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Distribution and depth profiles of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, and polychlorinated biphenyls in sediment collected from offshore waters of Central Vietnam



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ABSTRACT

Concentrations of PCBs and OCPs were measured in 35 surface sediment samples collected from offshore waters of Central Vietnam. The mean concentrations of PCBs, HCHs, and DDTs in surface sediments were 86.5, 37.0, and 44.5 pg g^{-1} , respectively. Additionally, nine PCDDs, eleven PCDFs, and twelve dl-PCBs were also examined in 19 sediment core samples collected from five locations. Concentration of PCDDs, PCDFs, and dl-PCBs ranged from 200 to 460, 0.39 to 2.9, and 1.6 to 22 pg g^{-1} , respectively. OCDD was detected at the highest concentration, ranged from 100 to 300 pg g^{-1} . Generally, the concentrations of PCDD/Fs at shallower depths were higher, meanwhile the depth profiles of dl-PCBs in sediment cores were different than the depth profiles of PCDD/Fs. The results suggest that the pollution of PCBs might be from many different sources leading to the variation between depths.

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Following the Stockholm Convention (2001), Vietnam government has issued a banned decree of the production and use of persistent organic pollutants (POPs) in general, including organochlorine pesticides (OCPs), polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzodifurans (PCDFs), polychlorinated biphenyls (PCBs), and dioxin-like polychlorinated biphenyls (dl-PCBs) (Signatory of the Stockholm Convention Citation, 2016). PCDD/Fs are chemicals generated unintentionally, while dl-PCBs were used for the purpose as additives in various industrial products. The toxicity of these chemicals was reported in many conferences, seminars, and scientific papers. Generally, OCPs, PCBs, PCDD/Fs, and dl-PCBs constitute five groups of relevant persistent organic pollutants with enhanced chronic toxicity.

The occurrence of OCPs and PCBs in surface sediments and soils collected from inlands in Vietnam has been reported in several earlier studies (Nhan et al., 1998, 2001; Minh et al., 2006, 2007a, 2007b; Toan et al., 2007b; Shiozaki et al., 2009; Romano et al., 2013). The PCBs concentrations were measured in soil samples ranging from 14,800 to

190,000 pg g^{-1} (Toan et al., 2007a), in sediment of the canals up to 40,000 pg g^{-1} (Nhan et al., 2001), and in sediment of the sewer system ranging from 1300 to 384,000 pg g^{-1} (Hoai et al., 2010) from Hanoi, Vietnam. The mean concentrations of HCHs and DDTs in surface soils in agricultural areas from Hanoi Vietnam were 8030 and 89,900 pg g^{-1} , respectively (Toan et al., 2007b). Further, Nhan et al. (1999) determined the concentration of HCHs, DDTs, and PCBs in sediments collected from the coast of the north of Vietnam ranged from 1200 to 33,700 pg g^{-1} , 6200 to 10,400 pg g^{-1} , and 470 to 28,100 pg g^{-1} , respectively.

In Vietnam, the aftermath of war, agricultural, and industrial production activities have resulted in a wide dispersion and distribution of POPs over time. The occurrence of these compounds in soil and sediment samples has been reported in some previous studies (Minh et al., 2003). The total concentration of 5 PCDDs and 5 PCDFs were found in soil collected from dumping sites in Hanoi ranging from 100 to 24,000 pg g^{-1} (mean: 3100 pg g^{-1}) and 15–26,000 pg g^{-1} (mean: 2900 pg g^{-1}), respectively (Kunisue et al., 2002). For sediment samples collected from Can Gio, Hue, and Hanoi, the total concentration of PCDDs and PCDFs were measured at respective level of 350–980 pg g^{-1} and 26–140 pg g^{-1} (Kishida et al., 2010).

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However, there are very few studies on the distribution of PCDD/Fs, and dl-PCBs in sea sediment samples in Vietnam. The total concentration of PCDDs and PCDFs in sediment samples collected from Central Vietnam coastal lagoons ranged from 192 to 2910 pg g^{-1} (Piazza et al., 2010).

In this study, the occurrence of PCBs and OCPs in surface sediments and PCDD/Fs, and dl-PCBs was examined in sediment cores collected from offshore waters of Central Vietnam. Understanding depth profiles of PCDD/Fs and dl-PCBs may provide insights into the fate and transport of these toxic chemicals in coastal offshore water, and help elucidating history of contamination.

Using the HQ14-11-78, 273 Regiment Military Zone 3, Vietnam ship (450 tons payload), sediment samples were collected from offshore between Nghe An to Thua Thien-Hue in Central Vietnam (Fig. 1). Coordinates of survey stations were determined by the SPS351 DGPS (Trimble, USA) set on the ship and connected to reference station was set at Thang Binh District (Quang Nam province). Sampling depth position was determined by the depth gauge F2000 (Japan) and corrected for national elevation system (0 m of continent) at Hon Dau (Hai Phong province). The ocean hoists collect sediment samples from surface to 20 cm depth.

Surficial sediment samples were collected by grab samples (from surface to 20 cm depth of sediments). Sediment core samples were collected by the gravity discharge tube gravity corer (10 cm in diameter), with lengths of cores from 20 to 160 cm. After taking up the core samples was transferred to a Teflon tube, cut by a plastic knife into 40 cm intervals and moved the outer layers (0.5 cm in thickness). Both types of surficial sediments and discharge tube samples were wrapped immediately in polyethylene bags. Then samples were immediately stored in cool boxes and moved to laboratories. Nineteen sediment samples were collected from 5 cores and assigned sample number of 236, 400, 465, 658, and 821. All sediment samples were air-dried and sieved with 2 mm sieve before analysis.

PCBs and OCPs in surface sediment samples were analyzed according to the method described by Minh et al. (2006) with some modifications. All glassware was washed in the following order: acetone, toluene, n-hexane, and backed at 400 °C for 12 h. A 25 g sediment sample was mixed with 150 mL acetone in a conical flask. The flask was shaken vigorously for 60 min in an electric shaker and sediment solution was filtered into a separating funnel containing 600 mL water and 100 mL n-hexane. The funnel was shaken for 20 min and kept in a stand for 8 h to entirely separate the aqueous from the n-hexane layers. The aqueous layer was discarded, and the n-hexane layer was washed three times with 100 mL water per wash. The volume of n-hexane in the final solution was measured for calculating recovery from the initial 100 mL, and then concentration by Kuderna-Danish (KD) apparatus to

approximately 10 mL, followed by further concentration by gentle nitrogen stream to 5 mL. The 5 mL solution was transferred to a multilayer column packed with silica gel, H_2SO_4 -absorbed silica gel and AgNO_3 -absorbed silica gel. Three layers in the column were packed in the following order: 0.5 g Na_2SO_4 , 0.5 g silica gel, 2 g H_2SO_4 -absorbed silica gel, 0.5 g silica gel, 2 g AgNO_3 -absorbed silica gel, and 0.5 g Na_2SO_4 . After transferring the sample solution into the multilayer column, the elution was made by passing 250 mL of 5% dichloromethane in hexane through the column. The collected mixture was concentrated by KD and blown down by nitrogen to exactly 5 mL. Four milliliters of this solution was collected by pipette for further cleanup by gel-permeation chromatography and separated by Florisil (Wako Chemicals, USA) chromatography column to obtain PCBs and OCPs fractions as previously described (Minh et al., 2003, 2007a, 2007b). PCBs, DDTs, HCHs were quantified by a gas chromatography–electron capture detection (GC-ECD, Shimadzu, Japan) using DB-1 fused silica capillary column (30 m \times 0.25 mm I.D. \times 0.25- μm film thickness). The column oven temperature was programmed from 70 °C (1 min) to 160 °C at a rate of 20 °C min^{-1} , held for 10 min, then increased to 270 °C at a rate of 2 °C min^{-1} , and held for 20 min. The PCBs (including PCB-28, PCB-52, PCB-101, PCB-118, PCB-138, PCB-143, PCB-180, and PCB-209), HCHs (α -, β -, δ -, γ -HCHs), and DDTs (p,p'-DDE, p,p'-DDD, o,p'-DDT, and p,p'-DDT) standards and internal standards used for quantification were purchased from Wellington, Ontario, Canada. Recovery of PCBs, HCHs, and DDTs obtained by this procedure were 79–107%, 82–102%, and 81–110%, respectively. The limited of quantification (LOQ) for PCBs was 2.0 pg g^{-1} , while for HCHs and DDT were 0.5 pg g^{-1} .

The methods for PCDD/Fs and dl-PCBs are those described by MOE, Government of Japan (2000) with slight modifications. A sediment sample of 8 g, with 0.5 g of active copper was mixed in a filter cup. Samples were Soxhlet extracted with 140 mL toluene for 16 h. The extracted solution was evaporated to 2 mL. Then the solution was transferred to a tube (30 mL), after 20 mL toluene solvent was added exactly. The solution was separated to two equal parts. Each part was spiked 1 mL of 0.2 ng mL^{-1} internal standard solution. Toluene solvent was changed with n-hexane. The cleanup step was performed by multilayer column (length: 40 cm; inner diameter: 2 cm). The multilayer column was prepared in the following sequence: Na_2SO_4 (2 g), silica gel (0.9 g), silica gel 2% KOH (3 g), silica gel (0.9 g), silica gel 44% H_2SO_4 (4.5 g), silica gel 22% H_2SO_4 (6 g), silica gel (0.9 g), silica gel 10% AgNO_3 (3 g), silica gel (0.9 g), and Na_2SO_4 (6 g). The sample was introduced to the cleanup column, eluted with 150 mL of n-hexane and the eluate was evaporated to 2 mL. The target compounds were cleaned up on the active carbon column (Kanto Chemical Co. Inc., Japan). The first step (F1) was eluted by 40 mL of mixture of n-hexane and dichloromethane (v:v, 3:1). The second step (F2) for PCDD/Fs analysis was eluted by 60 mL toluene. A half

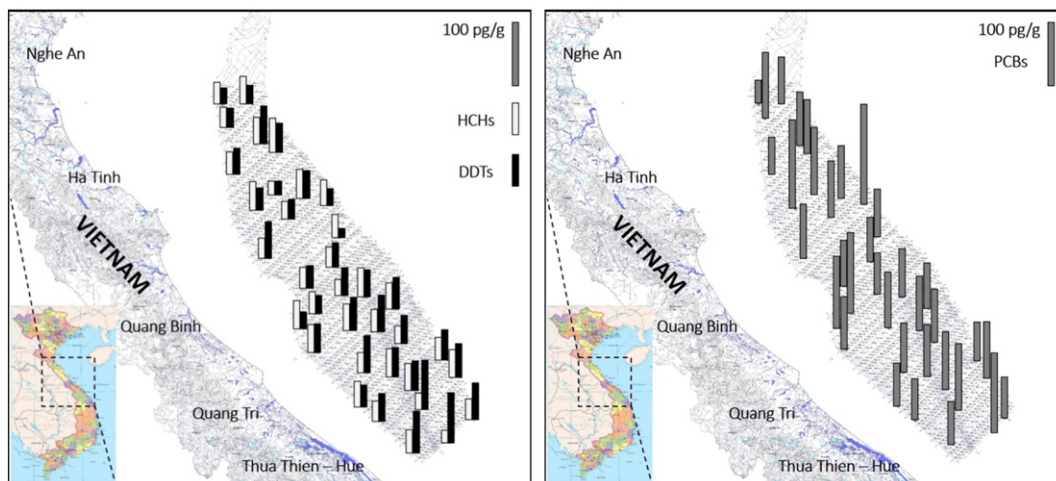


Fig. 1. Spatial distribution of the total of OCPs and PCBs in surface sediment collected from offshore waters of central, Vietnam (pg g^{-1} dry weight).

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