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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.

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 (Burreau et al., 2004). They may also be sorbed onto plastic resin pellets at concentration factors of up to ~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 (r2 > 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, χ and δ) were determined using an HP l 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

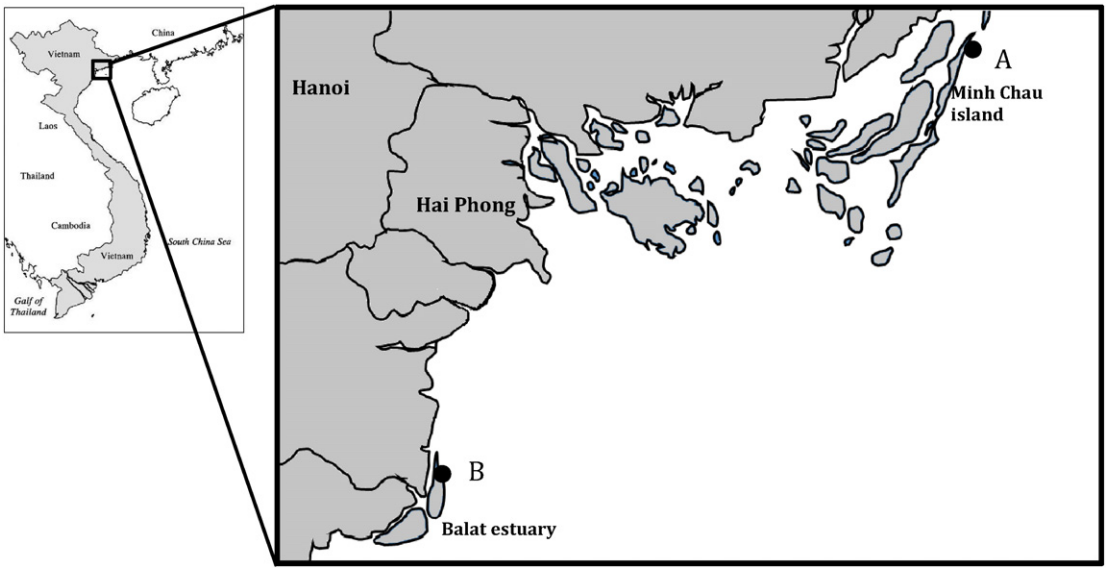

Fig. 1. Location of the two sampling sites in North Vietnam (A) Minh Chau and (B) Ba lat.