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Oil spill off the coast of Guimaras Island, Philippines: Distributions and changes of polycyclic aromatic hydrocarbons in shellfish

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ABSTRACT

The sinking of the *Solar 1* tanker caused serious heavy oil pollution around Guimaras Island, Philippines. In the present study, variations of parent polycyclic aromatic hydrocarbons (PAHs) and alkylated PAHs (alkPAHs) in some shellfish were investigated around Guimaras Island and other small islands from 3 months to 5 years after the spill. The total PAHs and alkPAHs in shellfish were detected in high concentrations at 448 and 33,666 ng/g dry weight, respectively, in November 2006. The concentrations of alkPAHs gradually decreased, while the parent PAHs in shellfish degraded more slowly than the alkPAHs, which was likely due to the persistent characteristics of PAHs. The risks based on European Union regulations were insignificant in 2008, but total PAHs in shellfish were still over 8 times higher at the investigated sites in November 2011 than that before the oil spill.

1. Introduction

On August 11, 2006, the *Solar 1* tanker was hit by a hard rainstorm and sank off the coast of Guimaras Island in the Philippines. About 2100 tons of heavy oil spilled from the tanker polluting about 200 km of shoreline, from Guimaras Island to Panay Island and Negros Island. A large extent of shoreline in Guimaras experienced particularly bad pollution, and the coastal fisheries were seriously damaged. Over 450 ha of protected mangroves with complex topography at Guimaras were also contaminated. In tropical areas, because of large amounts of sunshine and high temperatures, spilled oil is expected to degrade more rapidly than in cold areas, although it still is poorly studied.

Polycyclic aromatic hydrocarbons (PAHs) in the heavy oil are persistent in the environment (Michel and Zengel, 1998; Wetzel and van Vleet, 2003). Alkylated PAHs (alkPAHs) (generally with 1 to 4 methyl groups) are present in heavy oil along with the parent PAHs. The parent PAHs are often the emphasized contaminants, because some PAHs have potential mutagenic and carcinogenic properties (ex. Schneider et al., 2002; Jeffy et al., 2002) and other toxic properties. Our group also reported that the effects and accumulation potencies of PAHs in aquatic organisms have been reported previously (Koyama and Kakuno, 2004; Cheikyula et al., 2008; Pal et al., 2011; Kokushi et al., 2012; Song et al., 2012). On the other hand, studies on the toxicities of alkPAHs in aquatic organisms have been limited (Rhodes et al., 2005; Vondráček et al., 2007).

Although spilled oil degradation is governed by several factors such as evaporation, diffusion, photooxidation, and biodegradation, the degradation processes are very slow (Díez et al., 2007; Taylor and Reimer, 2008). However, in tropical areas, because of large amounts of sunshine, high temperatures, and frequent storms, spilled oil is expected to degrade faster than that in temperate and cold areas. Large scale oil spills have previously occurred, for instance during the war in the Persian Gulf (1991), the *Evikos-Orapin* in Singapore Straits (1997), and the *Jessica* at San Cristóbal, Galapagos Island (2001). There are only a few for which detailed investigations and studies of oil pollution were conducted.

In a previous study, we reported the concentrations of parent and alkylated PAHs in aquatic organisms 1 month after the oil spill (Uno et al., 2010). At that time, total PAHs (ΣPAHs) in shellfish were 38.0–3102 ng/g dry weight (DW) in a part of Guimaras Island, while those in fish were 11.9–52.3 ng/g DW. The pollution of *Solar 1* oil spill was very serious, and needed wide and continuous monitoring. Fortunately, we had investigated the PAH residues in bivalves

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collected from Panay Island, which is close to Guimaras, before the oil spill. The ΣPAHs was 2.9–4.2 ng/g wet weight at that time (Uno et al., 2007).

In the present study, we investigated the residue distributions and interannual variations for PAHs and alkPAHs in some species of shellfish collected at Guimaras Island and nearby small islands between 3 months and 5 years after the *Solar 1* oil spill. We additionally assessed the pollution levels at Guimaras Island 5 years after the oil spill by comparing the PAH concentrations found in bivalves at that time to levels observed before the accident.

2. Materials and methods

2.1. Collection of organisms

Samplings were carried out at 13 sites in November 2006 (Fig. 1 and Table 1). Then, following investigations were conducted only at selected sites (St. 1, 2, 3, 7, 8 and 9 in March; St. 2, 7, 8, and 9 in September 2007; St. 2, 7, and 9 in June 2008; and St. 2 and 9 in November 2011). The choices of sites were decided based on the last investigation, and, as a result, we performed the sampling at the sites with higher concentrations of parent and alkylated PAHs. *Modiolus* sp. (Biv-1), *Pinctada* sp. (Biv-2), *Crassostrea* sp. (Oyster) and *Clypeomorus* sp. (Snail) were collected from each sampling site. Biv-1 lives in widely ranging habitats, while Biv-2, Oyster, and Snail live in mangrove forests. After collection, all samples were stored at −20 °C until analysis was performed.

Table 1
Locations collected samples along Guimaras coasts.

Site no.	Latitude	Longitude
1	10.477	122.477
2	10.487	122.484
3	10.464	122.485
4	10.450	122.509
5	10.438	122.506
6	10.413	122.507
7	10.404	122.513
8	10.408	122.512
9	10.408	122.514
10	10.405	122.554
11	10.388	122.609
12	10.436	122.626
13	10.428	122.667

2.2. Extraction and measurement of parent and alkylated PAHs in shellfish

Analysis for parent and alkylated PAHs in shellfish were performed according to Uno et al. (2010). Shellfish samples of about 5 g were composed from multiple individuals in a species, and freeze-dried. Samples were ultrasonically extracted with dichloromethane-hexane mixture (1:1, v/v) from freeze-dried soft tissues of shellfish with about 2 g/wet weight. The extracts were saponified with ethanol added 1 M potassium hydroxide at 90 °C for 1 h. After saponification, target chemicals in the solution of potassium hydrate-ethanol were extracted by dichloromethane-hexane (1:3, v/v), cleaned with a silica-gel column (packed in a

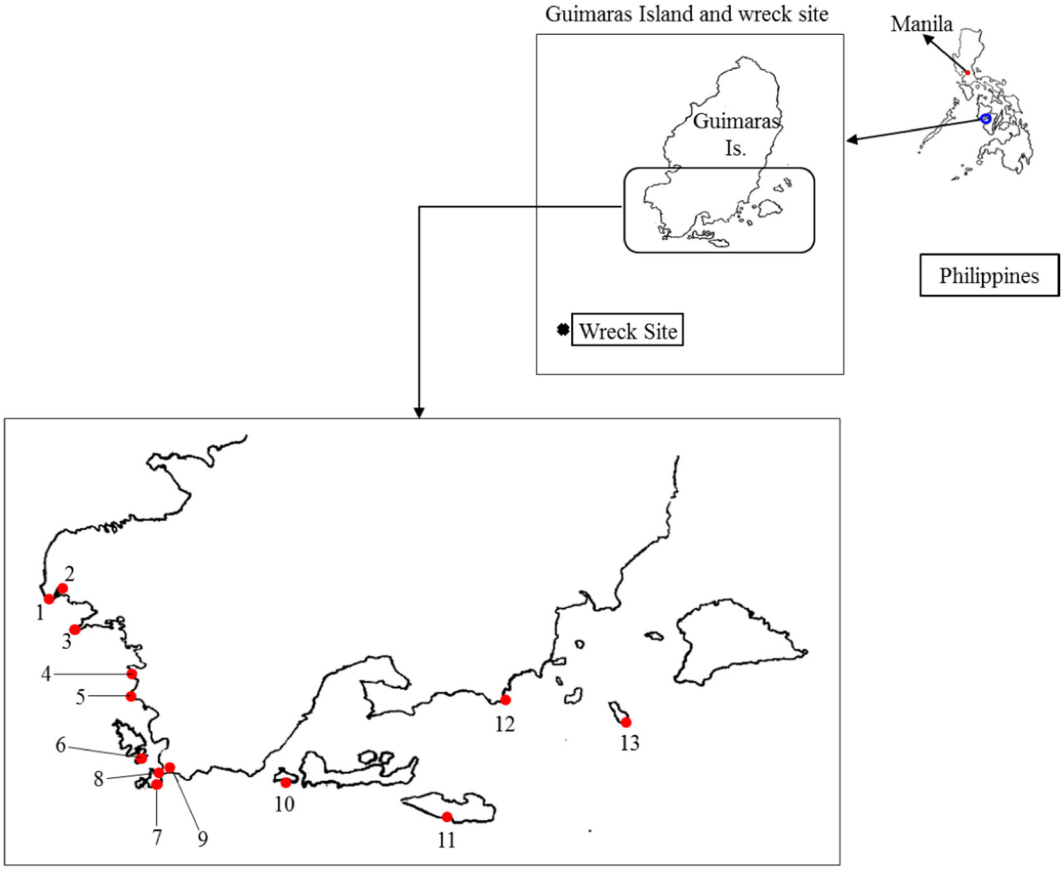

Fig. 1. Locations of the sampling sites where aquatic organisms were collected.