Marine Pollution Bulletin 130 (2018) 229–239

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

Transport of crude oil and associated microbial populations by washover events on coastal headland beaches

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ARTICLE INFO

Keywords:
PAHs
Gulf coast
MC252 oil
Biodegradation
Hurricanes
Crude oil

ABSTRACT

Storm-driven transport of MC252 oil, sand and shell aggregates was studied on a low-relief coastal headland beach in Louisiana, USA including measurement of alkylated PAHs and Illumina sequencing of intra-aggregate microbial populations. Weathering ratios, constructed from alkylated PAH data, were used to assess loss of 3-ring phenanthrenes and dibenzothiophenes relative to 4-ring chrysenes. Specific aggregate types showed relatively little weathering of 3-ring PAHs referenced to oil sampled near the Macondo wellhead with the exception of certain SRBs sampled from the supratidal environment and samples from deposition areas north of beach. Aggregates mobilized by these storm-driven washover events contains diverse microbial populations dominated by the class *Gammaproteobacteria* including PAH-degrading genera such as *Halomonas*, *Marinobacter* and *Idiomarina*. Geochemical assessment of porewater in deposition areas, weathering observations, and microbial data suggest that storm remobilization can contribute to susceptibility of PAHs to biodegradation by moving oil to beach microenvironments with more favorable characteristics. (149).

1. Introduction

Coastal headland beaches and barrier islands in Louisiana were impacted by MC252 crude oil released into the Gulf of Mexico from April 20, 2010 to July 15, 2010 (McNutt et al., 2011; Michel et al., 2013). These headland beaches are characterized by low relief with a poorly developed dune system fronting an area of mudflats, marshes and mangroves immediately north of the beach. Because of their position on the landscape and their morphology, these areas are susceptible to tidal events that completely inundate sections of the beach. These "washover" tidal events occur when cold fronts, tropical storms and hurricanes create storm surge at a higher elevation than the beach crest, resulting in the reworking and depositing of sand in the back marshes and mudflats (Georgiou et al., 2005). On Fourchon Beach, Louisiana, a ~14 km coastal headland beach impacted by the *Deepwater Horizon* oil spill (Elango et al., 2014; Lemelle et al., 2014; Urbano et al., 2013), these washover events have the potential to move oil from the subtidal and intertidal portions of the beach to the supratidal mudflats and marsh areas. These washover tidal events contribute to the rapid erosion of the beach, itself, which can occur at rates exceeding 12 m per year (McBride et al., 1992), one of the most rapidly eroding coastlines in the U.S.

Oil deposited on the beach during the *Deepwater Horizon* event consisted of various aggregates of oil, sand and shell termed surface residue balls (SRBs) or submerged oil mats (SOMs) (Georgiou et al., 2013; OSAT-II, 2011). SRBs are oil-sand aggregates, typically spherical or disk shaped, with an average mass and density on Fourchon Beach of 14.5 ± 3.0 g and 1.32 ± 0.11 g/cm^3, respectively (OSAT-II, 2011; Urbano et al., 2013). MC252 crude oil content in SRBs ranges from 4.2–12.8% gulf wide and 2.25–5.54% on Fourchon Beach (OSAT-II, 2011; Urbano et al., 2013). SOMs represent another aggregate form of oil found in the inshore surf zone in troughs between the sand bars or associated with offshore relict marsh platforms near these coastal headland beaches (Georgiou et al., 2013). These contiguous mats can also be buried on the intertidal and supratidal portions of the beach, itself, and then exposed during these erosional washover events. MC252 crude oil content in SOMs is higher than those found in SRBs, ranging from 9.4–16.8% gulf wide (OSAT-II, 2011) and 8.2–14.2% (Elango et al., 2014) on Fourchon Beach. Observations of post-storm oil deposition patterns suggest that beach dynamics can move these oil forms from the intertidal to the supratidal, and ultimately to the mudflats and marshes.

These transport processes may influence rates of weathering in crude oil not removed during response operations. In these oil:sand

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https://doi.org/10.1016/j.marpolbul.2018.03.008
Received 23 October 2017; Received in revised form 26 February 2018; Accepted 5 March 2018

aggregates, microbial degradation (Engel and Gupta, 2014; Lamendella et al., 2014; Leahy and Colwell, 1990; Prince et al., 2013) is expected to be a significant weathering mechanism for crude oil components such as polycyclic aromatic hydrocarbons (PAHs). Dissolution or solubilization of these high molecular PAHs would very inefficient with the small amount of flushing due to rainfall and occasional inundation from storms. Photodegradation would be limited due to the attenuation of the UV radiation a few hundred microns into the sand (GarciaPichel and Bebout, 1996), well away from where the oil is located in the aggregate. To understand factors affecting the rate and extent of biodegradation, post-spill microbial community structure in the Gulf of Mexico water column and on beach sands has been reported (Engel and Gupta, 2014; Hazen et al., 2010; King et al., 2015; Kostka et al., 2011; Lamendella et al., 2014) but information on microbial populations within the aggregates, themselves, is limited (Urbano et al., 2013). Microbial populations within the aggregates (Ebrahimi and Or, 2015; Sierra and Renault, 1996) could be important in initiating degradation processes as these materials are transported into different locations on the beach with different biogeochemical conditions. For microbial degradation, the physicochemical environment within the oil:sand aggregate (Urbano et al., 2013) and the surrounding environment can impact the rate and extent of degradation. Important characteristics include the availability of electron acceptors (e.g., oxygen or sulfate), the nutrient status (N and P), the temperature, and the moisture content within the aggregate (Camilli et al., 2010; Monrozier et al., 1991; Nam et al., 2003). The kinetics of PAH biodegradation under aerobic conditions (Haritash and Kaushik, 2009) are generally faster than biodegradation kinetics under anaerobic conditions (Coates et al., 1997) and a more extensive set of PAHs are biodegraded aerobically. Conditions within the aggregates that include sufficient oxygen, nutrients and moisture will increase biodegradation (Urbano et al., 2013). If aggregates are washed from zones on the intertidal and supratidal where more optimal conditions to exist to marsh and mudflat areas where oxygen is very limited, biodegradation will be slower and less extensive.

The objectives of this study are to understand the role of tidal and storm-driven washover events in transporting and distributing crude oil aggregates and their associated microbial populations. Our hypothesis is that washover events can move oil:sand aggregates across the beach profile (from subtidal to supratidal and finally to deposition in mudflat or marsh environments). These transport processes may increase the persistence of alkylated PAHs due to the lack of optimal physicochemical conditions in the deposition areas. At present, we have a limited understanding of anaerobic processes responsible for biodegradation of alkylated three ring and four ring PAHs (Fathepure, 2014). The present study evaluates the extent of weathering in different oil forms (SOM, SRBs and mudflat sediments) observed along the Fourchon Beach shoreline over a 2-year period from 2011 to 2013. This study also characterizes the microbial community in these aggregates using metagenomic techniques.

2. Materials and methods

2.1. Study location and sampling events

Samples were collected from a 14-km coastal headland beach, Fourchon Beach, in Port Fourchon, LA (Fig. 1). During *Deepwater Horizon* spill response, the beach was divided into 9 operational zones, approximately 1.6 km in length, numbered 1–9 from east to west (Pardue et al., 2014a). Washover events, driven by strong winter storms, tropical storms and hurricanes, regularly inundate the beach and form erosional channels, termed washover channels, through which water from the Gulf of Mexico flows into the marsh and mudflats north of the beach. During this process, sand is deposited into fan-shaped deposits, termed “washover fans”, at the edge of the marsh. Three different studies were conducted to understand the impact of washover events on oil mobilization and fate; (i) evaluation of specific oil transport scenarios from washover events that occurred from 2011 to 13, (ii) assessment of the presence of oil in washover fans from operational zones 1–3 and (iii) the distribution of PAHs as a function of depth within washover fans in Zones 1 and 5.

2.1.1. Washover event sampling

Weathered crude oil samples were collected and analyzed for PAHs and associated microbial populations after three specific storm-driven washover scenarios; (i) breakup and washover of an onshore SOM in Zone 7 immediately after Tropical Storm Lee (TS Lee) in September 2011, (ii) breakup and washover of an offshore SOM in Zone 4 during a frontal passage in April 2013 and (iii) SRBs resulting from repetitive washover events in Zone 1 from 2011 to 13. These samples were obtained from post-storm sampling surveys in areas with known oil distributions prior to the storm event. For scenario (ii) above, the sampling team was present during the washover event and sampled the oil in real time as it came ashore.

2.1.2. Washover fan survey zones 1, 2 and 3

A survey of the washover fans in Zones 1–3 was conducted from February to May 2012. During the survey, samples were collected from every washover fan encountered across the approximate 4.8 km distance for a total of 59 samples (Supplementary Information (SI), Fig. SD1). At every fan, a core was taken using a 15 cm dia × 30 cm depth aluminum core tube. The location of the core was at the approximate midpoint of the fan at the water-line of the marsh bounding the beach on the north. The top 15 cm of the core was sectioned and immediately composited after sampling and transported to the laboratory under ice for analysis of PAHs. During the survey, SRBs encountered in the washover channel adjacent to the fan were sampled as representative of the oil forms that are transported.

2.1.3. Depth distribution of PAHs in washover fans (zones 1 and 5)

Core samples were collected from 2 washover fans in Zones 1 and 5 to determine the depth distribution of MC252 crude oil components. On 12/15/2011, core samples were collected from a washover fan adjacent to Bay Champagne in Zone 5. Three cores, each 15 cm dia. × 70 cm depth were collected using an aluminum core tube. Core samples were cut into 7.5 cm sections and PAHs were extracted and analyzed for each section as described below. A second set of 17 cores were collected from a washover fan in Zone 1 (SI Fig. SD2) on 3/15/13 which had previously been breached during TS Lee in 2011. Core samples were collected using an AMS multi-stage core sampler fitted with a stainless steel extendable cylindrical sampler (6.5 cm dia. × 30.5 cm depth). Cores were collected from 3 transects across the washover fan. Each core was collected in a plastic liner, capped, and transported back to the laboratory. Recovery ranged from 20 to 25 cm. These cores were cut into 5 cm sections, extracted and analyzed for PAHs as described below.

2.2. PAH extraction and analysis

Approximately 10 g of SOM, SRB or washover fan sediment sample were used for extraction of crude oil components. Samples were mixed with sodium sulfate and magnesium sulfate to remove moisture, transferred into individual sample stainless steel cells and extracted with a Dionex Accelerated Solvent Extractor (ASE) 350 (Thermo Scientific, Waltham, MA). A 50:50 (v/v) mixture of hexane and acetone was used as the extraction solvent under 1700 psi at 100 °C. Approximately 60–80 mL of solvent extract volume was obtained and the sample volume was concentrated to 10 mL using a RapidVac N_2 Evaporation System (Labconco, Kansas City, MO). PAH analysis was performed using an Agilent 6890 N gas chromatograph equipped with a 5973 N mass selective detector. The GC conditions were: 1 μL of the sample, DB5 capillary column (30 m × 0.25 mm × 0.25 μm (film thickness)), helium carrier gas at a rate of 5.7 mL/min, temperature