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The Complexity of Spills: The Fate of the *Deepwater Horizon* Oil

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Abstract

The *Deepwater Horizon* oil spill was the largest, longest-lasting, and deepest oil accident to date in US waters. As oil and natural gas jetted from release points at 1,500-m depth in the northern Gulf of Mexico, entrainment of the surrounding ocean water into a buoyant plume, rich in soluble hydrocarbons and dispersed microdroplets of oil, created a deep (1,000-m) intrusion layer. Larger droplets of liquid oil rose to the surface, forming a slick of mostly insoluble, hydrocarbon-type compounds. A variety of physical, chemical, and biological mechanisms helped to transform, remove, and redisperse the oil and gas that was released. Biodegradation removed up to 60% of the oil in the intrusion layer but was less efficient in the surface slick, due to nutrient limitation. Photochemical processes altered up to 50% (by mass) of the floating oil. The surface oil expression changed daily due to wind and currents, whereas the intrusion layer flowed southwestward. A portion of the weathered surface oil stranded along shorelines. Oil from both surface and intrusion layers were deposited onto the seafloor via sinking marine oil snow. The biodegradation rates of stranded or sedimented oil were low, with resuspension and redistribution transiently increasing biodegradation. The subsequent research efforts increased our understanding of the fate of spilled oil immensely, with novel insights focusing on the importance of photooxidation, the microbial communities driving biodegradation, and the formation of marine oil snow that transports oil to the seafloor.

1. INTRODUCTION

This article introduces processes impacting the fate of oil spilled into the ocean and examines the processes affecting the fate of such oil, using *Deepwater Horizon* (DwH) as an example. DwH was an offshore drilling rig, and the accidental release lasted for months and occurred at depth, with oil accumulating in the water as well as on the sea surface and nearby shorelines. One main response measure was the addition of dispersants, both at the surface and at the leak site.

After providing a brief introduction to oil, the DwH spill, and the dispersant Corexit, this review describes the immediate distribution of the DwH oil upon its release. We then introduce the different weathering and transport processes driving the fate of the spilled oil and appraise their relative importance in different ecosystems of the Gulf of Mexico. Once crude oil is spilled, weathering continuously alters its chemical composition and thus its behavior. In this review, we define the term oil as an ever-changing mixture of compounds that includes chemical signatures linking them to the source crude oil. Thus, even when weathered, the altered oil mixture will still be called oil.

1.1. Oil Spill Research

By definition, oil spills are unplanned and unexpected. Since effective oceanographic field programs require logistical infrastructure and equipment, academic research into the consequences of an oil spill is somewhat handicapped even when a solid spill response plan is in place. In addition to the time requirements for logistics, pre-event environmental data are often either lacking or difficult to access, being produced as part of various programs mandated by governmental agencies (Ashton et al. 2020). In this respect, the DwH spill in 2010 in the northern Gulf of Mexico was no different. However, the DwH spill was the first significant deepwater-well blowout with live oil (i.e., a mixture of dissolved natural gas and oil), leaking at a depth of more than 1,000 m.

The long duration of the DwH spill—nearly three months passed until the leak could be closed—allowed for the establishment of an extensive sampling and analysis effort, which included NOAA's Natural Resources Damage Assessment process as well as research by other governmental agencies, industry, and academic scientists. A 10-year research program administered by the Gulf of Mexico Research Initiative was established to better understand the spill, the consequences for the Gulf of Mexico, and the fate of the released oil. All these research efforts notwithstanding, making accurate budget calculations that include the ultimate fate of all of the spilled oil is nearly impossible, with limited agreement even on the total amount of oil spilled (MacDonald 2010). Furthermore, because of the immense size of the impacted area, it was difficult to efficiently track the oil during the 87 days of the spill, with direct measurements of certain specific processes and pathways missing or insufficient, and with newly released oil continuously mixing with weathered oil that was released earlier (Joye 2015). Tracking efforts were further complicated by the chemical complexity of oil and natural gas and the weathering processes, combined with the biological complexity of organisms and ecosystems, in the physically dynamic Gulf of Mexico. Gradients caused by the discharge from the Mississippi River, the complex seafloor topography, the varied coastline structure (sandy beaches and muddy marshes), and the range of mediating measures applied led to high spatial and temporal variability of oil behavior.

1.2. Oil

Crude oil consists of tens of thousands of different hydrocarbon-type organic molecules that impart a unique quantitative signature to each reservoir. The relative composition determines the physical properties of an oil, such as its gas content, density, viscosity, surface tension, and dispersibility. Highly volatile compounds like methane are a gas at atmospheric pressure and go into solution at elevated pressure (e.g., at depths). Soluble compounds dissolve into the aqueous phase on a range of timescales. Most compounds of liquid oil are insoluble in water and may accumulate at the water–atmosphere interface, forming a surface slick; alternatively, under stormy conditions, insoluble compounds may be entrained into the water phase as dispersed oil droplets.

Crude oil molecules are generally classified into four categories: saturates (e.g., alkane hydrocarbons), aromatics [conjugated six-carbon-ring systems, such as benzene and polycyclic aromatic hydrocarbons (PAHs)], resins, and asphaltenes. Saturated hydrocarbons range from small molecules, such as methane (a one-carbon substance), to much larger molecules that contain dozens of carbon atoms linked together, with carbon numbers over 40. Some of the saturated multicyclic hydrocarbons, such as the hopanes and steranes, are particularly resistant to biodegradation and are known as petroleum biomarkers (Prince & Walters 2007, Prince et al. 1994). The resistance of biomarkers to weathering is an active area of research (Aeppli et al. 2014). Aromatic hydrocarbons can be modified with sulfur, nitrogen, and oxygen, which results in increased hydrocarbon water solubility and oxidation potential. This increases their bioavailability and thus poses a risk to living organisms (Honda & Suzuki 2020, Idowu et al. 2019). Resins and asphaltenes are high-molecular-weight compounds with aromatic and aliphatic molecular structures that can also contain NSO (nitrogen, sulfur, and oxygen) heteroatoms. They are solid, insoluble, and practically resistant to microbial degradation. Light crude oils generally contain only small quantities of these very large molecules. Refinery residues containing these types of insoluble compounds are commonly used as roofing tar and road asphalt, in addition to being components of marine fuel oils.

1.3. The *Deepwater Horizon* Accident

When the semisubmersible mobile offshore DwH rig, which was drilling an exploratory well, exploded on April 20, 2010, due to uncontrolled high-pressure release of gas and liquid oil, it killed 11, injured 17, sank, and caused a massive offshore deepwater oil spill. The site of the accident lies offshore, in Mississippi Canyon block 252, which is in the northern Gulf of Mexico, 65 km off the Louisiana coast, in the region of influence of the Mississippi River plume. For approximately three months, until mid-July, large amounts of live crude oil, including 50,000–70,000 barrels of liquid oil per day and 135 kg m^{-3} of gas per day, leaked from several points along the broken riser pipe at the seafloor and the blowout preventer valve at approximately 1,500-m depth (Camilli et al. 2010, McNutt et al. 2012, Reddy et al. 2011). By mass, the leaking fluid oil (Macondo oil) consisted of approximately 38% natural gas and 62% liquid oil (Reddy et al. 2011; Ryerson et al. 2011, 2012). The co-occurrence of gas (hydrocarbons that maintain a gaseous state at atmospheric pressure) and liquid oil is typical for a well blowout and is distinctive from most other oil spill scenarios. The Macondo oil is a light, sweet oil, with a density of approximately 0.84 g mL^{-1} at 15°C and a viscosity at 15°C of 7.1 mPa·s and 0.7 mPa·s for dead and live oil, respectively. Macondo oil has a relatively high content of low-molecular-weight hydrocarbons ($<n\text{-}C_{25}$) and a relatively low sulfur and asphaltene content (approximately 0.3% each) (French-McCay et al. 2018; Gros et al. 2016, 2017), and approximately 25% of compounds by mass are readily soluble (Ryerson et al. 2012). The specific release conditions during the DwH spill—i.e., high-speed jetting from various kinks in the riser pipe and at the blowout preventer—led to the partitioning of the spilled oil into two major locations, with peak oil concentrations at the sea surface and in deep intrusion layers that formed at depths between 900 and 1,300 m.

1.4. Response Efforts

Response efforts to mitigate the effects of an oil spill are unique to every spill, as they depend on the oil, the circumstances of the spill, and environmental and logistical considerations (Ventikos et al. 2004). A combination of measures was used during the DwH spill, including booming, mechanical recovery, in situ burning, and chemical dispersant application, and each changed the release dynamics of the spill and the fate of the oil (Özgökmen et al. 2016). Opening of the diversion channels, shoreline booming, and dispersant application were all implemented to minimize the amount of oil washed ashore.

Each response measure had different impacts. In situ burning of surface oil, which began on May 6 and ultimately included more than 400 burns by the time the spill ended in mid-July, left residues (char and soot) in the water (Passow & Stout 2020, Yan et al. 2016). At the end of May, a failed top-kill attempt to close the leak released large amounts of drilling mud at the site of the accident. Although not intended as a mediating measure, the release of fine clay-sized material is at times utilized to disperse, aggregate, and sink oil or harmful algae (Beaulieu et al. 2005, Chaeruh et al. 2005, Sengco & Anderson 2004). The opening of the Mississippi floodgates to create offshore flow inadvertently introduced nutrients, particles, and fresh water into the region.

The aerial application of dispersant began on April 24, only a few days after the accident. In total, approximately 1,070,000 gallons of the dispersants Corexit 9500A and 9527 were applied onto the surface slick, and approximately 770,000 gallons of Corexit 9500A were released at depths directly into the plume of the escaping oil (Lehr et al. 2010). The first deep dispersant application began in mid-May, but systematic injection of dispersant into the leak started after June 3, when the riser pipe was sheared, consolidating several leak points to one and allowing the installation of an oil capture device (TopHat #4) for partial direct recovery of discharging oil (Du & Kessler 2012, Dubinsky et al. 2013). Before June 3, oil escaped the riser pipe at several leak points, deep Corexit addition was spotty, and mechanical oil removal at depth was impossible. Increased oil recovery and the targeted deep Corexit application altered flow conditions and weathering dynamics after June 3, but comparisons between both periods are lacking, and existing estimates of the partitioning of oil between the surface and intrusion layer focus on the period after June 3, ignoring oil partitioning in April and May.

2. INITIAL DISTRIBUTION OF THE SPILLED *DEEPWATER HORIZON* OIL

To understand the fate of the spilled oil (see the sidebar titled What Was the Fate of the *Deepwater Horizon* Oil?), one must first look at the initial behavior of the oil exiting at the leak points. Liquid oil and gas escaped as a multiphase flow, forming a jet with extremely large buoyancy fluxes (Özgökmen et al. 2016, Zhao et al. 2015). The interactions of the liquid oil with dissolved gases and water led to the formation of oil droplets, gas bubbles, and methane hydrates, although the latter played a minor role during the DwH spill (Chen et al. 2014). Temperature at the leak point was 4.3°C with a pressure of 153 atm, representing a pressure drop of approximately 86 atm and a temperature drop of approximately 100°C between the reservoir and the blowout preventer. Natural gas exited the blowout preventer in its gaseous (methane) or liquid (e.g., ethane or propane) form, with dissolution into the aqueous phase starting immediately upon exposure to water (Gros et al. 2016, Natl. Acad. Eng. Sci. Med. 2019). The rapid changes in pressure and temperature determined the release dynamics of the escaping oil, especially droplet formation, and its further behavior (Malone et al. 2020). For example, at 153 atm, the solubility of n-C_1–C_4 hydrocarbons increases by a factor of 10–150 compared with their solubility at atmospheric pressure (Gros et al.

WHAT WAS THE FATE OF THE *DEEPWATER HORIZON* OIL?

The answer is complex, but in a nutshell, approximately 25% of the spilled oil was recovered or burned, 5–15% evaporated, and the remaining 60–70% spread and weathered within the Gulf of Mexico. As oil rose from its release point at 1,500-m depth, deep intrusion layers that were enriched in soluble compounds and tiny liquid oil droplets formed at approximately 1,000-m depth. Mostly insoluble, less volatile compounds formed a surface slick from which the volatile compounds evaporated. The dissolution of soluble compounds continued in both layers, with evaporation and photooxidation additionally weathering floating oil at the surface. Biodegradation is estimated to have removed up to 60% of the oil in the intrusion layer but was less efficient in weathering the floating oil. A significant amount of weathered oil was stranded, contaminating shorelines from Louisiana to Florida, or formed a "dirty bathtub ring" where the intrusion layer collided with the shelf break. Once the oil was stranded and not removed by cleanup crews, photodegradation and biodegradation rates decreased appreciably and depended on the environmental conditions. Sinking marine snow transferred oil from both the surface and the intrusion layer to depth, depositing significant amounts of oil onto the seafloor, where biodegradation rates were low under anoxic conditions. The fractions of the oil that entered food webs and were lost at sea are unknown.

2016), and gas bubbles are not stable (Aman et al. 2015). As n-C_1–C_4 hydrocarbons dissolved into the aqueous phase within the first 700 m of ascent, the remaining petroleum compounds formed into liquid droplets (Zhao et al. 2015).

Droplet size is a critical property, as it influences rising speed as well as the recoalescence, biodegradation, and dissolution rates. The droplet size distribution of dispersed oil is thus central for understanding the behavior of escaping oil and depends on the release conditions, including the energy of mixing, turbulence, and interfacial tension (Aman et al. 2015). Droplet size determines the ascending speed of droplets, with microdroplets (<100 μm) effectively remaining suspended because their buoyancy is nullified by flow resistance (John et al. 2016, Natl. Acad. Eng. Sci. Med. 2019). Large-diameter droplets (>1 mm) rise to the sea surface in a matter of hours, while small droplets (0.1–1 mm) require days. As a consequence, the extraction rate of soluble compounds is more significant during the ascent of small droplets compared with that of large ones, since the dissolution rate is higher in small droplets (high surface-to-volume ratio), and they take longer to reach the surface. A longer ascent time also means increased horizontal transport during ascent and a larger surface expression of the oil.

Small and large droplets rose to the surface and, although chemically altered during ascent, had a clear surface expression. Dissolved compounds and microdroplets were trapped at depths of 900–1,300 m, leading to the formation of deep intrusion layers (Diercks et al. 2010, Joye et al. 2011, Kessler et al. 2011, Ryerson et al. 2012, Valentine et al. 2010). Additional plumes formed in the water at shallower depths but were overall negligible in mass and not trackable. Elevated oil concentrations were also observed in the whole water column near the spill site, reflecting the plume of rising droplets, and at times in the subsurface layer, due to reentrained oil droplets from the surface slick (Payne & Driskell 2018).

2.1. Deep Intrusion Layers

The oil residing in the deep intrusion layer was enriched in gaseous and easily soluble compounds such as methane, ethane, and propane, which remained nearly completely at depth (Ryerson et al. 2011), along with the monoaromatic BTEX (benzene, toluene, ethylbenzene, and xylene) hydrocarbons (Valentine et al. 2012).

The deep intrusion layers, as identified by a negative oxygen anomaly caused by respiration, extended 500 km southwest and 120 km northeast from the spill site (Du & Kessler 2012). The oxygen anomaly covered an area of 72,000 km^2 and persisted until September 2010, i.e., for six to eight weeks after the leak was capped (Du & Kessler 2012). During this time, bioavailable hydrocarbons were successively utilized or diluted (Dubinsky et al. 2013). Gros et al. (2017) estimated that after early June, 24% of oil (by mass) remained in the deepwater intrusion as mostly dissolved (23%) or dispersed (<1%) microdroplets, implying that droplets contributed less than 5% to oil in the intrusion layer. By contrast, assessments by Ryerson et al. (2012) and Valentine et al. (2012) suggested that 50–70% of the spilled and unrecovered oil remained in the intrusion layer and that liquid droplets contributed approximately 30% of hydrocarbons, with dissolved compounds contributing the other 70%. Back-of-the-envelope calculations suggested that the low estimates of microdroplet concentrations in the intrusion layer are likely too low to account for the observed sedimentation of oil from this layer (Passow & Stout 2020, Valentine et al. 2014).

2.2. Sea Surface

The vast majority of rising droplets reached the sea surface within a 2-km radius of the spill site and within less than 10 h (Ryerson et al. 2012). When the droplets reached the surface, the drops formed or joined a surface slick and streamers of mostly emulsified liquid oil (water in oil emulsion). By the time oil reached the surface, most compounds less than n-C_8 were already lost due to dissolution during ascent (Stout et al. 2016). Volatile compounds that were still present evaporated, and the slick, now having lost most compounds less than n-C_{13}, was spread by winds and currents. Two days after surfacing, when most volatile substances had evaporated, n-C_{15}–C_{18} hydrocarbons were the most abundant by mass (Ryerson et al. 2012). The surface expression of the oil, which remained visible for a month after the spill, varied continuously in shape, size, thickness, and location (MacDonald et al. 2015).

2.3. Subsurface Layer

Wave action reentrained and (physically) dispersed oil into the upper 20–40 m below the slick, leading to elevated concentrations in this subsurface layer. During dispersion, dissolution of soluble compounds out of droplets was enhanced. Compared with floating surface oil, the dispersed oil was depleted more of its aromatic content in the form of two- and three-ringed PAHs than of its alkane and larger four- to six-ringed insoluble PAH content (Driskell & Payne 2018, Payne & Driskell 2018). During quiescent periods, dispersed oil droplets can resurface, reforming the slick, or can be diluted or oxidized before resurfacing, making the existence of such a subsurface layer a transient occurrence. The frequency of the formation of such a subsurface layer of increased hydrocarbon concentrations may be estimated from wind speeds. Wind speeds remained low during the DwH spill, reaching 8 m s^{-1} only once for a few days (MacDonald et al. 2015), suggesting that wave-induced natural dispersion of floating oil into the underlying water layer may have been minimal overall. However, newer models suggest that appreciable entrainment may be possible at 6 m s^{-1}, and the dispersant application onto the surface slicks would have greatly facilitated the dispersion of oil into the upper 20–40 m (Li et al. 2017), suggesting that entrainment of chemically dispersed oil occurred more regularly.

3. COREXIT APPLICATION AND THE FATE OF OIL

Chemical dispersants are amphiphilic; thus, they decrease the interfacial tension between oil and seawater and enable increased dispersion of tiny oil droplets into the near-surface water column for a given amount of turbulence or mixing (John et al. 2016). Application of a chemical dispersant therefore generally results in decreased droplet sizes, increased droplet concentrations, and faster dissolution compared with natural dispersion, which has consequences for transport and weathering (Khelifa et al. 2008).

Chemical dispersants are mixtures of synthetic chemicals containing surfactants as the main active compound, as well as solvents and additives (Comm. Underst. Oil Spill Dispersants Effic. Effects 2005). Solvents are present to promote the dissolution of surfactants and additives. Additives have a variety of purposes, such as increasing the dissolution of surfactants into an oil slick or increasing dispersant stability, and surfactants allow oil droplets to be entrained into the aqueous phase if the energy input (e.g., wave action) is sufficient (Comm. Underst. Oil Spill Dispersants Effic. Effects 2005). In general, dispersant effectiveness decreases with increasing oil viscosity and degree of oil weathering. The Corexit family, the most commonly stockpiled dispersants in the United States, contains mixtures of nonionic and anionic surfactants. Like most dispersants, Corexit works efficiently in seawater but not in fresh water. Corexit 9500 is slightly more effective with high-viscosity oils than Corexit 9527 and is also thought to be less harmful to organisms (Comm. Underst. Oil Spill Dispersants Effic. Effects 2005). The anionic surfactant dioctyl sodium sulfosuccinate (DOSS) is typical for Corexit formulas and was used as a marker for Corexit during the DwH spill (Gray et al. 2014, John et al. 2016, Kujawinski et al. 2011). However, dispersant analysis is difficult and expensive, and the presence of Corexit was not tracked routinely.

As Corexit was widely used during the DwH spill mediation, it may be assumed that field samples containing oil were also exposed to Corexit, whether tracked or not. To complicate matters further, the indirect effects of Corexit on oil distribution and weathering often cannot be distinguished from direct effects on organisms or organic matter distribution, even in experiments. For example, smaller oil droplet size due to Corexit addition might make the oil physically more bioavailable because the surface:volume ratio is increased, thereby permitting better access to the oxygen and nutrients required for biodegradation. However, the presence of Corexit may impact the ability of bacteria to degrade the oil, or favor bacteria that degrade Corexit, potentially leading to decreases in oil biodegradation rates (e.g., Kleindienst et al. 2015a,b). As a result, the specific impact of Corexit on the fate of the DwH oil is not always clearly identifiable. Below are some examples of Corexit influencing the fate of the DwH oil, but overall, this review addresses the fate of the DwH oil under ambient environmental and mediating measures.

The application of dispersant at great depth during the DwH accident was a first. DOSS could be traced in the deep intrusion layer in association with elevated hydrocarbon concentrations for 300 km from the wellhead and for 64 days after deepwater application ceased, suggesting that it remained associated with the oil and that removal processes were slow (Gray et al. 2014, Kujawinski et al. 2011). DOSS persisted for months to years in impacted coral communities and stranded oil samples from beaches, also suggesting low biodegradation rates (Perkins et al. 2016, White et al. 2014). In oil-contaminated surface samples, concentrations of DOSS varied widely, likely due to unknown matches between areal Corexit application in relation to sample collection. Additionally, DOSS may be photodegraded, but this has not been tested systematically (Gray et al. 2014).

The application of chemical dispersants onto the surface slick resulted in an estimated decrease of oil within the surface slick by 20% and an increase in the water-accommodated (dispersed plus dissolved oil) fraction of oil, leading to increased dissolution and decreased evaporation (MacDonald et al. 2015). The simultaneous almost 50% increase in the area covered by a thinner film of surface oil due to Corexit application (MacDonald et al. 2015) presumably increased the evaporation rate, because of rate limitations from diffusion within thick oil slicks (Natl. Acad. Eng. Sci. Med. 2019). Deep dispersant application likely altered droplet size distribution in the rising plume, although the debate about the extent of this effect is ongoing (Malone et al. 2020). Low evaporation during the DwH spill may thus be a consequence of the deep dispersant application, the flow dynamics under the release conditions, and/or the surface dispersant application; all of these factors likely contributed. As oil recovery also improved during the mediation effort that allowed targeted Corexit addition at depths, the respective effects of the different measures are difficult to separate (MacDonald et al. 2015, Özgökmen et al. 2016).

Reducing the amount of oil at the sea surface also reduced the amount of oil stranding on shorelines (Zeinstra-Helfrich et al. 2015). Biodegradation may be promoted by decreased droplet size due to dispersants, but the additional direct effects of the dispersant on microbes results in more complex outcomes (Baelum et al. 2012, John et al. 2016, Kleindienst et al. 2015b, Natl. Acad. Eng. Sci. Med. 2019). Furthermore, the presence of Corexit increased the desorption rate of heavily weathered oil from sediment when stranded oil–sediment agglomerations were resuspended (Duan et al. 2018). Dispersants also impact aggregation; on the one hand, the presence of Corexit appears to reduce the formation rate of aggregates, but on the other hand, the high concentration of oil droplets due to dispersants leads to high incorporation rates of droplets into aggregates. The overall consequence of these opposing trends for the sedimentation rate of oil varies (Passow et al. 2017).

4. WEATHERING

Here, we define weathering processes (see the sidebar titled What Is Weathering?) as those that chemically alter the oil mixture, and contrast them with transport processes that move oil without altering its chemistry or composition. The relative importance of individual weathering and transport processes depends on the environment, and together, they determined the fate of the DwH oil (**Figure 1**). Because weathering processes change the chemical composition of the remaining oil, careful compositional analysis may provide insights into the relative importance of different weathering processes that occurred in situ. In the case of the DwH spill, the continuous mixing of the daily released oil with older, weathered oil complicates the interpretation of compositional data.

4.1. Mixture Separation

Two major types of weathering processes may be distinguished: mixture separation and transformation. Mixture separation, such as evaporation, dissolution, or sorption to sinking particles,

WHAT IS WEATHERING?

The term weathering is used loosely to describe processes that alter the chemical composition and physical characteristics of oil released into the environment on timescales from seconds to decades. Specific molecules are lost from the oil mixture due to the evaporation of volatiles or the dissolution of soluble compounds into the aqueous phase (abiotic processes). Sorption to particles may selectively remove dissolved compounds from the aqueous phase. Photooxidation (abiotic) and biodegradation (biotic) are oxidation reactions that alter specific molecules. Whereas photochemically altered products are generally partially oxidized, biodegradation may lead to full oxidation to CO_2. Generally, low-molecular-weight hydrocarbons (e.g., $<n$-C_{25}) are more susceptible to weathering than larger ones, and the preferential removal of low-molecular-weight molecules increases the specific density and viscosity and decreases the dispersibility of oil as it weathers. Lightly weathered oil will have lost most of the volatile compounds, whereas heavily weathered oil may additionally have lost the soluble and most of the biodegradable compounds.

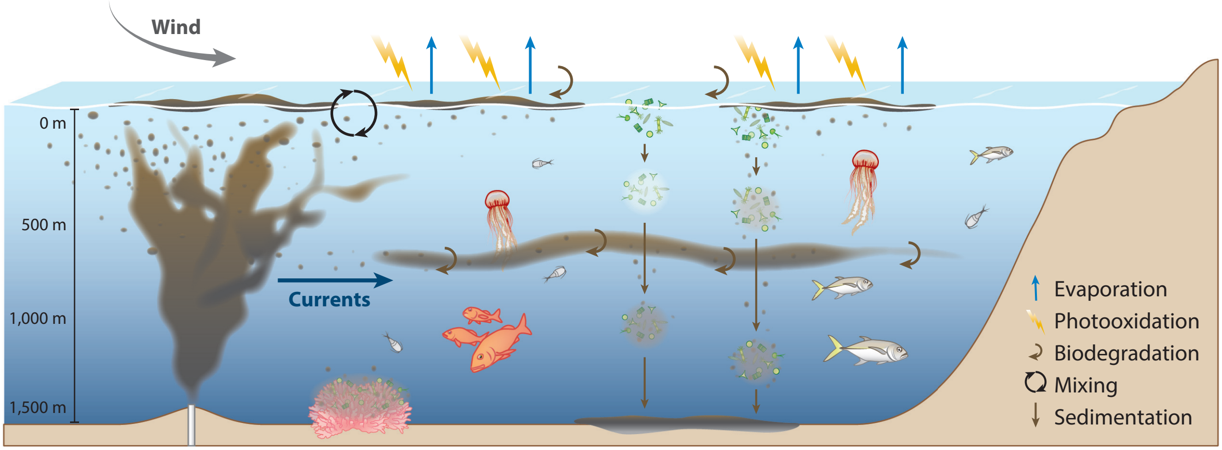

Figure 1

Two-dimensional schematic of the main transport and weathering processes of oil during the *Deepwater Horizon* spill (not to scale). In contrast to the depiction, the intrusion layer and the surface slick moved in different directions. Oil (*brown*) is visible in the released plume, as oil droplets, as floating surface slicks, and in the intrusion layer. Oil was also ingested into organisms (copepods, jellyfish, and fish) and trapped within sinking marine snow. Coral is shown covered in marine oil snow, which was also deposited onto the seafloor. Phytoplankton are shown in green.

translocates select compounds between environments, altering the chemical composition and physical properties of the remaining oil mixture. It removes the select compounds from one environment, but the molecular structure of a given compound (e.g., methane) stays chemically intact as it is translocated into the aqueous or gaseous phase.

4.1.1. Dissolution. Mixture separation includes the dissolution of soluble compounds trapped in an oil slick or droplets into the aqueous phase, resulting in the separation of the soluble compounds from the more insoluble ones. Low-molecular-weight alkanes ($<\sim n$-C_5) are the most soluble alkanes, but solubility also depends on environmental conditions such as pressure and temperature and on the physical manifestation (thick slick, thin film, or dispersed droplets) of the oil. During the DwH spill, the dissolution of soluble molecules and their extraction from rising oil droplets were high and drastically altered the composition of the oil ascending toward the sea surface (Ryerson et al. 2011). Methane, which contributed 20–30% by mass to the leaking oil, dissolved completely during ascent (Ryerson et al. 2011). The dissolution of semisoluble compounds continued as the oil aged, successively translocating these molecules into the aqueous phase (**Figure 1**). Microbial activity may also lead to water-soluble products. Additionally, leaching of hydrocarbon compounds out of buried oil deposits upon their redistribution intermittently led to elevated hydrocarbon concentrations in nearshore waters, even years after the spill (Turner et al. 2019).

4.1.2. Evaporation. Evaporation, which moves volatile compounds from the water–atmosphere interface into the atmosphere, occurs on timescales of seconds to days and is a function of the characteristics of the molecule, the environmental conditions, and the physical manifestation of the oil. As rising oil droplets burst at the sea surface, they stretch into a film, and the most volatile compounds evaporate immediately. However, when rising droplets join an established oil slick, evaporation is slower (Stout et al. 2016). During the DwH accident, undissolved and volatile hydrocarbons evaporated on average within 2–3 h, whereas a portion of the semivolatile fraction ($>n$-C_{11}) evaporated within 1–2 days. The prompt evaporation from the small area of oil surfacing (2 km^2) led to a narrow airborne plume downwind of the origin (Ryerson et al. 2011). Slower evaporation (10–100 h) of the hydrocarbons of intermediate volatility may have led to the formation of organic aerosols that occupied a wider plume as the source area at the sea surface increased with time (de Gouw et al. 2011). Once in the atmosphere, volatile hydrocarbons are rapidly oxidized (e.g., by hydroxyl radicals) on timescales of hours to days and may contribute to the formation of secondary volatiles and smog. The evaporated volatile and semivolatile compounds during the DwH spill represented approximately 14% of the surfacing mixture by mass (Ryerson et al. 2011). Overall, the magnitude of evaporation was appreciably less than expected based on the reservoir oil, mainly because of the many volatile compounds (n-C_1–C_6 alkanes and light aromatics) that dissolved before reaching the surface (Ryerson et al. 2011). Dissolution and evaporation together are thought to have reduced the mass of oil with a surface expression by approximately 36% (Ryerson et al. 2012), but budget estimates vary (Gros et al. 2017).

4.1.3. Sorption and coating. Particle–oil interactions with soluble or insoluble oil compounds are generally less well understood, but many soluble compounds sorb to surfaces (e.g., algae cells or other marine particles) and remain associated with them (Kowalewska & Konat 1997, Larson et al. 2018, Parsons et al. 2014), thereby translocating them from the dissolved to the particulate phase in water. Sorption of dissolved compounds to surfaces depends on the molecular characteristics of both the oil compound and the particle, making sorption a compound-specific process. Coating of particles and trapping of oil droplets in aggregates, however, is likely nonselective (Wirth et al. 2018), and we have categorized this as a form of transport (see below). Either way, association with the particulate phase will change the fate of the oil compound, as particles may sink or be trapped by filter feeders, whereas dissolved substances move with the water. Oil droplets, although liquid, may be retained by filters or coagulate, acting as particles in those cases (and chemists often refer to oil droplets as particulates).

4.2. Transformation Processes

Transformation processes, in contrast to mixture separation, alter the molecular structure of individual compounds due to full or partial oxidation via light or biological activity. Oxidation reactions are transformations that create distinct new molecules by adding oxygen. Photochemical reactions, which occur within days to weeks, generally lead to partial oxidation (Ward et al. 2018), whereas biodegradation (see the sidebar titled How Important Was Biodegradation During and After the *Deepwater Horizon* Oil Spill?) may lead to organic metabolites, biomass, or full oxidation

HOW IMPORTANT WAS BIODEGRADATION DURING AND AFTER THE *DEEPWATER HORIZON* OIL SPILL?

Nearly 90% of compounds from the DwH oil were degradable, and the microbial community composition changed in all oiled environments (i.e., in the water, in sediments, and onshore). However, effective biodegradation also depends on environmental conditions. Nutrients are required, which in the DwH case were frequently lacking in surface waters. Rapid biodegradation also requires oxygen, which became limiting when oil was buried ashore or at the seafloor, where much slower anoxic biodegradation dominated. Estimates suggest that roughly half of the oil compounds in the intrusion layer were biodegraded. Overall, biodegradation varied greatly between different environments, and reliable estimates of the total amount of oil fully biodegraded or metabolized are lacking.

to CO_2. Biodegradation often refers to microbial metabolic processes, but eukaryotes may also ingest and metabolize or store oil compounds.

4.2.1. Photochemical reactions.

Photooxidation was of primary importance for the DwH oil at the sea surface, where the slick was exposed to sunlight, and it is believed that at least half of the floating oil (by mass) was photooxidized (Ward et al. 2018). For example, PAHs and alkanes were converted to oxidized derivates (Stout et al. 2016). One study estimated that during the 102 days of surface oiling, the average daily oxygen uptake for partial photooxidation of oil compounds in the surface slick was 360 mmol O_2 (Ward et al. 2018). However, biodegradation may also result in partial oxidation and may have contributed to some extent. The organic oxidation products span a continuum of oil-soluble, interfacial (e.g., surfactant-like), and water-soluble compounds. The fate and effects of these oxidation products on organisms, food webs, and ecosystems await discovery.

Photooxidation was limited after oil was stranded on shorelines (Ward et al. 2018). Estimates suggest that, for oil stranded on beaches, 75% of the partial oxidation observed after six years happened within the first 10 days as oil floated at the sea surface, emphasizing that loss processes are greatly reduced once the oil is ashore (Ward et al. 2018). Oil-soluble and interfacial compounds typical for photooxidized oil constituted a large part of the oil that was washed ashore. The drastically reduced importance of photooxidation once oil was ashore is due in large part to the reduced exposure to sunlight, as oil was deposited as submerged oil mats or in thick oil layers, where light could only penetrate the upper 70 μm (Ward et al. 2018).

4.2.2. Biodegradation.

Microbial oxidation requires nutrient and trace element accessibility, as well as bioavailable substrates such as oil compounds (Sun & Kostka 2019). Oxygen availability promotes the biodegradation of oil, although hydrocarbon degradation may also occur under anoxic conditions, albeit slowly (Head et al. 2006, Joye et al. 2016b). Oil-degrading microbes, which can use specific hydrocarbons as a food source, are typically rare but respond with rapid growth to the input of oil. As specific substrates are depleted, the first-responder species are replaced with others, leading to a succession of dominating hydrocarbon degraders (Head et al. 2006, Joye & Kostka 2020). Additionally, microbes may respond to oil with the production of exudates that act as surfactants, emulsifiers, or a matrix for biofilm formation. Specific microbes may interact with individual hydrocarbon compounds or metabolites specializing in specific metabolic steps, creating collaborative networks that allow the joint utilization of complex organics such as oil (McGenity et al. 2012). Such collaborative microbial networks exist in biofilm-like structures populated by a diverse microbial community. Bacteria–oil aggregations (BOAs) represent biofilms formed not on a solid surface but at the oil–water interface (Burd et al. 2020).

In the Gulf of Mexico, microbial communities did respond strongly to the input of the DwH oil, with hydrocarbon-degrading species dominating communities in all contaminated environments (Joye & Kostka 2020, Joye et al. 2016b). Experiments confirmed that many of the compounds in the DwH oil, including those in the intrusion layer and the surface slick, were principally bioavailable, and conversion to biomass or to CO_2 was possible (e.g., Prince et al. 2017), with only an estimated 10% of the initial insoluble crude residues highly resistant to biodegradation (Joye & Kostka 2020, Joye et al. 2014). However, actual biodegradation rates varied widely depending on environmental conditions (**Figure 1**). In general, degradation rates were highest for low-molecular-weight compounds, but low- and medium-weight straight-chained alkanes (up to n-C_{25}) and two- and three-ringed PAHs were also biodegradable if conditions were right (Joye et al. 2016b). Hydrocarbons in the $>n$-C_{25} class, which include asphaltenes, resins, and oil-soluble photooxidation products, are insoluble and solid in nature and degrade orders of magnitude more slowly than $<n\text{-C}_{25}$ compounds.

4.2.2.1. Biodegradation at the sea surface. At the sea surface, nutrient limitation is thought to have inhibited biodegradation (Edwards et al. 2011), although the ability to fix nitrogen is common among some hydrocarbon-degrading microbes (Joye & Kostka 2020). Microbial activity, respiration rates, enzyme activity, and hydrocarbon respiration were stimulated inside an oil patch, suggesting degradation potential. However, although oil-degrading microbes dominated within the slick, no microbial bloom formed, and growth appeared to be severely phosphorus limited (Edwards et al. 2011). Indeed, biodegradation of hydrocarbons was found to be mostly negligible, based on the changes in the chemical composition of the floating oil (Ward et al. 2018). In this context, the large spatial and temporal heterogeneity in nutrient availability should be noted; the formation of large phytoplankton blooms in the area of influence of the Mississippi River suggests temporary and local nutrient availability at coastal sites. High spatial and temporal patchiness may have biased overall estimates.

Additionally, the observation in May 2010 of centimeter-sized BOAs at the edges of the surface slicks (Passow et al. 2012, Ziervogel et al. 2012) may indicate hot spots for microbial oil degradation (Gutierrez et al. 2018, Hatcher et al. 2018). Hydrocarbon-degrading bacteria form biofilms at the oil–water interface, a process that includes the production of biosurfactants that reduce interfacial tension (Omarova et al. 2019). When young, such biofilms may lead to increased dispersion of an oil slick upon agitation, but with age, they turn more rigid, decreasing the dispersibility of the oil slick (Omarova et al. 2019). Such biofilms breaking off from the main slick would explain the appearance of marine-snow-sized BOAs at the sea surface. Microbial and enzyme activity increased within such microbial aggregations (Arnosti et al. 2016), which may provide microenvironments with localized nutrient recycling and possibly sites for efficient biodegradation of oil compounds (Hatcher et al. 2018). High hydrocarbon degradation rates within the microenvironment of the BOAs, which eventually sink (Passow et al. 2012, Ziervogel et al. 2012), would remove evidence of biodegradation from the surface ocean.

4.2.2.2. Biodegradation of oil stranded on the coastline. The biodegradation rates of stranded oil vary widely depending on the environment. Biodegradation was visible within weeks in some stranded oil deposits (Stout et al. 2016); for example, at Pensacola Beach, where oil filtered into the sediment, the warm, well-aerated, and physically dynamic beach environment led to half-lives of less than a month (Kostka et al. 2020b). Sediment–oil agglomerates that are buried in sandy beaches lose approximately 85% of alkanes and 99% of PAHs within 3 years and may be completely biodegraded (<0.1% of their mass) after 30 years (Bociu et al. 2019). The sand embedded in sediment–oil agglomerates makes them porous, allowing oxygen, moisture, and nutrients to penetrate to the core, especially if they are buried in tidally impacted and ventilated beach sand, where rainfall, runoff, and decay add nutrients. Sediment–oil agglomerates lying on top of the sand biodegrade more slowly, but additional mechanical and photooxidative processes result in similar overall loss rates (Bociu et al. 2019). By contrast, the biodegradation of even PAHs and alkanes is extremely slow in oil mats buried in anaerobic layers of marsh sediments—on the order of multiple decades, with annual loss rates in Louisiana marshlands below detection between 2013 and 2018 (Turner et al. 2019). At Jimmy Bay, nearly all $n\text{-C}_9\text{–C}_{20}$ alkanes, as well as two- and three-ring PAHs, were lost after five years, but the levels of remaining oil (measured as total hydrocarbons) were still elevated compared with prespill levels (Duan et al. 2018). Even now, 10 years after the spill, oil originating from the accident occasionally washes up on beaches; this oil likely originates from sites less favorable to biodegradation (Gustitus & Clement 2017).

4.2.2.3. *Biodegradation in the deep intrusion layer.* Microbial degradation dominated oxidation in the deep intrusion layer and led to the removal of hydrocarbons within days to weeks (Camilli et al. 2010, Hazen et al. 2010, Joye et al. 2011, Kessler et al. 2011). In fact, the extension of the deep intrusion layer could be measured by the negative O_2 anomaly caused by respiration (Du & Kessler 2012). Budget calculations based on the removal of O_2 suggest that up to 60% of hydrocarbons in the intrusion layer were respired by September 20, when oxygen utilization dropped to zero (Du & Kessler 2012). Estimates of biodegradation rates vary widely (Camilli et al. 2010, Hazen et al. 2010). Methane degradation rates, which were directly measured, seemed inefficient (Crespo-Medina et al. 2014, Valentine et al. 2010); trace metal availability likely limited methane biodegradation (Shiller et al. 2017), but the quantitative role of methanotrophs is controversial (Dubinsky et al. 2013, Joye et al. 2016a).

The succession of biodegraders was studied in the intrusion layer, where three taxa dominated: Oceanospirillales, *Colwellia*, and *Cycloclasticus* (Kostka et al. 2020a, Redmond & Valentine 2011). A combination of field study and laboratory experiments revealed that the succession was driven by changes in hydrocarbon availability (Dubinsky et al. 2013, Hu et al. 2017): Within 2–3 weeks, the microbial abundance increased sixfold, and *Bermanella* (a member of the Oceanospirillales) contributed more than 33% to the microbial community, which utilized primarily linear low- and medium-weight *n*-alkanes (Kleindienst et al. 2016, Yang et al. 2016a). Later, when concentrations of linear alkanes were low, *Bermanella* contributed less than 2% to the community (Hu et al. 2017). The shift from alkane to aromatic degradation characterized the next stage and was accompanied by the dominance of species from the genera *Colwellia* and *Cycloclasticus* (Hu et al. 2017). Finally, the microbial community consisted of a diverse mix of microbes capable of degrading PAHs (Dubinsky et al. 2013, Hu et al. 2017). The continuous influx of new, nonweathered oil would likely have obfuscated such a succession in situ.

Some microbes formed biofilms around oil droplets, yielding micro-BOAs (~50–100 μm) (Baelum et al. 2012, Brakstad et al. 2015, Doyle et al. 2018, Kleindienst et al. 2015b, Wang et al. 2016). Within such BOAs, alkanes were degraded with half-lives of 0.6–9.5 days, whereas degradation of two- and three-ringed PAHs started after 9–18 days, and utilization of four- to six-ringed PAHs began after 18 days (Wang et al. 2016).

4.2.2.4. *Biodegradation in deep-sea sediments.* The microbial community at the seafloor also responded to the input of weathered oil (Yang et al. 2016b), but microbial activity and oil degradation rates were low to moderate, even when bioavailable oil compounds were present at high concentrations, largely because of oxygen limitation (Kostka et al. 2020b, Ziervogel et al. 2016b). The initial biodegradation depletes oxygen rapidly, and resupply by diffusion or bioturbation is generally slow or transient in deep-sea sediments. On timescales of months, the microbial community responded with elevated sulfate reduction and methanogenesis, implying anoxic oil degradation, which is appreciably slower than oxic biodegradation. Less than 1% of the DwH oil was removed in five months via this pathway (Orcutt et al. 2017). The composition of oil in sediments one year after the spill also points to slow but clear biodegradation, with labile *n*-alkanes and BTEX still present in sediments in May 2011 (Liu et al. 2012). Transient resuspension events induced by storms (e.g., after Hurricane Isaac) temporally reinvigorated biodegradation (Ziervogel et al. 2016a). The effects of hydrostatic pressure on biodegradation rates are only beginning to be explored (Kostka et al. 2020a).

4.2.3. Ingestion. Following the isotopic signature of the fossil carbon, the uptake of DwH oil could be traced through the planktonic, coastal, and mesopelagic food webs (Chanton et al. 2012, Graham et al. 2010, Quintana-Rizzo et al. 2015, Wilson et al. 2016). Grazing on bacteria that grew

HOW WAS THE *DEEPWATER HORIZON* OIL DISTRIBUTED WITHIN THE ENVIRONMENT?

Water movements such as currents, waves, and mixing shaped the distribution of oil at the surface and in the intrusion layer at 1,000 m. Wind additionally affected surface oil distribution. The buoyancy of large oil droplets led to their ascent and the accumulation of oil at the sea surface, while the association with ballasting particles transferred oil to depth, depositing it onto the seafloor. Whereas mixing and currents usually dilute oil, gravitational processes reconcentrate oil at the sea surface or the seafloor.

on DwH oil or on their predators transfers the isotopic signature of the fossil carbon (Δ^{14}C or δ^{13}C) up the food chain but provides no information on where in the trophic cascade the fossil carbon was metabolized. Eukaryotes may directly ingest oil compounds (e.g., via contaminated prey) or whole droplets (when filter feeding or drinking). Oil compounds were directly measured in organisms belonging to different trophic levels, including zooplankton such as crustaceans (Mitra et al. 2012) or salps (Lee et al. 2012) and fish (Murawski et al. 2014, Snyder et al. 2015).

5. TRANSPORT PROCESSES

Transport processes move oil within the marine environment, generally without altering its chemical composition (see the sidebar titled How Was the *Deepwater Horizon* Oil Distributed Within the Environment?). Physical transport processes include the horizontal movement of oil by currents and winds as well as the vertical mixing of oil, allowing the entrainment of oil (e.g., the dispersion of oil as droplets and the formation of emulsions). Transport processes also include the movement of oil due to gravitational forces. The buoyancy of oil droplets leads to their upward movement, whereas the association of droplets with sinking particles leads to their sedimentation. Interactions between marine particles and oil is a broad topic that has been insufficiently explored (**Figure 2**). Whereas the net effect of currents and mixing is dilution (i.e., a decrease in oil concentration), except if the oil is carried ashore, the gravitational processes result in the reconcentration of oil—e.g., during the surfacing of buoyant droplets or the deposition of sinking oil-containing marine snow or particles.

5.1. Physical Transport Processes

Circulation models are used to predict the likely physical distribution of oil at the sea surface and in the water and the likelihood of its stranding, and are used to aid in decision-making regarding response measures. Mesoscale circulation patterns (e.g., >10–20 km) in the northern Gulf of Mexico are complex, variable, and not easily predictable (Cardona & Bracco 2016); submesoscale dynamics also impact circulation patterns and appreciably affect the transport of oil (Luo et al. 2016). In a simplified way, currents in the northern Gulf of Mexico may be explained by a two-layer system, consisting of (*a*) an upper layer, which is dominated by the Loop Current and its eddies, and (*b*) currents below, which are influenced by topographic Rossby waves (large inertial waves) and seafloor topography. Both current systems display distinctly different circulation patterns (Cardona & Bracco 2016). Near-surface circulation along the north shelf of the Gulf of Mexico is additionally influenced by wind stress (Le Hénaff et al. 2012) and salinity gradients created by the discharge of the Mississippi River (Cardona & Bracco 2016, Kourafalou & Androulidakis 2013).

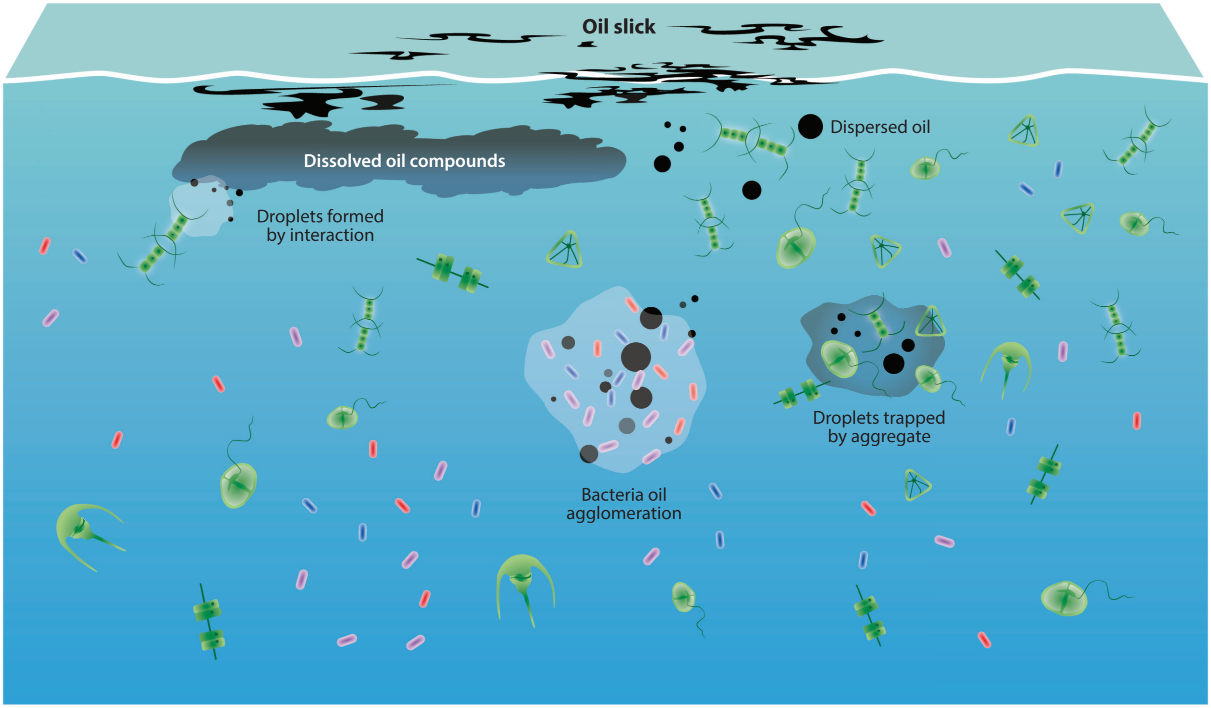

Figure 2

Artistic rendition of oil–particle interactions. Not only does oil distribute in the ocean in different phases (e.g., it exists as a surface slick, dispersed droplets, or dissolved substances), but it also interacts with a large variety of marine particles, microbes, and multicellular organisms. Oil may benefit microbes, which use it as a substrate, or it may impart injury (e.g., photosystem damage or skin lesions) or even lead to cell death. Some bacteria surround oil droplets, forming a biofilm that allows biodegradation of oil. Particles (e.g., phytoplankton) that coagulate inadvertently trap oil droplets within their aggregate matrix. Some microbes respond by releasing substances that may disperse or emulsify oil.

5.1.1. Transport of floating oil. The visible DwH oil surface expression occupied a continually changing area, with the highest surface oil concentrations near the spill site but extending far from the source (MacDonald et al. 2015). The total (cumulative) area impacted was nearly 150,000 km^2, reaching from Louisiana to the northern coastline of Florida (Kourafalou & Androulidakis 2013, Weisberg et al. 2016). The ever-changing daily surface expression of the oil, which was an order of magnitude smaller than the cumulative footprint (MacDonald et al. 2015), was determined primarily by the above-mentioned meso- and submesoscale circulation patterns and fronts (Luo et al. 2016). Only a small fraction of the floating oil was entrained in the Loop Current, and only during a transient event, because in 2010, the Loop Current was unusual in that it remained south of the spill site. Southerly winds moved oil northward, and riverine outflow was periodically strong in 2010, enhanced by the opening of the freshwater diversions along the lower Mississippi River (Kourafalou & Androulidakis 2013). An eddy to the east of the Mississippi mouth retained particles and oil slicks, and a strong eastward-flowing front along the shelf break transported surface oil to the Mississippi–Alabama–Florida shelf regions and into the DeSoto Canyon area (Weisberg et al. 2016).

5.1.2. Transport of oil in the deep intrusion layer. Most of the oil in the deep intrusion layer moved southwestward and was traceable for up to 400 km (Spier et al. 2013), but some northeastward movement also carried the intrusion layer into the DeSoto Canyon region (Bracco et al. 2016). Direct impingement of the intrusion layer along the continental slope led to the "dirty bathtub ring" at the depth of the intrusion (Hastings et al. 2016, Romero et al. 2015, Stout et al. 2017). A model tracer experiment revealed that even after one year, a significant fraction of tracer dye was still concentrated near the release site and in the south and southwest (Bracco et al. 2016). This is consistent with the finding that oil residues lingered for more than a year after the leak was sealed (Passow & Stout 2020), although by mid-September 2010, the deep intrusion layer was not detectable in water samples (Du & Kessler 2012).

5.1.3. Stranding. Model estimates suggest that 10–30% of the surface oil made landfall (Boufadel et al. 2014). Most oil strandings occurred after June. However, appreciable beaching occurred in May along the Chandeleur Islands and Louisiana's Mississippi River outlet marshes. Overall, the vast majority of all strandings (60–90%) occurred along the Louisiana shorelines, but oil also reached Mississippi, Alabama, and Florida coastlines, with a total of more than 2,113 km of shores oiled (Nixon et al. 2016). Approximately half of the impacted shorelines were beaches and half wetlands, with 40% and 30%, respectively, experiencing heavy oiling (Nixon et al. 2016).

By the time that oil arrived nearshore, it was largely heavily weathered, and a viscous emulsion that consisted of mostly insoluble, nonvolatile, high-molecular-weight compounds (largely alkanes $> \sim n\text{-C}_{14}$ and mostly the three- to six-ringed PAHs) as well as partially oxidized compounds (Bociu et al. 2019, Ward et al. 2018). Oil that washed onto sandy beaches formed puddles and filtered down into the sediment (50–70 cm) or interacted with sediment to form macroscopic sediment–oil agglomerates and sediment–oil mats (Gustitus & Clement 2017). Oil, sediment–oil agglomerates, and sediment–oil mats formed submerged deposits, which provided a source of continued contamination when leaching or being resuspended (Rouhani et al. 2017). Overall, oiling of beaches showed high spatial and temporal heterogeneity, reflecting the highly dynamic environment, where deposits are repeatedly buried, unearthed, remobilized, and transported along the shoreline or cross-shelf (Nixon et al. 2016).

When oil reached salt marshes, it was absorbed into sediments or stayed on the sediment and grass surfaces. For example, at the Jimmy Bay salt marshes (a heavily impacted area), concentrations increased from prespill background values of 4–90 mg of total hydrocarbons per kilogram of sediment to peak values of 510,000 mg per kilogram of sediment (Duan et al. 2018). Peak concentrations of target aromatics and alkanes in surficial marsh sediments 10 m from the shoreline of Louisiana marshes increased by two to three orders of magnitude (Turner et al. 2019). Initially, oil distribution was patchy, and concentrations decreased with distance from the shoreline. As oil was redistributed on timescales of months, distribution patterns became more even; patchiness decreased, and 100-m transects showed no gradients in oil concentration (Turner et al. 2019). This redistribution led to lower concentrations (by a factor of 100) compared with those from a year earlier.

5.2. Gravitational Processes

Gravitational forces also affect the distribution and fate of oil within the ocean. Oil, when released below the surface, may ascend due to buoyancy or sink toward the seafloor when associated with ballasting particles. The rapid upward movement of the escaping DwH oil, which determined its initial distribution and weathering, was described in Section 2. Different types of oil–particle interactions (Burd et al. 2020) that lead to sedimentation and deposition of oil at the seafloor, including the so-called MOSSFA (marine oil snow sedimentation and flocculent accumulation) events (see the sidebar titled What Is MOSSFA?), are discussed next.

WHAT IS MOSSFA?

MOSSFA stands for marine oil snow sedimentation and flocculent accumulation and describes the gravitational settling of oil in association with ballasting particles and its deposition onto the seafloor. The sedimentation of buoyant oil to the seafloor was not anticipated by responders and decision makers, but MOSSFA was a significant process during the DwH spill, depositing approximately 20% of the oil that was not reclaimed onto the seafloor. Different types of oil–particle associations led to MOSSFA events, including (*a*) the aggregation and sedimentation of large phytoplankton blooms that formed MOS; (*b*) the formation of BOAs, which are biofilm-like structures initiated by microbes in response to oil exposure; and (*c*) the formation of OPAs, where fine sediment particles, such as drilling mud, coat and penetrate oil droplets.

5.2.1. Oil–particle aggregates. Small sediment particles coat oil droplets, and depending on the relative contributions and densities of the oil and the sediment grains, such oil–particle aggregates (OPAs, previously called oil–mineral aggregates) (Omotoso et al. 2002, Stoffyn-Egli & Lee 2002) may sink rapidly (hundreds of meters per day). This mechanism is relatively well studied (Gong et al. 2014, Khelifa et al. 2008). OPA formation is important near shore, where sediment grains are mixed into the water column, but during the DwH spill, natural concentrations of fine sediment particles were too small, even in the turbidity layer, to initiate measurable OPA formation (U. Passow, unpublished data). However, OPA formation was important when drilling mud was added at the wellhead during attempts to end the spill. The seafloor deposition of olefins, a marker for the DwH drilling mud, suggests OPA formation and sedimentation in a 6.5-km^2 footprint around the DwH site (Stout & Payne 2017, Yan et al. 2016).

5.2.2. Marine oil snow and bacteria–oil aggregations. A significant fraction of the DwH oil sank to the seafloor during MOSSFA events (Burd et al. 2020, Daly et al. 2016), with the sinking oil originating from the surface (Stout & German 2018) or from the intrusion layer (Passow & Stout 2020). Different types of marine oil snow (MOS), including large BOAs (Baelum et al. 2012, Doyle et al. 2018, Kleindienst et al. 2015b), zooplankton-derived MOS (Lee et al. 2012, Mitra et al. 2012), and aggregates formed by the coagulation of marine particles (e.g., algae) and oil droplets (Passow 2016; Passow et al. 2017, 2019), led to different types of MOSSFA events (Burd et al. 2020, Passow & Ziervogel 2016). Whereas BOA formation is a microbial response to oil input, the formation of marine snow is independent of the presence of oil, although coagulation rate and aggregate characteristics are impacted by oil that is trapped within marine snow (Francis & Passow 2019, Wirth et al. 2018).

Sedimentation of phytoplankton MOS was important during the DwH spill, as evidenced by cosedimentation and codeposition of algae and oil (Burd et al. 2020, Larson et al. 2018, Passow & Stout 2020) and supported by modeling experiments (Francis & Passow 2019). The relative importance of large, mucus-rich BOAs (which had an isotopic carbon signature identical to that of the oil) for sedimentation is difficult to assess (Passow et al. 2012, Ziervogel et al. 2012), and the relationship between such marine-snow-sized BOAs and the small (<0.1 mm) BOAs as observed under intrusion-layer conditions (Baelum et al. 2012, Doyle et al. 2018, Kleindienst et al. 2015b) is currently unclear.

During their descent, the fate of sinking MOS, BOAs, and OPAs depends on several physical and biological processes, including horizontal advection by currents, physical and biological fragmentation, consumption, and microbial decomposition (Daly et al. 2020). Loss processes during sinking of MOS, BOAs, and OPAs are largely unexplored (Burd et al. 2020), although some models have attempted to integrate these processes (Dissanayake et al. 2018, Francis & Passow 2019). However, we know that attenuation of marine snow with depth due to bacterial degradation and grazing is high: Marine snow is a valuable food resource for organisms in the twilight zone.

5.2.3. Deposition at the seafloor. The MOSSFA events deposited a flocculent, oil-contaminated layer onto the seafloor (Chanton et al. 2015, Stout et al. 2017, Valentine et al. 2014), affecting pristine deep-sea environments and organisms such as corals (Fisher et al. 2014). When fresh, flocculent phytodetritus is deposited onto the deep seafloor, it commonly lasts hours to days before being grazed, degraded, bioturbated, resuspended, or redistributed (Lampitt et al. 1993). By contrast, MOSSFA events during the DwH spill frequently incapacitated infaunal communities and inhibited bioturbation because of the very high deposition rates (4–30 times higher than prespill rates) (Baguley et al. 2015, Larson et al. 2018, Montagna et al. 2013, Schwing et al. 2015). Thus, the flocculent layer prevailed for months to years, and bioturbation only resumed between 2013 and 2015 (Larson et al. 2018).

The documented footprint of the DwH oil deposition onto the seafloor was larger than 110,000 km^2, with the southern edge of the demarcated area not well defined due to a lack of sampling (Romero et al. 2017). DwH oil–contaminated seafloor was found more than 500 km from the site of the accident, and total hydrocarbon deposition was estimated to equal roughly 20% of the unrecovered spilled oil.

The footprint on the seafloor reflects the multidirectional transport of oil at the sea surface and in the intrusion layer and clearly reflects the two origins (Romero et al. 2017, Stout et al. 2017): At coastal, nearshore sites, concentrations of *n*-alkanes and biomarkers reached peak values, in accordance with the fact that the deposited material originated from the surface slick (Ward et al. 2018), whereas at the deep-sea sites, deposited PAHs were highest, indicating that deposited oil likely originated mainly from the deep intrusion layer (Romero et al. 2017, Stout & Payne 2017). These findings are consistent with sediment trap data (Passow & Stout 2020, Stout & German 2018). Additionally, pyrogenic PAHs stemming from incomplete combustion during the in situ burns, which were not conducted near shore, were found in deposits and traps at depths greater than 1,000 m (Romero et al. 2017, Stout et al. 2017).

The footprint of oil accumulated on the seafloor only partially matched the cumulative surface expression of floating oil. Oil deposition on the seafloor was high in the proximity of the spill site, under the intrusion layer, and at coastal stations, with the vast majority (90%) of the deposited oil accumulated at shallow sites (depth <200 m) (Romero et al. 2017). There are two explanations for this bias of the seafloor deposits toward coastal stations. First, very little is known about flux attenuation of MOS, but the loss rates of sinking marine snow with depth are high, with, on a global average, a 90% loss between the base of the euphotic zone and 1,000-m depth. Microbial degradation and grazing by deep-sea organisms are largely responsible for these losses. Accumulation of oil on the seafloor is thus expected to be much smaller at 1,500 m than at a shallower site, even if sedimentation rates at the base of the euphotic zone are similar at both sites. Second, MOSSFA events driven by phytoplankton blooms require that the aggregation and sedimentation of a phytoplankton bloom co-occur with the presence of oil. Daily map overlays of chlorophyll *a* concentrations of at least 10 $\mu g\ L^{-1}$ with the daily surface footprint of the floating oil indicate that conditions for phytoplankton-driven MOSSFA events were clearly biased toward coastal stations (**Figure 3**). Phytoplankton blooms occurred predominantly where Mississippi River outflow provided nutrients.

The observed small-scale patchiness in oil deposition is easily explained by the combination of physical redistribution and transport processes. Horizontal transport during sinking (Liu et al.

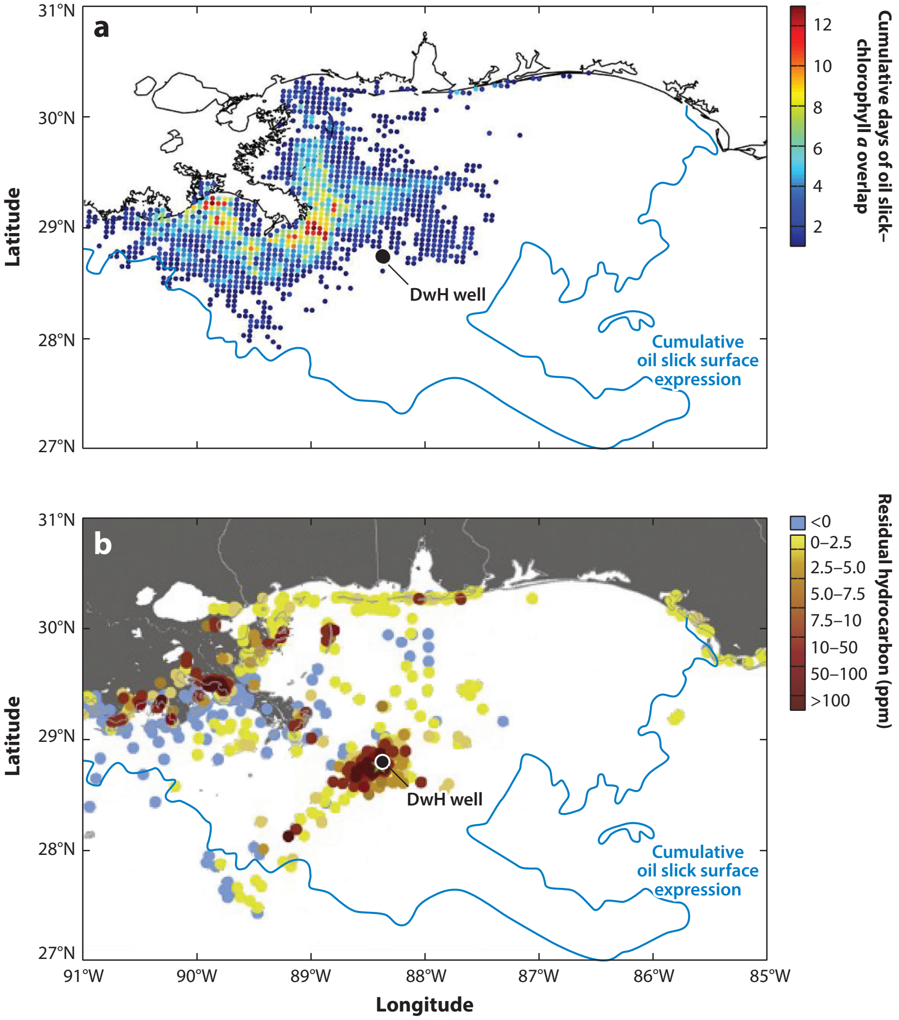

Figure 3

(*a*) Co-occurrence of high chlorophyll *a* concentrations and floating oil, which would potentially result in the sedimentation of oil due to phytoplankton aggregates. The colored circles indicate the cumulative number of days of overlap of the surface oil slick with the area of a chlorophyll concentration of at least 10 μg L^{-1}. The brown line shows the approximate cumulative expression of the oil slick at the sea surface during the DwH spill (as shown in MacDonald et al. 2015, figure 3), and the black circle is the location of the DwH well. The probability of a phytoplankton MOSSFA event is especially high in the light blue to red areas. The focus of predicted MOSSFA deposition on coastal sites is consistent with observed deposition patterns (see panel *b*). The model compared daily surface expression of DwH oil (MacDonald et al. 2015) with daily satellite-derived chlorophyll concentrations (Francis 2020) and calculated days of overlap in each binned area (5 km^2). (*b*) Residual hydrocarbon deposits on the seafloor (as shown in Romero et al. 2017, figure 4) along with the same surface expression of oil as in panel *a*. Abbreviations: DwH, *Deepwater Horizon*; MOSSFA, marine oil snow sedimentation and flocculent accumulation.

2018), as well as the resuspension and redistribution of settled MOS by bottom currents (Diercks et al. 2018), led to accumulation patterns favoring local valleys.

6. OUTLOOK: THE *DEEPWATER HORIZON* LEGACY AND RECOVERY

The intense research activity triggered by the accident has revealed details of the processes leading to the formation of a deep intrusion layer and to the occurrence of MOSSFA events, and has confirmed and emphasized the importance of photochemical oxidation as well as uncovered the genetic and functional relationships between microbial communities and biodegradation in different environments. Our understanding of the natural capacity of microbes to catalyze the bioremediation and to restore and maintain ecosystem balance has greatly expanded (Joye & Kostka 2020). Much has been learned regarding the response of organisms and ecosystem to such a large oil input. While many questions remain, these and other novel insights into the weathering and transport processes driving the fate of spilled oil allow the more accurate formulation of models that may help improve future response activities and plans.

Ten years after the accident, small amounts of DwH oil can still be found in some environments and has long ago disappeared in others. In the water, the hydrocarbon concentration fell below detection within weeks after the spill ended (but see Kolian et al. 2015), whereas the fossil carbon signal in material collected with sediment traps took several years to return to prespill values (Chanton et al. 2018, Giering et al. 2018) (**Figure 4**). Some of the weathered oil stranded in marshes or deposited onto the seafloor can still be found a decade later (Bociu et al. 2019, Gustitus & Clement 2017, Larson et al. 2018, Turner et al. 2019), but occurrences are patchy and greatly reduced. This review has focused on the fate of the spilled oil, but the impacts on organisms, ecosystems, and humans also span a wide range of timescales, with their own recovery times. The

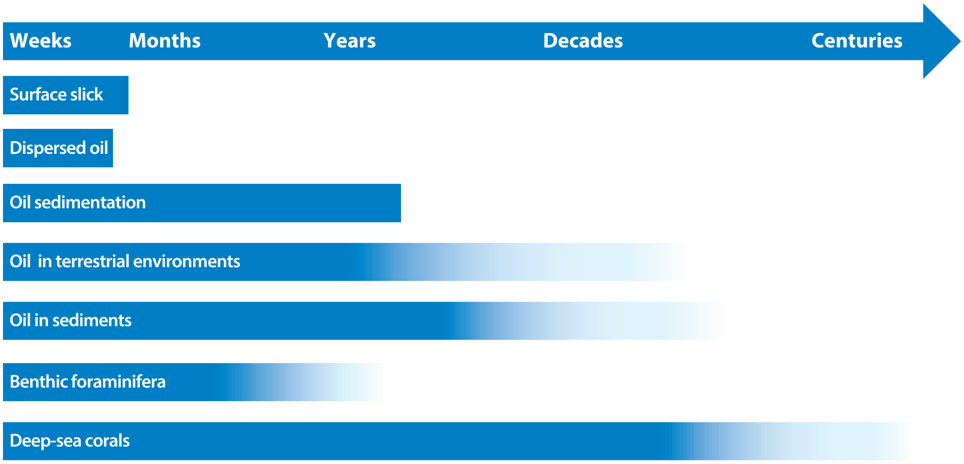

Figure 4

Estimated timescales of the persistence of measurable amounts of oil in different environments once the *Deepwater Horizon* spill ended, along with two examples of timescales of organism recovery from the impacts of the oil. The gradients denote the variability of recovery time depending on environment or species. The long recovery time of oil sedimentation, in comparison with dispersed oil in the water, suggests that although it was below detection limit, dispersed oil did remain in the water for years after the spill ended.

diversity of benthic infauna and foraminifera started to recover after one to three years (Schwing et al. 2018, Washburn et al. 2017), with other organisms taking longer (Lewis & Ricker 2020); for example, it is expected to take decades for dolphins to recover from long-term, sublethal effects such as chronic lung disease, impaired stress responses, and reduced reproductive success (Smith et al. 2017), and recovery of the slow-growing deep-sea corals to prespill values is predicted to take decades to centuries (Girard & Fisher 2018, Girard et al. 2019). The appreciable long-term decrease of deep-sea micronekton, which could have large impacts on food webs and productivity, is only just being discovered (T.T. Sutton, R.J. Milligan, A.B. Cook. K.M. Boswell, M. D'Elia, et al., manuscript in review). This raises the questions of whether the ecosystems of the Gulf of Mexico will return to prespill conditions or find a new state, and whether we would be able to identify a shift. The question of the recovery of the Gulf of Mexico from the spill is thus a matter of viewpoint and scale.

DISCLOSURE STATEMENT

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LITERATURE CITED

Aeppli C, Nelson RK, Radović JR, Carmichael CA, Valentine DL, Reddy CM. 2014. Recalcitrance and degradation of petroleum biomarkers upon abiotic and biotic natural weathering of *Deepwater Horizon* oil. *Environ. Sci. Technol.* 48:6726–34

Aman ZM, Paris CB, May EF, Johns ML, Lindo-Atichati D. 2015. High-pressure visual experimental studies of oil-in-water dispersion droplet size. *Chem. Eng. Sci.* 127:392–400

Arnosti C, Ziervogel K, Yang T, Teske A. 2016. Oil-derived marine aggregates – hot spots of polysaccharide degradation by specialized bacterial communities. *Deep-Sea Res. II* 129:179–86

Ashton BM, East RS, Walsh MM, Miles MS, Overton EB. 2020. *Studying and Verifying the Use of Chemical Biomarkers for Identifying and Quantitating Oil Residues in the Environment*. New Orleans, LA: US Dep. Interior Miner. Manag. Serv.

Baelum J, Borglin S, Chakraborty R, Fortney JL, Lamendella R, et al. 2012. Deep-sea bacteria enriched by oil and dispersant from the Deepwater Horizon spill. *Environ. Microbiol.* 14:2405–16

Baguley J, Montagna P, Cooksey C, Hyland J, Bang H, et al. 2015. Community response of deep-sea soft-sediment metazoan meiofauna to the Deepwater Horizon blowout and oil spill. *Mar. Ecol. Prog. Ser.* 528:127–40

Beaulieu SE, Sengco MR, Anderson DM. 2005. Using clay to control harmful algal blooms: deposition and resuspension of clay/algal flocs. *Harmful Algae* 4:123–38

Bociu I, Shin B, Wells WB, Kostka JE, Konstantinidis KT, Huettel M. 2019. Decomposition of sediment-oil-agglomerates in a Gulf of Mexico sandy beach. *Sci. Rep.* 9:10071

Boufadel MC, Abdollahi-Nasab A, Geng X, Galt J, Torlapati J. 2014. Simulation of the landfall of the Deepwater Horizon Oil on the shorelines of the Gulf of Mexico. *Environ. Sci. Technol.* 48:9496–505

Bracco A, Choi J, Joshi K, Luo H, McWilliams JC. 2016. Submesoscale currents in the northern Gulf of Mexico: deep phenomena and dispersion over the continental slope. *Ocean Model.* 101:43–58

Brakstad OG, Nordtug T, Throne-Holst M. 2015. Biodegradation of dispersed Macondo oil in seawater at low temperature and different oil droplet sizes. *Mar. Pollut. Bull.* 93:144–52

Burd AB, Chanton JP, Daly KL, Gilbert S, Passow U, Quigg A. 2020. The science behind marine-oil snow and MOSSFA: past, present, and future. *Prog. Oceanogr.* 187:102398

Camilli R, Reddy C, Yoerger D, Mooy BV, Jakuba M, et al. 2010. Tracking hydrocarbon plume transport and biodegradation at Deepwater Horizon. *Science* 330:201–4

Cardona Y, Bracco A. 2016. Predictability of mesoscale circulation throughout the water column in the Gulf of Mexico. *Deep-Sea Res. II* 129:332–49

Chaeruh SK, Tazaki K, Asada R, Kogure K. 2005. Interaction between clay minerals and hydrocarbon-utilizing indigenous microorganisms in high concentrations of heavy oil: implications for bioremediation. *Clay Miner.* 40:105–14

Chanton JP, Cherrier J, Wilson RM, Sarkodee-Adoo J, Bosman S, et al. 2012. Radiocarbon evidence that carbon from the Deepwater Horizon spill entered the planktonic food web of the Gulf of Mexico. *Environ. Res. Lett.* 7:045303

Chanton JP, Giering SLC, Bosman SH, Rogers KL, Sweet J, et al. 2018. Isotopic composition of sinking particles: oil effects, recovery and baselines in the Gulf of Mexico, 2010–2015. *Elem. Sci. Anthr.* 6:43

Chanton JP, Zhao T, Rosenheim BE, Joye S, Bosman S, et al. 2015. Using natural abundance radiocarbon to trace the flux of petrocarbon to the seafloor following the Deepwater Horizon oil spill. *Environ. Sci. Technol.* 49:847–54

Chen L, Levine JS, Gilmer MW, Sloan ED, Koh CA, Sum AK. 2014. Methane hydrate formation and dissociation on suspended gas bubbles in water. *J. Chem. Eng. Data* 59:1045–51

Comm. Underst. Oil Spill Dispersants Effic. Effects. 2005. *Understanding Oil Spill Dispersants: Efficacy and Effects*. Washington, DC: Natl. Acad. Press

Crespo-Medina M, Meile C, Hunter K, Diercks A, Asper V, et al. 2014. The rise and fall of methanotrophy following a deepwater oil-well blowout. *Nat. Geosci.* 7:423–27

Daly KL, Passow U, Chanton J, Hollander D. 2016. Assessing the impacts of oil-associated marine snow formation and sedimentation during and after the Deepwater Horizon oil spill. *Anthropocene* 13:18–33

Daly KL, Vaz AC, Paris CB. 2020. Physical processes influencing the sedimentation and lateral transport of MOSSFA in the NE Gulf of Mexico. In *Scenarios and Responses to Future Deep Oil Spills*, ed. S Murawski, C Ainsworth, S Gilbert, D Hollander, C Paris, et al., pp. 300–14. Cham, Switz.: Springer

de Gouw JA, Middlebrook AM, Warneke C, Ahmadov R, Atlas EL, et al. 2011. Organic aerosol formation downwind from the Deepwater Horizon oil spill. *Science* 331:1295–99

Diercks AR, Dike C, Asper VL, DiMarco SF, Chanton JP, Passow U. 2018. Scales of seafloor sediment resuspension in the northern Gulf of Mexico. *Elem. Sci. Anthr.* 6:32

Diercks AR, Highsmith RC, Asper VL, Joung D, Zhou Z, et al. 2010. Characterization of subsurface polycyclic aromatic hydrocarbons at the Deepwater Horizon site. *Geophys. Res. Lett.* 37:L20602

Dissanayake AL, Burd AB, Daly KL, Francis S, Passow U. 2018. Numerical modeling of the interactions of oil, marine snow, and riverine sediments in the ocean. *J. Geophys. Res. Oceans* 123:5388–405

Doyle SM, Whitaker EA, De Pascuale V, Wade TL, Knap AH, et al. 2018. Rapid formation of microbe-oil aggregates and changes in community composition in coastal surface water following exposure to oil and the dispersant Corexit. *Front. Microbiol.* 9:689

Driskell WB, Payne JR. 2018. Macondo oil in northern Gulf of Mexico waters – part 2: dispersant-accelerated PAH dissolution in the *Deepwater Horizon* plume. *Mar. Pollut. Bull.* 129:412–19

Du M, Kessler JD. 2012. Assessment of the spatial and temporal variability of bulk hydrocarbon respiration following the Deepwater Horizon oil spill. *Environ. Sci. Technol.* 46:10499–507

Duan J, Liu W, Zhao X, Han Y, O'Reilly SE, Zhao D. 2018. Study of residual oil in Bay Jimmy sediment 5 years after the Deepwater Horizon oil spill: persistence of sediment retained oil hydrocarbons and effect of dispersants on desorption. *Sci. Total Environ.* 618:1244–53

Dubinsky EA, Conrad ME, Chakraborty R, Bill M, Borglin SE, et al. 2013. Succession of hydrocarbon-degrading bacteria in the aftermath of the *Deepwater Horizon* oil spill in the Gulf of Mexico. *Environ. Sci. Technol.* 47:10860–67

Edwards B, Reddy C, Camilli R, Carmichael C, Longnecker K, Van Mooy B. 2011. Rapid microbial respiration of oil from the *Deepwater Horizon* spill in offshore surface waters of the Gulf of Mexico. *Environ. Res. Lett.* 6:9

Fisher CR, Hsing P-Y, Kaiser CL, Yoerger DR, Roberts HH, et al. 2014. Footprint of *Deepwater Horizon* blowout impact to deep-water coral communities. *PNAS* 111:11744–49

Francis S. 2020. Chlorophyll-a, Aqua MODIS, NPP, Gulf of Mexico, 2002-2012 (1 day composite). *ERDDAP*. **https://coastwatch.pfeg.noaa.gov/erddap/griddap/erdMGchla1day.graph**

Francis S, Passow U. 2019. Transport of dispersed oil compounds to the seafloor by sinking phytoplankton aggregates: a modeling study. *Deep-Sea Res. I* 156:103192

French-McCay DP, Horn M, Li Z, Jayko K, Spaulding ML, et al. 2018. Modeling distribution, fate, and concentrations of *Deepwater Horizon* oil in subsurface waters of the Gulf of Mexico. In *Oil Spill Environmental Forensics Case Studies*, ed. SA Stout, Z Wang, pp. 683–735. Oxford, UK: Elsevier

Giering SLC, Yan B, Sweet J, Asper V, Diercks A, et al. 2018. An ecosystem baseline of particle flux in the Northern Gulf of Mexico. *Elem. Sci. Anthr.* 6:264

Girard F, Cruz R, Glickman O, Harpster T, Fisher CR. 2019. In situ growth of deep-sea octocorals after the Deepwater Horizon oil spill. *Elem. Sci Anthr.* 7:12

Girard F, Fisher CR. 2018. Long-term impact of the Deepwater Horizon oil spill on deep-sea corals detected after seven years of monitoring. *Biol. Conserv.* 225:117–27

Gong Y, Zhao X, Cai Z, O'Reilly SE, Hao X, Zhao D. 2014. A review of oil, dispersed oil and sediment interactions in the aquatic environment: influence on the fate, transport and remediation of oil spills. *Mar. Pollut. Bull.* 79:16–33

Graham WM, Condon RH, Carmichael RH, D'Ambra I, Patterson HK, et al. 2010. Oil carbon entered the coastal planktonic food web during the Deepwater Horizon oil spill. *Environ. Res. Lett.* 5:045301

Gray JL, Kanagy LK, Furlong ET, Kanagy CJ, McCoy JW, et al. 2014. Presence of the Corexit component dioctyl sodium sulfosuccinate in Gulf of Mexico waters after the 2010 Deepwater Horizon oil spill. *Chemosphere* 95:124–30

Gros J, Reddy C, Nelson R, Socolofsky S, Arey J. 2016. Simulating gas–liquid–water partitioning and fluid properties of petroleum under pressure: implications for deep-sea blowouts. *Environ. Sci. Technol.* 50:7397–48

Gros J, Socolofsky SA, Dissanayake AL, Jun I, Zhao L, et al. 2017. Petroleum dynamics in the sea and influence of subsea dispersant injection during *Deepwater Horizon*. *PNAS* 114:10065–70

Gustitus SA, Clement TP. 2017. Formation, fate, and impacts of microscopic and macroscopic oil-sediment residues in nearshore marine environments: a critical review. *Rev. Geophys.* 55:1130–57

Gutierrez T, Teske A, Ziervogel K, Passow U, Quigg A. 2018. Microbial exopolymers: sources, chemico-physiological properties, and ecosystem effects in the marine environment. *Front. Microbiol.* 9:1822

Hastings DW, Schwing PT, Brooks GR, Larson RA, Morford JL, et al. 2016. Changes in sediment redox conditions following the BP DWH blowout event. *Deep-Sea Res. II* 129:167–78

Hatcher PG, Obeid W, Wozniak AS, Xu C, Zhang S, et al. 2018. Identifying oil/marine snow associations in mesocosm simulations of the Deepwater Horizon oil spill event using solid-state ^{13}C NMR spectroscopy. *Mar. Pollut. Bull.* 126:159–65

Hazen TC, Dubinsky EA, DeSantis TZ, Andersen GL, Piceno YM, et al. 2010. Deep-sea oil plume enriches indigenous oil-degrading bacteria. *Science* 330:204–8

Head IM, Jones DM, Roling WFM. 2006. Marine microorganisms make a meal of oil. *Nat. Rev. Microbiol.* 4:173–82

Honda M, Suzuki N. 2020. Toxicities of polycyclic aromatic hydrocarbons for aquatic animals. *Int. J. Environ. Res. Public Health* 17:1363

Hu P, Dubinsky EA, Probst AJ, Wang J, Sieber CMK, et al. 2017. Simulation of *Deepwater Horizon* oil plume reveals substrate specialization within a complex community of hydrocarbon degraders. *PNAS* 114:7432–37

Idowu O, Semple KT, Ramadass K, O'Connor W, Hansbro P, Thavamani P. 2019. Beyond the obvious: environmental health implications of polar polycyclic aromatic hydrocarbons. *Environ. Int.* 123:543–57

John V, Arnosti C, Field J, Kujawinski E, McCormick A. 2016. Oil spill remediation: fundamental concepts, rationale for use, fate, and transport issues. *Oceanography* 29(3):108–17

Joye SB. 2015. Deepwater Horizon, 5 years on. *Science* 349:592–93

Joye SB, Bracco A, Özgökmen TM, Chanton JP, Grosell M, et al. 2016a. The Gulf of Mexico ecosystem, six years after the Macondo oil well blowout. *Deep-Sea Res. II* 129:4–19

Joye SB, Kleindienst S, Gilbert JA, Handley K, Weisenhorn P, et al. 2016b. Responses of microbial communities to hydrocarbon exposures. *Oceanography* 29(3):136–49

Joye SB, Kostka JE. 2020. *Microbial genomics of the global ocean system*. Rep., Am. Soc. Microbiol., Washington, DC. **https://doi.org/10.1002/essoar.10502548.1**

Joye SB, MacDonald IR, Leifer I, Asper V. 2011. Magnitude and oxidation potential of hydrocarbon gases released from the BP oil well blowout. *Nat. Geosci.* 4:160–64

Joye SB, Teske AP, Kostka JE. 2014. Microbial dynamics following the Macondo oil well blowout across Gulf of Mexico environments. *BioScience* 64:766–77

Kessler JD, Valentine DL, Redmond MC, Du M, Chan EW, et al. 2011. A persistent oxygen anomaly reveals the fate of spilled methane in the deep Gulf of Mexico. *Science* 331:312–15

Khelifa A, Fingas M, Brown C. 2008. *Effects of dispersants on oil-SPM aggregation and fate in US coastal waters*. Rep., Coast. Resp. Res. Cent., Durham, NH

Kleindienst S, Grim S, Sogin M, Bracco A, Crespo-Medina M, Joye SB. 2016. Diverse, rare microbial taxa responded to the *Deepwater Horizon* deep-sea hydrocarbon plume. *ISME J.* 10:400–15

Kleindienst S, Paul JH, Joye SB. 2015a. Using dispersants after oil spills: impacts on the composition and activity of microbial communities. *Nat. Rev. Microbiol.* 13:388–96

Kleindienst S, Seidel M, Ziervogel K, Grim S, Loftis K, et al. 2015b. Chemical dispersants can suppress the activity of natural oil-degrading microorganisms. *PNAS* 112:14900–5

Kolian SR, Porter SA, Sammarco PW, Birkholz D, Cake EW Jr., Subra WA. 2015. Oil in the Gulf of Mexico after the capping of the BP/*Deepwater Horizon* Mississippi Canyon (MC-252) well. *Environ. Sci. Pollut. Res.* 22:12073–82

Kostka JE, Joye SB, Overholt W, Bubenheim P, Hackbusch S, et al. 2020a. Biodegradation of petroleum hydrocarbons in the deep sea. In *Deep Oil Spills: Facts, Fate, and Effects*, ed. SA Murawski, CH Ainsworth, S Gilbert, DJ Hollander, CB Paris, et al., pp. 107–24. Cham, Switz.: Springer

Kostka JE, Overholt WA, Rodriguez-R LM, Huettel M, Konstantinidis K. 2020b. Toward a predictive understanding of the benthic microbial community response to oiling on the northern Gulf of Mexico Coast. In *Scenarios and Responses to Future Deep Oil Spills: Fighting the Next War*, ed. SA Murawski, CH Ainsworth, S Gilbert, DJ Hollander, CB Paris, et al., pp. 182–202. Cham, Switz.: Springer

Kourafalou VH, Androulidakis YS. 2013. Influence of Mississippi River induced circulation on the Deepwater Horizon oil spill transport. *J. Geophys. Res. Oceans* 118:3823–42

Kowalewska G, Konat J. 1997. Distribution of polynuclear aromatic hydrocarbons (PAHs) in sediments of the southern Baltic Sea. *Oceanologia* 39:83–104

Kujawinski EB, Kido Soule MC, Valentine DL, Boysen AK, Longnecker K, Redmond MC. 2011. Fate of dispersants associated with the Deepwater Horizon oil spill. *Environ. Sci. Technol.* 45:1298–306

Lampitt RS, Hillier WR, Challenor PG. 1993. Seasonal and diel variation in the open ocean concentration of marine snow aggregates. *Nature* 362:737–39

Larson RA, Brooks GR, Schwing PT, Holmes CW, Carter SR, Hollander DJ. 2018. High-resolution investigation of event driven sedimentation: northeastern Gulf of Mexico. *Anthropocene* 24:40–50

Le Hénaff M, Kourafalou VH, Paris CB, Helgers J, Aman ZM, et al. 2012. Surface evolution of the Deepwater Horizon oil spill patch: combined effects of circulation and wind-induced drift. *Environ. Sci. Technol.* 46:7267–73

Lee RF, Köster M, Paffenhöfer G-A. 2012. Ingestion and defecation of dispersed oil droplets by pelagic tunicates. *J. Plankton Res.* 34:1058–63

Lehr B, Bristol S, Possolo A. 2010. *Oil budget calculator: Deepwater Horizon*. Rep., Coast. Resp. Res. Cent., Durham, NH. **http://www.restorethegulf.gov/sites/default/files/documents/pdf/OilBudgetCalc_Full_HQ-Print_111110.pdf**

Lewis CG, Ricker RW. 2020. Overview of ecological impacts of deep spills: *Deepwater Horizon*. In *Deep Oil Spills: Facts, Fate, and Effects*, ed. SA Murawski, CH Ainsworth, S Gilbert, DJ Hollander, CB Paris, et al., pp. 344–54. Cham, Switz.: Springer

Li Z, Spaulding ML, French-McCay D. 2017. An algorithm for modeling entrainment and naturally and chemically dispersed oil droplet size distribution under surface breaking. *Mar. Pollut. Bull.* 119:145–52

Liu G, Bracco A, Passow U. 2018. The influence of mesoscale and submesoscale circulation on sinking particles in the northern Gulf of Mexico. *Elem. Sci. Anthr.* 6:36

Liu Z, Liu J, Zhu Q, Wu W. 2012. The weathering of oil after the *Deepwater Horizon* oil spill: insights from the chemical composition of the oil from the sea surface, salt marshes and sediments. *Environ. Res. Lett.* 7:035302

Luo H, Bracco A, Cardona Y, McWilliams JC. 2016. Submesoscale circulation in the northern Gulf of Mexico: surface processes and the impact of the freshwater river input. *Ocean Model.* 101:68–82

MacDonald IR. 2010. *Deepwater* disaster: how the oil spill estimates got it wrong. *Significance* 7:149–54

MacDonald IR, Garcia-Pineda O, Beet A, Daneshgar Asl S, Feng L, et al. 2015. Natural and unnatural oil slicks in the Gulf of Mexico. *J. Geophys. Res. Oceans* 120:8364–80

Malone K, Aman ZM, Pesch S, Schlueter M, Krause D. 2020. Jet formation at the spill site and resulting droplet size distribution. In *Deep Oil Spills: Facts, Fate, and Effects*, ed. SA Murawski, CH Ainsworth, S Gilbert, DJ Hollander, CB Paris, et al., pp. 43–64. Cham, Switz.: Springer

McGenity TJ, Folwell BD, McKew BA, Sanni GO. 2012. Marine crude-oil biodegradation: a central role for interspecies interactions. *Aquat. Biosyst.* 8:10

McNutt MK, Camilli R, Crone TJ, Guthrie GD, Hsieh PA, et al. 2012. Review of flow rate estimates of the *Deepwater Horizon* oil spill. *PNAS* 109:20260–67

Mitra S, Kimmel DG, Snyder J, Scalise K, McGlaughon BD, et al. 2012. Macondo-1 well oil-derived polycyclic aromatic hydrocarbons in mesozooplankton from the northern Gulf of Mexico. *Geophys. Res. Lett.* 39:L01605

Montagna P, Baguley J, Cooksey C, Hartwell I, Hyde LJ, et al. 2013. Deep-sea benthic footprint of the Deepwater Horizon blowout. *PLOS ONE* 8:e70540

Murawski SA, Hogarth WT, Peebles EB, Barbeiri L. 2014. Prevalence of external skin lesions and polycyclic aromatic hydrocarbon concentrations in Gulf of Mexico fishes, post-Deepwater Horizon. *Trans. Am. Fish. Soc.* 143:1084–97

Natl. Acad. Eng. Sci. Med. 2019. *The use of dispersants in marine oil spill response*. Washington, DC: Natl. Acad. Press

Nixon Z, Zengel S, Baker M, Steinhoff M, Fricano G, et al. 2016. Shoreline oiling from the Deepwater Horizon oil spill. *Mar. Pollut. Bull.* 107:170–78

Omarova M, Swientoniewski LT, Mkam Tsengam IK, Blake DA, John V, et al. 2019. Biofilm formation by hydrocarbon-degrading marine bacteria and its effects on oil dispersion. *ACS Sustain. Chem. Eng.* 7:14490–99

Omotoso OE, Munoz VA, Mikula RJ. 2002. Mechanisms of crude oil-mineral interactions. *Spill Sci. Technol. Bull.* 8:45–54

Orcutt BN, Lapham LL, Delaney JM, Sarode N, Marshall KS, et al. 2017. Microbial response to oil enrichment in Gulf of Mexico sediment measured using a novel long-term benthic lander system. *Elem. Sci. Anthr.* 5:18

Özgökmen TM, Chassignet EP, Dawson CN, Dukhovskoy D, Jacobs G, et al. 2016. Over what area did the oil and gas spread during the 2010 Deepwater Horizon oil spill? *Oceanography* 29(3):96–107

Parsons ML, Turner RE, Overton EB. 2014. Sediment-preserved diatom assemblages can distinguish a petroleum activity signal separately from the nutrient signal of the Mississippi River in coastal Louisiana. *Mar. Pollut. Bull.* 85:164–71

Passow U. 2016. Formation of rapidly-sinking, oil-associated marine snow. *Deep-Sea Res. II* 129:232–40

Passow U, Stout SA. 2020. Character and sedimentation of "lingering" Macondo oil to the deep-sea after the Deepwater Horizon oil spill. *Mar. Chem.* 218:103733

Passow U, Sweet J, Francis S, Xu C, Dissanayake AL, et al. 2019. Incorporation of oil into diatom aggregates. *Mar. Ecol. Prog. Ser.* 612:65–86

Passow U, Sweet J, Quigg A. 2017. How the dispersant Corexit impacts the formation of sinking marine oil snow. *Mar. Pollut. Bull.* 125:139–45

Passow U, Ziervogel K. 2016. Marine snow sedimented oil released during the Deepwater Horizon spill. *Oceanography* 29(3):118–25

Passow U, Ziervogel K, Asper V, Diercks A. 2012. Marine snow formation in the aftermath of the Deepwater Horizon oil spill in the Gulf of Mexico. *Environ. Res. Lett.* 7:11

Payne JR, Driskell WB. 2018. Macondo oil in northern Gulf of Mexico waters – part 1: assessments and forensic methods for *Deepwater Horizon* offshore water samples. *Mar. Pollut. Bull.* 129:399–411

Perkins MJ, Joye SB, Field JA. 2016. Selective quantification of DOSS in marine sediment and sediment-trap solids by LC-QTOF-MS. *Anal. Bioanal. Chem.* 409:971–78

Prince RC, Butler JD, Redman AD. 2017. The rate of crude oil biodegradation in the sea. *Environ. Sci. Technol.* 51:1278–84

Prince RC, Elmendorf DL, Lute JR, Hsu CS, Haith CE, et al. 1994. 17α(*H*),21β(*H*)-hopane as a conserved internal marker for estimating the biodegradation of crude oil. *Environ. Sci. Technol.* 28:142–45

Prince RC, Walters CC. 2007. Biodegradation of oil and its implications for source identification. In *Oil Spill Environmental Forensics*, ed. Z Wang, SA Stout, pp. 349–79. Burlington, MA: Academic

Quintana-Rizzo E, Torres JJ, Ross SW, Romero I, Watson K, et al. 2015. δ^{13}C and δ^{15}N in deep-living fishes and shrimps after the Deepwater Horizon oil spill, Gulf of Mexico. *Mar. Pollut. Bull.* 94:241–50

Reddy CM, Arey JS, Seewald JS, Sylva SP, Lemkau KL, et al. 2011. Composition and fate of gas and oil released to the water column during the *Deepwater Horizon* oil spill. *PNAS* 109:20229–34

Redmond MC, Valentine DL. 2011. Natural gas and temperature structured a microbial community response to the *Deepwater Horizon* oil spill. *PNAS* 109:20292–97

Romero I, Schwing P, Brooks G, Larson R, Hastings D, et al. 2015. Hydrocarbons in deep-sea sediments following the 2010 Deepwater Horizon blowout in the northeast Gulf of Mexico. *PLOS ONE* 10:e0128371

Romero I, Toro-Farmer G, Diercks A-R, Schwing P, Muller-Karger F, et al. 2017. Large-scale deposition of weathered oil in the Gulf of Mexico following a deep-water oil spill. *Environ. Pollut.* 228:179–89

Rouhani S, Baker M, Steinhoff M, Zhang M, Oehrig J, et al. 2017. Nearshore exposure to Deepwater Horizon oil. *Mar. Ecol. Prog. Ser.* 576:111–24

Ryerson TB, Aikin KC, Angevine WM, Atlas EL, Blake DR, et al. 2011. Atmospheric emissions from the Deepwater Horizon spill constrain air-water partitioning, hydrocarbon fate, and leak rate. *Geophys. Res. Lett.* 38:L07803

Ryerson TB, Camilli R, Kessler JD, Kujawinski EB, Reddy CM, et al. 2012. Chemical data quantify *Deepwater Horizon* hydrocarbon flow rate and environmental distribution. *PNAS* 109:20246–53

Schwing PT, O'Malley BJ, Hollander DJ. 2018. Resilience of benthic foraminifera in the northern Gulf of Mexico following the Deepwater Horizon event (2011–2015). *Ecol. Indic.* 84:753–64

Schwing PT, Romero IC, Brooks GR, Hastings DW, Larson RA, Hollander DJ. 2015. A decline in benthic foraminifera following the Deepwater Horizon event in the northeastern Gulf of Mexico. *PLOS ONE* 10:e0128505

Sengco MR, Anderson DM. 2004. Controlling harmful algal blooms through clay flocculation. *J. Eukaryot. Microbiol.* 51:169–72

Shiller AM, Chan EW, Joung DJ, Redmond MC, Kessler JD. 2017. Light rare earth element depletion during *Deepwater Horizon* blowout methanotrophy. *Sci. Rep.* 7:10389

Smith CR, Rowles TK, Hart LB, Townsend FI, Wells RS, et al. 2017. Slow recovery of Barataria Bay dolphin health following the *Deepwater Horizon* oil spill (2013–2014), with evidence of persistent lung disease and impaired stress response. *Endanger. Species Res.* 33:127–42

Snyder SM, Pulster EL, Wetzel DL, Murawski SA. 2015. PAH exposure in Gulf of Mexico demersal fishes, post-*Deepwater Horizon*. *Environ. Sci. Technol.* 49:8786–95

Spier C, Stringfellow WT, Hazen TC, Conrad M. 2013. Distribution of hydrocarbons released during the 2010 MC252 oil spill in deep offshore waters. *Environ. Pollut.* 173:224–30

Stoffyn-Egli P, Lee K. 2002. Formation and characterization of oil-mineral aggregates. *Spill Sci. Technol. Bull.* 8:31–44

Stout SA, German CR. 2018. Characterization and flux of marine oil snow settling toward the seafloor in the northern Gulf of Mexico during the *Deepwater Horizon* incident: evidence for input from surface oil and impact on shallow shelf sediments. *Mar. Pollut. Bull.* 129:695–713

Stout SA, Payne JR. 2017. Footprint, weathering, and persistence of synthetic-base drilling mud olefins in deep-sea sediments following the Deepwater Horizon disaster. *Mar. Pollut. Bull.* 118:328–40

Stout SA, Payne JR, Emsbo-Mattingly SD, Baker G. 2016. Weathering of field-collected floating and stranded Macondo oils during and shortly after the Deepwater Horizon oil spill. *Mar. Pollut. Bull.* 105:7–22

Stout SA, Rouhani S, Liu B, Oehrig J, Ricker RW, et al. 2017. Assessing the footprint and volume of oil deposited in deep-sea sediments following the *Deepwater Horizon* oil spill. *Mar. Pollut. Bull.* 114:327–42

Sun X, Kostka JE. 2019. Hydrocarbon-degrading microbial communities are site specific, and their activity is limited by synergies in temperature and nutrient availability in surface ocean waters. *Appl. Environ. Microbiol.* 85:e00443–19

Turner RE, Rabalais NN, Overton EB, Meyer BM, McClenachan G, et al. 2019. Oiling of the continental shelf and coastal marshes over eight years after the 2010 *Deepwater Horizon* oil spill. *Environ. Pollut.* 252:1367–76

Valentine DL, Fisher GB, Bagby SC, Nelson RK, Reddy CM, et al. 2014. Fallout plume of submerged oil from *Deepwater Horizon*. *PNAS* 111:15906–11

Valentine DL, Kessler JD, Redmond MC, Mendes SD, Heintz MB, et al. 2010. Propane respiration jump-starts microbial response to a deep oil spill. *Science* 330:208–11

Valentine DL, Mezić I, Maćešić S, Črnjarić-Žic N, Ivić S, et al. 2012. Dynamic autoinoculation and the microbial ecology of a deep water hydrocarbon irruption. *PNAS* 109:20286–91

Ventikos NP, Vergetis E, Psaraftis HN, Triantafyllou G. 2004. A high-level synthesis of oil spill response equipment and countermeasures. *J. Hazard. Mater.* 107:51–58

Wang J, Sandoval K, Ding Y, Stoeckel D, Minard-Smith A, et al. 2016. Biodegradation of dispersed Macondo crude oil by indigenous Gulf of Mexico microbial communities. *Sci. Total Environ.* 557–58:453–68

Ward CP, Sharpless CM, Valentine DL, French-McCay DP, Aeppli C, et al. 2018. Partial photochemical oxidation was a dominant fate of *Deepwater Horizon* surface oil. *Environ. Sci. Technol.* 52:1797–805

Washburn TW, Reuscher MG, Montagna PA, Cooksey C, Hyland JL. 2017. Macrobenthic community structure in the deep Gulf of Mexico one year after the Deepwater Horizon blowout. *Deep-Sea Res. I* 127:21–30

Weisberg RH, Zheng L, Liu Y, Murawski S, Hu C, Paul J. 2016. Did Deepwater Horizon hydrocarbons transit to the west Florida continental shelf? *Deep-Sea Res. II* 129:259–72

White HK, Lyons SL, Harrison SJ, Findley DM, Liu Y, Kujawinski EB. 2014. Long-term persistence of dispersants following the Deepwater Horizon oil spill. *Environ. Sci. Technol. Lett.* 1:295–99

Wilson RM, Cherrier J, Sarkodee-Adoo J, Bosman S, Mickle A, Chanton JP. 2016. Tracing the intrusion of fossil carbon into coastal Louisiana macrofauna using natural ^{14}C and ^{13}C abundances. *Deep-Sea Res. II* 129:89–95

Wirth M, Passow U, Jeschek J, Hand I, Schulz-Bull DE. 2018. Partitioning of oil compounds into marine oil snow: insights into prevailing mechanisms and dispersant effects. *Mar. Chem.* 206:62–73

Yan B, Passow U, Chanton JP, Nöthig E-M, Asper V, et al. 2016. Sustained deposition of contaminants from the *Deepwater Horizon* spill. *PNAS* 113:E3332–40

Yang T, Nigro LM, Gutierrez T, D'Ambrosio L, Joye SB, et al. 2016a. Pulsed blooms and persistent oil-degrading bacterial populations in the water column during and after the Deepwater Horizon blowout. *Deep-Sea Res. II* 129:282–91

Yang T, Speare K, McKay L, MacGregor BJ, Joye SB, Teske A. 2016b. Distinct bacterial communities in surficial seafloor sediments following the 2010 Deepwater Horizon blowout. *Front. Microbiol.* 7:1384

Zeinstra-Helfrich M, Koops W, Murk AJ. 2015. The NET effect of dispersants—a critical review of testing and modelling of surface oil dispersion. *Mar. Pollut. Bull.* 100:102–11

Zhao L, Boufadel MC, Adams E, Socolofsky SA, King T, et al. 2015. Simulation of scenarios of oil droplet formation from the Deepwater Horizon blowout. *Mar. Pollut. Bull.* 101:304–19

Ziervogel K, Dike C, Asper V, Montoya J, Battles J, et al. 2016a. Enhanced particle fluxes and heterotrophic bacterial activities in Gulf of Mexico bottom waters following storm-induced sediment resuspension. *Deep-Sea Res. II* 129:77–88

Ziervogel K, Joye SB, Arnosti C. 2016b. Microbial enzymatic activity and secondary production in sediments affected by the sedimentation pulse following the Deepwater Horizon oil spill. *Deep-Sea Res. II* 129:241–48

Ziervogel K, McKay L, Rhodes B, Osburn C, Dickson-Brown J, et al. 2012. Microbial activities and dissolved organic matter dynamics in oil-contaminated surface seawater from the Deepwater Horizon oil spill site. *PLOS ONE* 7:e34816