

## **Marine snow formation and fluxes after crude oil spills: Review of findings from the Deepwater Horizon oil spill study.**

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### **Abstract**

PS5-05 (689531): The Deepwater Horizon oil spill is the largest in US history in terms of oil released and the amount of dispersants applied. It is also the first spill in which the incorporation of oil and/or dispersant into marine snow was directly observable. Marine snow formation, incorporation of oil (MOS – marine oil snow) and subsequent settling to the seafloor, has been termed MOSSFA: Marine Oil Snow Sedimentation and Flocculent Accumulation. This pathway accounts for a significant fraction of the total oil returning back to the sea floor. GOMRI funded studies have determined that important drivers of MOSSFA include, but are not limited to, an elevated and extended Mississippi River discharge, which enhanced phytoplankton production and suspended particle concentrations, zooplankton grazing, and enhanced mucus formation (operationally defined as EPS, TEP, marine snow). Efforts thus far to understand the mechanisms

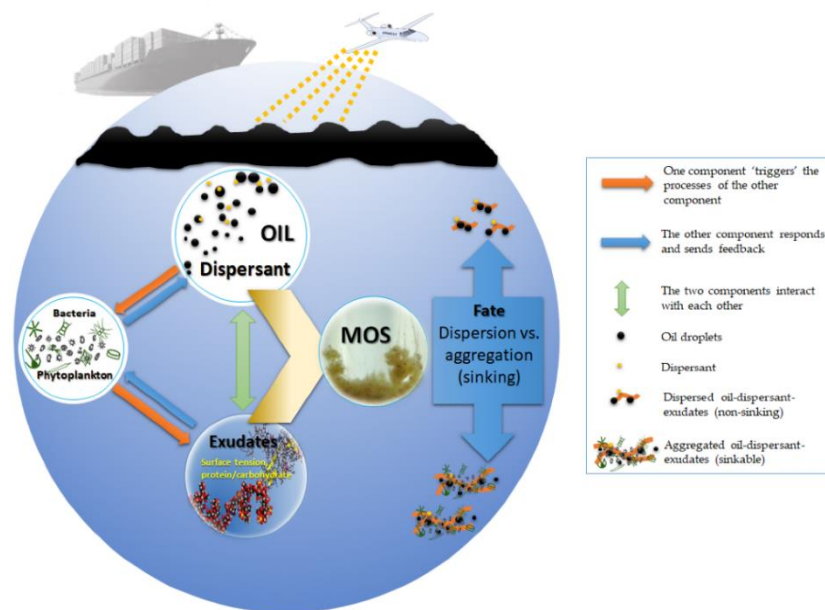
driving these processes are being used to aid in the development of response strategies. These include modeling efforts towards predicting plume dynamics. Although much has been learned during the GOMRI program (reviewed herein and elsewhere), there are still important unknowns that need to be addressed. Understanding of the conditions under which significant MOSSFA events occur, the consequences to the biology, the sinking velocity and distribution of the MOSSFA as well as its ultimate fate are amongst the most important consideration for future studies. Also important is the modification of the oil and dispersant within the MOS and its transport as part of MOSSFA. Ongoing studies are needed to further develop our understanding of these complex and interrelated phenomena.

Crude oils are some of the most complex and diverse organic mixtures found in nature, containing thousands of different compounds. The low molecular weight components are more susceptible to processes such as evaporation, dissolution, and biodegradation, while the heavier molecular weight, more hydrophobic compounds tend to adhere to living organisms or particulates, and persist (Overton et al. 2016, 2020; Adhikari et al. 2017; Ward et al. 2018). The polycyclic aromatic hydrocarbons (PAHs) and other compounds determine the acute and chronic toxicity of spilled oil to living organisms (Duran & Cravo-Laureau 2016). The explosion of the Deepwater Horizon (DwH) drilling rig in the northern Gulf of Mexico on April 20, 2010 that killed 11 people released an estimated 4.1 million barrels of oil over 87 days (but the total volume *actually* released was difficult to determine). While a large fraction reached the sea surface; oil remained in the water column, or reached sediments and the coast (Reddy et al. 2012; Overton et al. 2016; Murray and Boehm 2017). The composition of the DwH crude oil favored its removal through natural

degradation (biodegradation, photo-oxidation), evaporation, dissolution, and dispersal processes (Reddy et al. 2012; Overton et al. 2016; Ward et al. 2018). Given most petroleum hydrocarbons are highly insoluble, biodegradation can only take place at the hydrocarbon-water interface. Dispersion or emulsification of oil thus increases the bioavailability of oil products to biodegradation. While it is known that weathering and hydrodynamic forces affect both the distribution and properties of the released oil, and that opportunistic microbes play an important role in its degradation, but prior to the DwH oil spill there were few studies examining the interactions between oil, microbes and their exudates. Hence, the environmental fate (sinking versus dispersion versus aggregation), transport (exudates) and effects (on microbes) of spilled oil remain to be fully elucidated.

Here we focus on the effort of GOMRI investigators to *understand how the introduction of hydrocarbons to the ocean triggers production of exopolymeric substances (EPS) that may protect microorganisms from the oil, emulsify the oil, or both, therefore altering its fate and transport* (i.e., degradation, dispersion or sedimentation) (Fig. 1). EPS are a group of chemically heterogeneous polymers that include micro-gels, transparent exopolymer particles (TEP) and marine snow (reviewed in Quigg et al. 2016; Decho and Gutierrez 2017). In the presence of hydrocarbons, microbes release these exudates to protect, aid their attachment, emulsify and or solubilize oil products, thus increasing the bioavailability of diverse oil components (e.g., Head et al. 2006; McGenity et al. 2012; Doyle et al. 2018, 2020). Particulate exudates, like TEP, may be sticky, promoting coagulation of marine particles and providing the matrix called marine snow (Passow 2002, 2016; Silver 2015; Quigg et al. 2016, 2020). After the DwH spill, large marine snow (> 0.5 mm) formed with elevated hydrolytic enzyme activities in association with the upper water column oil layer (Ziervogel et al. 2012; Passow et al. 2012). The mucus-rich exudates

harbored a very distinct community of interacting microbes, with a specific functionality that was different from those in the surrounding seawater (Ziervogel et al. 2012; Arnosti et al. 2016). These mucous-like flocs contained fossil carbon with the same  $^{13}\text{C}$  signature as the oil (Passow et al. 2012). The addition of the dispersant Corexit appeared to retard and reduce the formation of such mucous flocs in the presence of oil (Passow et al. 2017). MOS formation and its subsequent settling to the seafloor has been termed MOSSFA (Daly et al. 2016). Many excellent reviews (e.g., Daly et al. 2016; Passow 2016; Passow and Ziervogel 2016; Quigg et al. 2016, 2020; Burd et al. 2020; special issue of Oceanography: <https://tos.org/oceanography/issue/volume-29-issue-03>), chapters (e.g., Murawski et al. (2020a,b) and GOMRI synthesis activities (<https://gulfresearchinitiative.org/gomri-synthesis/products/>) have captured our understanding developed since the DwH – the reader is referred to these and the references therein for more details than possible here. Including all the details, particularly those important to IOSC attendees is not feasible herein.



**Figure 1.** Schematic of potential interactions between the microbial community, their exudates, the oil plus/minus dispersant. Binding between oil and exudates impact the oil's fate (dispersion versus aggregation), which in turn determines its distribution into the water column or to sediments. The microbial community composition depends on oil and dispersant availability, which in turn determines the characteristics of microbial exudates, and dictates the method by which microbes interact with the oil (directly or via exudates).

Effort was devoted towards defining the important drivers of dispersion versus aggregation of the oil-dispersant-exudate complex as well as sinking versus dispersion (Fig. 1). The critical questions addressed were (1) which microbes (bacteria, phytoplankton) are producing EPS/TEP/marine snow in response to oil spills, (2) what are the mechanisms by which EPS/TEP/marine snow aids in aggregation and/or dispersion of oil, and (3) how do oil-degrading microbes respond to dispersants and how does the presence of dispersants affect the resulting ternary system (oil-dispersant-EPS) in order to understand the role of EPS/TEP/marine snow in oil spill response.

First, oil degrading microbes are ubiquitous in marine waters, although they usually contribute only a small portion of a pre-spill community (Head et al. 2006; Baelum et al. 2012). The release of hydrocarbons as a result of a spill triggers a complex cascade of microbial responses, whereby not a single species dominates, but complex microbial consortia develop (Head et al. 2006; Baelum et al. 2012; Kleindienst et al. 2015; Arnosti et al. 2016; Doyle et al. 2018, 2020). After the DWH spill, responding oil degraders comprised largely of Gammaproteobacteria in the genus *Colwellia* and the order Oceanospirillales (Hazen et al. 2010; Mason et al. 2012; Baelum et al. 2012; Dubinsky et al. 2013; Gutierrez et al., 2013, 2016; Yang et al., 2016). Studies show genera of *Marinobacter*, *Alcanovorax*, *Cycloclasticus*, and other putative hydrocarbon oxidizers increase in relative abundance within 12-24 hours of oil addition in surface waters of the Gulf of Mexico (Doyle et al. 2018, 2020). In mesocosm studies, an increase in community diversity due to the outgrowth of several aliphatic- and aromatic-hydrocarbon degrading species was observed in response to the addition of only oil, while prokaryotic community diversity was reduced in mesocosms containing oil+dispersant and exhibited slower rates of succession (Doyle et al. 2018, 2020). Concurrently, enhanced extracellular enzyme activities and EPS production was observed

by Kamalanathan et al. (2018a) in the mesocosms containing dispersed oil, similar to Ziervogel et al. (2012) and Kleindienst et al. (2015), suggesting these observations are universal in nature. Further monitoring of enzyme activities over a longer period (16 days) offered an insight into the microbial utilization of dissolved organic matter during oil exposure, that is, a switch from oil to EPS-polysaccharide as the oil was depleted (Kamalanathan et al. 2020a).

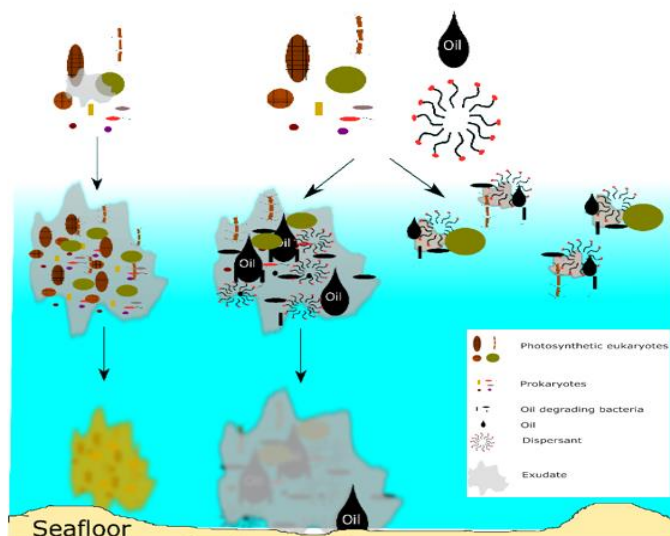
The inter-relationships between eukaryotic heterotrophs, photoautotrophs, their grazers and fungi that transform and degrade oil were reported (Bretherton et al. 2019a; Finkel et al. 2019). The very different impact of chemically dispersed oil vs dispersed oil exposure on microbial eukaryotes revealed that the former has a more negative impact on phytoplankton physiology than the latter alone. Mesocosm studies found that dinoflagellates species decreased in relative abundance in response to both oil and chemically dispersed oil, chrysophytes (predominantly mixotrophs and heterotrophs) increased in relative abundance when exposed to oil, while chlorophytes and heterotrophic euglenozoa increased in relative abundance under chemically dispersed oil treatments (Bretherton et al. 2019a; Finkel et al. 2019). Potential oil-degrading eukaryotic groups, such as Basidiomycota (fungi), increased in relative abundance when exposed to oil and decreased when exposed to chemically dispersed oil (Bretherton et al. 2019a). The impact of oil and chemically dispersed oil on diatoms is highly species specific (Bretherton et al. 2020). The diatom *Pseudo-nitzschia* released more of the neurotoxin domoic acid into the water in the presence of chemically dispersed oil versus oil alone (Bretherton et al. 2019b), leading to the suggestion that oil spills could trigger toxic phytoplankton blooms (Bretherton et al. 2019b).

Lab studies were used to examine which bacteria produce EPS in response to oil and/or dispersant. Some bacteria increase EPS production and modify the protein to carbohydrate (P:C) ratio; however no clear pattern was found (Bacosa et al. 2018; Santschi et al. 2020; Shiu et al.

2020a). Chiu et al. (2019) showed that the P:C ratio can be used to determine aggregation behavior of EPS and MOS, with Santschi et al. (2020) then proposing to use the P:C ratio as a key factor to estimate MOS aggregation potential. Less is known about the response of phytoplankton, with studies focused on the molecular mechanism governing differential responses (Bretherton et al. 2018; 2019a,b; 2020; Kamalanathan et al. 2018a,b, 2019, 2020a,b). Generally, chemically dispersed oil induces large decreases in photosynthetic function and reduced growth rates. We hypothesize this is because the increased bioavailability of oil increases membrane damage and proton leakage or it may be related to the dispersant specifically (Bretherton et al. 2018, 2020; Kamalanathan et al. 2020a,b). Oxidative stress induced by oil exposure plays a central role in selection against sensitive species of phytoplankton due to severe damage to the light harvesting complex (therefore their ability photosynthesize) and chloroplast membrane lipids. The negative impacts of oil exposure is not caused exclusively by PAH as previously hypothesized, but alkanes exert a similar effect (Kamalanathan et al. 2020b). The anti-oxidative abilities of phytoplankton appears to counter the oxidative stress caused by oil exposure; this is one of the major factors determining the resistant or sensitive nature of their response. Increased membrane damage and cell breakage would increase the concentration of protein and carbohydrates in the water column and facilitate MOS formation.

In order to develop an understanding of the mechanisms by which EPS aids in aggregation and/or dispersion of oil, findings from previous studies provided a strong foundation to test hypotheses. The biosurfactant qualities of EPS are similar to those of dispersants used for oil spill response, that is, EPS has hydrophilic and hydrophobic moieties that increase the bioavailability of some oil components to microbes (Head et al. 2006; Ding et al. 2009; McGenity et al. 2012; Fu et al. 2014; Wirth et al. 2018). EPS produced by the bacteria *Halomonas* sp. has amphiphilic

properties, interacting easily with hydrophobic substrates like hydrocarbons, leading to their solubilization and biodegradation (Gutierrez et al. 2013). EPS may facilitate attachment of bacteria (*Pseudomonas putida*) to PAHs (McGenity et al. 2012). Bacteria may affect EPS production by phytoplankton and enhance formation of aggregates and microalgal biofilms (Grossart et al. 2006; Gärdes et al. 2011), while EPS produced by phytoplankton can influence hydrocarbon degradation, including its emulsification (McGenity et al. 2012). In this way, EPS with entrained oil droplets (Fig. 2) grows to form mucus-rich aggregates that act as an energy and carbon source to other members of the microbial community (Baelum et al. 2012; Gutierrez et al. 2013). These complex networks of microbes utilizing the different components of oil and their metabolites develop into hotspots of activity (Ziervogel et al. 2012; Arnosti et al. 2016; Doyle et al. 2018) and serve as transport vehicle for hydrocarbons to the seafloor. These processes influence both remineralization and mobilization of oil-derived carbon, and consequently its fate, transport, and effects.



**Figure 2.** Marine snow formation and sedimentation and MOSSFA processes. Exudates are released naturally by phytoplankton and bacteria through active secretion or by cell lysis. These exudates are sticky in nature and flocculate on collision with each other to form marine snow, which over time lose their buoyancy and sediment to the seafloor. During an oil spill, marine snow interacts with oil, leading to the formation of MOS. Oil in MOS affects the microbial community composition by enhancing the abundance of oil degrading microbes. Similar to marine snow, the buoyancy of MOS changes with time, and leads to its sedimentation to the seafloor (MOSSFA). The addition of dispersant has varying effects on the sinking of MOS: either enhancing flocculation and sedimentation or formation of smaller MOS that remains buoyant and disperses.

Ding et al. (2009) and Shiu et al. (2020b) found that very small amounts of amphiphilic EPS (or polystyrene particles with hydrophobic surfaces) can greatly accelerate the formation of micro-gels, which are thought to be the precursor for TEP and marine snow. Sunlight is known to



inhibit or disrupt the aggregation process of marine colloids by cleavage of high molecular weight compounds into smaller, less stable fragments; however, EPS proteins excreted by bacteria can form aggregates via photo-oxidation induced cross-linking (Sun et al. 2019). Experiments on a well-characterized protein-containing EPS from the marine bacterium *Sagittula stellate* showed that proteins (via cross-linking of fragments/radicals) are the active component of EPS for aggregate formation. Marine snow aggregates did not form for a non-protein containing EPS from the phytoplankton *Amphora sp.*, supporting this assertion (Sun et al. 2017, 2018, 2019). Sun et al. (2017) also showed that hydroxyl radical and peroxide played critical roles in the photo-oxidation processes, and salt and  $\text{Ca}^{2+}$  assisted the aggregation process that leads to marine snow formation. For the first time, it was shown that light-induced free radicals, i.e., reactive oxygen species (ROS), mediated this chemical crosslinking process. The work also provides new insights into polymer assembly, marine snow formation, and the fate/ transport of organic carbon and nitrogen in the ocean.

The unique molecular characteristics of MOS harvested from controls relative to that from dispersed oil treatments reveals the oil containing particle extracts have formulas with low O/C and H/C ratios indicating solubilization of less oxygenated oil components due to degradation processes in mesocosms (Wozniak et al. 2019). CHO-containing molecular formulas were very different in dispersed oil treatments from the Macondo oil (very little CHO containing formulas), but very similar to oil exposed to weathering processes on Gulf of Mexico beaches (Reddy et al. 2012). Oil degradation, demonstrated for surface slicks and oiled sands, has now been reported within complex MOS aggregates. These results collective suggest that oil degradation is rapid and facilitated by the microbial community on time scales in agreement with estimated hydrocarbon half-lives (Wade et al., 2017), changes in oil composition (Shi et al. 2020; Morales-DeDevitt et al.

2020) and rapid micro-aggregate formation (Doyle et al., 2018). Similar phenomena reported in roller table experiments (e.g., Fu et al. 2014; Wirth et al. 2018). The rapid degradation process of oil trapped within MOS needs to be investigated with greater time resolution as thus far, little is known about how the oil in MOS is altered (e.g., Wirth et al. 2018). This is important given that the degradation profoundly effects toxicological and hence ecological impacts of the material.

Dispersants interfere with, alter, marine snow or aggregate formation by dispersing EPS micro-gels (Chiu et al. 2017, 2019; Passow et al. 2012, 2017, 2019). We have just begun to understand the complexity of interactions between the dispersant Corexit, oil and organic matter, including exudates (micro-gels EPS, TEP), which lead to seemingly opposing observations to the question whether Corexit application promotes or hinders MOS formation. On one hand, Corexit application leads to increased oil concentrations in the aqueous phase, thereby increasing the probability that oil droplets are incorporated into marine snow (Fu et al. 2014; Rahsepar et al. 2017; Hatcher et al. 2018; Passow et al. 2019). On the other hand, Corexit also disperses exudates, which are required for the formation of marine snow, thus inhibiting its formation (Chiu et al. 2017, 2019; Passow 2016; Passow et al. 2017, 2019). Corexit application to oil may result in the formation of fewer, but oil-richer aggregates, or may hinder marine snow formation completely (Chiu et al. 2017; Passow et al. 2017). In the first scenario sedimentation rates of oil will be increased, although sedimentation rates of marine particles will decrease. However, if Corexit disperses exudates efficiently, MOS formation will be inhibited and no sedimentation would be observed (see Fig. 2). In contrast to MOS, the formation of oil-sediment aggregations is independent of exudates, and not negatively affected by exudate dispersion. The dispersant effect on oil-sediment aggregations remains unresolved.

Further, the addition of Corexit was found to increase the rate at which micro-aggregates (10-200  $\mu\text{m}$ ) of microbes, oil and dispersant, form (Doyle et al. 2018). This is hypothesized to be a result of increased dissolved oil in the water, not a specific response by microbes to Corexit, and it is unknown if these micro-aggregates are related to MOSSFA or a separate phenomenon. Others found that exposure to dispersant alone enhanced EPS production, which in-turn stimulated marine snow formation (Duran Suja et al. 2019; van Eenennam et al. 2016). However, the secreted EPS was mostly protein rich and attributed to bacteria, therefore these contrasting observation suggests the initial bacterial community may be a key component in determining marine snow formation.

Studies also found dispersants influenced which putative hydrocarbon degraders increased in relative abundance in the overall microbial community (Doyle et al. 2020). While the response was different with oil alone versus oil plus Corexit, functional redundancy was apparent - there were putative oil degraders present, regardless of treatment. In one case, different operational taxonomic units of *Marinobacter* were present with high relative abundance in oil only versus oil plus dispersant. This indicates that response is likely variably at the species or strain level at times. Doyle et al. (2018), Bera et al. (2019) and others have shown that bacteria respond differently to different portions of hydrocarbons from the same source - when dissolved only versus dissolved plus particulate hydrocarbons were used in exposure experiments, there were statistically different memberships in putative hydrocarbon oxidizers following exposure.

P:C ratios of the bulk EPS were found to be consistently higher in treatments with Corexit than in oil-alone and control treatments, suggesting Corexit and oil stimulated the production of extracellular protein production more than that of polysaccharides (Xu et al., 2018b, 2019a,b; Shiu et al. 2020a). It appears that the hydrophobic interaction between hydrocarbons and proteins leads to a selective partitioning with hydrophilic polysaccharides preferentially associating with sinking

MOS, as proteins associating more with the colloidal oil-laden EPS gels become buoyant and dispersed in the water column. Polysaccharides, as one of the major constituents of EPS, are mostly hydrophilic. Acid polysaccharides such as uronic acids contain carboxyl groups and provide binding sites for divalent (e.g.,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) or trivalent (e.g.,  $\text{Fe}^{3+}$ ) ions providing bidentate inner-sphere coordination sites that can cause supra-macromolecular aggregation and  $\text{Ca}^{2+}$  bridging for structural stability (Verdugo et al. 2004). Proteins, as the other major EPS component, are amphiphilic and mediate the stability and aggregation of the 3-D networks of biopolymers, through hydrophobic and electrostatic interactions (Ortega-Retuerta et al. 2009), as well as light-induced cross-linking (Sun et al., 2017). The hydrophobic domains of EPS (consisting mainly of proteins) absorb organic pollutants, such as hydrocarbons (Liu et al. 2007). Moreover, the elevated P:C ratios of aggregates suggest that photo-reactions and subsequent crosslinking of the microbially derived proteinaceous fragments/ROS, are responsible for aggregate formation, in both the colloidal and particulate phases (Xu et al. 2018a,b). P:C ratios appear to determine the relative hydrophobicity of EPS and thus their aggregation potential (Verdugo et al. 2004; Xu et al. (2018 a,b, 2019a,b).

Schwehr et al. (2018) found microbial exudates serving as biosurfactants. The effectiveness of a dispersing agent increases with the magnitude of the interfacial tension (IFT) reduction to critical micelle concentration (CMC). Dispersant application lowers the oil/water IFT to promote entrainment of oil droplets into the water column (see Fig. 6 in Quigg et al. 2016). The efficiency of the CMC is constrained to  $< 2$  fold the concentration needed for Corexit to form critical micelles. Corexit is more effective, i.e., lowers the surface tension more than the EPS constituents; however, EPS can emulsify oil at far lower concentrations, thus beginning a process of gel growth and marine snow/MOS formation through  $\text{Ca}^{2+}$  bridging, ROS radical formation, enzymatic cleavage

radical formation, oxidation and polymerization pathways. The characterization of EPS may be useful for industry and management programs to compare against manufactured dispersant products. The implication is that the surface tension reduction in the EPS emulsifiers and subsequent gel formation mechanisms are more efficient for marine snow/MOS sedimentation efficiency while the Corexit application leads to an emulsion that does not appear to enhance biodegradation. This hypothesis was consistent with observations of lower enzymatic and lower dissolved oxygen (Kamalanathan et al. 2019), lower diversity and abundance of phytoplankton (Bretherton et al. 2019a), lower biodegradation of n-alkane and biomarker comparisons (Wade et al. 2017), less oxidative change in molecular composition of the exudates (Wozniak et al. 2019); and lower sedimentation efficiency through carbon mass balance and  $^{14}\text{C}$  analysis (Xu et al. 2018a). Similar findings of lower oil biodegradation rates and lower oxygen consumption was observed by Rahsepar et al. (2017) in marine oil snow on addition of Corexit. Santschi et al. (2020) summarized the most recent literature to describe the relationship between the P:C ratio of EPS and a number of biophysical properties related to biopolymer aggregation (e.g., relative hydrophobicity, surface activity and surface tension, attachment efficiency, light-induced chemical crosslinking) and sedimentation efficiency of marine snow. The significance of the P:C ratio of colloidal and particulate matter for “marine scavenging” of pollutants is elaborated in the study.

Responders with knowledge of the physical properties of the Macondo oil executed their preplanned efforts and kept a majority of the oil from reaching the more sensitive coastal areas. Information gathered during the GOMRI program can serve to improve predictive models for risk assessment, to develop potential interventions to reduce the environmental impact and to formulate better responding/management plans for future oil spill incidents, especially for offshore, deep

water spills and for natural resource injury assessment, as this simultaneous process gauges oil and response induced injury to facilitate restoration. The research established the interactive mechanisms of oil/dispersants, marine microbes, EPS and various environmental factors that could critically determine the fate, transport and effect of oil. This information will serve as the basis in establishing improved predictive models for risk assessment and to develop potential interventions to reduce the environmental impact and to formulate better response/management plans for future oil spill incidents. Nonetheless, much more work is required to robustly determine the relative sensitivity of microbes under a wider range of environmental conditions and how these changes in conditions will alter outcomes, specifically the formation of MOS and MOSSFA events.

The MOSSFA pathway accounted for a significant fraction (~5-31%) of the oil transported to the sea floor (Valentine et al. 2014; Fu et al. 2014; Chanton et al. 2015; Yan et al., 2016; Romero et al. 2017; Xu et al. 2018a). This transported dispersant to the seafloor but less is known about this (see eg. Passow et al. 2017). The region-wide sinking and flocculent accumulation of MOS on the sediment surface changed redox conditions, slowed down the biodegradation of the oil, and increased the spatial and temporal impacts on the benthic community and habitat suitability (e.g., van Eenennaam et al. 2018, 2019; Schwing et al 2017, 2018a,b; Rohal et al. 2020). Based on sediment records, it appears that MOSSFA also occurred during the IXTOC I blowout, but perhaps not in other significant spills (Vonk et al. 2015). For both these events, numerical models simulate MOS formation, sinking, and predict the time evolution of physical properties and spatial distribution of MOS (e.g., Dissanayake et al. 2018; Murk et al. 2020). Such models can be used during response and planning activities associated with oil spills in the marine environments.

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