

Earth Surface Processes and Environmental Sustainability in China

Occurrence and possible sources of organochlorine pesticides in soils of Ningbo, East China

Jianfeng TANG^{1,2*}, Jiayong FENG³, Gang LI^{1,2}, Mengqi LIANG², Ruijie WANG^{1,2} and Chao CAI¹

¹ Key Lab of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China.

Email: jftang@iue.ac.cn

² Ningbo Urban Environment Observation and Research Station Environment, Chinese Academy of Sciences, Ningbo 315800, China.

³ Ningbo Environmental Monitoring Center, Ningbo 315012, China.

*Corresponding author

ABSTRACT: Study of the residues of organochlorine pesticides (OCPs) can assist with the assessment of the contamination level, distribution, sources, transportation and trends in these selected OCPs. We investigated the concentration levels of hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethane (DDT), chlordane, which is currently used, and the never-used aldrin and dieldrin in the Ningbo region – a heavily industrialised and urbanised area in East China. OCPs were found in all soil samples and were predominantly composed of HCHs, DDT and its metabolites. The concentration of DDTs in the soil ranged from 2.2 to 566.6 ng g⁻¹, with a mean value of 55.6 ± 94.8 ng g⁻¹, while the concentration of HCHs ranged from 2.7 to 28.2 ng g⁻¹, with an average of 4.6 ± 2.9 ng g⁻¹. Aldrin, dieldrin, endrin and heptachlor were detected at much lower concentrations. The wide distribution of these never-used OCPs indicates that they have been input via long-range atmospheric processes. The results from analysis suggest that HCHs and DDTs were mainly derived from use in historical times.

KEY WORDS: DDTs, HCHs, OCPs.

Organochlorine pesticides (OCPs) are synthetic compounds with great chemical stability in the environment and represent an important group of persistent organic pollutants (POPs). These compounds are ubiquitous in the environment (Hu *et al.* 2014) and have had widespread use since the middle of last century. Thus, there has been extensive environmental concern about OCPs due to their toxicity, persistence, bioaccumulation and biomagnification. In addition, some of the OCPs are believed to act as endocrine disruptors affecting the hormone regulation of the human body (Hu *et al.* 2014; Qu *et al.* 2015; Wei *et al.* 2015).

As a large agricultural country, China has been a major producer and consumer of OCPs since the 20th Century, thus resulting in high levels of pesticides and their metabolites in various environmental media (Briz *et al.* 2011; Zhang *et al.* 2011). The assortment of OCPs included hexachlorocyclohexane (HCH), dichlorodiphenyltrichloroethane (DDT), aldrin, dieldrin, heptachlor (HEPT), endrin and others. DDTs and HCHs were two pesticides that were largely produced and used in China during 1951–1983, and production reached 0.4 million and 4.9 million t, respectively, which accounted for 33% and 20% of the global production, respectively (Briz *et al.* 2011; Wei *et al.* 2015). The next are chlordane, hexachlorobenzene (HCB) and HEPT. China produced 363 and 3522 tons per year, respectively, until they were banned in 2004 in China (Fang *et al.* 2017). At present, endosulfan is still

produced and used on crops in China (Fang *et al.* 2017). HEPT was never applied in the purified form, but it is a component of technical chlordane (Zhang *et al.* 2011).

Soils are still an important reservoir for OCPs due to their persistence, even though the production of some OCPs was stopped 30 years ago. Even though numerous countries have withdrawn from the use of OCPs over the last few decades, these compounds still persist at considerable levels in the environment (Hu *et al.* 2014). Many researchers have investigated OCPs in Chinese soils, and the total concentrations of DDTs and HCHs vary from ND (not detected) to 2910 ng g⁻¹ and from ND to 131 ng g⁻¹ (Cai *et al.* 2008). Recent observations have revealed large concentrations of OCPs within China and new inputs of DDTs are still entering the environment, mainly of the dicofol-type (Qiu *et al.* 2005; Zhang *et al.* 2008; Jiang *et al.* 2009; Liu *et al.* 2009; Yang *et al.* 2010). In addition, most of the published studies focus on DDTs and HCHs, and the study for other OCPs, such as endosulfan, chlordane and HEPT, are scarce. The State Department of China has proposed an action plan for soil pollution prevention (ten chapters for soil). Therefore, an investigation into the distribution of OCPs in soils can provide valuable data about soil contamination.

OCPs occur in the E of China in many industrial urban areas and also where there is agriculture. It is important to understand the sources, pathways and fates of OCPs in China. Ningbo is one of the most developed cities on the Yangtze



River Delta (YRD) in the Zhejiang Province, where OCPs were extensively used from the 1950s to the 1980s (Tieyu *et al.* 2005; Zhang *et al.* 2012). As a result, there is a history of OCP pollution in this area, and identifying the sources of the various contaminants is complicated. The levels of OCPs in agricultural soils in Zhejiang Province were high (the levels of most of the soil samples were $>1 \text{ ng g}^{-1}$ dry weight (dw)); the highest concentration of endosulfan was 42 ng g^{-1} dw (Zhang *et al.* 2012). Ningbo is a major commercial, industrial and transportation centre, and there is little information on OCP residue levels in topsoils in this region. The objective of this study was to investigate the current status of residues for OCPs in the Ningbo region and to evaluate their possible sources.

1. Materials and methods

1.1. Materials

Standards of HCHs, DDTs, trans-chlordane (TC), cis-chlordane (CC), aldrin, dieldrin, endrin, HEPT and endrin were purchased from Supelco (Bellefonte, PA, USA). All solvents used in extraction and analysis procedures were HPLC grade.

1.2. Sampling

Sampling was carried out in the Ningbo region, in the S of YRD, in September 2012. Ningbo is located in Zhejiang Province, comprising a total area of about 9695 km^2 , and is one of the most industrialised and urbanised regions in China. A total of 83 surface soil samples were collected in Ningbo (Fig. 1). Each soil sample consisted of 3–5 subsamples, collected between depths of 0 and 20 cm. All soil samples were collected using a hand auger, and then stored in polyethylene bags. The soil samples were kept at 20°C until analysis. To investigate the pollution impact of various human activities, land use was classified into five types: farmland soils, vegetable soils, industrial areas, residential areas and traffic areas (Fig. 1).

1.3. Extraction and preparation

Soil samples were freeze-dried for 24 h, pulverised and sieved through a stainless steel 80-mesh. The samples ($\sim 10 \text{ g}$) were then placed in an accelerated solvent extraction cell (ASE 300, Dionex, Canada); the extraction was then carried out using acetone and dichloromethane (1:1) at a temperature of 100°C and a pressure of 1500 psi. The extraction was repeated twice for each sample. The combined extracts were solvent exchanged with 10 ml of hexane and then concentrated to about 3 ml using a rotary evaporator. The extracts were then purified by a silica/aluminium column. The fraction was reduced to exactly 1 ml by volume under a gentle stream of ultra-pure nitrogen gas.

1.4. Instrumentation

An analysis of the OCPs was performed using Agilent 6890 series gas chromatography (GC), equipped with a 7863 auto-sampler and a micro-electron capture detector (ECD, Agilent Technologies, Palo Alto, CA, USA). Analytes were separated with an Agilent HP-1701 type capillary column ($30 \text{ m} \times 0.25 \text{ mm} \times 0.25 \mu\text{m}$, Agilent Technologies). Nitrogen was used as a carrier gas at 0.9 ml min^{-1} under the constant flow mode. A $1 \mu\text{l}$ sample was injected into the GC using a splitless mode. The oven was programmed from 60 to 160°C (2-min hold time) at a rate of $20^\circ\text{C min}^{-1}$, then to 260°C at a rate of 4°C min^{-1} , and held for 5 min. Injector and detector temperatures were maintained at 200°C and 280°C , respectively. Identity was established using retention time comparisons and the retention window was set at $\pm 0.5\%$. Calibration curves were based on area using internal standards.

1.5. Quality control and quality assurance

For each set of ten field samples, a procedural blank and a matrix sample spiked with standards were used to determine the accuracy. Each sample was analysed in duplicate unless otherwise stated. In addition, surrogate standards were added to each of the samples. The recovery ratios for the surrogates in the samples conform to the reported ranges by the US Environmental Protection Agency. The average recoveries were $78 \pm 9\%$ and $86 \pm 8\%$ for tetrachloro-*m*-xylene and decachlorobiphenyl. The recoveries of OCPs ranged from 72% to 109%. Reported OCP concentrations were corrected according to the recoveries of the standards.

1.6. Data analysis

Data analysis was performed using SPSS 16.0 (SPSS Inc., IL, USA). Concentrations were log transformed before statistical analysis. The levels of statistical significance were set at $P < 0.05$.

2. Results and discussion

2.1. OCP residues in soils

The concentration levels of individual OCPs in all soil samples are given in Table 1. The compounds identified included HCHs, cyclodienes (aldrin, dieldrin, endrin, HEPT), DDTs and its decomposition products. The results show that HCHs and DDTs were the predominant contaminants in the soils of the study area and were detected in all soil samples. The concentration of \sum DDTs was much greater than that of \sum HCHs, which ranged from 2.2 to 566.6 ng g^{-1} , with a mean value of $55.6 \pm 94.8 \text{ ng g}^{-1}$, while the HCHs ranged from 2.7 to 28.2 ng g^{-1} , with an average of $4.6 \pm 2.9 \text{ ng g}^{-1}$. The average concentrations of DDT in all soil samples followed the order of *p,p'*-Dichlorodiphenyldichloroethylene (DDE) (26.0 ng g^{-1}), *p,p'*-DDT (12.4 ng g^{-1}), *o,p'*-DDT (8.6 ng g^{-1}) and *p,p'*-Dichlorodiphenyldichloroethane (DDD) (8.6 ng g^{-1}). The average concentrations of other OCPs (including TC, CC, aldrin, dieldrin, endrin, HEPT) were all below 1.1 ng g^{-1} .

Aldrin was found in all 83 samples, with levels ranging from 0.27 to 0.81 ng g^{-1} ; dieldrin was found in 67 of the 83 samples, with levels ranging from under the limit of detection to 1.43 ng g^{-1} . As well as endrin, the concentrations of detectable endrin in 74 samples ranged from 0.33 to 20.2 ng g^{-1} . The results were generally in agreement with those of Zhang *et al.* (2012). Aldrin, dieldrin and endrin were never industrially produced or used in China (The People's Republic of China 2009). The wide distribution of these never-used OCPs indicates that they have been input to China via long-range atmospheric transport. Further studies are needed to understand the environmental behaviour and fate of the never-used OCPs in China.

Chlordane production began in the 1960s in China, and the pesticides were mainly produced by about nine manufacturers in East China over the following 40 years (Zhang *et al.* 2012). Technical chlordane has been used mostly as an agricultural pesticide; its total production and application amount was about 9000 t in China (Zhang *et al.* 2012). TC and CC were the most abundant among the chlordane isomers. TC and CC were found in all the soil samples. Levels of chlordane in samples ranged from 0.59 to 2.82 ng g^{-1} for TC and from 0.31 to 2.48 ng g^{-1} for CC. The concentrations were $<5.0 \text{ ng g}^{-1}$ for TC and CC in all of the soil samples, which were in agreement with those across China (Zhang *et al.* 2012). Even though chlordane was largely used in agricultural soils and the residues were mainly from the use of technical chlordane as a termiticide in the study area, the concentration of TC and CC

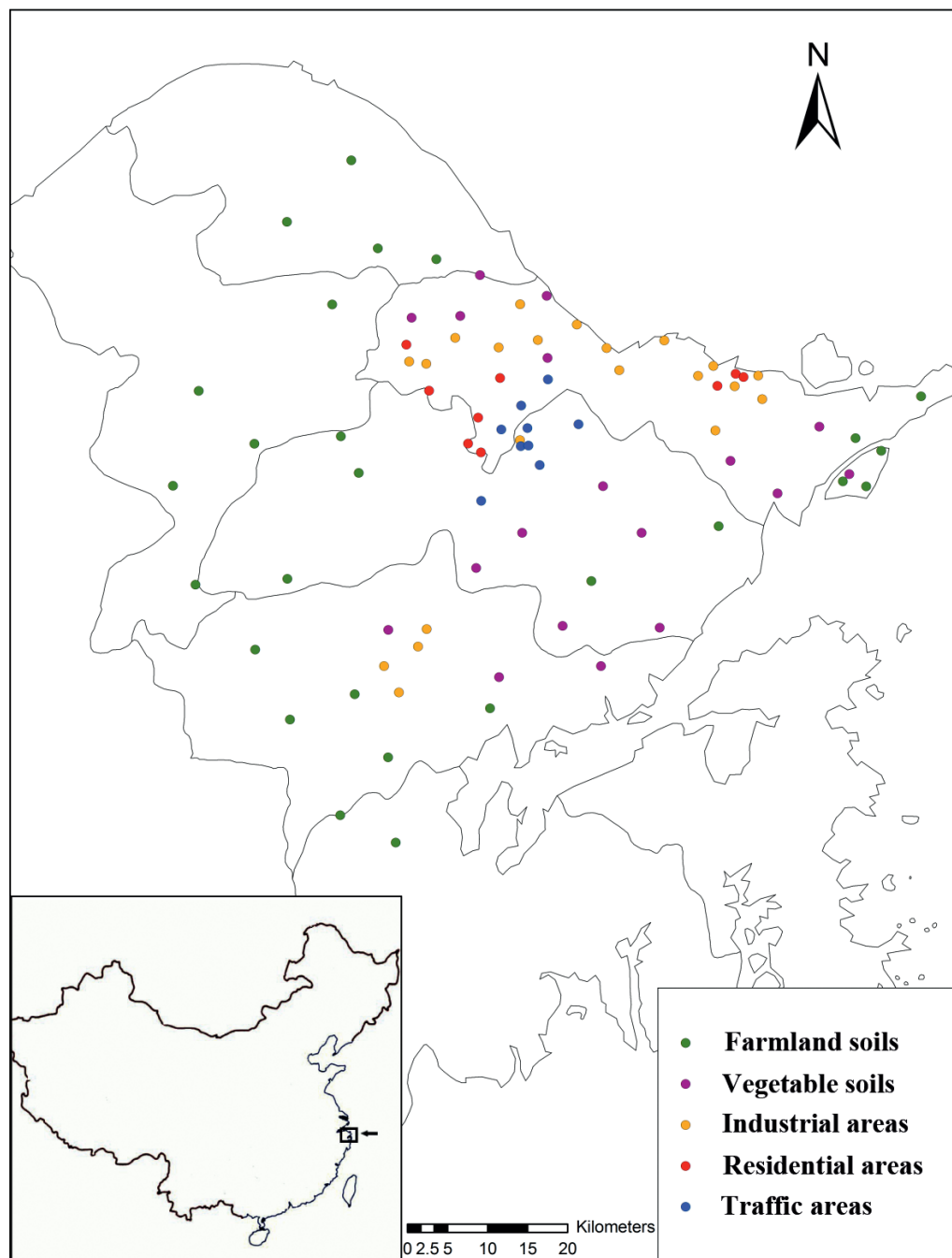


Figure 1 Map of the study area and sampling sites.

in the study area was slightly higher than in the Pearl River Delta, China ($0.24\text{--}0.89\text{ ng g}^{-1}\text{ dw}$) (Li *et al.* 2006).

Technical HEPT contains $\sim 72\%$ HEPT and $\sim 28\%$ related compounds; HEPT epoxide is a soil oxidation product of HEPT (Zhang *et al.* 2012). The mean soil concentration of HEPT in Ningbo was 0.86 ng g^{-1} , and ranged from 0.77 to 1.37 ng g^{-1} . Residues of heptachlor epoxide (HEPX) ranged from 0.20 to 2.58 ng g^{-1} , with a mean value of 0.73 ng g^{-1} .

2.2. Comparison of OCPs under five land use types

No significant differences were found in five types of soils for most of the tested OCPs such as aldrin, HEPT, HEPX, TC and CC. The data are summarised in supplementary Table 1 (available at <https://doi.org/10.1017/S1755691018000506>). As for DDTs, the mean concentrations in surface soils followed the sequence: traffic areas > residential areas > farmland soils > vegetable soils > industrial areas. The mean concen-

tration of DDTs in traffic areas was 3.5 times greater than farmland and vegetable soils, and more than ten times than that in industrial areas. The highest DDT level in traffic areas was probably due to the historical use of these areas. For HCHs, the mean concentration of HCHs in residential areas was slightly greater than under the four land uses.

2.3. Pollution assessment

In the past, HCHs and DDTs were the most popular pesticides used in many countries and have received much more attention. They can be used as representative compounds among OCPs to evaluate the pollution status of OCPs from different countries and areas. China's environmental quality standard for soils containing OCPs has only HCHs and DDTs (GB 15618–1995). According to this standard, the limits for both HCHs and DDTs in soils are 50 , 500 and 1000 ng g^{-1} , corresponding to Class I, II and III, respectively. In this study, the quality of

Table 1 The concentrations of OCPs in surface soils of Ningbo (in ng g^{-1} dw). Abbreviations: SD = standard deviation; nd = not detected.

| Compounds | Range | Mean \pm SD | Median |
|---------------|------------|-----------------|--------|
| α -HCH | 0.48–7.3 | 0.99 \pm 0.83 | 0.82 |
| β -HCH | 0.71–14.0 | 1.6 \pm 1.5 | 1.3 |
| γ -HCH | 0.45–2.6 | 0.64 \pm 0.28 | 0.57 |
| δ -HCH | 1.0–4.35 | 1.4 \pm 0.41 | 1.28 |
| Σ -HCH | 2.66–28.2 | 4.6 \pm 2.9 | 4.29 |
| p,p'-DDT | 0.95–213.6 | 12.4 \pm 26.9 | 3.7 |
| o,p'-DDT | 0.39–62.5 | 8.6 \pm 11.9 | 3.6 |
| p,p'-DDE | 0.51–321.5 | 26.0 \pm 61.0 | 9.4 |
| p,p'-DDD | 0.25–63.6 | 8.6 \pm 12.2 | 3.5 |
| Σ -DDT | 2.2–566.6 | 55.6 \pm 94.8 | 22.5 |
| TC | 0.59–2.82 | 0.72 \pm 0.25 | 0.68 |
| CC | 0.31–2.48 | 0.40 \pm 0.25 | 0.36 |
| HEPT | 0.77–1.37 | 0.86 \pm 0.95 | 0.83 |
| HEPX | 0.20–2.58 | 0.73 \pm 0.36 | 0.72 |
| Aldrin | 0.27–0.81 | 0.32 \pm 0.10 | 0.29 |
| Deldrin | nd–1.43 | 0.39 \pm 0.25 | 0.42 |
| Endrin | nd–20.2 | 1.03 \pm 2.9 | 0.47 |

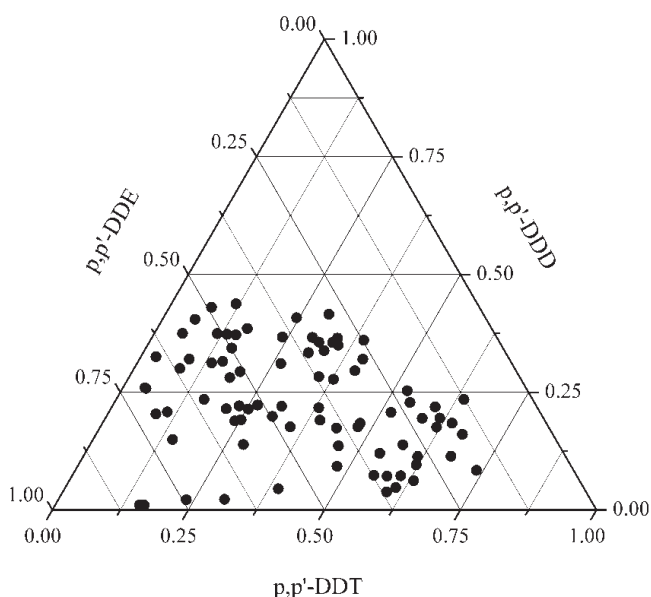


Figure 2 Ternary diagram of the relative amounts of the metabolic form of DDT in the soil samples collected from Ningbo, China.

soils was accordingly classified as no contamination with HCH and DDT concentrations $< 50 \text{ ng g}^{-1}$; low contamination with HCH and DDT concentrations between 50 and 500 ng g^{-1} ; moderate contamination with HCH and DDT concentrations between 500 and 1000 ng g^{-1} ; and high contamination with HCH and DDT concentrations $> 1000 \text{ ng g}^{-1}$. In this study, we found that HCHs in all the soil samples were $< 50 \text{ ng g}^{-1}$. Therefore, the levels of HCHs in samples from the Ningbo region could be considered as non-contaminated. In addition, it was found that the concentrations of DDTs were $< 50 \text{ ng g}^{-1}$ in 75.9% of the samples; those between 50 and 500 ng g^{-1} were in 22.9% of the samples; and only one sample had DDT levels that exceeded 500 ng g^{-1} . Therefore, the levels of DDTs in most samples from the study area of Ningbo could be considered as not contaminated. However, the DDTs in several samples were between 50 and 1000 ng , which can be regarded

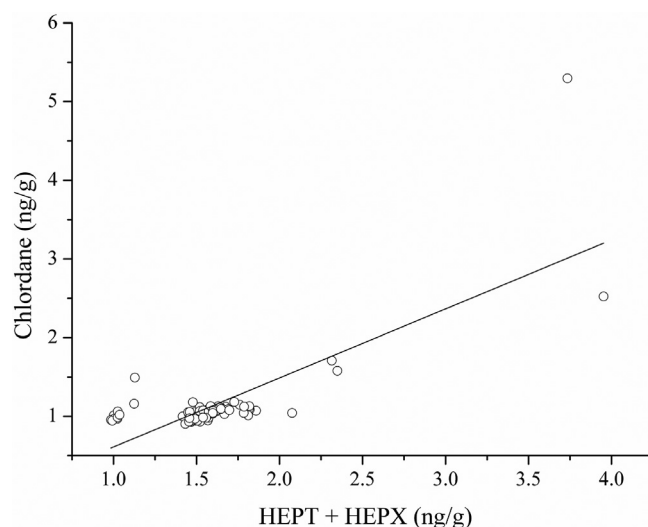


Figure 3 Linear relationship ($y = -0.27 + 0.88x$; $R = 0.57$; $P < 0.01$) between the residues of chlordane and the sum of HEPT and HEPX residues.

as having low contamination, and one sample from the traffic areas was $> 500 \text{ ng g}^{-1}$, which indicates moderate contamination.

In general, according to the Chinese Environmental Quality Standard for soils (GB 15618–1995), the HCHs levels in all the samples and DDTs levels in most of the samples could be categorised as not contaminated, and the levels of DDTs in some samples could be regarded as having low contamination; only one sample from the traffic areas could be categorised as moderately contaminated with DDTs.

2.4. Sources of OCPs

Composition differences of HCH isomers, DDT congeners and other individual compounds in the environment can indicate different contamination sources of OCPs. In general, both technical HCH mixtures and pure lindane were used. Commercial HCH products mainly include technical HCHs and lindane. The composition of typical technical HCHs produced in China contains 71% of α -HCH, 6% β -HCH, 14% γ -HCH and 9% δ -HCH, and lindane consists of 99% γ -HCH (Tao *et al.* 2008). China had been a major producer and consumer of technical HCHs since the 1950s, until its ban on production and agricultural use was enforced in 1983. Even after the ban on technical HCHs, lindane was still used in forest management between 1991 and 2000 (Zhang *et al.* 2011).

The ratio of α -HCH/ γ -HCH is between 5 and 7 in the technical HCHs and nearly zero in lindane (Zhang *et al.* 2011), so the ratio of α -HCH/ γ -HCH can be used to identify the possible HCH source. A ratio of α -HCH/ γ -HCH > 3 indicates the input of technical HCH and long-range transport, while a ratio close to (or less than) 1 is characteristic of a lindane source (Ge *et al.* 2013). Our results show that the overall ratio of α -HCH/ γ -HCH in all the soil samples was in the range of 0.97–2.78, with a mean value of 1.45, which indicates that the HCH residues at these sites were from the mixed historical use of technical HCH and lindane. The ratio of α -HCH/ β -HCH is another useful indicator to assess whether the technical HCHs were recently applied in the region or not (Tao *et al.* 2008). The ratio of α -HCH/ β -HCH of all the samples was < 4.6 (with mean values of 0.7), whereas the ratio in the technical mixture was 11.8. This is consistent with the ban on the production and application of technical HCHs in 1983 in China.

Since DDT was the dominant component in certain commercial products, such as those used in agriculture (95.2%),

Table 2 Comparison of HCHs and DDTs in surface soils from other regions in China. Abbreviation: nd = not detected.

| Area | \sum HCH | | \sum DDTs | | Reference |
|-------------|-------------|-------|-------------|-------|-----------------------------|
| | Range | Mean | Range | Mean | |
| Zhangzhou | 0.72–30.16 | 9.79 | 0.64–78.07 | 3.86 | Yang <i>et al.</i> (2012) |
| Beijing | 0.25–197.0 | 10.54 | 5.94–1039 | 162.0 | Li <i>et al.</i> (2008) |
| Tianjin | 1.3–1095 | 45.8 | | | Gong <i>et al.</i> (2004) |
| Shanghai | nd–2.4 | 1.0 | 18–142 | 67.0 | Nakata <i>et al.</i> (2005) |
| Guangzhou | 0.19–42 | 4.39 | 3.58–831 | 81.4 | Chen <i>et al.</i> (2005) |
| Hong Kong | | 6.19 | | 0.52 | Zhang <i>et al.</i> (2006) |
| Harbin | 0.136–51.8 | 7.12 | 0.072–287.2 | 5.425 | Wang <i>et al.</i> (2009) |
| Haihe Plain | 0.02–349 | 3.9 | 0.4–2350 | 63.6 | Tao <i>et al.</i> (2008) |
| Tibet | 0.2–5.4 | 1.97 | nd–2.83 | 1.22 | Fu <i>et al.</i> (2001) |
| YRD Plain | nd–99.0 | 0.02 | nd–600 | 46.0 | Hu <i>et al.</i> (2014) |
| Ningde | 0.45–151.21 | 14.51 | 0.51–241.30 | 29.61 | Qu <i>et al.</i> (2015) |

and since it can be degraded to DDE or DDD in the soil (depending on its redox conditions), the ratio of DDT/DDE or DDT/ \sum DDTs can be used as an indicator to identify recent input of technical DDT (Tao *et al.* 2008). In the soil samples collected, the average percentages of *p,p'*-DDT, *p,p'*-DDE and *p,p'*-DDD were 22.3, 46.8 and 15.5%, respectively, with *p,p'*-DDE being the dominant compound in most of the samples. Among the 83 soil samples, only eight were exceptions, with a ratio of *p,p'*-DDT/*p,p'*-DDE > 3.0. The ratio of *p,p'*-DDT/*p,p'*-DDE in this study ranged from 0.04 to 5.1, with an average of 1.1. Hence, it is clear that fresh applications of DDT in this area were very unlikely, with the exception of a few isolated sites (six samples from industrial areas, one from farmland soils and one from traffic areas suggest a fresh input of DDTs).

The ratio of *p,p'*-DDT/*o,p'*-DDT can also provide further information on whether the residues are caused by technical DDT or by technical dicofol (Qiu *et al.* 2005). Generally, *p,p'*-DDT/*o,p'*-DDT ranged from 3 to 5 in technical DDT and from 0.01 to 0.77 in technical dicofol (Qiu *et al.* 2005). The present study gives the average ratio of *p,p'*-DDT/*o,p'*-DDT as 2.7 (ranging from 0.1 to 15.3) and was much closer to that of the technical DDT than dicofol, indicating that the application of dicofol made a small contribution to the DDTs contamination in the soils in this region compared to other input sources.

The relative amounts of the different metabolites in DDT are presented in Figure 2. Greater percentages of *p,p'*-DDT, *p,p'*-DDE or *p,p'*-DDD might be attributed to a technical DDT input, aerobic transformation of DDT and anaerobic transformation, respectively (Ge *et al.* 2013). Most of the sites have values in the middle of this range, reflecting mixed modes of DDT metabolite transformations in the study area.

Technical chlordane is generally used as an insecticide, herbicide and termiticide, and is still being used in China. TC and CC were the most abundant among the chlordane isomers. In general, the ratio of CC/TC in technical chlordane mixtures is about 0.77. Since TC is easier to degrade than CC in the environment, a ratio of CC/TC > 1.0 is generally indicative of aged chlordane; in most of the sites the results showed that the ratio of CC/TC was < 1.0, which indicates that there had still been fresh inputs of chlordane.

The purified form of HEPT is a component of technical chlordane and it has not been used as an agricultural pesticide in China (Bidleman *et al.* 2002). The relationships between the concentrations of HEPT + HEPX and chlordane at each site are depicted in Figure 3. The results reveal that the total amount of HEPT and HEPX correlated with the amount of

chlordane (Fig. 3; $r = 0.58$). This significant correlation indicates that the residues of HEPT and HEPX in the study area were mainly from the local application of chlordane.

In summary, composition analyses indicated that the OCPs in soils of the Ningbo region mainly came from historical usage for agriculture. There are still some inputs such as chlordane.

2.5. A comparison with HCHs and DDTs in other regions in China

The levels of HCHs and DDTs in other regions in China, as published in the literature, were compared with those of the present study (Table 2). The contamination levels of \sum HCHs in the present study were higher than those reported from the surface soils in other study areas of China, such as the Haihe Plain in Northern China (Tao *et al.* 2008), a typical alluvial plain of the YRD region (Hu *et al.* 2014), and those from soil samples collected in pristine areas in Tibet (Fu *et al.* 2001) as well as the agricultural soils from Shanghai (Nakata *et al.* 2005). The results of this study are comparable to those soils collected from Guangzhou and Hong Kong (Chen *et al.* 2005; Zhang *et al.* 2006). In this study, the levels of HCHs were lower than the levels in the surface soil samples collected in Tianjin, Beijing, Wuxi, Zhangzhou and Ningde (Gong *et al.* 2004; Li *et al.* 2008; Dan *et al.* 2012; Qu *et al.* 2015).

The concentration levels of DDTs have also been reported. The levels detected in the present study were significantly less than those reported from urban parks in Beijing and the contaminated agricultural soil samples collected in Guangzhou, but are compatible with those collected from urban soils in Tianjing (Gong *et al.* 2004) and agricultural soils collected on the Haihe Plain (Tao *et al.* 2008). The contamination levels of DDTs in this study were much greater than those in Zhanzhou, Harbin, Shanghai, Hong Kong, Wuxi and Tibet (Fu *et al.* 2001; Nakata *et al.* 2005; Li *et al.* 2008; Xu *et al.* 2009; Dan *et al.* 2012).

3. Concluding remarks

The high detection frequencies of OCPs in soil samples from Ningbo, East China, suggest that they were widespread in surface soils of the study area, with DDTs and HCHs being the most predominant of these compounds. The OCPs that were never industrially produced or used in China (aldrin and dieldrin) were also detected in all the soil samples, which indicates that they have been continuously applied in China via long-range atmospheric transport, but further study is

needed. A comparison of the observed concentrations of OCP residues from the different sampling places in China indicates comparatively smaller amounts of OCP residues in this area than in other cities. It was also found that concentrations of HCHs in all of the soil samples and DDTs in most of the soil samples were less than the Grade I (50 ng g^{-1}) permitted in the 'Environmental Quality Standard for Soils in China' (GB 15619-1995). According to the ratio between DDT and its metabolites, the residues of OCPs in most samples were derived from historical application. The HCH residues in this area were mainly from the mixed historical use of technical HCH and lindane. The results provide a foundation for the future monitoring of POPs and similar chemicals.

4. Acknowledgements

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5. Supplementary material

Supplementary material is available online at <https://doi.org/10.1017/S1755691018000506>.

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