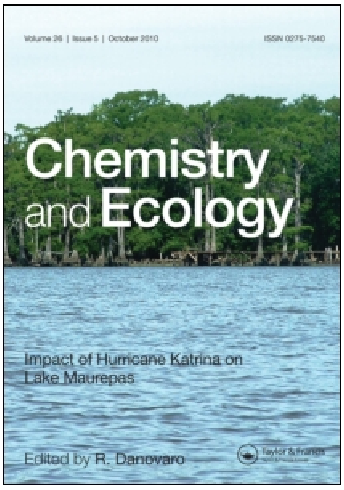

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Organochlorine pesticides (HCHs and DDTs) in soils along the north coastal areas of the Bohai Sea, China

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A systematic survey of organochlorine pesticides (OCPs) including hexachlorocyclohexane isomers (α-HCH, β-HCH, γ-HCH, δ-HCH and ΣHCH) and dichlorodiphenyltrichloroethane metabolites (p,p'-DDT, p,p'-DDE, o,p'-DDT, p,p'-DDD and $\sum$DDT) in soils along the north coastal areas of the Bohai Sea, China, has been lacking. In this study, 31 representative surface soil samples were collected along the north coastal and riverine areas of the Bohai Sea to characterise the potential for adverse effects of $\sum$HCH, $\sum$DDT and their individual isomers and transformation products. Concentrations of ΣHCH and ΣDDT in soils ranged from less than the limit of detection (<LOD) to 3.0×10^1 ng · g^{-1} dw (mean: 3.5 ng · g^{-1} dw) and <LOD to 2.6×10^2 ng · g^{-1} dw (mean: 1.7×10^1 ng · g^{-1} dw), respectively. Compared with studies of OCPs in soils from other locations, concentrations of HCHs and DDTs observed in this study were moderate. Concentrations of OCPs observed in soils were generally less than proposed reference values. HCH residues were a mixture of historical technical HCH and current lindane sources. The pattern of DDTs was consistent with historical releases of technical DDTs. Selected soil physicochemical properties did not explain the sorption and/or partitioning of HCHs or DDTs.

Keywords: soil contamination; organochlorine pesticides; relative proportions; risk assessment; sources

1. Introduction

Organochlorine pesticides (OCPs) such as hexachlorocyclohexane isomers (α-HCH, β-HCH, γ-HCH, δ-HCH, and the sum of these isomers, $\sum$HCH) and dichlorodiphenyltrichloroethane metabolites (p,p'-DDT, p,p'-DDE, o,p'-DDT, p,p'-DDD, and the sum of these transformation products, $\sum$DDT) have been extensively used worldwide in agriculture, forestry and public health applications [1,2]. Some of these compounds are resistant to transformation and/or degradation by biological processes, photolysis and hydrolysis, and can be bioaccumulated [3]. From 1950

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to 1983, an estimated total of >4 million tons of ΣHCH and 0.46 million tons of ΣDDT were produced in China, with most of the production applied in agriculture [4]. Although production of technical HCH and DDT has been officially banned in China since 1983, they have still been detected in some soils in China [5–8]. Soil can play an important role in the global fate and distribution of pollutants because it has been identified as a sink from which they can be released into water or air [9]. OCPs in soils can enter aquatic environments through effluents, atmospheric deposition and run-off [10]. Their occurrence in soils at relatively great concentrations is thought to be one of the major sources of OCPs to aquatic environments [11], which can cause adverse effects in wildlife or the people who eat it.

The Bohai Sea, together with nearby coastal and riverine areas, is a major commercial artery and region of significant urbanisation and industrialisation [12]. Its proximity to Beijing, the capital of China, makes it one of the busiest seaways in the world. Benefiting from rich resources and convenient transportation, the economic contribution of the Bohai Sea amounts to one tenth of the gross national product of China, such that economists call the zone around the Bohai Sea ‘the golden necklace’ of Northern China. Several rivers, including the Yellow River, Liaohe River, Haihe River, Luanhe River and Dalinhe River, discharge into the Bohai Sea. The drainage areas of these five rivers are important to both agricultural and chemical production [13]. Rapid social and economic development has caused to the area to become increasingly urbanised, but also has led to the area being contaminated with persistent organic pollutants (POPs) [14]. Because the Bohai Sea is semi-enclosed, water exchange with the Pacific Ocean is relatively slow. Thus, pollutants can easily be accumulated, and the Bohai Sea is considered to be one of the most polluted marine environments in China [15].

Studies of contaminants in seawater, sediment and marine organisms have been conducted in the coastal areas of the Bohai Sea [16], but few studies have reported concentrations of HCHs and DDTs in soils along the north coastal and riverine regions of the Bohai Sea. Therefore, baseline information on concentrations of OCPs in soils was needed to better understand the current status and trends in their concentrations. The results presented here were collected as part of a large systematic investigation of concentrations and environmental risks of HCHs and DDTs in soils from the north coastal and riverine areas of the Bohai Sea. Because most previous studies have been conducted in coastal areas, and considered primarily inland sources of OCPs, this study of rivers and upper regions of the Bohai Sea provides novel and current information. The primary objectives of this study were to: (1) determine concentrations of HCHs and DDTs in soils from north coastal areas of the Bohai Sea; (2) elucidate the relative proportions and possible sources of HCHs and DDTs in the soils; and (3) evaluate the potential environmental risk to the local marine ecosystem and human health posed by the target residues, and inform management strategies.

2. Materials and methods

2.1. *Study area and sample collection*

This study focused on the north coastal and riverine areas of the Bohai Sea, China, including the following eight regions (metropolitan areas): Tangshan (TS), Qinhuangdao (QH), Huludao (HL), Jinzhou (JZ), Panjin (PJ), Yinkou (YK), Dalian (DL) and Dandong (DD) (Figure 1). The Bohai Sea is a semi-enclosed 7.8×10^4 km^2 bay of the north-west Pacific Ocean. In May 2008, 31 soil samples were collected along the north coastal and riverine areas of the Bohai Sea, China. The sampling design was developed to focus on possible loadings and sources from inland areas. Thus, in each region, soils were collected near the upstream, middle and downstream, and coastal areas of the Bohai Sea. The drainage areas of these rivers include both agricultural use and the production of chemicals.

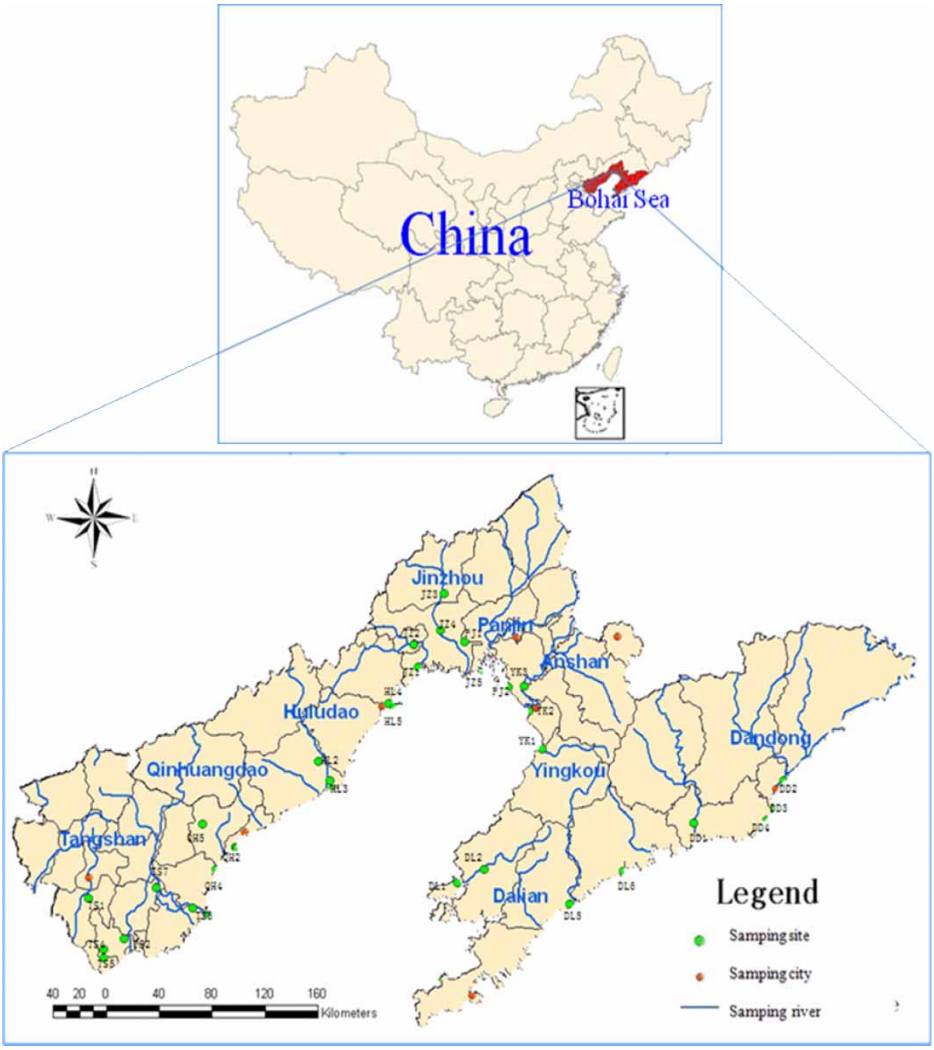

Figure 1. Study area and sampling stations along the north coastal and riverine areas of the Bohai Sea, China.

The locations of the sampling sites are shown in Figure 1. Throughout the survey, a global positioning system (GPS) was used to locate and record the sampling sites. Each sample was made up of a mixture of five subsamples collected from five spots of an area of $\sim 100 \times 100\,\mathrm{m}^2$ (at the four corners and the centre). All soil subsamples were collected at a depth of 0–20 cm using a stainless steel shovel. Grass, twigs and other sundries were removed from the surface of each sampling location before the sample was collected. Wet soil samples ($\sim$1 kg) were transported to the laboratory in polyethylene zip-lock bags, lyophilised, sieved through a 2 mm mesh and stored at 4 °C in pre-cleaned glass jars until analysis.

2.2. *Physicochemical properties of soils*

Total organic carbon (TOC) was analysed using a Universal CHNOS Elemental Analyser (Elementar Vario EL III, Germany). Soil pH was measured using a pH meter and 10 g of air-dried soil suspended in 25 mL deionised water.

2.3. *Extraction, clean-up and quantification*

OCPs were extracted from soil with n-hexane and dichloromethane (J.T. Baker, HPLC grade). Solid-phase extraction (6 mL; Supelco, Bellefonte, PA, USA) cartridges containing 1 g of Florisil were washed with 10 mL of n-hexane before use. Each soil sample (5.0 g dw) was mixed with 1 g anhydrous sodium sulphate. One millilitre of 2,4,5,6-tetrachloro-m-xylene (Supelco, Bellefonte, PA, USA) at a concentration of $0.08\,\mu g \cdot mL^{-1}$ was added as a surrogate standard. The mixture was extracted twice with 35 mL hexane:dichloromethane (1:1, v/v) by ultrasonication for 60 min and then centrifuged. Extracts were concentrated to $\sim$2 mL by rotary evaporation, and further purified with solid-phase extraction cartridges loaded with 1 g silica gel and 1 g anhydrous sodium sulphate. Anhydrous sodium sulphate (analytical grade) and silica gel (60 mesh, for column chromatography) were activated at 180 °C for 24 h before use. Analytes were eluted with 20 mL hexane:dichloromethane (7:3, v/v). The eluent was concentrated by N_2 to a final volume of 1 mL for gas chromatography (GC) analysis. Reagent blanks were also analysed concurrently with the experimental samples. Separate subsamples of several grams of each soil were dried for 8 h at 80 °C to determine moisture content.

An Agilent 6890 gas chromatograph (GC) equipped a ^{63}Ni electron capture detector (μECD) (Agilent Technologies, Wilmington, DE, USA) was used for the identification and quantification of HCH isomers and DDT metabolites. The column used was an HP-5 silica capillary column with $30\,m \times 0.32\,mm$ i.d $\times\ 0.25\,\mu m$ film thickness. The GC-μECD was operated in split-less injection mode with ultrapure nitrogen as the carrier and make-up gas. The injector and detector temperatures were 220 and 300 °C, respectively. The oven temperature was initially set at 100 °C with a 2 min hold, ramped at $10\,°C \cdot min^{-1}$ to 160 °C with a 2 min hold, $4\,°C \cdot min^{-1}$ to 230 °C with a 5 min hold, $10\,°C \cdot min^{-1}$ to 270 °C with a 2 min hold. The four HCH isomers (α, β, γ, δ) and four DDT homologues (p,p'-DDE, p,p'-DDD, o,p'-DDT, and p,p'-DDT) were identified by retention time matched to standards and were quantified based on peak area. Concentrations of $\sum$HCH and $\sum$DDT were calculated by summing the four HCH isomers and four DDT metabolites, respectively, and were reported on a dry soil basis.

For quality assurance and control, procedural blanks and matrices spiked with standard solution were analysed. None of the target compounds was detected in the procedural blanks. All solvents used were distilled in glass (PR grade) and checked for interferences or contamination prior to use. Extraction efficiency, as indicated by recovery of the surrogate standards (2,4,5,6-tetrachloro-m-xylene), was $75 \pm 10\%$. LOD were described as three times thre signal-to-noise ratio (S/N). The detection limit ranged from $5.0 \times 10^{-2}\,ng \cdot g^{-1}$ dw to $2.0 \times 10^{-1}\,ng \cdot g^{-1}$ dw. A standard solution containing α-, β-, γ-, δ-HCH isomers, p,p'-DDE, p,p'-DDD, o,p'-DDT and p,p'-DDT at $1\,mg \cdot g^{-1}$ of each compound was obtained from the National Research Center for Certified Reference Materials (CRM) of China. The recovery of CRM spiked to soil ranged from 73.3 to 96.2%. GC analysis was repeated twice for each replicate sample and the relative standard deviation (RSD) of replicate analyses was $<10\%$.

2.4. *Data analysis and multivariate statistical approach*

Concentrations of HCHs and DDTs in soils were described by minimum, maximum, difference between mean, median, standard deviation and coefficient of variation (CV). The software used for mapping the sampling sites was ArcGis (ESRI, USA). Relationships between concentrations of individual HCHs and DDTs and soil properties were investigated using principal component analysis (PCA) and Spearman's correlation analysis for the non-normal distributed data using SPSS 11.5 for Windows. PCA is a multivariate statistical tool used to reduce a set of original variables and to extract a small number of latent factors for analysing relationships among the observed variables [17]. PCA can also be used to identify potential or even likely sources of contaminants and their degradation behaviour [17,18]. Samples were grouped by use of varimax rotation and the retention of principal components having eigenvalues >1. Variables with similar characteristics were grouped into factors. The first and second principal component factors often explained $>70\%$ of the variance. Thus, the first two factors were used to determine possible sources.

3. Results

3.1. *Concentrations of HCHs and DDTs and soil properties*

Concentrations of HCHs and DDTs in soils and selected physicochemical properties of the soils are presented (Table 1). The mean concentrations of $\sum$HCH and $\sum$DDT were 3.5 and $1.7 \times 10^1\,\mathrm{ng \cdot g^{-1}}$ dw, ranging from <LOD to $3.0 \times 10^1\,\mathrm{ng \cdot g^{-1}}$ dw and <LOD to $2.6 \times 10^2\,\mathrm{ng \cdot g^{-1}}$ dw, respectively. Not all HCH or DDT compounds or isomers were detected in all samples. In general, the concentrations of $\sum$HCH were less than those of $\sum$DDT. This was consistent with previous measurements of concentrations of $\sum$HCH and $\sum$DDT in soils in the vicinity of Guanting Reservoir, China [19], soils of the Haihe Plain, China [4] and agricultural soils of central Germany [18]. HCH is more water soluble and has a greater vapour pressure, biodegradability, lesser K_{ow} and K_{d} values, relative to DDT [20], which may account for the relatively small amount of HCH residues in soils.

The large CV values ($>100\%$) of HCH and DDT concentrations indicated spatial heterogeneity of HCHs and DDTs in the study area. This type of patchy distribution is often observed for concentrations of organic residues in soils and leads to non-normal frequency distributions. In fact, a log-normal or positively skewed distribution is common and for this distribution the mean often equals the standard deviation, which results in a CV value of 100%. Mean values of soil tital organic carbon (TOC) and pH were approximate to their median values, with low CV values ($<80\%$), which indicated that soil TOC and pH had normal frequency distribution.

3.2. *Relative proportions of HCH and DDT components*

3.2.1. *Isomers of HCH*

HCHs are among the most widely used and most readily detected OCPs in environmental samples. HCH has been used either as a technical mixture (a formulation dominated by the α-isomer) or as lindane (a formulation containing almost 100% of the γ-isomer) [21]. Technical HCH is a mixture of isomers with different chemical and physical properties. The typical technical HCH generally contains 55–80% α-HCH, 5–14% β-HCH, 8–15% γ-HCH and 2–16% δ-HCH, respectively [22]. Among the HCH isomers, α-HCH is more likely to partition into the air and transport for long distances, whereas β-HCH is more resistant to hydrolysis and environmental degradation and is the dominant isomer in soils, animal tissue and fluids [23].

In this study, with the exception of station PJ1, the α-, β-, γ- and δ-HCH isomers were quantifiable in all samples. However, the patterns of relative compositions differed among regions (Figure 2). The differences in concentrations among locations depended on the properties of HCHs and soil properties, including TOC, pH, texture, and microbiota, that define the rates of dissipation. Neither $\sum$HCH nor any of its isomers were detected in soil from Panjin (station PJ1). The soil was from a paddy, which indicated that HCH residues may have been carried away by run-off or degraded subsequent to being prohibited from use in agriculture in 1983. When detectable, individual α, β, γ, and δ isomers of HCH were found to contribute ~2, 86, 7 and 5% to $\sum$HCH, respectively. These relative proportions of isomers were different from the originally

Table 1. Concentrations ($ng \cdot g^{-1}$ dw) of HCHs and DDTs in soils from north coastal and riverine areas of the Bohai Sea, soil properties and soil guideline values for corresponding compounds suggested by China, the Netherlands and the USA are also given.

								Reference values		
OCPs	Mean	SD	Min.	Max.	Median	CV (%)	Detection rate (%)	China	Neth	USA
α-HCH	7.0×10^{-2}	1.7×10^{-1}	<LOD	8.8×10^{-1}	0.0	242	3.9×10^{1}		3.0	2.5
β-HCH	3.0	5.4	<LOD	2.9×10^{1}	1.3	178	9.7×10^{1}		9.0	1.0
γ-HCH	2.4×10^{-1}	5.4×10^{-1}	<LOD	2.1	0.0	225	7.1×10^{1}		5.0×10^{-2}	5.0×10^{-2}
δ-HCH	1.9×10^{-1}	2.7×10^{-1}	<LOD	1.2	0.0	142	5.5×10^{1}			
$\sum$HCH	3.5	5.6	<LOD	3.0×10^{1}	1.9	157	9.7×10^{1}	5.0×10^{1}	1.0×10^{1}	
p,p'-DDE	7.8	3.2×10^{1}	<LOD	1.8×10^{2}	6.3×10^{1}	405	7.7×10^{1}			
p,p'-DDD	6.0×10^{-1}	1.2	<LOD	5.7	0.0	206	6.1×10^{1}			
o,p'-DDT	1.4	3.9	<LOD	1.7×10^{1}	0.0	269	6.8×10^{1}			
p,p'-DDT	6.6	1.2×10^{1}	<LOD	6.8×10^{1}	2.0	187	8.7×10^{1}			
$\sum$DDT	1.7×10^{1}	4.7×10^{1}	<LOD	2.6×10^{2}	5.5	286	9.0×10^{1}	5.0×10^{1}	1.0×10^{1}	2.5
Ratio1	1.5×10^{-1}	4.1×10^{-1}	0.0	1.9	0.0					
Ratio2	5.2×10^{-1}	6.5×10^{-1}	0.0	2.7	3.6×10^{-1}					
TOC	8.6×10^{-1}	6.1×10^{-1}	1.9×10^{-1}	3.1	7.3×10^{-1}	7.1×10^{1}				
pH	6.8	9.6×10^{-1}	5.0	8.3	6.9	1.4×10^{1}				

Notes: Mean, SD, minimum, maximum and median were calculated assuming less than limits of detection $<\text{LOD} = 0$ for statistical purposes. $\sum\text{HCH} = \alpha\text{-HCH} + \beta\text{-HCH} + \gamma\text{-HCH} + \delta\text{-HCH}$, $\sum\text{DDT} = p,p'\text{-DDE} + p,p'\text{-DDD} + o,p'\text{-DDT} + p,p'\text{-DDT}$. $\text{Ratio1} = \alpha\text{-HCH}/\gamma\text{-HCH}$; $\text{Ratio2} = (p,p'\text{-DDE} + p,p'\text{-DDD})/p,p'\text{-DDT}$; CV (%), coefficient of variation; China, *Chinese Environmental Quality Standard for Soils* (GB15618-1995); Neth, *Netherlands Target Values for Soil Remediation* (2000); USA, *Recommended Ecological Screening Values for Soil* (2001).

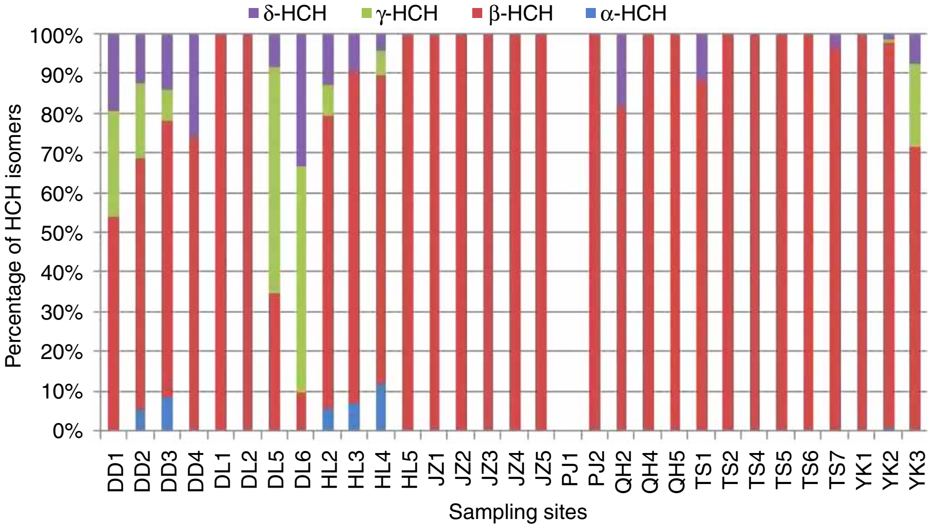

Figure 2. Relative proportions of HCH components in soils from different sampling sites.

manufactured technical mixture. β-HCH has the lowest water solubility and vapour pressure, and is the most stable and relatively resistant to microbial degradation [22]. It is these properties of β-HCH relative to other HCHs that most likely accounted for the greater proportion of β-HCH in the samples. In addition, after a long period of weathering, α- and γ-HCHs could be transformed into β-HCH [20]. The relatively high proportion of β-HCH (86% of $\sum$HCH) observed indicated that the HCH was mainly due to historical use as a pesticide [24], because HCH has not been used in this region recently [25]. The mean ratio of α-HCH/γ-HCH was 1.5×10^{-1}, with a maximum of 1.9 (Table 1), and the ratios were 0 in 84% of the samples, much less than the α/γ-HCH ratio found in the technical HCH mixture. This indicates the possible input of lindane in the past few years or that more α-HCH likely evaporates into atmosphere from soil in a timely manner [26].

3.2.2. *Metabolites of DDT*

Because the use of DDT in China was banned in the early 1980s, the presence of DDT in the soils was predominantly from historical usage. DDT can be transformed in the environment to DDE by both abiotic and biological processes [27]. Ratios of various degradation products may, therefore, reflect some of the effects of the environmental conditions on the degradation process [27]. DDT can be biodegraded to DDE under aerobic conditions and to DDD under anaerobic conditions; the relative concentration of the parent DDT compound and its metabolites, DDD and DDE, can therefore be used as indicative indices for assessing the possible pollution sources [28].

Relative proportions of constituents of the DDT complex observed in this study varied significantly among locations (Figure 3). The order of contribution of individual constituents to $\sum$DDT was: p,p'-DDT $>$ p,p'-DDE $>$ o,p'-DDT $>$ p,p'-DDD, with p,p'-DDT accounting for the greatest proportion of $\sum$DDT (65%). Because concentrations of DDT in the environment decrease over time and the major metabolites are expected to be DDE and DDD, the ratio of (p,p'-DDE + p,p'-DDD)/p,p'-DDT can be used as an indicator of resident time of p,p'-DDT in the environment [29]. A ratio > 1 is normally expected for aged mixtures in the environment and values < 1 indicate relatively recent inputs of DDT [30]. The mean ratio of (p,p'-DDE + p,p'-DDD)/p,p'-DDT

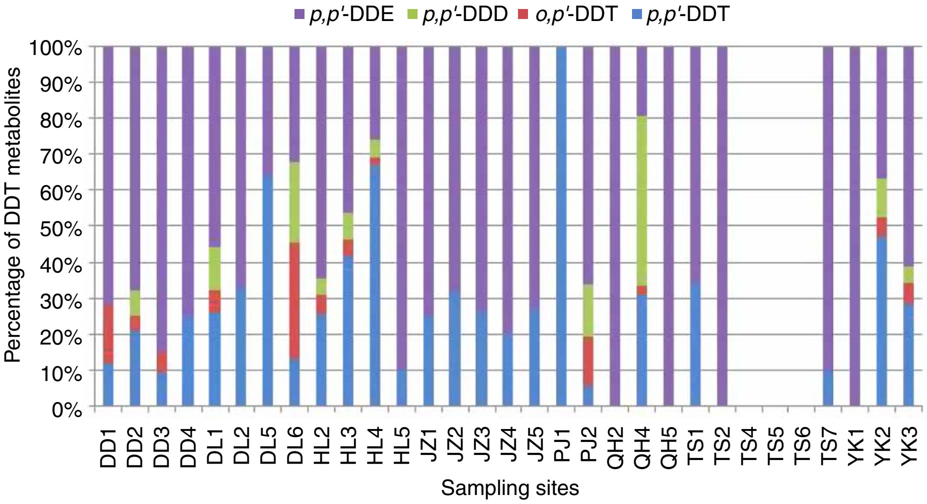

Figure 3. Relative proportions of DDT components in soils from different sampling sites.

was 5.2×10^{-1}, with a maximum of 2.7 (Table 1), and 81% of the whole samples showed ratios of <1. The relatively high proportion of p,p'-DDT found in soils from the areas around the Bohai Sea indicates slow degradation of historical DDT or a recent input of DDT at some sampling sites [27]. In particular, the greatest proportion of p,p'-DDT was found in soils collected from Qinghuangdao (stations QH2 and QH5), Tangshan (station TS2) and Yingkou (station YK1). This could be explained by the fact that after cessation of the use of DDT in agriculture, input from weathered agricultural soils and atmospheric deposition may have served as continuing sources of DDT residue [31,32]. The relatively greater proportion of DDE (26% of $\sum$DDT) found in the soils from these areas was related to the longer persistence of DDE than of DDD or the parent DDT. In addition, during the dry season, because of the aerobic soil conditions, active oxidation was expected to have caused transformation of p,p'-DDT to p,p'-DDE [24]. The reference soil samples (stations TS4, TS5 and TS6) located in Tangshan contained non-detectable concentrations of DDT transformation products. The land uses of the three sampling sites were all fallow land, and the lower concentrations of DDTs in these soils reflected the absence of local pollution sources near the sampling sites and were consistent with the lesser use of DDTs in these stations.

4. Discussion

4.1. *Comparison to other studies and risk assessment*

To understand the contamination status of HCHs and DDTs in soils along the north coastal and riverine areas of the Bohai Sea, average $\sum$HCH and $\sum$DDT concentrations in surface soils from different locations were compared. Mean concentrations of $\sum$HCH in the study area were greater than concentrations reported for soils from the Tibet plateau, China (range: 1.8×10^{-1} to $5.4\,\text{ng} \cdot \text{g}^{-1}$ dw, mean: $2.0\,\text{ng} \cdot \text{g}^{-1}$ dw) [33], the Pearl River Delta, China (range: 5.0×10^{-2} to $2.4 \times 10^{1}\,\text{ng} \cdot \text{g}^{-1}$ dw, median: $2.9\,\text{ng} \cdot \text{g}^{-1}$ dw) [5], mountain Andean Lakes (range: 1.0×10^{-3} to $4.0 \times 10^{-1}\,\text{ng} \cdot \text{g}^{-1}$ dw) [34] and James Ross Island (range: 5.0×10^{-1} to $1.3\,\text{ng} \cdot \text{g}^{-1}$ dw) [35], but were less than those of central Germany (range: 4.6 to $1.2 \times 10^{1}\,\text{ng} \cdot \text{g}^{-1}$ dw, mean: $7.5\,\text{ng} \cdot \text{g}^{-1}$ dw) [19] and Romania (range: 2.8 to $9.0 \times 10^{1}\,\text{ng} \cdot \text{g}^{-1}$ dw, mean: $2.6 \times 10^{1}\,\text{ng} \cdot \text{g}^{-1}$ dw) [36].

Concentrations of $\sum$DDT in the study area were greater than those in soils from the Taihu Lake region, China (range: 3.0×10^{-1} to $5.3\,\mathrm{ng \cdot g^{-1}}$ dw) [6], the mountain Andean lakes (range: 1.0×10^{-1} to $1.1\,\mathrm{ng \cdot g^{-1}}$ dw) [34] and James Ross Island (range: 5.1×10^{-1} to $3.7\,\mathrm{ng \cdot g^{-1}}$ dw) [35], but were significantly less than those of the Pearl River Delta, China (range: 5.2×10^{-1} to $4.1 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw, median: $2.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw) [5], central Germany (range: 2.4×10^{1} to $1.7 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw, mean: $7.2 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw) [19] and Romania (range: 9.0 to $1.9 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw, mean: $6.3 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw) [36]. In general, concentrations of $\sum$HCH and $\sum$DDT in soils along the coastal areas of the Bohai Sea were less than those from most other areas.

It is difficult to accurately confirm the contamination and assess the potential for effects of HCHs and DDTs in soils. Despite studies having been conducted, there are no consensus soil quality guidelines available for HCHs and DDTs. To assess risk, concentrations of HCHs and DDTs in soils were compared with the soil quality reference values recommended in China [37], the Netherlands [38] and the USA [39] (Table 1). According to the Guideline of Chinese Environmental Quality Standard for soils, the maximum permitted concentration of both $\sum$HCH and $\sum$DDT in the soil should be less than the second grade ($5.0 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw) in order to ensure the safety of agricultural products and prevent food contamination by $\sum$HCH and $\sum$DDT, which can be detrimental to human health. The first grade ($5.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw) was set by the Guideline of Chinese Environmental Quality Standard in terms of protecting regional ecology and maintaining the natural environmental background. Concentrations of $\sum$HCH in all samples were less than the first grade of Chinese guidelines, but in two locations, TS7 and YK2, were 1.4×10^{1} and $2.9 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw, respectively, which are higher than Dutch target standards ($1.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw), although corresponding concentrations were less than its intervention values ($2.0 \times 10^{3}\,\mathrm{ng \cdot g^{-1}}$ dw) [38]. Mean concentrations of α-HCH were less than the suggested soil guidelines of the Netherlands and the USA [39]. As for γ-HCH, the mean concentration was greater than the suggested limit of the Dutch and US target values, while for β-HCH, the mean concentration was also greater than the suggested limit of the US target values, but less than the suggested limit of the Netherlands. Critical concentrations of α-HCH, β-HCH and γ-HCH in soils have been suggested [40]. These values, which represent a hazard concentration for 50% of the soil invertebrate community, have been suggested to be $1.0 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw, $4.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw and $1.0 \times 10^{4}\,\mathrm{ng \cdot g^{-1}}$ dw, respectively, for these three isomers of HCH. Concentrations of HCHs in soil around our study area were unlikely to have adverse effects on soil invertebrate communities. Concentrations of $\sum$DDT in 30 samples were less than the first grade of China guideline ($5.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw), whereas $\sum$DDT in one sample (station HL4) was between the first and the second grade ($2.6 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw). Compared to foreign guidelines, there were 17 soil samples that exceeded the US target values ($2.5\,\mathrm{ng \cdot g^{-1}}$ dw) and 11 soil samples that exceeded the Dutch target values ($1.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw), but none of the concentrations of $\sum$DDT exceeded intervention values ($4.0 \times 10^{3}\,\mathrm{ng \cdot g^{-1}}$ dw) [38]. The critical concentration of DDT in soils was estimated by use of a simplified food web model with three trophic levels [41]. Maximum permissible concentrations of DDT in soils were $1.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw to protect plants and invertebrates, $1.1 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw to protect small birds and mammals, and $1.9 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw to protect birds and animals that eat birds, respectively. The concentration of $\sum$DDT in only one soil from HL4 exceeded $1.9 \times 10^{2}\,\mathrm{ng \cdot g^{-1}}$ dw. Concentrations of $\sum$DDT in 10 soils exceeded $1.0 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw, and concentrations in 9 soils exceeded $1.1 \times 10^{1}\,\mathrm{ng \cdot g^{-1}}$ dw. Therefore, concentrations of $\sum$DDT in soils around our study area may have the potential to cause ecological risks to birds and soil organisms. However, because the screening levels of soil quality criteria are meant to be conservative and thus protect wildlife, exceeding these values does not mean that effects will be observed, rather that it cannot be concluded that no effects would be expected. This result suggests that more extensive, site-specific evaluations should be conducted.

4.2. *Source identification*

4.2.1. *PCA*

When PCA was performed for HCH, 75.70% of total variance was explained by the first two factors. The first principal component (PC1), accounting for 47.05% of total variance, had large positive coefficients for α-HCH (0.515), β-HCH (0.343), γ-HCH (0.914) and δ-HCH (0.814). Because the γ-HCH content of lindane is ∼99.9%, PC1 explained primarily the sources of lindane. The second principal component (PC2), which explained 28.65% of total variance, was characterised by absolute loadings of α-HCH (0.556) and β-HCH (0.762), which were statistically significant, whereas loadings of γ-HCH (−0.472) and δ-HCH (−0.176) were relatively small. The major loading of β-HCH and α-HCH indicated that PC2 was mostly related to the degradation of 'old' technical HCH.

For DDT, the first two factors accounted for 93.75% of the total variance. Four DDT metabolites were all included in the first factor (PC1), which explained 81.61% of the total variance. Most of p,p'-DDE (0.938), p,p'-DDD (0.923), p,p'-DDT (0.961) and more than half of o,p'-DDT (0.780) were involved. According to the composition and degradation rules of technical DDT, PC1 was mostly related to the degradation of technical DDT. The second factor (PC2), which accounted for 12.14%, was dominated by o,p'-DDT (0.625) and little negatively related to p,p'-DDE (−0.196), p,p'-DDD (−0.105) and p,p'-DDT (−0.215). For technical DDT, o,p'-DDT was a second major transformation product compared with other products. Thus, this factor can be interpreted as representing transformation of 'old' technical DDT.

4.2.2. *Correlation analysis*

Multiple factors can affect concentrations of HCHs and DDTs in soils. Previously, soil physicochemical properties such as soil TOC and pH have been found to be key factors in determining the concentrations of pollutants in many studies [26,42,43]. These parameters affect both the current concentration in the soil system and possible adsorption onto various soil components [44]. To better explore the sources and fates of HCHs and DDTs in the study area, and to examine whether TOC and pH were major factors controlling the residue of HCHs and DDTs, nonparametric correlation analysis was performed. According to the values of Spearman's correlation coefficients (R), typical components were selected to reflect the overall HCH and DDT contamination level, and check whether there was good correlation between individual compounds and soil property variables. The results of correlation analysis between OCPs (HCHs and DDTs and all isomers) and soil properties of TOC and pH are listed in Table 2. Although the distribution of concentrations of HCHs and DDTs was largely different in the study area, there existed positive correlations between HCHs and DDTs. The positive correlations are consistent with HCHs and DDTs having similar and/or common sources, as well as comparable environmental behaviour among regions. Earlier studies of HCHs and DDTs in Beijing urban soils [45], Tianjing chemical industrial park soils [8] and remote Austrian forest soils [46] showed similar results. Significant negative correlations among α-HCH ($p < 0.05$), γ-HCH ($p < 0.05$), δ-HCH ($p < 0.01$) and p,p'-DDE ($p < 0.01$) with pH were observed. pH can affect the concentrations of HCHs and DDTs in soil by influencing the microbiological activity in the soil [47]. A significant positive correlation between p,p'-DDE and TOC was found ($p < 0.05$), which suggests partitioning of p,p'-DDE to soil organic matter [25]. Greater organic carbon content in soil can also allow a larger microbial biomass and thus induce the degradation or absorption of DDTs [48]. As a result, TOC could affect concentrations of p,p'-DDE in soils [42]. Such relationships were not observed for HCH isomers and other DDT metabolites, indicating that soil TOC was not likely to be a significant factor affecting the accumulation of HCHs and other DDT metabolites in soils from

Table 2. Relationship between OCPs (individual HCHs and DDTs) and selected soil properties.

	α-HCH	β-HCH	γ-HCH	δ-HCH	∑HCH	p,p'-DDE	p,p'-DDD	o,p'-DDT	p,p'-DDT	∑DDT	TOC	pH
α-HCH	1											
β-HCH	0.550^{**}	1										
γ-HCH	0.492^{**}	0.191	1									
δ-HCH	0.606^{**}	0.306	0.743^{**}	1								
∑HCH	0.542^{**}	0.829^{**}	0.573^{**}	0.663^{**}	1							
p,p'-DDE	0.544^{**}	0.714^{**}	0.477^{**}	0.465^{**}	0.776^{**}	1						
p,p'-DDD	0.599^{**}	0.557^{**}	0.577^{**}	0.492^{**}	0.723^{**}	0.708^{**}	1					
o,p'-DDT	0.521^{**}	0.591^{**}	0.395^{*}	0.329	0.684^{**}	0.745^{**}	0.930^{**}	1				
p,p'-DDT	0.574^{**}	0.860^{**}	0.385^{*}	0.452^{*}	0.851^{**}	0.720^{**}	0.694^{**}	0.682^{**}	1			
∑DDT	0.564^{**}	0.828^{**}	0.418^{*}	0.464^{**}	0.860^{**}	0.809^{**}	0.773^{**}	0.769^{**}	0.970^{**}	1		
TOC	0.302	0.264	0.171	0.338	0.322	0.369^{*}	0.184	0.175	0.334	0.348	1	
pH	-0.462^{*}	-0.169	-0.422^{*}	-0.504^{**}	-0.28	-0.490^{**}	-0.342	-0.267	-0.237	-0.301	-0.441^{*}	1

Note: **Correlation is significant at the 0.01 level (two-tailed); *correlation is significant at the 0.05 level (two-tailed).

the study area. Overall, the above results suggest that the use of limited soil geochemical factors would be insufficient to assess the ability of soil to sorb HCHs and DDTs in the study area [17], because degradation of HCHs and DDTs is complex and thus could be affected by many factors that were not considered at this time.

5. Conclusions

The results of this study elucidated the concentrations, compositional properties, environmental risk and sources of HCHs and DDTs in soils along the north coastal and riverine area of the Bohai Sea, China. Among the isomers of HCH and metabolites of DDT, β-HCH and p,p'-DDT were found to be predominant. Compared with earlier studies of OCPs in soils, the pollution caused by HCHs and DDTs in the areas studied was generally found to be moderate at the time samples were collected. Compared with the reference values suggested by China, the Netherlands and the USA, the concentrations of $\sum$HCH in soils were at safe levels, although the DDT residues would pose adverse biological effects in several sites with elevated concentrations of $\sum$DDT. PCA and correlation analysis indicated that the HCHs and DDTs seemed to share common sources of origin, as well as similar environmental behaviour. These results indicated that HCH pollution was a mixture of historical technical HCH and current lindane sources, while DDT inputs were due only to historical technical DDT use. In general, soil properties such as TOC and/or pH alone could not fully explain the adsorption of target OCPs into soil, suggesting rather complex behaviours between soil properties and target OCP residues.

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References

[1] K. Kannan, N. Kajiwara, B.J. Le Boeuf, and S. Tanabe, *Organochlorine pesticides and polychlorinated biphenyls in California sea lions*, Environ. Pollut. 131 (2004), pp. 425–434.

[2] R. Singh, *Comparison of organochlorine pesticide levels in soil and groundwater of Agra, India*, Bull. Environ. Contam. Toxicol. 67 (2001), pp. 126–132.

[3] G. Bakan and S. Ariman, *Persistent organochlorine residues in sediments along the coast of mid-Black Sea region of Turkey*, Mar. Pollut. Bull. 48 (2004), pp. 1031–1039.

[4] S. Tao, W.X. Liu, Y. Li, Y. Yang, Q. Zuo, B.G. Li, and J. Cao, *Organochlorine pesticides contaminated surface soil as reemission source in the Haihe Plain, China*, Environ. Sci. Technol. 42 (2008), pp. 8395–8400.

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

[6] K. Feng, B.Y. Yu, D.M. Ge, M.H. Wong, X.C. Wang, and Z.H. Cao, *Organo-chlorine pesticide (DDT and HCH) residues in the Taihu Lake Region and its movement in soil–water system I. Field survey of DDT and HCH residues in ecosystem of the region*, Chemosphere 50 (2003), pp. 683–687.

[7] W.Y. Hu, Y.L. Lu, G. Wang, T.Y. Wang, W. Luo, Y.J. Shi, X. Zhang, and W.T. Jiao, *Organochlorine pesticides in soils around watersheds of Beijing reservoirs: a case study in Guanting and Miyun Reservoirs*, Bull. Environ. Contam. Toxicol. 82 (2009), pp. 694–700.

[8] G. Wang, Y.L. Lu, J. Li, T.Y. Wang, J.Y. Han, W. Luo, Y.J. Shi, and W.T. Jiao, *Regional differences and sources of organochlorine pesticides in soils surrounding chemical industrial parks*, Environ. Monit. Assess. 152 (2009), pp. 259–269.

[9] P. Shegunova, J. Klanova, and I. Holoubek, *Residues of organochlorinated pesticides in soils from the Czech Republic*, Environ. Pollut. 146 (2007), pp. 257–261.

[10] R.B. Zhou, L.Z. Zhu, and Q.X. Kong, *Levels and distribution of organochlorine pesticides in shellfish from Qiantang River, China*, J. Hazard. Mater. 152 (2008), pp. 1192–1200.

[11] M.T. Wan, J.N. Kuo, and J. Pasternak, *Residues of endosulfan and other selected organochlorine pesticides in farm areas of the Lower Fraser Valley, British Columbia, Canada*, J. Environ. Qual. 34 (2005), pp. 1186–1193.

[12] M. Liu, Y. Yang, L. Hou, S. Xu, D. Ou, B. Zhang, and Q. Liu, *Chlorinated organic contaminants in surface sediments from the Yangtze Estuary and nearby coastal areas, China*, Mar. Pollut. Bull. 46 (2003), pp. 672–676.

[13] Y. Wang, R. Yang, and G. Jiang, *Investigation of organochlorine pesticides (OCPs) in mollusks collected from coastal sites along the Chinese Bohai Sea from 2002 to 2004*, Environ. Pollut. 146 (2007), pp. 100–106.

[14] W.X. Liu, J.L. Chen, X.M. Lin, and S. Tao, *Distribution and characteristics of organic micropollutants in surface sediments from Bohai Sea*, Environ. Pollut. 140 (2006), 4–8.

[15] P. Zhang, J. Song, and H. Yuan, *Persistent organic pollutant residues in the sediments and mollusks from the Bohai Sea coastal areas, North China: an overview*, Environ. Int. 35 (2009), pp. 632–646.

[16] W.Y. Hu, T.Y. Wang, J.S. Khim, W. Luo, W.T. Jiao, Y.L. Lu, J.E. Naile, C.L. Chen, X. Zhang, and J.P. Giesy, *HCH and DDT in sediments from marine and adjacent riverine areas of North Bohai Sea, China*, Arch. Environ. Contam. Toxicol. 59 (2010), pp. 71–79.

[17] B. Skrbic and N. Durisic-Mladenovic, *Principal component analysis for soil contamination with organochlorine compounds*, Chemosphere 68 (2007), pp. 2144–2152.

[18] M. Manz, K.D. Wenzel, U. Dietze, and G. Schuurmann, *Persistent organic pollutantsin agricultural soils of central Germany*, Sci. Total. Environ. 277 (2001), pp. 187–198.

[19] T.Y. Wang, Y.L. Lu, Y.J. Shi, J.P. Giesy, and W. Luo, *Organochlorine pesticides in soils around guanting reservoir, China*. Environ. Geochem. Health 29 (2007), pp. 491–501.

[20] R.Q. Yang, G.B. Jiang, Q.F. Zhou, C.G. Yuan, and J.B. Shi, *Occurrence and distribution of organochlorine pesticides (HCH and DDT) in sediments collected from East China Sea*, Environ. Int. 31 (2005), pp. 799–804.

[21] L. Chrysikou, P. Gemenetzis, A. Kouras, E. Manoli, E. Terzi, and C. Samara, *Distribution of persistent organic pollutants, polycyclic aromatic hydrocarbons and trace elements in soil and vegetation following a large scale landfill fire in northern Greece*, Environ. Int. 34 (2008), pp. 210–225.

[22] R.Q. Yang, A.H. Lv, J.B. Shi, and G.B. Jiang, *The levels and distribution of organochlorine pesticides (OCPs) in sediments from the Haihe River, China*, Chemosphere 61 (2005), pp. 347–354.

[23] K.L. Willett, E.M. Ulrich, and R.A. Hites, *Differential toxicity and environmental fates of hexachlorocyclohexane isomers*, Environ. Sci. Technol. 32 (1998), pp. 2197–2207.

[24] V.D. Toan, V.D. Thao, J. Walder, H.R. Schmutz, and C.T. Ha, *Contamination by selected organochlorine pesticides (OCPs) in surface soils in Hanoi, Vietnam*, Bull. Environ. Contam. Toxicol. 78 (2007), pp. 195–200.

[25] Y.F. Zhu, H. Liu, Z.Q. Xi, H.X. Cheng, and X.B. Xu, *Organochlorine pesticides (DDTs and HCHs) in soils from the outskirts of Beijing, China*, Chemosphere 60 (2005), pp. 770–778.

[26] F. Gao, J.Y. Jia, and X.M. Wang, *Occurrence and ordination of dichlorodiphenyltrichloroethane and hexachlorocyclohexane in agricultural soils from Guangzhou, China*, Arch. Environ. Contam. Toxicol. 54 (2008), pp. 155–166.

[27] D. Yuan, D. Yang, T.L. Wade, and Y. Qian, *Status of persistent organic pollutants in the sediment from several estuaries in China*, Environ. Pollut. 114 (2001), pp. 101–111.

[28] R.A. Doong, C.K. Peng, Y.C. Sun, and P.L. Liao, *Composition and distribution of organochlorine pesticide residues in surface sediments from the Wu-Shi River estuary, Taiwan*, Mar. Pollut. Bull. 45 (2002), pp. 246–253.

[29] X.H. Qiu, T. Zhu, L. Jing, H.S. Pan, Q.L. Li, G.F. Miao, and J.C. Gong, *Organochlorine pesticides in the air around the Taihu Lake, China*, Environ. Sci. Technol. 38 (2004), pp. 1368–1374.

[30] K. Jaga and C. Dharmani, *Global surveillance of DDT and DDE levels in human tissues*, Int. J. Occup. Med. Environ. Health 16 (2003), pp. 7–20.

[31] R. Gioia, J.H. Offenberg, C.L. Gigliotti, L.A. Totten, S. Du, and S.J. Eisenreich, *Atmospheric concentrations and deposition of organochlorine pesticides in the US Mid-Atlantic region*, Atmos. Environ. 39 (2005), pp. 2309–2322.

[32] S. Villa, M. Vighi, V. Maggi, A. Finizio, and E. Bolzacchini, *Historical trends of organochlorine pesticides in an Alpine glacier*, J. Atmos. Chem. 46 (2003), pp. 295–311.

[33] S. Fu, S.G. Chu, and X.B. Xu, *Organochlorine pesticide residue in soils from Tibet, China*, Bull. Environ. Contam. Toxicol. 66 (2001), pp. 171–177.

[34] F. Borghini, J.O. Grimalt, J.C. Sanchez-Hernandez, R. Barra, C.J.T. Garcia, and S. Focardi, *Organochlorine compounds in soils and sediments of the mountain Andean lakes*, Environ. Pollut. 136 (2005), pp. 253–266.

[35] J. Klanova, N. Matykiewiczova, Z. Macka, P. Prosek, K. Laska, and P. Klan, *Persistent organic pollutants in soils and sediments from James Ross Island, Antarctica*, Environ. Pollut. 152 (2008), pp. 416–423.

[36] A. Covaci, C. Hura, and P. Schepens, *Selected persistent organochlorine pollutants in Romania*, Sci. Total Environ. 280 (2001), pp. 143–152.

[37] GB15618-1995. Science & Technology Department of State Environmental Protection Administration, *Environmental Quality Standard for Soils*, Beijing, China Standard Press, 1995, p. 2 (in Chinese).

[38] The Netherlands Ministry of Housing, *Spatial Planning and Environment's Circular on Target Values and Intervention Values for Soil Remediation*, 2000, pp. 11–12. Available at http://www2.minvrom.nl/Docs/internationaal/annexS_I2000.pdf.

[39] US Environmental Protection Agency, *Recommended Ecological Screening Values for Soil (WSRC-TR-98-00110)*, US Environmental Protection Agency, Washington, DC, 2001, pp. 3–4. Available at http://www.epa.gov/region4/waste/ots/epatab4.pdf.

[40] A. Urzelai, M. Vega, and E. Angulo, *Deriving ecological risk-based soil quality values in the Basque Country*, Sci. Total Environ. 247 (2000), pp. 279–284.

[41] R.H. Jongbloed, T.P. Traas, and R. Luttik, *A probabilistic model for deriving soil quality criteria based on secondary poisoning of top predators. 2. Calculations for dichlorodiphenyltrichloroethane (DDT) and cadmium*, Ecotoxicol. Environ. Safety 34 (1996), pp. 279–306.

[42] H.B. Zhang, Y.M. Luo, Q.G. Zhao, M.H. Wong, and G.L. Zhang, *Residues of organochlorine pesticides in Hong Kong soils*, Chemosphere 63 (2006), pp. 633–641.

[43] X.J. Wang, X.Y. Piao, J. Chen, J.D. Hu, F.L. Xu, and S. Tao, *Organochlorine pesticides in soil profiles from Tianjin, China*, Chemosphere 64 (2006), pp. 1514–1520.

[44] H. Zhang, Y.L. Lu, R.W. Dawson, Y.J. Shi, and T.Y. Wang, *Classification and ordination of DDT and HCH in soil samples from the Guanting Reservoir, China*, Chemosphere 60 (2005), pp. 762–769.

[45] X.H. Li, Y.F. Zhu, X.F. Liu, S. Fu, X.B. Xu, and H.X. Cheng, *Distribution of HCHs and DDTs in soils from Beijing City, China*, Arch. Environ. Contam. Toxicol. 51 (2006), pp. 329–336.

[46] P. Weiss, G. Lorbeer, and S. Scharf, *Regional aspects and statistical characterisation of the load with semivolatile organic compounds at remote Austrian forest sites*, Chemosphere 40 (2000), pp. 1159–1171.

[47] K.D. Wenzel, M. Manz, A. Hubert, and G. Schuurmann, *Fate of POPs (DDX, HCHs, PCBs) in upper soil layers of pine forests*, Sci. Total Environ. 286 (2002), pp. 143–154.

[48] F. Wang, X. Jiang, Y.R. Bian, F.X. Yao, H.J. Gao, G.F. Yu, J.C. Munch, and R. Schroll, *Organochlorine pesticides in soils under different land usage in the Taihu Lake region, China*, J. Environ. Sci. 19 (2007), pp. 584–590.