



Temporal and spatial changes in persistent organic pollutants in Vietnamese coastal waters detected from plastic resin pellets



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ABSTRACT

Plastic resin pellets collected at Minh Chau island and Ba Lat estuary between 2007 and 2014 in Vietnam were analyzed for dichloro-diphenyl-trichloroethanes (DDTs), polychlorinated biphenyls (PCBs) and hexachlorocyclohexanes (HCHs). The study was carried out as part of the International Pellet Watch program for monitoring the global distribution of persistent organic pollutants (POPs). Higher levels of DDTs compared to PCBs indicated agricultural inputs rather than industrial discharges in the region. Most POP concentrations on both beaches decreased over the period, with the exception of HCH isomers. Though the concentration of DDTs showed a drastic decline on both beaches between 2007/2008 and 2014, DDTs accounted for 60–80% of total DDTs, suggesting that there is still a fresh input of these chemicals in the region. This study strongly recommends further investigations to track temporal and spatial patterns of POP levels in the marine environment using plastic resin pellets.

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1. Introduction

Persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs) and organochlorine compounds (OCs), have been of great concern in recent decades because they are persistent and toxic, causing an array of adverse effects, including death, disease, and birth defects among humans and animals (Fry and Toone, 1981). Though not soluble in water after environmental release, POPs are readily absorbed into fatty tissue where concentrations can become significantly magnified to many times background levels (Bureau et al., 2004). They may also be sorbed onto plastic resin pellets at concentration factors of up to $\sim 10^6$ relative to ambient seawater (Mato et al., 2001). Plastic resin pellets are an industrial raw material with a disk-like or cylindrical shape, with diameters of < 5 mm; they are unintentionally released into the environment from the manufacturing and transport industries. Because of their environmental persistence and buoyancy, POPs may be sorbed onto their surface during environmental transport from the original source, and end up on beaches globally (Mato et al., 2001). The impacts of these materials on marine environments are widespread, and are succinctly reviewed by Derraik (2002). For example, many seabirds ingest

the pellets, mistaking them for prey, where they cause injury and inhibit the digestion of food (Derraik, 2002; Ryan et al., 2009); a further concern is the transfer of plastic-derived chemicals from ingested plastics to the tissues of birds (Tanaka et al., 2013). Consequently, POPs can bio-accumulate in marine organisms and tend to bio-magnify in top consumers such as predatory birds, mammals, and even humans through the food chain (Tanaka et al., 2013; Minh et al., 2002; Minh et al., 2004). Concentrations of pollutants in resin pellets were found to be consistent with those in mussels (Endo et al., 2005), suggesting a potential use of resin pellets for monitoring pollution in seawater. Thus, an increasing number of monitoring works have recently used plastic pellets as proxies for POP monitoring in marine environments (Zhang et al., 2015; Ryan et al., 2012; Hirai et al., 2011; Mato et al., 2001). International Pellet Watch (IPW) is a volunteer-based global monitoring program launched in 2005, concerned with the impact of contaminated resin pellets in the marine environment. The monitoring work of the IPW program led to the publication of the first global map based on such samples, showing strong regional patterns in concentrations of different pollutants (Ogata et al., 2009). Additionally, Ryan et al. (2012) demonstrated the potential of resin pellets for tracking temporal patterns in the abundance of POPs in marine environments. Zhang et al. (2015) suggested that pollutants from the pellets were likely to reflect the primary types of contaminants within the adjacent terrestrial environment, such as those arising from industrial development and

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agricultural activities. Thus, monitoring of temporal and spatial changes in the concentrations of these compounds is critically important for marine environmental risk assessment.

Vietnam is an agricultural country located across subtropical and tropical regions. The Red River Delta in the north is the one of two major agricultural production areas. Due to low costs and high insecticidal efficiency, large amounts of OCs have been applied in agriculture to increase crop yields (Nhan et al., 1998). Additionally, malaria became a serious problem during the late 1980s and early 1990s (Hung and Thiemann, 2002); over 90% of severe cases and deaths occurred in mountainous, forested and largely ethnic minority areas (Erhart et al., 2007). Hence, a huge quantity of insecticides has been widely sprayed for malaria vector control and agricultural purposes. Application of such chemicals in the environment can influence global pollution, as Vietnam is located at the center of the Southeast Asian region as well as in a high-temperature region. The use of dichloro-diphenyl-trichloroethane (DDT) was officially banned in Vietnam in 1995 (Sinh et al., 1999), and initial monitoring studies showed that POPs levels decreased after the ban (Toan et al., 2007; Nishina et al., 2010). However, recent studies have detected trace levels of POPs in the environment (Hoai et al., 2011), therefore, questions remain about whether DDTs are still locally in use and whether the ban has worked effectively over the last two decades. Although many approaches have been applied to assess POP pollution in Vietnam recently, earlier studies were limited in their spatial and temporal extent, particularly in the marine environment. While plastic resin pellets haven't been considered to be good proxies for POP monitoring in the marine environment (Endo et al., 2005; Ogata et al., 2009), no such work has been undertaken in Vietnam thus far. Thus, the objective of this study is to track temporal and spatial changes in DDT, PCB and hexachlorocyclohexane (HCH) concentrations in Vietnamese coastal regions.

2. Materials and method

Following a call for pellets by IPW (Takada, 2006), beached plastic resin pellets were collected at two beaches: Minh Chau Island and Balat estuary (downstream area of the Red River Delta), in North Vietnam (Fig. 1). Minh Chau is a remote island located in Bai Tu Long Bay; there are no agricultural or industrial activities on the island, whereas Ba Lat estuary is downstream of the Red River Delta, an area of paramount agricultural and economic importance in North Vietnam. Samples were collected, from the high-tide line of the sandy beaches, using soap-rinsed fingers. Around 80–100 pellets were collected from

each beach, in the interval between 2007 and 2014, to track temporal changes in POPs in the nearly 20 years since the ban. The pellets then were wrapped in aluminum foil, put into paper envelopes and sent to the Laboratory of Organic Geochemistry (LOG) at the Tokyo University of Agriculture and Technology via air mail for chemical analysis.

Chemical analysis followed IPW protocols as described in detail by Ogata et al. (2009). POPs were extracted from pellets by soaking in hexane. PCBs were quantified by comparing the integrated peak area of the quantification ion with that of the injection internal standard ($m/z = 186 < 256, 288/290 < 360, 427/429/431 < 464$) as derived from calibration lines drawn for individual chlorinated biphenyls (CBs) using standard solutions for calibration (2.5, 5, 10, 20 and 40 ppb; Wellington Laboratories). All calibration lines for each CB showed high linearity ($r^2 > 0.99$). The sum of all congeners quantified (i.e., CB#66, 101, 110, 149, 118, 105, 153, 138, 128, 187, 180, 170 and 206) is expressed as P13 PCBs in this study. DDT and dichlorodiphenyldichloroethane (DDD) and four HCH isomers (α, β, γ and δ) were determined using an HP 1 electron capture detector fitted with an HP 7890 gas chromatograph (GC-ECD), and quantified by comparing the integrated height of the peaks of standard solutions for calibration (DDT, DDD and four HCH isomers; 20 ppb each).

Recovery was tested by spiking the aliquots of the extracts with authentic standards; recoveries were $>95\%$. A procedural blank was run with every set analyzed (five pools). Analytical values <3 times the corresponding blank were considered to be below the limit of quantification (LOQ). The smallest LOQs were 0.07 ng/g for P13 PCBs, 0.1 ng/g for DDT, 0.04 ng/g for DDE, 0.07 ng/g for DDD, and 0.4 ng/g for HCHs.

3. Results

All median concentrations of POPs detected in pellets are presented in Table 1. Among the target organochlorine pesticides measured, DDT compounds were the predominant contaminant, with concentrations ranging from 12.3 to 558 ng/g-pellet; PCBs were in the concentration range of 4.0 to 24.0 ng/g-pellet. HCHs were present at relatively low concentrations ranging from 0.44 to 1.44 ng/g-pellet. The spatial distribution of POPs in the pellets indicated that the concentrations of POPs in Ba Lat were higher than those in Minh Chau, except for PCBs in 2014. These results suggest that contamination is closely related to human activities. Furthermore, temporal data indicates that the median concentrations of pellets in both beaches decreased from 2007 to 2014, except for the isomers of HCH. The HCHs varied differently between the beaches; α -HCH tended to increase between Minh Chau and Ba



Fig. 1. Location of the two sampling sites in North Vietnam (A) Minh Chau and (B) Ba lat.

Table 1

Median concentrations (ng/g-pellet) of PCBs, DDTs, HCHs in pellet from Mich Chau and Ba Lat beaches.

POPs	Minh Chau beach		Balat beach		Heskett et al. (2012) ^a
	2007	2014	2008	2014	
PCBs ^b	13.0	8.0	24.0	4.0	<10
DDTs	DDT	132	7.8	357.0	10.0
	DDD	25.30	3.41	186.7	3.52
	DDE	5.76	1.20	14.40	0.50
	Total	163	12.3	558	14.1
HCHs	α	0.01	0.05	0.21	0.79
	β	0.39	0.20	0.82	0.13
	γ	0.15	0.08	0.34	0.10
	δ	0.68	0.11	0.07	0.21
	Total	1.23	0.44	1.44	1.23

The significance of bold is to highlight the total concentration of each pollutant.

^a Global background levels in pellets.

^b Σ 13PCBs = sum of 13 PCB congeners (CB-66, 101, 110, 149, 118, 105, 153, 138, 128, 187, 180, 170, 206).

Lat beaches, while δ -HCH showed the opposite trend (Fig. 2). Though DDTs showed a drastic decline on both beaches from high concentrations in 2007 and 2008 to similar low concentrations in 2014, DDT accounted for 60–80% of total DDTs, suggesting that there is still a fresh input of these species in the region (Fig. 3).

4. Discussion

Relatively high levels of DDTs were found in pellets at both beaches in 2007 and 2008, although the DDT usage ban became effective in Vietnam in 1995. The high levels of DDTs from Ba Lat could be explained by the absorption of these compounds from the surrounding environment, as the Ba Lat estuary is downstream of intensive agricultural activities along the Red River Delta, which was a significant emission source of DDT pollutants throughout the 1970s–1990s (Nhan et al., 1998; Hong et al., 2008). A number of studies have shown high DDT residues in sediments (Hong et al., 2008), migrant birds (Minh et al., 2002) and even human breast milk (Minh et al., 2004) in North Vietnam. Unlike Ba Lat, Minh Chau Island is a remote marine area and the beach faces toward open sea. There were no agricultural activities on the island. Limited information on DDT residues in biota and abiota are available for this island, although small amounts of DDTs may have been used for mosquito control from time to time. DDT concentrations in Minh Chau

were found to be much lower than those in Ba Lat, but still higher than those in other countries (Ogata et al., 2009); Minh Chau island is located near the Chinese border area, where relatively high POP levels have been found (Minh et al., 2008). Cross-boundary transport of POPs by coastal currents has been blamed for such high concentrations in coastal border areas in North Vietnam (Minh et al., 2008). Thus, the high concentrations of DDTs in pellets from Minh Chau Island might be due to contamination from land sources during long-range transport to the island, rather than the Minh Chau ambient environment.

On the other hand, the slow degradation, evaporation and burial in sediments of DDTs is a reason for the detection of high levels of compounds in pellets, even given limited DDT input. This finding was consistent with Hung and Thiemann (2002) and Minh et al. (2002), who reported that DDTs were still detected in river surface water and marine organisms; however, DDT concentrations showed a decreasing trend and were below the allowable limit in surface waters (Toan et al., 2007). Later investigations showed that no recent DDT and HCH inputs were found in the environment a decade after the ban was issued (Toan et al., 2007; Nishina et al., 2010). These results were somewhat consistent with the uniformly drastic decline of compounds in pellets from both beaches in 2014, suggesting that the ban has been moderately effective in reducing the environmental burden of these compounds. Beside the ban, the Vietnamese government has worked to reduce and gradually stop reliance on DDT usage for agricultural purposes through national control programs and campaigns (Hoai et al., 2011). Among these, encouragement and guidance to use alternative organic pesticides was one long-term effective program (Berg, 2001). The alternative pesticides included validamycin, propiconazole and hexaconazole; insecticides included fenobucarb, cartap and lambda-cyhalothrin; herbicides were fenoxaprop-P-ethyl, 2,4D, pretilachlor (Berg, 2001). Moreover, development of integrated rice-fish farming also had a significant impact in assisting farmers to use fewer pesticides and gain higher yield (Berg, 2001). Other campaigns widely applied throughout the country, such as '3 Reductions, 3 Gains', '1 Must Do, 5 Reductions, and 4 Rights' also achieved initial successes in reducing DDT usage (Hoai et al., 2011).

Although decreasing drastically over the period, concentrations of DDT and its metabolites, DDD and dichlorodiphenyldichloroethylene (DDE), found in pellets from beaches in 2014 were still twofold or threefold higher than global background pollution levels (Heskett et al., 2012). Moreover, DDT showed the highest median concentration (accounting for 60–80% of total DDTs), followed by DDD (18–20%) and DDE (<10%) (Fig. 2), which indicated that there is still a current input

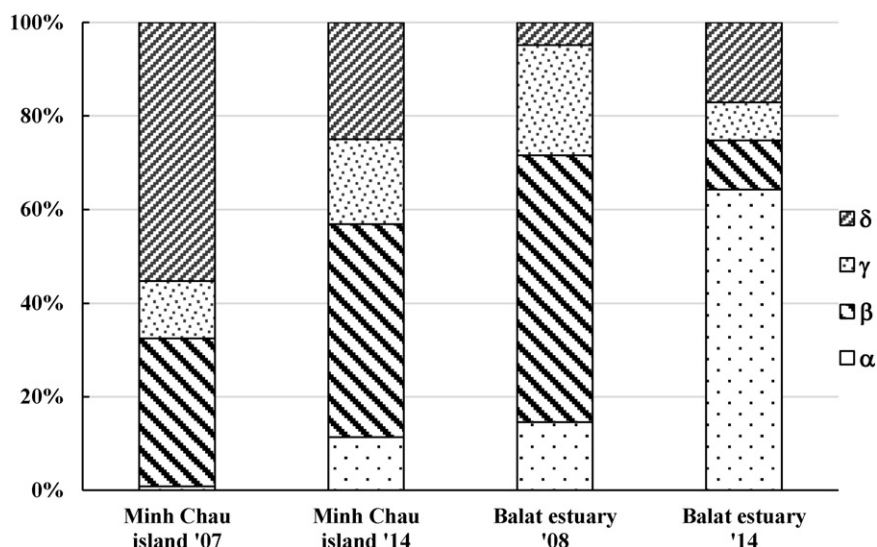


Fig. 2. Relative composition of HCHs in pellets from Minh Chau and Ba lat.

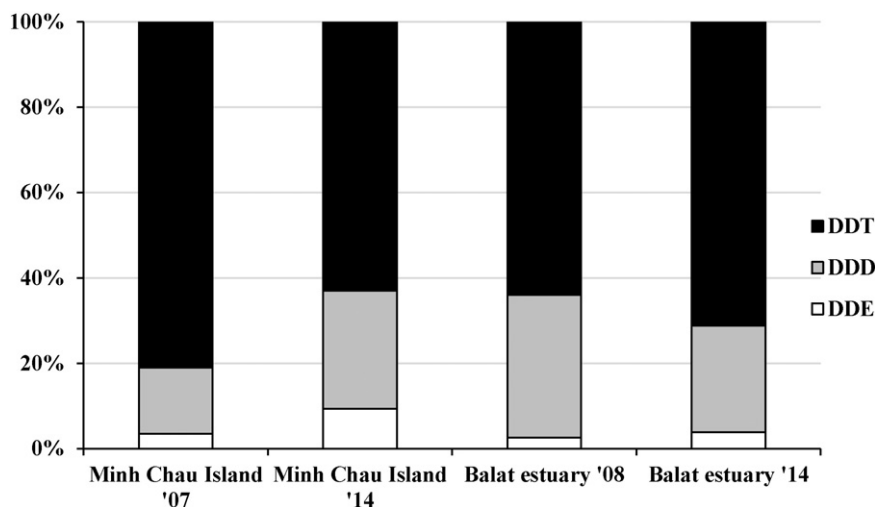


Fig. 3. Relative composition of DDTs in pellets from Minh Chau and Ba lat.

of DDT in the region. This finding may possibly be attributed to illegal use and malaria control. According to Hoai et al. (2011), DDTs and other pesticides, such as fenobucarb, trichlorfon, cyfluthrin, and cypermethrin, were concomitantly found at relatively high levels in soils, vegetables and fish nearby paddy fields in the Hanoi region, where there is a wide plain along the midstream zone of the Red River Delta. Additionally, despite these successes, malaria control remained a high priority for some areas in Vietnam. Recently, an increasing incidence of malaria in Hanoi and adjacent areas, including tens of infection cases and several deaths, has been attributed to resumed use of DDTs in the environment for malaria vector control (Manh et al., 2011). Thus, the runoff of chemicals from spraying areas could reach the Balat estuary during rainy seasons.

In Vietnam, the strategy for economic growth has shifted gradually from agriculture to industry, since a range of political and economic reforms were (Doi Moi) launched in 1986. Rapid industrialization has contributed to increasing PCB emissions in urban areas recently (Minh et al., 2008). The sources of these compounds have thus much received attention in several studies (Minh et al., 2002, 2004, Hoai et al., 2010, 2011). PCBs were found at high concentrations in rivers around urban or suburban areas where industrial services were concentrated, such as Hanoi, Haiphong and Quangninh (Hong et al., 2008; Hoai et al., 2010). In particular, high levels of PCBs in sediments were detected, up to 384 ng/g in midstream zones along tributaries of the Red River, such as the Nhue, To Lich, and Lu rivers (Hoai et al., 2010). However, concentrations of PCBs showed an apparent decreasing trend from the city canals toward the river and coastal areas (Minh et al., 2008). Residues in sediments collected from downstream and coastal areas were much lower than those in sediments from the original sources in urban areas (Minh et al., 2008). In fact, Minh Chau island and Ba Lat estuary were not industrial areas, and the levels of PCBs in these sites were slightly higher than global background pollution levels during 2007 and 2008, respectively. These levels were much lower than those reported in developed countries (Ogata et al., 2009) and China, where heavy industrial activity was present for decades (Zhang et al., 2015). Furthermore, judging by Hong et al. (2008), the sources of PCBs in sediment from Ba Lat were much lower compared to industrial areas along the north coast of Vietnam, and much lower than those reported from industrialized temperate regions such as the US, the Mediterranean, and South Korea (Hong et al., 2008). Furthermore, in this study, a 40–80% decline was found over the period, which was comparable to the global background levels in pellets (Heskett et al., 2012), suggesting no or limited input of PCBs in the region. This could relate to significant efforts by the Vietnam government to reduce the emission of PCBs into the environment through the PCBs Management Project, launched in 2010.

The project originated from the National Implementation Plan for the Stockholm Convention, ratified by the Prime Minister in 2006. The critical objective was to eliminate the usage of PCBs in equipment and facilities in 2020 and safely dispose of PCBs by 2028 (PCB-WB project, 2010).

Unlike PCBs and DDTs, HCHs are much less persistent in the environment (Hong et al., 2008). In this study, the low levels of HCHs found in pellets were similar to those found in many other locations around the world (Ogata et al., 2009) and the global background in remote oceanic islands (Heskett et al., 2012). Moreover, the decreasing trend found for most HCHs was best explained by decreasing usage of this pesticide over the last few decades in Vietnam. The decreasing trends also reflected the worldwide ban on the usage of HCH pesticides and the decreased retention of HCHs close to their sources due to rapid evaporation of HCH isomers after use in tropical regions with high temperatures (Tuduri et al., 2006). Ogata et al. (2009) identified Lindane, a pesticide that contains mainly γ -HCH, as the most likely source of this material in many locations in the world. γ -HCH (Lindane) was the one of most common and extensively used insecticides in the Red River Delta during the 1980s and 1990s. However, the decreasing trend in the γ - isomer of HCH in this study was consistent with a report on the levels of this compound in water and sediment by Minh et al. (2008), which suggested no input of this insecticide into the Red River watershed. Unlike the trend for the other HCH compounds, the α -HCH isomer tended to increase in the Ba Lat estuary in this study (Fig. 3). Although the α -isomer was the most volatile of the HCH isomers in this subtropical region (ATSDR, 2005), the increase in α -HCH meant that a technical HCH has been most recently used in the region.

In general, concentrations of three POPs analyzed from the pellets have decreased at both sites sampled, suggesting that concentrations in Vietnamese coastal waters have decreased over the last decade. However, a small fresh input of DDT and an increasing α -HCH isomer in pellets are required to continuously monitor changes in environmental burdens of these compounds across the Vietnamese coast. These results also strongly support the value of global monitoring of POPs using beached plastic resin pellets, as called for by the IPW.

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