Distribution of Organochlorine Pesticides in the Northern South China Sea: Implications for Land Outflow and Air–Sea Exchange

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The South China Sea (SCS) is surrounded by developing countries in Southeast Asia, where persistent organic pollutants (POPs), such as organochlorine pesticides (OCPs), are still used legally or illegally, and are of concern. Yet little is known about the distribution of OCPs in the water and atmosphere over SCS, as well as their air–sea equilibrium status and time trends. In this study, ship-board air samples and surface seawater collected in the northern SCS between September 6 and 22, 2005 were analyzed for selected OCPs. The measured OCP concentrations in the atmosphere over the northern SCS were influenced by proximity to source regions and air mass origins. The highest atmospheric OCP concentrations were found at sampling sites adjacent to continental South China. OCPs in surface seawater showed significant spatial variations, with the highest concentration observed in a water sample from off Vietnam. The coastal currents were suggested to play a key role in the delivery of waterborne OCPs in the northern SCS. Time trend, land outflow, and air–sea exchange of selected OCPs in the SCS were investigated, by comparison of this dataset with historical data.

Introduction

Organochlorine pesticides (OCPs) are typical persistent organic pollutants (POPs). They are of worldwide concern owing to their persistence, bioaccumulation, and potential negative impacts on humans and wildlife. OCPs are subject to global redistribution through the environment via long range atmospheric transport (LRAT), or by ocean currents and animal migration (*1*).

Large water bodies such as oceans and seas play an important role in the global biogeochemistry of POPs (*2*), either acting as a sink (*3*) or as a source for POPs in the environment. Diffusive exchange of POPs across the air–water interface may alter direction as a consequence of

global/regional reduction of POP sources (*4*). The evaluation of air–sea equilibrium status of POPs in different regions is therefore critical to understanding their global source/sink contributions (*4, 5*).

The South China Sea (SCS) is a marginal sea in Southeast Asia, surrounded by tropical–subtropical developing countries which are considered potentially important source regions of OCPs (*6, 7*), including China (South), Vietnam, Thailand, Indonesia, Philippines, and Malaysia. Compared to developed countries, developing countries surrounding SCS historically had higher agricultural applications of OCPs, such as technical DDT and HCH. High ambient temperatures also lead to a high evaporation loss of pesticides after their application in these low latitude regions. The agricultural application of technical DDT was banned in China in 1983, the Philippines in 1989, Vietnam in 1993, and Malaysia in 1999 (*1*). However, DDT can still be used for health services, e.g., in malaria/mosquito control (*8*), for which there is an exemption under the Stockholm Convention.

Recent monitoring data showed that OCPs occurred at high concentrations in the environment of these countries (*8–12*). However, there have been very few reports (*13*) on the concentrations, time trends, potential sources and transmission pathways, and the air–sea equilibrium status of OCPs in the regionally important SCS.

Materials and Methods

Sample Collection. A total of 32 8-h day/night air samples were taken from September 6–22, 2005 during a Shiyan III cruise which circumnavigated the SCS in a zigzag from Guangzhou to eastern SCS, Hainan Island, and back to Guangzhou—a distance of approximately 6500 km. The cruise line is depicted in Figure 1. A high-volume (Hi-Vol) air sampler was placed windward on the foredeck of the ship. Approximately 140 m³ of air was drawn through a glass fiber filter (GFF) (20.3 × 25.4 cm, Whatman), and subsequently through a polyurethane foam (PUF) plug, 6.5 cm (i.d.) × 7.5 cm (thickness), density 0.030 g cm⁻³. Prior to sampling, the GFFs were baked at 450 °C for 12 h, and PUF plugs were Soxhlet extracted for 24 h each with methanol and acetone, and 48 h with dichloromethane (DCM). After sampling, loaded GFFs were wrapped with prebaked aluminum foils and sealed with double layers of polyethylene bags. The PUFs were placed in solvent-rinsed glass jars with Teflon lined lids, and then transported to the laboratory and stored at −20 °C. Meteorological data were recorded *in situ*. Table SI1 (in the Supporting Information) lists necessary information about the sampling locations and environmental parameters for all the air samples.

Surface water samples (each of 100 L) were collected at 9 stations (Figure 1, locations and water temperatures listed in detail in Table SI2), with a metal bucket and stored in four 25 L glass bottles. The water was immediately filtered through pre-combusted (4 h at 450 °C) 150 mm diameter GFFs (Gelman Type A/E, nominal pore size 1 μm). After adding known amounts of 2,4,5,6-tetrachloro-*m*-xylene (TC*m*X) and PCB209 (decachlorobiphenyl) as recovery surrogates, dissolved-phase pesticides were then reclaimed by passing the filtrate over a mixed solid-phase extraction (SPE) resin (Amberlite XAD-2 and XAD-4, 1:1, V/V) glass column (25 mm i.d.; 400 mm length) at a flow rate of approximately 1 L min⁻¹.

Sample Treatment and Analytical Procedure. GFFs and PUFs were spiked with 20 ng of TC*m*X and PCB209 as surrogates, and Soxhlet-extracted with DCM (Merck Inc.) for 48 h. Activated copper was used to remove elemental

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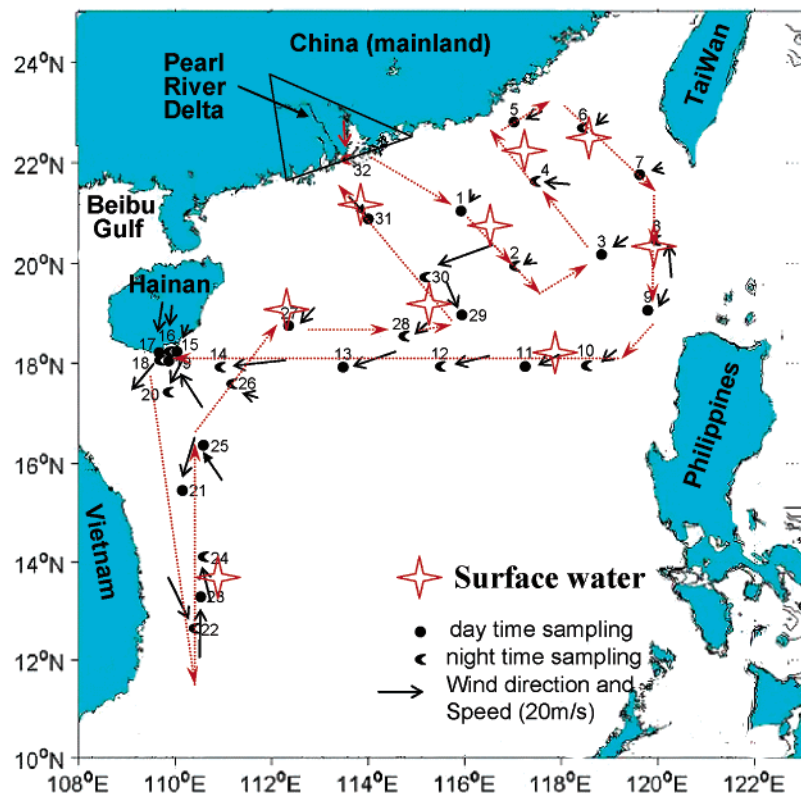§ Lancaster University.

FIGURE 1. Mean sample locations and broad origins of the air masses.

sulfur. The extract was concentrated and further purified on an 8 mm i.d. alumina/silica column packed with neutral alumina (6 cm, 3% deactivated), neutral silica gel (10 cm, 3% deactivated), 50% (w/w) sulfuric acid silica (10 cm), and anhydrous sodium sulfate (1 cm). Before use, neutral alumina, neutral silica gel, and anhydrous sodium sulfate were Soxhlet-extracted for 48 h with DCM, and baked for purification. The column was eluted with 50 mL of DCM/hexane (1:1) to yield the OCP fraction. The fraction was reduced in volume and solvent exchanged to 25 μL of dodecane containing a known quantity of pentachloronitrobenzene (PCNB) as an internal standard.

Each XAD resin column was eluted with 50 mL of methanol (MeOH) followed by 50 mL of dichloromethane at a flow rate of 5 mL/min. The XAD resin was removed into a flask and ultrasonically extracted with (2:1) DCM/MeOH (3 × 100 mL). The combined extracts were back-extracted 3 times using saturated NaCl solution and DCM. Then the DCM fraction was washed with pure water and dried with 15 g of pre-combusted anhydrous Na_2SO_4. The extract for each sample was further reduced to approximately 1 mL under a gentle nitrogen stream. Concentrated extracts were fractionated with an alumina/silica gel column, using the procedure described for air samples.

GC−MS analysis was carried out on an Agilent-5975 GC-MSD system with a CP-Sil 8 CB capillary column (50 m length × 0.25 mm i.d., 0.25 μm film thickness), operating under selected ion monitoring (SIM) mode. 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}. Split/splitless injection of a 1 μL sample was performed. The injector temperature was at 250 °C. Pyrolytic degradation (hot needle discrimination) of DDT at the GC inlet was checked daily to be less than 15%.

Quality Control. Method blanks (solvent) and field blanks (PUF and XAD) were analyzed. In addition, surrogate standards were added to each of the samples to monitor procedural performance. The recovery ratios for the surrogates in the samples conform to the ranges reported by the US EPA. For air samples, the average recoveries were 76 ± 11% and 85 ± 7% for TCmX and PCB209, respectively. For water samples, the average recoveries were 66 ± 15% and 75 ± 20% for TCmX and PCB209, respectively. The concentrations of OCPs were corrected for the surrogate recoveries.

Back Trajectories. Air mass origins were determined for the cruise samples using the HYSPLIT transport and dispersion model from the NOAA Air Resources Laboratory (*14*, *15*). Two back trajectories (BTs) were performed, for the start and end of each sampling episode. BTs were traced for 5 days at 00:00 coordinated universal time (UTC) and 8:00 UTC during day time sampling and at 12:00 and 20:00 UTC during night time sampling at 500 and 1000 m above sea level. Samples are shown with a general indication of the direction of their air mass origin/surface wind patterns in Figure 1. At the beginning of sampling schedule times, the SCS was dominated by the northeasterly monsoon. BTs showed air masses flowed from the East China Sea and western Pacific along the East Asian coast over the boundary of Asian continent or through the nearby vicinity of Taiwan Island and Philippines (samples 1−14). During the latter half of the cruise, several typhoon and tropical cyclones occurred in/ near the SCS with the largest wind speed of 23.5 m/s. Air masses circled in the SCS region with high wind speeds and passed over nearby Malaysia, the Philippines, southern China, and Vietnam (samples 15−32).

Results and Discussion

Concentrations and Time Trends. The organochlorine pesticides that occurred most frequently in the SCS air samples were HCHs, DDTs, chlordane, and α-endosulfan. Table SI3 provides detailed information about the OCP concentrations in the air samples, and Table 1 presents a summary of the concentrations of selected OCPs in the vapor phase.

TABLE 1. Summary of Atmospheric (Gas Phase) Concentrations of OCPs over the Northern SCS

pg m^{-3}	mean	median	range
α-HCH	67 ± 33	60	27–206
γ-HCH	771 ± 310	743	343–1950
TC	510 ± 652	205	75–2650
CC	453 ± 619	131	58–2470
p,p′-DDE	51 ± 50	28	11–190
o,p′-DDT	196 ± 141	153	61–794
p,p′-DDT	58 ± 65	28	9–234
α-Endosulfan	131 ± 112	111	9–435

The atmospheric concentrations of α-HCH and γ-HCH ranged from 27 to 206 (67 ± 33) pg m^{-3} and from 343 to 1950 (771 ± 310) pg m^{-3}, respectively. These levels were higher than those observed over the open Atlantic Ocean and the Indian Ocean recently (16, 17). Compared with previous data reported by Iwata et al. (13), these α-HCH concentrations had declined by between ∼4–30 times over the last 10–15 years. This decrease of atmospheric α-HCH is likely to be related to the recent restricted usage of technical HCH in this area. Unlike α-HCH, γ-HCH concentrations did not show any significant decline compared to values measured in 1989 (13). Lindane is still in use as an insecticide and this may be responsible for the continued high levels. The α-/γ-HCH ratios varied between 0.03 and 0.23 in the air, indicative of ongoing lindane (γ-HCH) usage.

The mean DDT concentrations were 196, 58, and 51 pg m^{-3} for o,p′-DDT, p,p′-DDT, and p,p′-DDE, respectively. These were higher than those reported for the open Indian Ocean and Atlantic Ocean (16, 17), and even higher than those reported a decade ago in the adjacent areas by Iwata et al. (13). Most notably, the o,p′-DDT levels were ∼9 times higher. High total DDT concentrations, dominated by o,p′-DDT, indicate inputs from dicofol, which is in use in several countries of Southeast Asia, including China (9, 18, 19). However, some samples displayed relatively low o,p′-DDT/p,p′-DDT values (of <2), which are unlikely to be from dicofol-DDT (19). This may indicate that either technical DDT or DDT-containing products are still in use in some parts of the region. For instance, DDT for Malaria control (8) and as additives in antifouling paints for fishing ships in China (20).

High concentrations of *trans*-chlordane (TC) and *cis*-chlordane (CC) were observed over the northern SCS, with concentrations ranging from 75 to 2650 pg m^{-3} and from 58 to 2470 pg m^{-3}, respectively. These concentrations were comparable to those in the Pearl River Delta in South China (20), and higher than those observed in the Indian Ocean (16). Compared to previous data published by Iwata et al. (13), the levels did not show significant declines. Clearly, chlordane is still in extensive use as a termiticide in the SCS region (13).

The concentrations of α-endosulfan have a mean value of 131 pg m^{-3} (ranges from 9 to 435 pg m^{-3}). The high levels comparable to that reported in relevant application regions possibly indicated the continental outflow from upwind agricultural areas (20–22).

Spatial Variation and Continental/Land Outflow. As shown in Figure 2, significant difference in the spatial distribution of OCPs was observed. The concentrations of DDTs were generally much higher at sites close to land than those in the open sea. HCH concentrations were relatively uniform with high/low values of 7.6 and 5.7 for α- and γ-isomer. For chlordane, relatively higher concentrations were observed at some sampling sites, and the reason was unknown. The lowest concentrations of α-endosulfan were observed at sites with the highest DDT and chlordane concentrations, implying different source regions and transport pathway for α-endosulfan.

At the beginning of the sampling campaign (from site 1 to 12, c. f. Figure 1), a northeasterly (monsoon) wind dominated the air circulation. Back trajectory analysis revealed that the air masses flowed from the East China Sea and western Pacific along the coast, or passed over Taiwan Island and the Philippines. During this period, relatively consistent DDT levels were observed, and the ratios of o,p′-DDT/p,p′-DDT ranged from 6.4 to 11. This is indicative of dicofol-DDT inputs, likely received en route from the eastern China region, where large amounts of DDT-containing dicofol are produced and applied (13, 19). Higher concentrations of α-endosulfan were also observed during this period, which is consistent with the fact that eastern China is a very important cotton growing area, where endosulfan is extensively used for caterpillar control.

From September 13 to 15, there was a strong tropical cyclone, and the ship stayed in Sanya harbor. The 6 air samples collected at the harbor (samples 15–20) displayed very high concentrations of DDTs, with p,p′-DDT dominating the composition (Figure 2), and p,p′-DDT/p,p′-DDE ratios >1. The ratios of o,p′-DDT/p,p′-DDT were close to 2, much lower than that of dicofol-DDT (6–8) (19). These observations provide evidence of a strong local "fresh" DDT source other than dicofol application, most likely from DDT additives in antifouling paint for fishing ships anchored in the harbor (20). DDT anti-fouling paints are still used in coastal regions of China to prevent the adhesion of barnacles to the hulls of fishing ships. Samples 29 and 32 downwind of the Pearl River estuary (see Figure 1) also displayed similar technical DDT signatures, supporting this suggestion and consistent with reports on the atmosphere of Hong Kong (20). Interestingly, a similar DDT composition was observed in sample 10, close to the Philippines.

In the second half of the sampling campaign (samples 21–32, Figure 1), several typhoons and tropical cyclones took place in/near the SCS, with the highest wind speed reaching 23.5 m s^{-1}. These air masses circulated in the SCS region at high speeds, passing over nearby Malaysia, the Philippines, South China and Vietnam. During this time, the o,p′-DDT/p,p′-DDT ratios ranged from 2 to 6, indicating a mixed source from dicofol and anti-fouling paints. Relatively high concentrations of α-endosulfan were observed in samples 23 and 25 off Vietnam during this period, which may indicate potential atmospheric outflow from this country.

As shown in Figure 2, α-HCH was generally uniform in the atmosphere over the northern SCS, except for samples 31 and 32 off the Pearl River estuary, implying an outflow of α-HCH from continental mainland China. γ-HCH concentrations were also generally uniform, despite its higher concentrations than other OCPs. This is consistent with the widespread application of lindane in the SCS region.

Based on the above discussion, it can be concluded that the OCPs in the atmosphere over the SCS were largely controlled by the origins of the air masses, in particular those from continental/land outflow from the surrounding countries/regions. This implies that POPs over this marginal sea are controlled by advection from ongoing land-based sources, a different scenario from over open oceans, where dynamic air–sea exchange of POPs may play an important role in influencing atmospheric concentrations (17, 23).

Concentrations in Seawater. The detailed analytical results for the water samples are given in Table SI2. ΣDDT concentrations ranging from 97 to 1620 pg L^{-1} (mean 470) were observed in the SCS seawater samples. These are higher than those reported for open oceans (24). Ueno et al. used skipjack tuna as a biomonitor of OCPs in oceans, and found the highest DDT levels in the SCS (25). Compared with those reported by Iwata et al., DDT concentrations in water appear to have increased by up to 70 times during the last 15 years
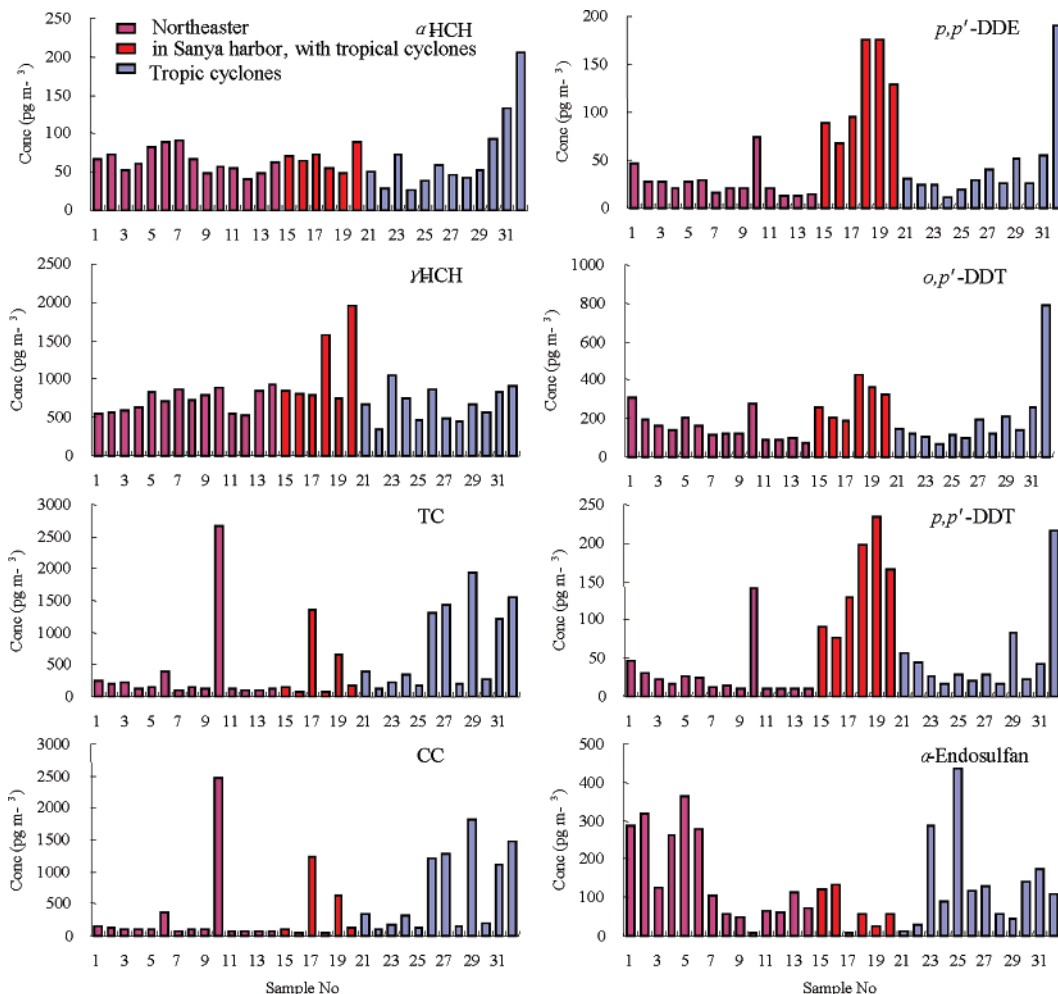
FIGURE 2. Gaseous concentrations of selected OCPs over the northern SCS.

(*13*). The (*p,p′*-DDE + *p,p′*-DDD)/*p,p′*-DDT and *o,p′*-DDT/ *p,p′*-DDT ratios in the seawater ranged from 0.58 to 6.6 and from 0.17 to 2.0, respectively, indicating ongoing fresh inputs of DDT in the SCS region. While the *o,p′*-DDT/*p,p′*-DDT may indicate a currently increasing input of dicofol-type DDT to the northern SCS (*19*), the increase of (*p,p′*-DDE + *p,p′*-DDD)/ *p,p′*-DDT ratio indicated an input of "weathered" DDT, which may have built up in soils and was transported/leached into the rivers and reached coastal waters along with the emerging fast economic development and land transformation taking place in the surrounding developing countries/regions (*26*). The coastal currents occurring strongly in the fringe of the marginal SCS, in particular when northeasterly wind prevails here, may play a key role in the transport of land derived/ riverine pollutants to the SCS.

The concentration of ΣHCH (sum of α-HCH, β-HCH, γ-HCH, and δ-HCH) ranged from 633 to 3760 pg L^{-1} (Figure 3). These were lower than those reported for Southeast Asia (ranges from 430 to 42 200 pg L^{-1}) (*27*), but much higher than those in the open oceans, such as the Indian Ocean and the Atlantic Ocean (*17*). The mean concentrations of α-HCH and γ-HCH were 612 pg L^{-1} (252−1310 pg L^{-1}) and 461 pg L^{-1} (150−1310 pg L^{-1}), respectively, which were 2 and 5 times higher than those in 1989 (*13*). The ratios of α-HCH/γ-HCH in the surface seawater samples averaged 1.49 (±0.34), up to 3 times lower than those in 1989, indicating a shift in the predominant source of HCH, from technical HCH to lindane (*13*).

The concentrations of TC and CC ranged from 5 to 93 (mean 27 ± 27) pg L^{-1} and from 15 to 218 (mean 91 ± 66)

pg L^{-1}, respectively. These levels were also higher than those monitored in the same area in 1989 (*13*). Only 2 of 10 samples had detectable levels of α-endosulfan (samples 306 and 502, see Figure 3), and the concentrations of α-endosulfan were 39 and 266 pg L^{-1}, respectively, which were comparable to those reported for the Bering and Chukchi Sea (*24*).

Spatial Distribution of OCPs in Surface Seawater. The highest concentration of ΣDDT was observed in a sample collected from the water off Vietnam (site FB, Figure 3), and *p,p′*-DDT dominated the composition. It was reported that DDT levels up to 324 ng L^{-1} were found in water samples from the Red and Duong rivers in Vietnam, with *p,p′*-DDT concentrations higher than those of *p,p′*-DDE (*8*). The riverine outflow of DDTs may be related to site FB by the perennial surface current running across Beibu Gulf (Figure 1) and flowing southward along the coast of Vietnam (*28*). Similarly, relatively high levels and fresh input of DDT were found in the samples to the west of the Pearl River estuary, and the entryway of Taiwan Strait to the SCS (Figure 3), both located along the coast and along the coastal current route. It is therefore suggested that the coastal currents may play a key role in the dissipation and transport of DDTs in the waters of the northern SCS.

The highest HCH concentration was also monitored in the water off Vietnam. The relatively high proportion of γ-HCH compared to β-HCH (Figure 3) is indicative of current lindane use in Vietnam (*8*).

The most abundant components in the technical chlordane are TC and CC. In a previous study, a TC/CC ratio of 1.18 in the technical mixture was reported (*29*). TC is reported
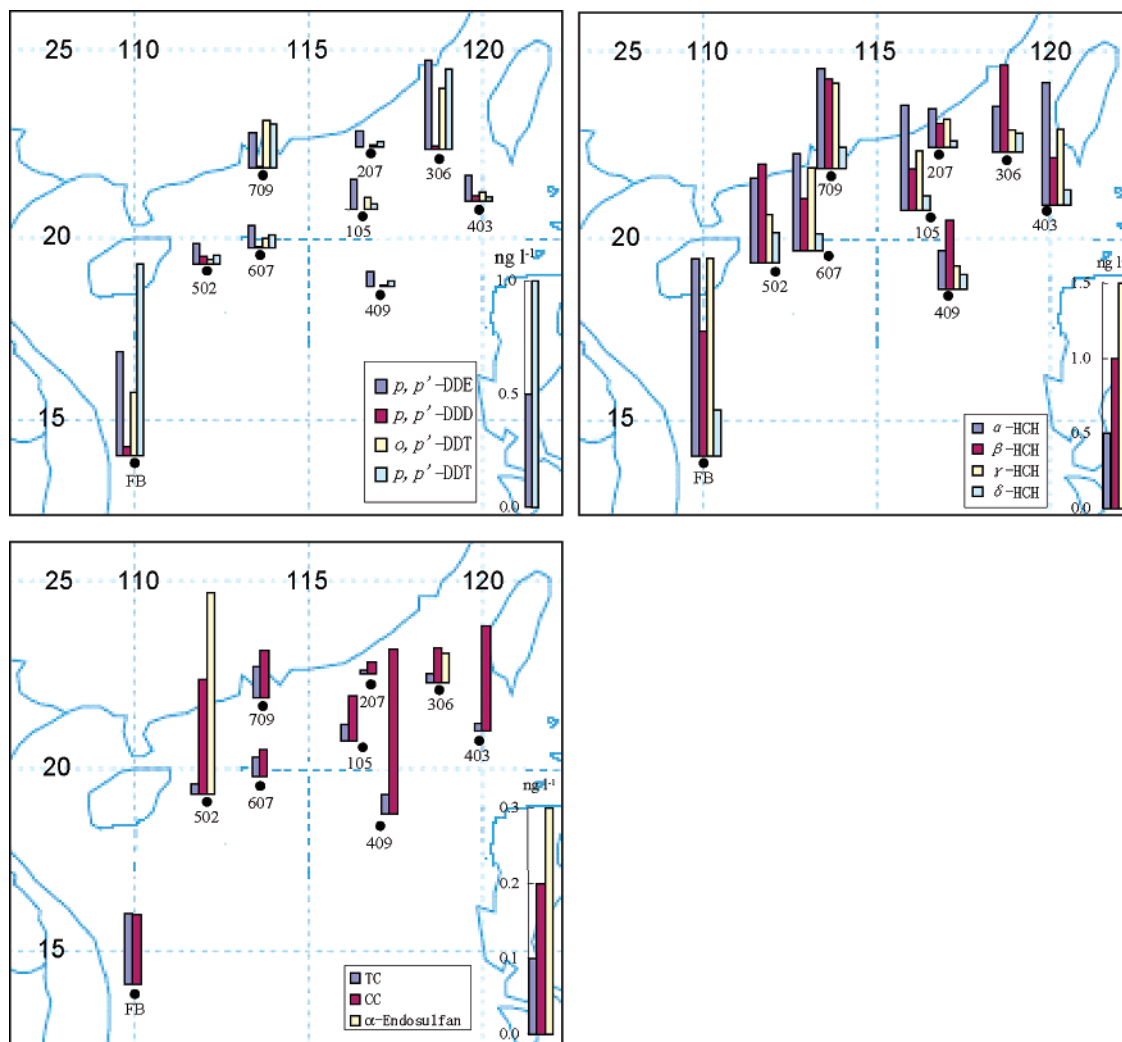
FIGURE 3. Surface water (dissolved phase) concentrations of selected OCPs in the northern SCS.

to be more labile to photochemical reaction than CC (*30*). Relatively high values of TC/CC were observed in the proximity to Vietnam and South China (see Figure 3), suggesting "fresher" chlordane around the northern SCS than those sites far away from land.

Air/Water Exchange of Selected OCPs. The driving force for OCP volatilization is the difference in fugacities between water and air. Fugacities in water (f_w) and air (f_a) were calculated from (*31*)

$$f_w = C_w H \qquad (1)$$

$$f_a = C_a RT \qquad (2)$$

where C_w and C_a are the concentrations of the chemical in water and air (mol m^{-3}), R is the gas constant (8.314 Pa m^3 mol^{-1} K^{-1}), H is the Henry's law constant (Pa m^3 mol^{-1}), and T is absolute temperature (K). For these calculations an average temperature of 302.2 K based on the average seawater temperature (29.2 ± 0.5 °C, $n = 9$) was used, along with temperature-corrected H values for selected OCPs (*32, 33*). Results were expressed as the fugacity fraction (*ff*) (*34*)

$$ff = f_w/(f_w + f_a) \qquad (3)$$

where $ff = 0.5$ at water−air equilibrium, $ff > 0.5$ indicates net volatilization from the water, and $ff < 0.5$ indicates net deposition to the water.

It should be noted that there is substantial uncertainty associated with the determination of the direction of net air−sea exchange. A detailed analysis of the uncertainties associated with the determination of air−sea gaseous exchange fluxes of organic chemicals was given by Hoff (*35*) and Bruhn et al. (*36*). Briefly, the uncertainty in the calculated *ff* values was predominantly based on the error in the measured C_w, C_a, and H values. A reasonable estimation of the error of the C_a and C_w that considers the random sampling and analytical error is from 0.15 to 0.30 (*36*). The uncertainty associated with H is generally considerably larger (*32, 33, 36*). So application of this method to estimate the exchange direction of organic contaminants across air−sea interface should be cautious. In this study, uncertainty (45%) in the calculated *ff* values was determined on the basis of propagation of error in the measured C_w, C_a, and H values.

The fugacity fraction (*ff*) for HCH in the SCS demonstrated that air/water flux of α-HCH was near equilibrium at all sample sites, except for sample site FB near Vietnam, where the flux was from water to air, whereas gas deposition of γ-HCH occurred across the air−sea surface of the SCS (refer to Figure SI1). This was similar to the Arctic and Antarctic, where α-HCH was found to be near equilibrium and γ-HCH demonstrated a potential of air to water exchange (*i.e.*, deposition) (*37, 38*). Compared with the results from 15 years ago in the same study area (*13*), the flux direction of α-HCH has changed from net deposition to equilibrium or volatil-

ization, with γ-HCH consistently undergoing net deposition over this time period.

Different air−sea exchange scenarios for DDT isomers were observed. The air/water flux of p,p'-DDE was generally from water to atmosphere, whereas that of o,p'-DDT and p,p'-DDT varied among geographic locations, from water to atmosphere in the proximity to land and net deposition in the remote ocean. The net volatilization of p,p'-DDE may imply the impact of historical usage of DDT around this region, while the results of o,p'-DDT and p,p'-DDT reflect the impact of current source of the fresh DDT in the SCS region.

The gas exchange fluxes of TC and CC are predominantly from air to water, suggesting the impact of land outflow on the seawater. However, there were several exceptional sample sites for CC in the more remote ocean, where the gas exchange flux was from water to atmosphere.

Implications for Future Studies. Many POPs can migrate from warmer places to colder places over the global scale (1). Most developing countries are situated in low latitude regions (LLR). Considering the combination of higher temperatures, ongoing (legal and illegal) usage, and later production/ application bans of pesticidal POPs in LLR developing countries/regions, it is likely that the current global/regional sources of these compounds have shifted to LLR. Our results show that, even after a long period of regional/global control/ reduction, the OCP levels in the SCS are still high compared to other seas and historical data. The terrestrial/aquatic outgassing and LRAT of these chemicals is therefore of concern in the global context, and deserves further study.

Acknowledgments

Supporting Information Available

Table SI1: Information on sampling location, sampling times, and meteorological parameters; Table SI2: Dissolved phase concentrations (pg L^{-1}) and isomer ratios of OCPs in the surface water, water temperature; Table SI3: Gas-phase concentrations and isomer ratios of OCPs in the atmosphere over the northern SCS (pg m^{-3}); Figure SI1. Water/air fugacity fraction (ff) for selected OCPs in the SCS water during September 2005 (fractions >0.5 indicate transfer from water to air, fractions <0.5 indicate transfer from air to water). This material is available free of charge via the Internet at http:// pubs.acs.org.

Literature Cited

(1) UNEP. *Regional Based Assessment of Persistent Toxic Substances: Global Report 2003*; United Nations Environment Programme: Nairobi, Kenya, 2004.

(2) Dachs, J.; Lohmann, R.; Ockenden, W. A.; Mejanelle, L.; Eisenreich, S. J.; Jones, K. C. Oceanic biogeochemical controls on global dynamics of persistent organic pollutants. *Environ. Sci. Technol.* **2002**, *36*, 4229−4237.

(3) Lohmann, R.; Jurado, E.; Pilson, M. E. Q.; Dachs, J. Oceanic deep water formation as a sink of persistent organic pollutants-art. no. L12607. *Geophys. Res. Lett.* **2006**, *33*, 12607−12607.

(4) Jantunen, L. M.; Kylin, H.; Bidleman, T. F. Air−water gas exchange of alpha-hexachlorocyclohexane enantiomers in the South Atlantic Ocean and Antarctica. *Deep-Sea Res., Part I* **2004**, *51*, 2661−2672.

(5) Breivik, K.; Wania, F. Mass budgets, pathways, and equilibrium states of two hexachlorocyclohexanes in the Baltic Sea environment. *Environ. Sci. Technol.* **2002**, *36*, 1024−1032.

(6) Tanabe, S. Fate of Toxic-Chemicals in the Tropics. *Mar. Pollut. Bull.* **1991**, *22*, 259−260.

(7) Loganathan, B. G.; Kannan, K. Global Organochlorine Contamination Trends-an Overview. *Ambio* **1994**, *23*, 187−191.

(8) Hung, D. Q.; Thiemann, W. Contamination by selected chlorinated pesticides in surface waters in Hanoi, Vietnam. *Chemosphere* **2002**, *47*, 357−367.

(9) Jaward, T. M.; Zhang, G.; Nam, J. J.; Sweetman, A. J.; Obbard, J. P.; Kobara, Y.; Jones, K. C. Passive air sampling of polychlorinated biphenyls, organochlorine compounds, and polybrominated diphenyl ethers across Asia. *Environ. Sci. Technol.* **2005**, *39*, 8638−8645.

(10) Luo, X.; Mai, B.; Yang, Q.; Fu, J.; Sheng, G.; Wang, Z. Polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides in water columns from the Pearl River and the Macao harbor in the Pearl River Delta in South China. *Mar. Pollut. Bull.* **2004**, *48*, 1102−1115.

(11) Wong, C. K. C.; Leung, K. M.; Poon, B. H. T.; Lan, C. Y.; Wong, M. H. Organochlorine hydrocarbons in human breast milk collected in Hong Kong and Guangzhou. *Arch. Environ. Contam. Toxicol.* **2002**, *43*, 364−372.

(12) Iwata, H.; Tanabe, S.; Sakai, N.; Nishimura, A.; Tatsukawa, R. Geographical distribution of persistent organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes. *Environ. Pollut.* **1994**, *85*, 15−33.

(13) Iwata, H.; Tanabe, S.; Sakai, N., Tatsukawa, R Distribution of persistent organochlorines in the oceanic air and surface seawater and the role of ocean on their global transport and fate. *Environ. Sci. Technol.* **1993**, *27*, 1080−1098.

(14) Rolph, G. D. *Real-time Environmental Applications and Display System (READY) Website*; NOAA Air Resources Laboratory: Silver Spring, MD, 2003; http://www.arl.noaa.gov/ready/hysplit4.html.

(15) Draxler, R. R.; Rolph, G. D. NOAA Air Resources Laboratory; S. S., MD, Ed.; 2003.

(16) Wurl, O.; Potter, J. R.; Obbard, J. P.; Durville, C. Persistent organic pollutants in the equatorial atmosphere over the open Indian Ocean. *Environ. Sci. Technol.* **2006**, *40*, 1454−1461.

(17) Jaward, F. M.; Barber, J. L.; Booij, K.; Dachs, J.; Lohmann, R.; Jones, K. C. Evidence for Dynamic Air−water Coupling and Cycling of Persistent organic Pollutants over the Open Atlantic Ocean. *Environ. Sci. Technol.* **2004**, *38*, 2617−2625.

(18) Taylor, M. D.; Klaine, S. J.; Carvalho, F. P.; Barcelo, D.; Everaarts, J. *Pesticide Residues in Coastal Tropical Ecosystems*; Taylor and Francis: London, 2003.

(19) Qiu, X. H.; Zhu, T.; Yao, B.; Hu, J. X.; Hu, S. W. Contribution of dicofol to the current DDT pollution in China. *Environ. Sci. Technol.* **2005**, *39*, 4385−4390.

(20) Li, J.; Zhang, G.; Guo, L.; Xu, W.; Li, X.; Lee, C. S. L.; Ding, A.; Wang, T. Organochlorine pesticides in the atmosphere of Guangzhou and Hong Kong: Regional sources and long-range atmospheric transport. *Atmos. Environ.* **2007**, *41*, 3889−3903.

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

(22) Yeo, H. G.; Choi, M.; Sunwoo, Y. Seasonal variations in atmospheric concentrations of organochlorine pesticides in urban and rural areas of Korea. *Atmos. Environ.* **2004**, *38*, 4779−4788.

(23) Wania, F.; Axelman, J.; Broman, D. A review of processes involved in the exchange of persistent organic pollutants across the air-sea interface. *Environ. Pollut.* **1998**, *102*, 3−23.

(24) Yao, Z. W.; Jiang, G. B.; Xu, H. Z. Distribution of organochlorine pesticides in seawater of the Bering and Chukchi Sea. *Environ. Pollut.* **2002**, *116*, 49−56.

(25) Ueno, D.; Takahashi, S.; Tanaka, H.; Subramanian, A. N.; Fillmann, G.; Nakata, H.; Lam, P. K. S.; Zheng, J.; Muchtar, M.; Prudente, M.; Chung, K. H.; Tanabe, S. Global pollution monitoring of PCBs and organochlorine pesticides using skipjack tuna as a bioindicator. *Arch. Environ. Contam. Toxicol.* **2003**, *45*, 378−389.

(26) Zhang, G.; Parker, A.; House, A.; Mai, B.; Li, X.; Kang, Y.; Wang, Z. Sedimentary Records of DDT and HCH in the Pearl River Delta, South China. *Environ. Sci. Technol.* **2002**, *36*, 3671−3677.

(27) Wurl, O.; Obbard, J. P. Chlorinated pesticides and PCBs in the sea-surface microlayer and seawater samples of Singapore. *Mar. Pollut. Bull.* **2005**, *50*, 1233−1243.

(28) Morton, B.; Blackmore, G. South China Sea. *Mar. Pollut. Bull.* **2001**, *42*, 1236−1263.

(29) Jantunen, L. M. M.; Bidleman, T. F.; Harner, T.; Parkhurst, W. J. Toxaphene, chlordane, and other organochlorine pesticides in Alabama air. *Environ. Sci. Technol.* **2000**, *34*, 5097−5105.

(30) Halsall, C. J.; Bailey, R.; Stern, G. A.; Barrie, L. A.; Fellin, P.; Muir, D. C. G.; Rosenberg, B.; Rovinsky, F. Y.; Kononov, E. Y.; Pastukhov, B. Multi-year observations of organohalogen pesticides in the Arctic atmosphere. *Environ. Pollut.* **1998**, *102*, 51−62.

(31) Falconer, R. L.; Bidleman, T. F.; Gregor, D. J. Air−water gas exchange and evidence for metabolism of hexachlorocyclohexanes in Resolute Bay, N.W.T. *Sci. Total Environ.* **1995**, *160−161*, 65−74.

(32) Shen, L.; Wania, F. Compilation, evaluation, and selection of physical-chemical property data for organochlorine pesticides. *J. Chem. Eng. Data* **2005**, *50*, 742−768.

(33) Xiao, H.; Li, N. Q.; Wania, F. Compilation, evaluation, and selection of physical-chemical property data for alpha-, beta-, and gamma-hexachlorocyclohexane. *J. Chem. Eng. Data* **2004**, *49*, 173−185.

(34) Harner, T.; Bidleman, T. F.; Jantunen, L. M. M.; Mackay, D. Soil-air exchange model of persistent pesticides in the United States cotton belt. *Environ. Toxicol. Chem.* **2001**, *20*, 1612−1621.

(35) Hoff, R. M. An error budget for the determination of the atmospheric mass loading of toxic chemicals in the Great Lakes. *J. Great Lakes Res.* **1994**, *20*, 229−239.

(36) Bruhn, R.; Lakaschus, S.; McLachlan, M. S. Air/sea gas exchange of PCBs in the southern Baltic Sea. *Atmos. Environ.* **2003**, *37*, 3445−3454.

(37) Sahsuvar, L.; Helm, P. A.; Jantunen, L. M.; Bidleman, T. F. Henry's law constants for [alpha]-, [beta]-, and [gamma]-hexachlorocyclohexanes (HCHs) as a function of temperature and revised estimates of gas exchange in Arctic regions. *Atmos. Environ.* **2003**, *37*, 983−992.

(38) Dickhut, R. M.; Cincinelli, A.; Cochran, M.; Ducklow, H. W. Atmospheric concentrations and air−water flux of organochlorine pesticides along the western Antarctic Peninsula. *Environ. Sci. Technol.* **2005**, *39*, 465−470.