Marine Pollution Bulletin 128 (2018) 115–125

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Distribution of hydrocarbons in seabed sediments derived from tsunami-spilled oil in Kesennuma Bay, Japan

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ARTICLE INFO

Keywords:
Sediment
N-hexane extracts (NHE)
Polycyclic aromatic hydrocarbons (PAH)
Petroleum hydrocarbons
Tsunami

ABSTRACT

As a result of the 2011 Great East Japan earthquake, about 11,000 kL of fuel oil was spilled into Kesennuma Bay, Miyagi Prefecture. This oil either accumulated in seabed sediments or was burned in a marine fire on the sea surface. We investigated spatial and temporal variations in the concentrations of oil and hydrocarbons. The maximum concentrations of n-hexane extract (NHE), total petroleum hydrocarbon (TPH), and polycyclic aromatic hydrocarbons (PAHs) in the sediments in 2012 were 8000, 1200, and 16.2 mg/kg (DW), respectively. NHE and TPH concentrations were high in the inner and west bay, and PAHs concentrations were high in the east bay. NHE and PAH concentrations didn't change; however, TPH concentrations decreased significantly with time. The total NHE in the sediment across the whole bay was estimated at 1685 kL and there was still about 13% of the original amount in the sediment in December 2014.

1. Introduction

As a result of the huge earthquake and tsunami that occurred in East Japan on 11 March 2011, 22 fuel oil tanks on the coast of Kesennuma Bay, Miyagi, were destroyed. Fritz et al. (2012) reported the maximum tsunami height in the Kesennuma Bay narrows reaches 9 m, and maximum tsunami outflow currents of 11 m/s were measured.

About 11,000 kL of marine fuel oil was spilled into the sea (Yamamoto et al., 2012; Sakai, 2013; Yokoyama et al., 2014). Sakai (2012) reported that most of the spilled oil was flushed to the outside of the bay by waves; however, some was also mixed with sand and suspended mud particles and was subsequently deposited on the seabed. In addition, a marine fire occurred on the sea surface, producing polycyclic aromatic hydrocarbons (PAHs) that adhered to the suspended particles and accumulated on the seabed. The seabed sediments in Kesennuma Bay have been extensively and seriously polluted by this spilled oil (Sakai, 2012). While the straight-chain hydrocarbon components of the heavy oil decompose easily, PAHs persist for long periods of time (Mori and Sotoyama, 1994; Reddy et al., 2002). Furthermore, some of these PAHs may have effects on health; for example, benzo(*a*)pyrene has carcinogenic properties (Blumer, 1971), and USDOH ATSDR (1995) reported toxicological profile for PAHs in detail. Ohwada (1998) reported that sediments become anaerobic in the presence of oil with consequences for benthos. It is also known that PAHs from sediments persist in shellfish (Blumer et al., 1970).

Aquaculture, such as seaweed, oysters, and scallop culturing, was a thriving activity in Kesennuma Bay before the earthquake. The fish catches in 2011 and 2013 were 29% and 66%, respectively, less than the fish catch in 2010 before the tsunami (Kesennuma City Fish Market, 2015). In this study, we investigated the distribution and temporal changes in the concentrations of the oil deposited on the seabed of Kesennuma Bay because of the tsunami, by examining the concentrations of n-hexane extract (NHE), total petroleum hydrocarbons (TPHs), and PAHs. We also (1) explored the drivers for changes in the concentrations of these compounds, (2) estimated the total amount of oil deposited on the seabed of Kesennuma Bay, and (3) examined the effects of oil on marine benthic organisms.

2. Materials and methods

2.1. Sites description and sediments sampling

Sediments in Kesennuma Bay, Miyagi, Japan, were sampled eight times in February, March, and December of 2012, February, July, and November of 2013, and July and December 2014 at the sampling station shown in Fig. 1. Sampling was carried out at stations 1–20 from February 2012 to November 2013, and also in December 2014. An additional station 40, close to where the fuel tank sank into the seabed,

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https://doi.org/10.1016/j.marpolbul.2017.12.018
Received 6 November 2016; Received in revised form 24 November 2017; Accepted 6 December 2017

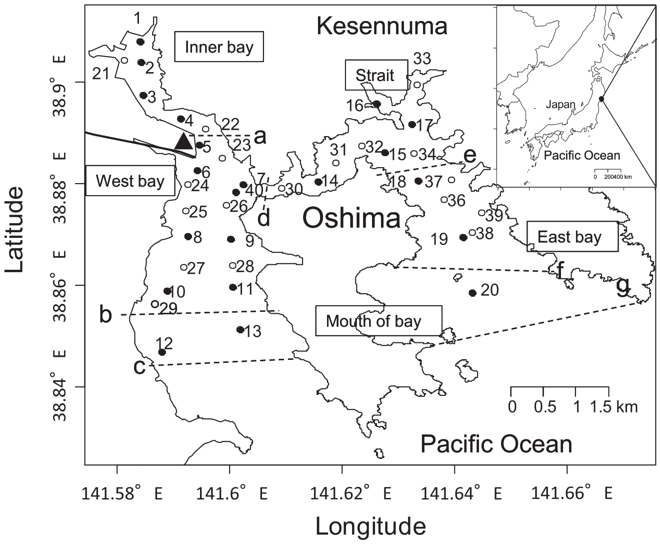

Fig. 1. Sampling stations in Kesennuma Bay. Filled circles (Sta. 1–20, 40) show sampling stations where sediments were obtained in all sampling events. Blank circles (Sta. 21–39) indicate sampling stations added in July 2014. The black triangle near station 5 represents the place of destroyed oil terminal.

was added after December 2012. Samples were only collected from site 10 in February 2012, because of the low silt and clay contents of the sediment. Furthermore, stations 21–39 were added in July 2014. We could not collect sediment samples at stations 19 and 35 in July 2014 because there was a lot of gravel on the seabed. Kesennuma Bay is divided into five water areas: the inner bay, west bay, strait, east bay, and mouth of the bay, as shown in Fig. 1. The inner bay is north of line (a) (stations 1–4, 21, and 22), and the west bay is between line (a) and line (b) (stations 5–11, 23–28, 40). The strait is between line (d) and line (e) (stations 14–17, 30–34). The east bay is between line (e) and line (f) (stations 18–20, 36–39). The mouth of the bay is comprised of two areas between line (b) and line (c) (stations 12 and 13) and between line (f) and line (g) (station 20). The inner bay, west bay, strait, east bay, and the mouth of the bay cover areas of 1.42 4.62, 3.03, 3.96, and 8.55 km^2, respectively, and the water depths in each area are 6.5–11.5, 12.3–31.1, 7.8–32.1, 16.4–39.2, and 31.5–33.6 m, respectively.

The sediment samples were collected with an Ekman-Birge bottom grab sampler (20 cm square) and a core sampler with a 5-cm diameter. The Ekman-Birge bottom grab sampler was dropped from a boat at each sampling station, and about 1.5 L of seabed surface sediment (ca. 3–4 cm thick) was collected. The sediment samples were homogenized in a tray on board, stored in plastic containers, frozen, and transported back to the laboratory. Five replicate samples were collected at stations 2, 8, and 17 in July 2014, while three replicate samples were collected at all stations in December 2014. One sample was collected at each site in the other sampling events. There were some observation points that showed a large SD of measurement values (PAHs etc.) shown in the following section, but we judged that there was no problem for the discussion of this article. Core samples were collected by scuba divers at stations 2, 4, 7, 9, 14, and 17 in December 2012, and at stations 2, 7, 14, 17, and 19 in July 2014. The cores collected in December 2013 were 5 cm in diameter and 30 cm deep, while those collected in July 2014 were between 15 and 50 cm deep. One sample was collected from each station. The core samples were cut into 5-cm intervals.

2.2. Analysis and methods

The total oil concentrations in sediments are reported as NHE (Ministry of Environment, Japan, 2012). The concentrations of NHE were determined as follows. Approximately 20–30 g of each dry sediment sample was transferred into an extraction thimble. First, n-hexane was added to samples and extracted in a Soxhlet extraction apparatus for 5–6 h, after which samples were left overnight. The temperature of n-hexane extraction in the Soxhlet apparatus was 80 °C. Sodium sulfide was added to 500 mL of the extract in order to dehydrate. The extracts were distilled and concentrated to approximately 2 mL. The extracts were moved to an evaporation container, weighed, and volatilized on the heating board, the temperature of which was maintained at approximately 80 °C. The extracts in the evaporation containers were then put in a dryer and heated at 80 ± 5 °C for 30 min. Finally, these were cooled for approximately 30 min in a desiccator, and then weighed.

TPH analysis followed the Guidelines for Countermeasures against Oil Contamination (Ministry of the Environment, Japan, 2006). Approximately 10 g of the wet sediment samples were transferred into a conical flask with a stopper and weighed, and 30 g of anhydrous sodium sulfate were added in order to dehydrate. Sulfur disulfide was added to the sediments in the conical flask, and the sediments were extracted by penetration for 3 min and left to rest for 2 h.

The abovementioned operation was repeated three times and extracts with a volume of 100 mL were obtained. Finally the extracts were filtered through a membrane filter with a pore size of 0.45 μm and analyzed with a gas chromatography equipped with flame ionization detector (GC-FID). TPHs in the extracts were identified and quantified by a gas chromatograph equipped with a FID (HP6890, Agilent Technologies, CA, USA). A slightly polar fused silica capillary column (HT-5(SGE), Shimadzu Co. Ltd., Kyoto, Japan) that was 30 m long, 0.53 mm in diameter, and with a liquid film thickness of 0.5 μm was used. A 1-μl aliquot of the condensed samples was injected into the injector in splitless mode. The temperature of the GC was held at 35 °C for 5 min and then increased to 360 °C at a rate of 10 °C/min.

Six sets of the sediments samples (those collected after December 2012) were analyzed for PAHs and TPHs, and all the samples were analyzed for NHEs. Samples were analyzed for 16 nonalkylated PAHs designated as priority substances by the United States Environmental Protection Agency (USEPA), including naphthalene (Naph), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fle), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*cd*]pyrene (IdP), dibenz[*a*,*h*]anthracene (DbA), benzo[*ghi*]perylene (BghiP), and four kinds of alkylated PAHs, namely 3-methylphenanthrene (3MP), 2-methylphenanthrene (2MP), 9-methylphenanthrene (9MP), and 1-methylphenanthrene (1MP). To determine the PAH concentrations, sediment samples were subjected to batch-extraction with acetone and dichloromethane. Approximately 3 g of wet sediment samples were transferred into 50 mL glass centrifuge tubes and weighed. First, 7 mL of acetone was added to the samples and sonicated in a sonicator bath for 10 min. After centrifugation, acetone extracts were removed from centrifugation tubes with glass pipettes and transferred to other centrifugation tubes. Acetone was added again to the residual sediment samples, and the process described above was repeated. After the acetone extraction was completed, a further extraction was carried out with dichloromethane. The first and second acetone extracts and dichloromethane extracts were combined in a centrifuge tube. The same volume of ultra-pure water was added and they were centrifuged to separate the aqueous and organic layers. The bottom organic layer was recovered with a glass pipette, and 10 mL was poured into glass syringe columns filled with anhydrous sodium sulfate. Glass syringe columns were connected to a manifold in which the glass tubes were set out to receive drops of dehydrated extracts. Excess organic solvents were removed under a stream of nitrogen gas and replaced by 0.5 mL of added n-decane. The resulting n-decane extracts were spiked with deuterium-labeled PAH surrogates and then were subjected to chromatography in a silica gel column (ca. 0.5 g of silica gel corresponding to a volume of 3 mL). The first extract fraction was eluted with 4.5 mL of n-hexane. Subsequently, the second fraction for aromatic hydrocarbon analysis was eluted with 9 mL of 5% (vol/vol) acetone-containing hexane. The