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What Happened to All of the Oil?

By Uta Passow and Robert D. Hetland

ABSTRACT. The explosion of the Deepwater Horizon platform in the Gulf of Mexico in 2010 caused an oil spill that was unique in that it originated at great depth and persisted for an extended period of time, resulting in release of a very large quantity of oil and gas into the environment. What happened to all of this oil and gas? This paper briefly discusses the various physical, chemical, and biological processes that affected the fate and distribution of the spilled petrocarbon: some of the spilled oil was directly removed by mitigating measures, some was rapidly biodegraded, and some was deposited on the seafloor. Part of what remained entered food webs or contaminated shorelines. Consolidation of different estimates of the diverse distribution pathways provides a "guesstimate" budget that assesses the fate of the spilled petrocarbon after it partitioned between the deep plume and the sea surface.

Photo credit: Luke McKay

INTRODUCTION

In April 2010, an explosion at the Deepwater Horizon (DWH) platform in the Gulf of Mexico led to an oil spill that was extraordinary in its volume (~5,000,000 barrels of oil and 7.7 billion standard cubic feet of natural gas), duration (87 days), and depths of release (1,500 m) (McNutt et al., 2012). Estimates of oil and gas concentrations and the sizes of impacted areas, however, vary widely because of the uneven distribution of oil compounds in the environment and because of the difficulty in sampling marine habitats. What happened to all of this oil? The chemical complexity of oil, which consists of thousands of distinct chemical compounds, combined with the complexity of the physical, chemical, and biological processes that determine the fate of the spilled oil and gas, preclude a simple answer. Mitigating response actions further influenced the oil distribution and had some unintended consequences that altered the fate of the oil. In this paper, we provide an overview of the main distribution pathways of the spilled petroleum and the associated processes that influenced their transport and transformation in the marine environment.

THE EFFECTS OF MITIGATING MEASURES ON OIL DISTRIBUTION

Dispersant Addition. A number of measures were taken to mitigate the effect of the released oil. The dispersant Corexit was added both at the leak depth and onto the surface slick. Dispersants reduce the surface tension of the oil-water interface, leading to smaller oil droplet sizes and increased dispersion of oil in water. At the surface, the addition of Corexit reduced the thickness of the oil carpet while increasing the affected surface area and the number of oil droplets in the upper mixed layer (Garcia-Pineda et al., 2013; MacDonald et al., 2015).

The unprecedented addition of Corexit (about 1.84 million gallons) at the source of the oil leak at >1,400 m depth also appreciably affected the distribution and fate of the spilled oil (Kujawinski et al., 2011; Gray et al., 2014). The dispersant increased the fraction of oil that was spreading within the water column, and the oil that did reach the sea surface was displaced laterally, a few kilometers from the site of the blowout (Chan et al., 2015). The primary reason for adding dispersants directly to the leak at depth was so that responders, required to work directly above the wellhead, would be less affected by the surface expression of the oil (Socolofsky et al., 2015). The decreased droplet size (Zhao et al., 2015) and increased fraction of subsurface oil was also meant to reduce coastal impacts of the surface oil slick, but the success of this strategy is controversial (Paris et al., 2012). Additionally, the reduced droplet size due to dispersant addition was thought to promote biodegradation of oil, but this concept has recently been challenged based on experiments that show Corexit suppresses microbial oil degradation (Kleindienst et al., 2015).

Direct Recovery. Efforts to collect oil directly at depth at the leak site and to remove oil from the surface, either by skimming or via in situ burns, removed about 25% of all petroleum spilled (Lehr et al., 2010; Lubchenco et al., 2012). In situ burning via more than 400 fires, while removing oil, aerosolized some oil compounds and added burn residue, including char and soot, to the water. Some burn residue is heavier than seawater, so it sinks directly near the site of its production. Char and soot particles, however, may linger in the water for months until collected by sinking marine snow (Yan et al., 2016).

Opening of Diversionary Channels. The release of water from diversionary channels of the Mississippi River, meant to prevent oil from entering Louisiana marshes, appeared to keep oil out of the areas where freshwater was released (Bianchi et al., 2011). However, this water release also led to additional input of nutrients and clay, which may have, directly and indirectly, increased sedimentation of oily marine snow in, for example, the DeSoto Canyon area (Brooks et al., 2015).

Addition of Drilling Mud. Drilling mud, a dense fluid, was pumped into the wellhead in unsuccessful attempts to stop the leak. Drilling mud is heavy and sinks rapidly, taking oil along with it in the form of a sediment-oil suspension. The footprint of this event was restricted to a circle of about 6–7 km diameter around the leak site (NRDA, 2015), although fine drilling mud residue was found to be transported much farther (Yan et al., 2016).

PHYSICOCHEMICAL DISTRIBUTION PATHWAYS

The different compounds of the Macondo crude oil partitioned in the environment depending on their specific characteristics (Socolofsky et al., 2016, in this issue): gas and lighter compounds preferentially evaporated or dissolved, whereas the heavier compounds remained behind, transported and modified by currents at many different scales (see Box 1; Figure 1). Associations with biogenic particles and suspended minerals further impacted the distribution of the different oil compounds. Once released, the petroleum formed three distinct features: (1) a rising plume between the leak and the sea surface, (2) a subsurface plume (or intrusion layer) at about 1,100 m depth, and (3) a massive oil slick at the surface (Thibodeaux et al., 2011).

Rising Plume. A buoyant plume composed of oil and gas, and the added Corexit, formed above the riser pipe leak. The turbulent plume entrained background ocean water that included organisms and organic matter as it rose. Within the plume, the behavior of the released petrocarbons was affected by the specific physicochemical properties of the individual compounds and by marine particles and environmental conditions (e.g., pressure, temperature, turbulence). Physicochemical partitioning due to pressure or temperature changes during ascent, and biological activity, all changed the properties of the rising oil plume. While rising, the plume spread laterally due to mixing and entrainment, forming a cone, but remained predominantly within a 1–2 km radius around the leak (Ryerson et al., 2012). Roughly one-half of the oil and gas from this rising plume was partitioned into a deep, subsurface intrusion layer; the rest rose to the sea surface.

Deep Intrusion Layer or Deep Plume. The deep intrusion layer formed predominately between 1,000 m and 1,400 m depth, where the mixture of buoyant petrocarbons and entrained, denser seawater became neutrally buoyant (Socolofsky et al., 2011). About 50% of the total discharged petroleum, including most soluble petrocarbons as well as small oil droplets, were trapped within the intrusion layer (Diercks et al., 2010; Reddy et al., 2011; Ryerson et al., 2012; Spier et al., 2013).

Diffuse subsurface plumes of oil compounds were also observed at other depths, for example, at 400 m, but were less pronounced. Physical transport of the subsurface plumes differed from those at the surface, with currents moving the main intrusion layer at ~1,100 m up to 400 km to the southwest (Spier et al., 2013). Oil trapped in the deep plume was too deep to reach nearshore habitats, but ran aground when encountering upsloping seafloor, where it infiltrated the sediments and left a contaminated layer analogous to a dirty bathtub ring (Hastings et al., 2014; Romero et al., 2015).

Surface Oil Slick. About half of the spilled oil and gas reached the sea surface, creating a large oil slick. About 14% of the oil compounds that reached the surface evaporated from the surface slick within hours to days (Ryerson et al., 2012). The footprint of the surface slick continuously changed in size and location (MacDonald et al., 2015), as physical processes on

Box 1. Key Concept: Physical Transport of Oil

Currents transport and modify oil at many different scales. At the smallest scales (millimeters to meters), energetic turbulent motions near the wellhead can break a body of oil into droplets. This process is complex, as oil and gas mixtures within the water will rise and generate their own turbulent wakes, creating feedback between the oil and gas droplets and the turbulence. At slightly larger scales (a few meters to a kilometer) near the wellhead, an expanding vertical plume will bring oil and gas toward the surface, entraining background water to balance mass within the expanding plume. Because the entrained water may be denser than the water above, the plume may reach a level of neutral buoyancy. At this point, a subsurface intrusion layer may form. At even larger scales (hundreds of meters to a few kilometers) larger "submesoscale" turbulence may help to disperse the plume. The submesoscale turbulence is strongest in the ocean's surface mixed layer, within the upper 10–50 m, and strongly influences any surface oil slick. These flow features cause the "swirls" seen in satellite images of surface oil slicks and have the effect of spreading the slick out over a broader region than would otherwise be affected. At the largest scales (tens to hundreds of kilometers), mesoscale circulation features transport the oil. These circulation features are typically strongest near the surface and the bottom of the water column, and generally follow bathymetric features.

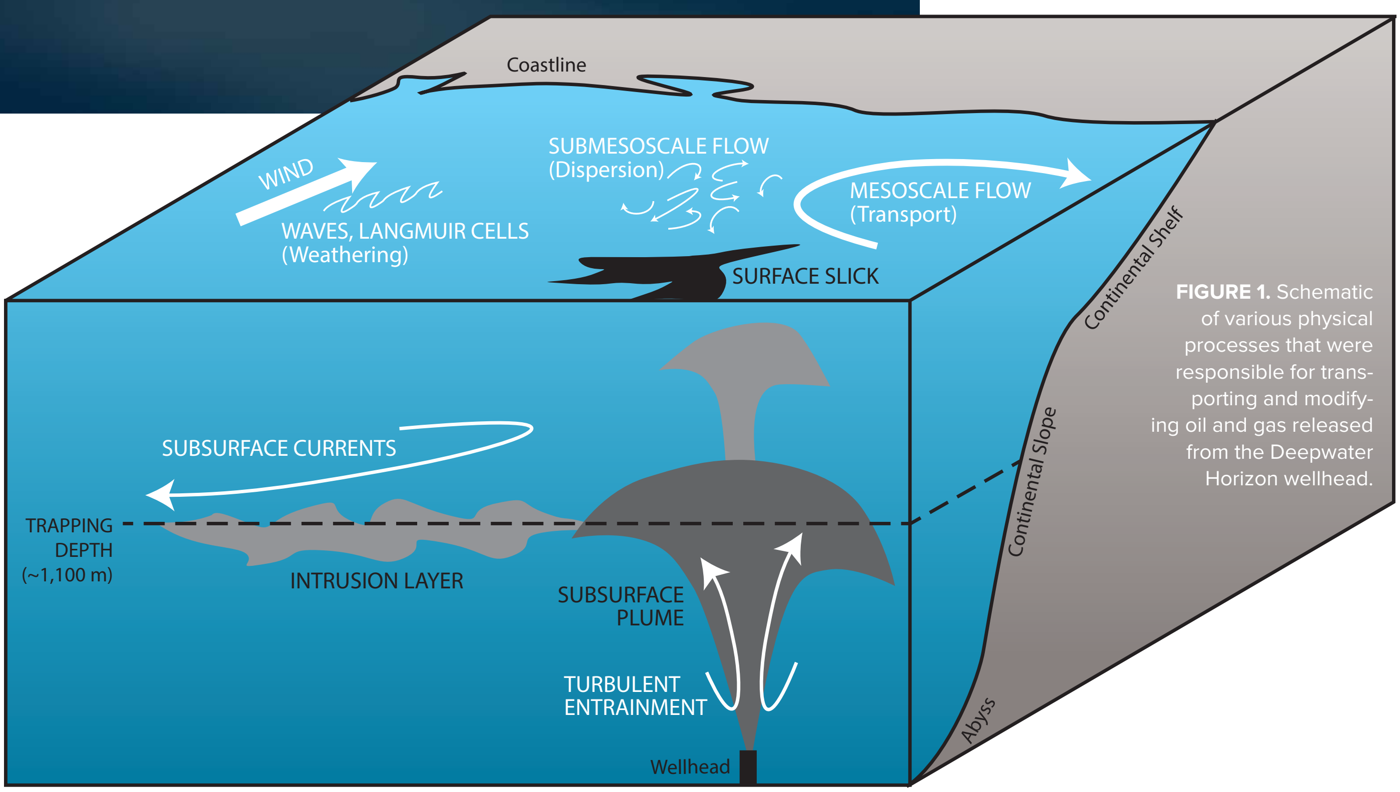

FIGURE 1. Schematic of various physical processes that were responsible for transporting and modifying oil and gas released from the Deepwater Horizon wellhead.

scales from millimeters to hundreds of kilometers impacted the distribution of the oil (Reed et al., 1999). The cumulative area where oil was detected via synthetic aperture radar (SAR) imaginary was about 112,115 km^2. This area represents an upper estimate of the area where oil could have sedimented to the seafloor and organisms could have been impacted directly. The average area covered with oil on any day during the spill was an order of magnitude smaller (White et al., 2016, in this issue). Wind and surface currents continuously moved the oil slick and pushed a large fraction of it toward the coast along the northern Gulf of Mexico (Le Hénaff et al., 2012). Özgökman et al. (2016, in this issue) discuss in more detail the different physical processes that transported the oil on the sea surface.

Mixing and wave action (Delvigne and Sweeney, 1988; Johansen et al., 2015), combined with the airborne release of Corexit onto the slicks, led to the re-entrainment of small but biologically significant amounts of oil into the upper mixed layer, down to ≥20 m. The fate of this water-accommodated fraction of oil differed from that of the oil slick itself, but it is difficult to track. Although physical and chemical dispersion and dilution of oil in the water column as well as biodegradation resulted in low concentrations of oil by the time the spill ended in mid-July, some fraction of the oil lingered in the water for many months after the leak was sealed, as inferred from its continued sedimentation for months thereafter (Yan et al., 2016).

OIL REACHING NEARSHORE HABITATS

Wind and currents transported surface slicks into nearshore environments where they affected more than 1,800 km of shoreline and predominantly contaminated marshes (45% of the impacted shoreline) and beaches (51% of the impacted shoreline; Michel et al., 2013; Stout et al., 2016). Oiling was categorized as moderate to heavy for 33% of this affected coastline. Before reaching these coastal ecosystems, the oil had traveled 80–100 km on the sea surface and was heavily weathered. Mostly it stranded in a thick viscous emulsion. Specifically, oil made landfall around the Mississippi delta and along the Mississippi and Alabama coastlines (Le Hénaff et al., 2012). At sandy beaches or marsh soils, oil enters the pore water, seeping into cracks and holes, and making it nearly impossible to estimate the total amount of oil hidden in these shallow environments (Mendelssohn et al., 2012). In subtidal habitats, oil mixed with sediment may form a viscous emulsion that sticks to mud, sand, plants, animals, and rocks. Some of the oil washed up on beaches as tar balls, patties, or oil mats. The ultimate fate of petroleum reaching the shoreline is largely degradation, either by photochemical processes or due to microbial activity of bacteria and fungi (Joye et al., 2014). The supply of oxygen and nutrients probably controlled the microbial degradation of oil that reached shore. Oil that washed ashore also entered the terrestrial ecosystems and food webs (Cornwall, 2015).

Cleanup efforts removed oil on 73% of oiled beaches, but natural recovery was deemed less damaging than invasive cleanup procedures for most oiled marshes (Michel et al., 2013). Despite cleanup and natural weathering processes, some oil persisted for years, and was remobilized and redistributed during storms (Dalyander et al., 2014). After two years, 39% of oiled beaches were still contaminated (Michel et al., 2013).

OIL IN THE OPEN OCEAN: PLANKTON-OIL INTERACTIONS

Organisms encountering oil in the various affected ecosystems were not only impacted by the oil (Buskey et al., 2016, in this issue) but also directly influenced its distribution and fate (Figure 2). Degradation of oil compounds by bacteria is best understood, but petroleum also entered and resided in food webs, and was packaged into sinking marine snow.

Microbial Degradation of Oil. Certain bacteria can degrade specific compounds of oil and natural gas, effectively removing or altering them (Joye et al., 2016, in this issue). The microbial response to the spill changed the abundance and composition of bacteria at the surface, in the deep plume, and at the seafloor (Yang et al., 2012), indicating that oil was degraded in all habitats. In the deep plume, increased respiration due to elevated bacterial activity resulted in a small

Box 2. Key Terms

BIODEGRADATION: Microorganisms utilize oil as a source of energy, generating new organic compounds or CO_2 in the process.

BIOTURBATION: Reworking and mixing of sediments by benthic organisms.

COREXIT: A dispersant that reduces the surface tension between oil and water and promotes the formation of tiny oil droplets that scatter in water rather than forming a surface oil film.

DISPERSION OF OIL: The distribution of oil droplets into the water, for example, by wave action or with the aid of dispersants.

DISSOLUTION: Water-soluble compounds of oil dissolve in water.

EMULSIFICATION OF OIL: Formation of a mousse that consists of tiny oil droplets within water.

EVAPORATE: Release into the air, as for example, the volatile fraction of oil.

FLOC: Loose layer of material on the seafloor, formed by settled marine snow.

MARINE SNOW: Large (> 0.5 mm) composite particles that sink to the ocean floor. (Oil-containing marine snow is termed marine oil snow or MOS.)

OIL: A liquid consisting of tens of thousands of chemically distinct compounds, each with different physical-chemical properties that influence its behavior and fate.

PHOTOOXIDATION: A sunlight-induced process where oxygen reacts with the oil, changing it chemically.

but detectable oxygen deficit, reflecting the rapid (days to weeks) removal of the more bioavailable compounds of the spilled oil and gas (Camilli et al., 2010; Hazen et al., 2010; Valentine et al., 2010; Joye et al., 2011; Kessler et al., 2011). Net degradation in the surface layer could not be quantified the same way, because at the surface, utilized oxygen is rapidly replenished from the atmosphere. As the oil compounds that formed the surface slick were less bioavailable than those in the intrusion layer, and because nutrient limitation in surface waters inhibited microbial degradation, it is assumed that microbial degradation of the surface slick was slower, although photodegradation of oil at the surface may have increased its bioavailability. Additionally, the formation of cooperative biodegradation networks within microbial oil snow allowed, similar to biofilms, the efficient degradation of oil from the surface via close interactions between different microbes. The quantitative importance of this process still awaits exploration (Joye et al., 2014; Passow and Ziervogel, 2016, in this issue). Oil-degrading bacteria also thrive in some copepod fecal pellets, so copepods feeding on oil products may also contribute to the degradation of petroleum (Størdal et al., 2015).

Incorporation of Oil into Food Webs. The bacterial blooms resulting from the DWH spill led to increased grazer populations that efficiently moved oil- and methane-derived carbon into the planktonic food web (Graham et al., 2010; Chanton et al., 2012; Cherrier et al., 2013). Fossil carbon from the Macondo oil may be tracked because its isotopic signal differs from that of modern marine organic carbon. When zooplankton or nekton take up oil (Lee et al., 2012; Mitra et al., 2012), specific petrocarbons bioaccumulate (Almeda et al., 2013a,b), partitioning preferentially into lipid-rich tissue, and persisting within organisms (Meador, 2003). Organisms ingest oil compounds, either directly as oil droplets or inadvertently via oil-coated food particles, or via respiratory surfaces. Oil compounds or signs of their presence were found in animals from all exposed habitats and at all levels of the food web (Cornwall, 2015). Although the fraction of the spilled oil that bioaccumulated in organisms is presumably small, its effect is disproportionately large due to the toxic and mutagenic properties of some of these substances (Paul et al., 2013; Garr et al., 2014).

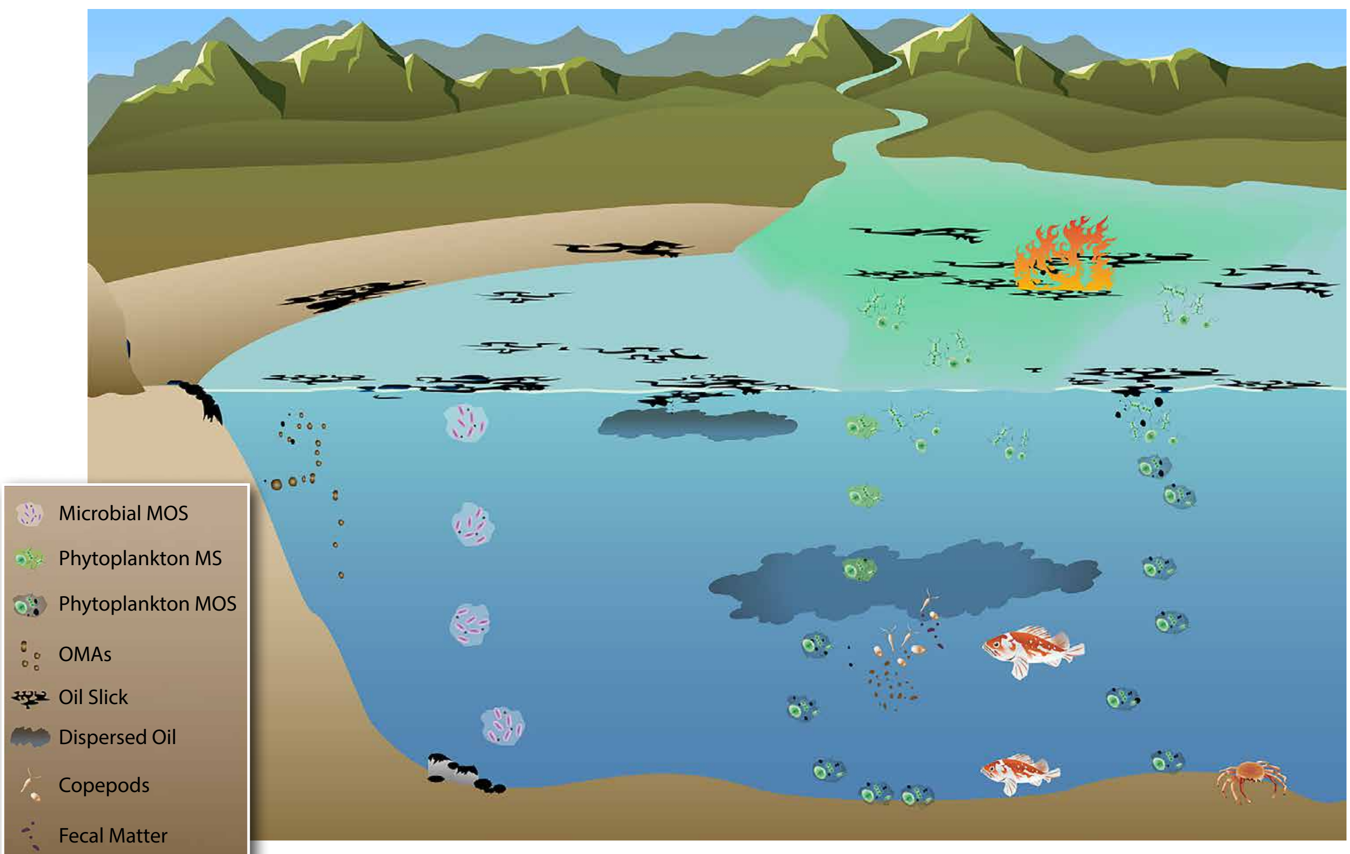

FIGURE 2. Depiction of biologically mediated oil distribution pathways during the Deepwater Horizon spill. Oil that stranded along coastlines was incorporated into ecosystems there, and oil that floated at the sea surface or in the deep plume interacted with plankton and mineral particles in the water column. Microbial degradation removed oil compounds in all habitats. Marine oil snow (MOS) formation transported petrocarbon to the seafloor, at times collecting oil during transit through the subsurface plume. In the presence of minerals, near the shelf slope, for example, oil-mineral aggregations may have formed, also leading to the settling of petrocarbon. Organisms from all habitats incorporated oil. Riverine effluent, as well as in situ burning further impacted oil fate.

Marine Oil Snow Formation and Sedimentation. Large phytoplankton aggregation events and zooplankton or bacterial activity may lead to the formation of oily marine snow, termed marine oil snow (MOS; Passow and Ziervogel, 2016, in this issue). Phytoplankton aggregates, consisting of diatoms or the cyanobacteria *Trichodesmium*, which are both common in the Gulf of Mexico, may incorporate significant amounts of petroleum either during their formation, or later while sinking through oil-rich plumes. Zooplankton feeding in dilute oil suspensions concentrate and repackage oil into sinking MOS that consists of discarded feeding structures or feces (Lee et al., 2012). Deepwater Horizon-derived MOS, whether formed from zooplankton or bacterial activity or via phytoplankton aggregation, sank rapidly (hundreds of meters per day), transporting oil toward the seafloor (Passow et al., 2012).

DEPOSITION OF OIL ON THE SEAFLOOR

Oil and large amounts of loose flocculent material (called floc) accumulated on the seafloor during and after the DWH spill (Chanton et al., 2014; Valentine et al., 2014; Brooks et al., 2015; Romero et al., 2015; Yan et al., 2016). Although direct fallout in association with drilling mud transported some petroleum in the immediate vicinity of the well to depth, sinking MOS was the primary cause of oily floc deposition. The combination of processes leading to MOS formation, sedimentation, and deposition at the seafloor has been dubbed MOSSFA, for marine oil snow sedimentation and flocculent accumulation (Daly et al., 2016).

Estimates based on oil compounds and isotopic signatures of material accumulated at the seafloor suggest that about 2%–15% of all the spilled petroleum was deposited at the seafloor; however, this estimate is almost certainly too low, providing a lower limit only (Passow and Ziervogel, 2016, in this issue).

Upon arrival at the seafloor, MOS covered corals (White et al., 2012; Hsing et al., 2013) and formed a 0.5–1.2 cm thick, loose layer of floc (Brooks et al., 2015). The floc layer caused reduced bioturbation and changed sediment redox conditions (Hastings et al., 2014; Brooks et al., 2015). Natural recovery of benthic deep-sea ecosystems after oil deposition is thought to be slow, in part because of low temperatures. Four years after the DWH spill, the Macondo oil footprint on the seafloor was still quite extensive, at about half of its original size (Montagna et al., 2013; Stout et al., 2015).

BUDGET CONSIDERATIONS

Here, we attempt an overall budget to address the question of what happened to all the oil. This guesstimate budget (Figure 3) consolidates best estimates of different physical and biological processes governing the fate of the oil. The budget is based on a total release of 5,000,000 barrels of petroleum (oil and gas), as derived by McNutt et al. (2012); the partitioning of oil and gas among evaporated, dissolved, and undissolved petrocarbon mixtures at the surface or in the deep plume (Ryerson et al., 2012); and estimates of microbial utilization (Du and Kessler, 2012; Joye et al., 2016, in this issue) and sedimentation (Passow and Ziervogel, 2016, in this issue).

About 833,000 barrels of the spilled oil were collected directly at the wellhead at depth (Lubchenco et al., 2012), and roughly 390,000 barrels were removed from the ocean surface either by direct recovery via skimming or by in situ burning (Lehr et al., 2010; Lubchenco et al., 2012). Most of the gaseous compounds, dominated by methane, were trapped in the deep plume and utilized by bacteria within days to weeks. One-third to up to almost half of the liquid oil compounds are thought to have been trapped in the deep plume as well, but these approximations rely on estimates of the amount of petroleum at the surface, which may be too low, because the amounts of oil in both thin sheens and thick carpets are difficult to quantify accurately. Microbial utilization of oil compounds in the plume based on the oxygen deficit imply that some of the oil droplets trapped in the deep plume were also utilized by bacteria, removing another 6% of all released petroleum, assuming that all of the dissolved petroleum was utilized first. Another significant fraction (~10%) of the oil in the deep plume sank, as the fingerprints of deposited oil compounds suggest (Valentine et al., 2014).

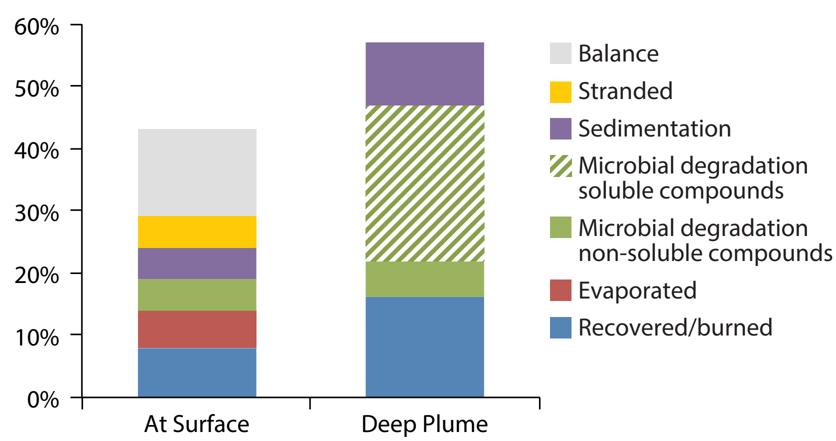

FIGURE 3. Guesstimate budget to assess what happened to the 5,000,000 barrels of released oil. It is still uncertain whether more or less than half of the spilled petrocarbon remained in the deep plume. Here, it is assumed that 57% remained at depth vs. 43% reaching the surface, because estimates of the fate of petrocarbon from the deep plume add up to this amount. Oil from the deep plume was largely biodegraded, recovered, or deposited as sediment on the seafloor. Budget uncertainties are larger for the petrocarbon at the surface, and the 5% values for microbial degradation, sedimentation, and stranding are placeholders only. See text for further detail.

Microbial degradation of surface oil has not been quantified, but may be assumed to be lower than that of the undissolved oil in the deep plume. Sedimentation rates of oil, as measured with shallow traps, imply that sedimentation of surface oil was a significant loss process, but quantification is problematic (Passow and Ziervogel, 2016, in this issue). Likewise, the total amount of oil that stranded along shorelines, or was lost due to weathering and photodegradation, has not been quantified. In lieu of a good estimate, our budget assumes that 5% each of the spilled petroleum was (1) microbially degraded at the surface, (2) incorporated into sinking MOS at the surface, and (3) transported to shorelines. The large unexplained residual (Figure 3: balance of 14%) implies that each of these approximations may easily be low by a factor of two. The fraction of oil compounds residing in the food chain (living organisms) is ignored in this budget, as it is assumed to be quantitatively minor.

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