G., Parker, A., House, A., Mai, B., Li, X., Kang, Y., Wang, Z., 2002. Sedimentary records of DDT and HCH in the Pearl River Delta, South China. Environ. Sci. Technol. 36, 3671–3677.

Zhu, C., Wang, Z.H., Xue, B., Yu, P.S., Pan, J.M., Wagner, T., Pancost, R.D., 2011. Characterizing the depositional settings for sedimentary organic matter distributions in the lower Yangtze River–East China Sea shelf system. Estuar. Coast. Shelf Sci. 93, 182–191.