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Profiles of environmental contaminants in hawksbill turtle egg yolks reflect local to distant pollution sources among nesting beaches in the Yucatán Peninsula, Mexico

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ABSTRACT

Knowledge of spatial variation in pollutant profiles among sea turtle nesting locations is limited. This poses challenges in identifying processes shaping this variability and sets constraints to the conservation management of sea turtles and their use as biomonitoring tools for environmental pollutants. We aimed to increase understanding of the spatial variation in polycyclic aromatic hydrocarbon (PAH), organochlorine pesticide (OCP) and polychlorinated biphenyl (PCB) compounds among nesting beaches. We link the spatial variation to turtle migration patterns and the persistence of these pollutants. Specifically, using gas chromatography, we confirmed maternal transfer of a large number of compounds (n = 68 out of 69) among 104 eggs collected from 21 nests across three nesting beaches within the Yucatán Peninsula, one of the world's most important rookeries for hawksbill turtles (*Eretmochelys imbricata*). High variation in PAH profiles was observed among beaches, using multivariate correspondence analysis and univariate Peto-Prentice tests, reflecting local acquisition during recent migration movements. Diagnostic PAH ratios reflected petrogenic origins in Celestún, the beach closest to petroleum industries in the Gulf of Mexico. By contrast, pollution profiles of OCPs and PCBs showed high similarity among beaches, reflecting the long-term accumulation of these pollutants at regional scales. Therefore, spatial planning of protected areas and the use of turtle eggs in biomonitoring needs to account for the spatial variation in pollution profiles among nesting beaches.

1. Introduction

Global anthropogenic pollution of the marine environment by organic contaminants such as persistent organic pollutants (POPs) and polycyclic aromatic hydrocarbons (PAHs) is an issue of great concern. Due to their widespread use and capacity for long-range transport, POPs are ubiquitous in the environment and have infiltrated aquatic systems worldwide (D'ilio et al., 2011). Polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbons (PAHs) can accumulate within tissues, organs and fluids of all of the 7 main sea turtle species: loggerhead (*Caretta caretta*), green (*Chelonia mydas*), leatherback (*Dermochelys coriacea*), hawksbill (*Eremochelys imbricata*), flatback (*Natator depressus*), kemp's ridley (*Lepidochelys kempi*), and olive ridley (*Lepidochelys olivacea*) turtles (D'ilio et al., 2011; Camacho et al., 2013a, 2014; Gardner et al., 2003; Keller et al., 2013; Ylitalo et al., 2017). Chronic accumulation of these pollutants in sea turtles has been linked to health problems such as immune, endocrine and reproductive disruption (Aguirre et al., 1994; Camacho et al., 2013b; Keller et al., 2004a). Consequently, minimizing exposure of sea turtles to POPs and PAHs is of critical importance for their conservation.

The transfer of POPs and PAHs from mothers to their offspring is of particular concern for sea turtle conservation as it can affect hatchling success and supply eggs with pollutants even before offspring come into contact with the environment. PCBs, OCPs and PAHs can concentrate in adipose tissues and transfer from female sea turtles to their offspring via egg yolk (Corsolini et al., 2000; Alava et al., 2006; Guirlet et al., 2010; van de Merwe et al., 2010). Significant positive correlations between the concentration of ΣPCBs, p,p'-DDE, ΣPBDEs and ΣChlordanes in the blood of leatherback turtles and their eggs have been observed (Stewart et al., 2011). Likewise, for green turtle, significant correlations were detected for ΣPCBs, ΣPBDEs, γ-hexachlorocyclohexane, *trans*-chlordane and mirex between maternal blood and eggs, between eggs and hatchling blood and between maternal and hatchling blood (van de Merwe et al., 2010). Lipids stored in the egg yolk provide a vital energy supply during egg development and during the "Lost Year" (the period

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between hatching and arrival of juveniles at their foraging grounds, in which migratory movements are vaguely known, Hamann et al., 2002; Miller et al., 1985). PCB and OCP contamination in freshwater snapping turtles (*Chelydra sepertina*) increased embryonic mortality and deformities (Bishop et al., 1994; de Solla et al., 2008). Similarly, increasing POP concentrations in green turtle eggs were negatively correlated with hatchling mass:length ratio (van de Merwe et al., 2010). POPs and PAHs present in the yolk can thus affect the survival and development of eggs and hatchlings (Keller et al., 2013; Alava et al., 2006, 2011).

The implementation of conservation efforts to protect nesting beaches against PCB, OCP and PAH pollution is complicated as a diversity of local to global environmental factors (i.e. oceanic currents, atmospheric circulation, temperature, precipitation patterns, salinity and organic matter content) and biological factors of sea turtles (e.g. lipid content of tissues, body condition, trophic status, age and sex) influence the uptake and transport of PCBs, OCPs and PAHs (Keller et al., 2013). Feeding presents one of the main uptake routes of pollutants in sea turtles (van de Merwe et al., 2010; McKenzie et al., 1999). However, sea turtles can undertake long migrations and display a diversity of feeding preferences throughout their life (Jones and Seminoff, 2013). Hence, concentrations of contaminants can, to a large extent, be influenced by the contamination at distant foraging locations (Alava et al., 2011).

Mexican coastal and territorial waters provide reproductive nesting beaches and foraging areas for all 7 species of sea turtles. Particularly for hawksbill turtles, beaches along the Yucatán Peninsula provide one of the biggest nesting areas in the Caribbean and Gulf of Mexico and among the most important in the world (Gorham et al., 2014; Pérez-Castañeda et al., 2007; Wood et al., 2012). Hawksbill turtles are critically endangered worldwide since 1996 (Mortimer and Donnelly, 2008). They are highly valuable because of their long evolutionary history, their historical and cultural interaction with indigenous cultures (Frazier et al., 2005), their socio-economic importance for tourism, education and research and their ecological role in coastal and marine ecosystems (Frazier et al., 1993). Hawksbill nesting populations in the Yucatán Peninsula have been monitored since at least 1977 (Garduño-Andrade, 2000). However, baseline knowledge regarding pollution levels in hawksbill turtles has not been monitored in this area. Moreover, knowledge of the maternal transfer of POPs and PAHs and the variability in their concentrations and composition within nesting populations is limited for hawksbill turtles worldwide.

Few studies have reported POP concentrations and profiles in tissues of hawksbill turtles, often with limited sample replication (Table 1). Moreover, POPs in eggs of hawksbills are particularly poorly studied and often only focus on specific compounds. Gaining additional information from comparable studies is complex as different geographic locations, life stages, sexes and tissues are investigated for different compound mixtures and with varying measurement units (such as dry weight, wet weight, whole egg content or yolk contents only). Nonetheless, current research suggests that hawksbill turtles have a high potential to bioaccumulate POPs given their trophic level (as spongivorous) relative to other sea turtles species at the same location (Malarvannan et al., 2011; García-Besné et al., 2015; Hermanussen et al., 2008; Keller et al., 2012). The trophic level of hawksbills is under debate as knowledge on their diet is scare and a number of studies have hinted at other items contributing to their diet as well as sponges (Jones and Seminoff, 2013). In contrast to the suggested high bioaccumulation potential relative to trophic level, hawksbill turtles contained lower levels of PCBs and OCPs in blood compared to green turtles on Cape Verde (Camacho et al., 2014). However, the composition of individual compounds within each of the POP groups differed among species, and individuals from the different species came from different foraging locations which could have influenced their POP compositions (Camacho et al., 2014). Similarly, PCB levels were lower in egg albumen from hawksbill than green turtles on Guadaloupe Island, albeit again the composition of individual compounds differed (Dyc et al., 2015).

The overall paucity of data on PAHs in hawksbills is a critical gap in current understanding regarding the potential risk that these compounds may contribute to this species (Table 1). Camacho et al. (2014) reported higher ΣPAHs in green than in hawksbill blood plasma samples, with di- and tri-cyclic PAHs (suggesting petrogenic origins) predominant in both species. However, results are hard to generalize based on this one study. Moreover, studies on PAH contaminations that focus on eggs of hawksbills are lacking. PAHs can be released to the marine environment from a variety of sources, such as industry discharges, urban runoff, vessel operation, tanker accidents, atmospheric deposition and oil exploration and production (Van Metre et al., 2000). Although some PAHs with high octanol-water coefficient (Kow, Camacho et al., 2012) are lipophilic compounds, previous studies have shown that PAHs do not biomagnify in teleost fish and other vertebrate species (Mackay et al., 1998; Murawski et al., 2014; Ylitalo et al., 2017). This is because these animals can rapidly metabolize PAHs to more polar compounds that are then secreted into bile for elimination (Gray, 2002; da Silva et al., 2006; Beyer et al., 2010; Roscales et al., 2011). Hence, PAHs measured in tissues of sea turtles reflect recent, local contamination. In contrast, OCPs and PCBs have been shown to accumulate over longer periods and represent contaminants from larger geographical scales in sea turtles (Alava et al., 2011; Keller et al., 2004b). PAHs can be found in moderate levels in benthic invertebrates and marine sediments of coastal areas close to urbanization and industries (Soclo et al., 2000). This raises the question about the potential exposure of nesting sea turtles to PAHs when they are close to the coastline.

To support conservation management of hawksbill turtles at their nesting beaches, this study aimed to increase understanding of the presence and spatial variation in PAH, OCP and PCB pollution within the Yucatán Peninsula. Specifically, for PAHs, OCPs and PCBs, we aimed to: a) establish a high replication baseline of contamination levels across 3 distinct nesting beaches in this area, b) establish maternal transfer of individual compounds as evidenced by their presence in freshly laid eggs and c) interpret variations in pollution profiles among beaches relative to the persistence of pollutants in the environment and the migration of turtles.

Table 1
Peer reviewed studies on polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), perfluoroalkyl compounds (PFCs) and polybrominated diphenyl ethers (PBDEs) in hawksbill turtle tissues.

Life stage	PAHs	OCPs	PCBs	PFCs	PBDEs	Sampled tissue	Sample size	Location	Ref.
Juveniles		X	X		X	Blood	3	Ishigaki Islands, Japan	Malarvannan et al., 2011
Juveniles				X		Blood	5	Florida, US	Keller et al., 2012
Juveniles	X	X	X			Blood	13	Cape Verde	Camacho et al., 2014
Juvenile					X	Blood, fat, muscle, liver	1	Queensland, Australia	Hermanussen et al., 2008
Nesting females		X	X			Dermis	3	Guadaloupe, French West Indies	Dyc et al., 2015
Eggs		X	X		X	Egg yolk	1	Guadaloupe, French West Indies	Dyc et al., 2015
Nesting females		X	X			Blood	24	Campeche, Mexico	García-Besné et al., 2015
Eggs		X	X			Total egg content	28	Campeche, Mexico	García-Besné et al., 2015
Nesting females		X				Blood	30	Campeche, Mexico	Tremblay et al., 2017