

## Recent Studies on Fate and Degradation of Hydrocarbons Dispersed Subsea

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## ABSTRACT

The goal of applying dispersants as an oil spill response technique whether at the surface or subsea is to minimize surface oil impacts to people, wildlife, and shorelines and to facilitate rapid dilution and natural degradation of the dispersed oil in the water column. Thus, reliable estimates of the fate and degradation of oil, dispersed oil, and, for subsea releases, gas are key considerations when selecting response techniques. The American Petroleum Institute (API) has sponsored research on various aspects of subsea dispersant injection for over 4 years. Three of the most recent of those studies further advanced our understanding of the fate and biodegradation of hydrocarbons dispersed subsea and are discussed in this paper.

An effort to evaluate the latest dispersed oil biodegradation studies and biodegradation modeling algorithms resulted in an overview of current state-of-the-science for characterizing biodegradation processes in far field oil spill models and recommendations on improving these modeling practices. Another project examined the current state-of-the-science on oil sedimentation processes including “marine snow” formation in the context of oil spills and dispersant use. It was conducted in order to better understand dynamics, fate, and environmental impacts of oil sedimentation from the perspective of Net Environmental Benefit Analysis, NEBA (aka Spill Impact Mitigation Assessment). The third study conducted numerical modeling to predict the fate of light hydrocarbons with and without subsea dispersant use and to estimate the

changes in air quality near a well site. The goal of this effort was to evaluate whether subsea dispersant injection can reduce surface volatile hydrocarbon concentrations in the vicinity of well-control operations to protect responders' health and safety.

These and other API projects advanced our understanding of the scientific and environmental aspects of subsea dispersant use and provide a scientific basis for inclusion of this technique into contingency plans.

## INTRODUCTION

Oil spill response strategies for offshore spills can include well control, natural attenuation, monitoring, containment, mechanical on-water recovery, surface and subsea dispersants, in-situ burning, as well as shoreline protection and recovery. For offshore subsea releases, injection of dispersants subsea at the wellhead offers some significant benefits, including direct access to the freshest and non-emulsified oil in a high turbulence environment, ability to reduce the volume of required dispersant by injecting it directly into the oil stream, ability to safely operate day and night under a much wider range of weather conditions, and availability of a large water volume to rapidly decrease the concentration of a dispersed oil plume. To advance the science of subsea dispersant injection and provide a strong basis for inclusion of this technique into contingency plans, the American Petroleum Institute (API) has sponsored research on various aspects of subsea dispersant injection for over 4 years (Nedwed, 2014; Nedwed, 2015; Broje and Nedwed, 2016). This comprehensive effort included several studies on subsea dispersant injection effectiveness, oil fate and effects, subsea plume monitoring, and numerical modeling.

The following are three recent studies funded by the API that focused on the fate and degradation of oil, dispersed oil, and gas:

- Comparison of Oil Biodegradation Modeling Algorithms
- Literature Review of Oil Sedimentation Processes Including Marine Snow Formation
- Effects of Subsea Dispersants Injection on Atmospheric Concentrations of Volatile Organic Compounds Above Well Sites

This work has further advanced the scientific basis for selecting response techniques that would result in maximum environmental protection and facilitate the fastest ecosystem recovery.

#### COMPARISON OF OIL BIODEGRADATION MODELING ALGORITHMS

Biodegradation of hydrocarbons in the marine environment is an important fate pathway. Biodegradation is a biologically-mediated chemical reaction that converts hydrocarbon molecules to other hydrocarbons or to other compounds (e.g., carbon dioxide and water). Accurate prediction of degradation processes by oil fate and transport models can significantly improve predictions of oil mass balance and overall evaluation of long-term environmental impacts. Biodegradation rate studies have shown both first-order and higher-order rate laws, and have found that the biodegradation rates depend on the temperature, oxygen and nutrients availability, biological community structure and abundance, the state of the oil and the surface area of the oil/water interface for degradation of liquid oil. Studies also confirmed successful biodegradation of oil even in cold dark deepwater environments (Prince, et al., 2013; Hazen, et al., 2016, Wang, et al., 2016).

Development of numerical algorithms for modeling of oil spill fate and behavior have been ongoing for decades (Zhao, et al., 2014; Johansen, 2003; Reed, et al., 1999; ASCE 1996; Spaulding, 1988). Recent modeling advances include refined algorithms for the fate and transport of oil and gas from blowouts in the deep sea. One of the recently completed projects sponsored by API compared numerical models and different algorithms used to simulate oil fate and behavior under blowout scenarios and has resulted in a review by Socolofsky et al., 2015. A new API project is building on this earlier work and aims at investigating the underlying technical basis used to characterize degradation processes across different commonly used oil fate models and identifying opportunities to facilitate greater consistency in quantifying these processes that reflect current state-of-the-art. The degradation sub-models incorporated into four commonly used oil spill models for far-field transport, fate and exposure (SIMAP, OSCAR, GNOME/ADIOS, and MIKE) were evaluated. This effort identified the equations used to describe degradation processes including their dependence on environmental conditions, oil composition, and other parameters. A summary of the project findings is provided below.

Hydrocarbons can be present in the water either in a dissolved form, as droplets in the water column, or a slick on the water surface. Degradation of dissolved hydrocarbons is easier to model as this scenario doesn't involve a defined oil/water interface. A common approach for modeling the decay of dissolved hydrocarbons used in all reviewed models is given by the first-order rate law where the concentration of individual hydrocarbons reduces exponentially with time, as a function of a biodegradation rate constant.

Both slicks and droplets are defined by their oil/water interface and defining a two-phase concentration of oil in water becomes more challenging. The simplest approach is to assume that the mass of hydrocarbon in the liquid oil phase is also degraded following a first-order rate

process resulting in an exponential reduction of mass as a function of time as well as a different rate constant. Since this particular rate constant depends on parameters related to the surface area of the oil/water interface it is desirable to develop models that account for the size of the oil/water interface such that the rate constant is independent of the slick or droplet size or shape. One such approach is proposed by Thrift-Viveros et al. (2015) and is being implemented in the new version of the General NOAA Operational Modeling Environment (PyGNOME). Other evaluated models use different empirical rate constants for oil within different substrates (droplet, slick, sediments, etc.).

All evaluated models treat the oil as a mixture of many hydrocarbons, usually organized into groups of similar molecules. These groups are called pseudo-components, and their properties vary from oil to oil because they depend on the whole oil mixture, which is different for every oil. For simple hydrocarbon molecules (e.g., methane), biodegradation converts the hydrocarbon to carbon dioxide and water; however, for more complex molecules, degradation of one hydrocarbon may produce other hydrocarbons. Some models account for this by allowing the degradation pathway for each pseudo-component to lead to generation of other pseudo-components. A research version of the OSCAR model is an example. Other models select rate constants that are meant to represent the complete degradation pathway from the parent molecule through to carbon dioxide and water.

Next, a comparison of four selected models was conducted to evaluate the sensitivity of oil fate predictions to a range of degradation modeling approaches and to compare predicted degradation rates with the actual degradation rates reported in recent studies and field measurements. Building on a previous model inter-comparison study (Socolofsky, et al., 2015), a scenario with realistic but idealized ambient conditions (ambient velocity, turbulent diffusivity,

temperature) and discharge conditions (oil and gas flow rates, release depth, treatment with chemical dispersants, etc.) relevant to a deepwater oil release was simulated with a Lagrangian tracking model LTRANS (North et al., 2015)....

*/Placeholder for a one page of final results from this model comparison that will be available in the final draft of this paper. Modeling is expected to be completed in December/*

## LITERATURE REVIEW OF OIL SEDIMENTATION PROCESSES INCLUDING MARINE SNOW FORMATION

The natural formation of particles, consisting of macroscopic aggregates of detritus, living organisms and inorganic matter, and their precipitation mainly from the euphotic zone, to deep water of the ocean has been studied for many years. A good overview of this process has been provided in Alldredge and Silver, 1988. The classical concept of “marine snow” includes mucus-producing organisms, including phytoplankton (diatoms), casting off mucus structures from zooplankton (larvacean houses) and fecal zooplankton materials. These structures are inhabited by bacteria which also may produce exopolymeric substances. In addition, biominerals, lithogenic components of terrestrial origin and detritus contribute to “marine snow” structures (Alldredge and Silver 1988). Although bacterial biomass is involved in these processes, bacteria have not been described as a primary source for generation of “marine snow” in unpolluted seawater. However, during the Macondo response another mechanism was suggested, in which oil stimulated propagation of biomass, mainly consisting of bacterial communities, and it was suggested that this particulate aggregated material of oil and biomass was sinking at rates comparable to those in uncontaminated water. These aggregates were

referred to as "flocs" or "flocculates" (Hazen et al., 2010), "floating biofilms" and "oil-associated MS" (Passow, 2014), "aggregates", "oil aggregates", "oil gels" or "marine snow" (Passow et al., 2012; Ziervogel et al., 2012; Shanks and Trent, 1979).

Evaluation of a long-term fate and environmental impacts of oil is an important component of the Net Environmental Benefit Analysis (aka Spill Impact Mitigation Assessment). This analysis evaluates potential impacts of oil on ecosystems as a function of different response options and allows selecting a response technique, or a combination of response techniques, resulting in better environmental protection and fastest ecosystem recovery. To facilitate this analysis API has undertaken a literature review to examine the current state-of-the-science relating to the drivers of oil sedimentation processes including "marine snow" formation in order to better understand dynamics, fate, and environmental impacts resulting from these processes.

The literature review highlighted that while oil sedimentation through its adhesion to mineral particles is reasonably well understood, currently the scientific literature indicates some fundamental differences in understanding how "marine snow" is formed in the presence of hydrocarbons and whether or not localized dispersant use at the surface or subsea may affect this process. While these processes require further investigation in order to be fully incorporated into response options and environmental impact analyses, available information suggests that formation of the flocs does not prevent biodegradation of oil in the water column. Research by Brakstad et al., 2015a and Brakstad et al., 2015b confirmed that dispersed oil flocs are active sites of oil biodegradation, which will progress as the flocs continue to increase in size. Active biodegradation of oil (whether with or without dispersants) in these flocs was not considered by many authors and may have resulted in misinterpretation of laboratory experiments as well as field observations. Many estimates of the amount of oil on the seabed resulting from the

Macondo release used conservative oil tracers (e.g., hopane). Converting sediment hopane concentrations to whole oil could significantly overestimate the mass of oil on the seabed if the oil has been significantly biodegraded. Key observations made during the API literature review project as well as further discussion of the oil biodegradation process is provided in the following paragraphs.

Oil can be dispersed into the water as small oil droplets, either by turbulence of the gas and oil flow into the sea at the wellhead or facilitated by addition of dispersant subsea or at the sea surface. Partially water-soluble chemical compounds, such as the BTEX and lower molecular weight PAHs, partition from the oil droplets and dissolve into the water column. These dissolved compounds undergo rapid biodegradation in the water. The remaining oil containing the higher molecular weight PAHs and other compounds would more slowly, but progressively biodegrade to eventually leave a recalcitrant residue consisting of the higher molecular weight, and insoluble, PAHs and other oil components such as asphaltenes and resins. This residue has very low aquatic toxicity, is present at very low concentrations, and essentially is not bio-available. One of the remaining oil components would be hopane, which is often used as an oil biomarker because it doesn't biodegrade as fast as other hydrocarbons. As an example, hopane is present in the Macondo oil in concentrations of around 68.8 ppm (Stout, 2015), representing a very small fraction of the original oil. Its presence in weathered or biodegraded oil doesn't indicate that other, lighter hydrocarbons originally contained in oil are also present, nor can it serve as an indicator of residual toxicity or its potential environmental impacts.

Valentine et al., 2014 used hopane as a proxy for whole oil being present on the seabed, while acknowledging that "*the estimate does not account for biodegradation or dissolution of other petroleum hydrocarbons.*" They estimated that  $1.8 \pm 1.0 \times 10^6$  g of hopane (0.8 to 2.8



tonnes) was deposited via “marine snow” over a seabed area of 3,200 km<sup>2</sup>. This was further extrapolated to represent approximately 80,000 to 620,000 barrels of whole crude oil. Not taking the biodegradation of oil, or the dissolution of partially water-soluble compounds from the oil, into account means that the results of the analysis can easily be misunderstood, since oil compounds would have both dissolved and biodegraded aerobically in the water column before deposition, and also biodegraded aerobically and anaerobically after deposition on the seabed. The small proportion of recalcitrant oil compounds that remained, including hopane, are only evidence that hydrocarbons are present, not of the actual volume of oil deposited on the seabed. The Valentine et al., 2014 paper has been cited in many subsequent papers as evidence of the “marine snow” hypothesis indicating that considerable amounts of oil were deposited on the seabed. The actual paper contains several key statements that clarify what has been presented, but these points were not referenced in some of the subsequent papers (Stout et al., 2015).

Stout et al., 2015 presented data from work conducted with sediment samples collected in 2010-2011 and 2014. Chemical analysis was used to determine the probability of the hydrocarbons in samples originating from Macondo crude oil. The measured concentrations were interpolated over large areas to estimate a mass of oil on the seabed. In a similar way to the study by Valentine et al., 2014, the authors of this paper expressed the amount of severely weathered and biodegraded oil residue, detected on the seabed by the presence of hopane and total PAH concentrations, as the equivalent amount of ‘fresh’ Macondo crude oil released from the wellhead resulting in estimations of 219,000 to 247,000 barrels of oil, which some later reports used to infer possible environmental impacts of the spill.

However, in another report Stout, 2015 shows that the hydrocarbons on the sea floor were a severely weathered and biodegraded residue of the original oil, with a significantly different

chemical composition. This report used analytical chemistry methods (GC-FID and GC-MS) to determine the concentrations in sediments of Total Petroleum Hydrocarbons (TPH), selected alkanes, the sum of 50 PAH, alkylated PAH and sulphur-containing aromatics (TPAH50), single PAH targets (51 PAH and alkylated PAH) and petroleum biomarkers. The degree of weathering of the Macondo oil residue was quantified based upon mass depletion relative to the conservative internal marker of hopane. Oil residues in samples taken close to the wellhead (within 1 mile) were relatively lightly weathered and found in combination with varying amounts of SBM (Synthetic Based Mud) indicating that the crude oil may have been deposited on the seabed during the attempts to kill the well. In the sediments further than 1 mile from the wellhead, mass losses of 98 % n-alkanes and  $94 \pm 3$  % or more of TPAH50 were found, when compared to the source oil. The residual n-alkanes (nC30-nC40) have high melting points and their physical form are as waxy solids at the prevailing seabed temperature of 4-5°C, which makes them resistant to biodegradation. The residual PAH included higher molecular weight compounds like fluoranthenes/pyrenes, naphthobenzothiophenes, ohrrosyne/triphenylene and chrysenes. The report emphasized severe weathering of this residue on the seabed suggesting that degradation processes were “super-activated” within the chemically and naturally dispersed oil droplets in the deep-sea plume. The proliferation of bacteria, and the associated super-activity, was likely promoted by the small droplet size of the oil within the deep-sea plume which, having a large surface area to mass ratio, allowed for accelerated dissolution and biodegradation (Stout, 2015).

While other studies (Valentine et al., 2014, Stout et al., 2015) have used the presence of a recalcitrant biomarker (hopane) as a proxy to estimate the amount of ‘oil’, a report by Stout, 2015 is the first to clearly establish that the dispersed oil in the water column was severely weathered/biodegraded and was deposited on the seabed after it had already lost a proportion of

its chemical components. The material analyzed on the seabed was a wax-rich, severely weathered residue derived from the Macondo crude oil. It confirms that the estimated quantities of 'oil' deposited on the seabed, ranging from 80,000 to 620,000 barrels of oil (Valentine et al., 2014, Chanton et al., 2014, Stout et al., 2015) need to be re-interpreted in light of the finding that only a small percentage of the original oil volume could have been deposited on the seabed as a highly weathered and biodegraded residue. Potential environmental impacts of this residue should be reviewed from the perspective of not just volume, but also its composition, bioavailability and concentration in the environment. Residue that remained after severe biodegradation consisted of compounds that were too insoluble and too resistant to biodegradation to be further depleted, i.e., they were not bioavailable. Most of the potentially acutely toxic PAH compounds had been biodegraded, leaving only the highest molecular weight PAHs, such as C1-C4 fluoranthenes/Pyrenes, C1-C4-Naphthobenzothiophenes and C1-4-Chrysenes present in the residue. The physical nature of the residue would be as a high-viscosity, tarry semi-solid, perhaps encapsulated in solid wax. The data from the toxicity studies that have employed fresh or lightly weathered Macondo crude would not be applicable for evaluation of impacts of these residues. The scattered nature and low concentrations of these residues at a distance from the well should also be taken into account.

Many of the NEBA analyses conducted for offshore spills over the years as well as past field experiences indicate that actions which may reduce overall environmental impacts of the spill include preventing oil from spreading into uncontaminated areas (especially sensitive near shore and coastal areas), removing oil from the water surface, preventing it from coming into contact with concentrated populations of organisms (especially those sensitive to oil and slower to recover), diluting it to concentrations below levels of environmental concern, and reducing its

persistence in the environment (e.g. by facilitating natural weathering and biodegradation) (HDR, 2015; Lewis, and Aurand, 1997; AURIS, 1994; NRC 2005). These are the factors that often lead to a selection of dispersant use (surface or subsea) as a preferred response technique delivering these desired outcomes. While existing literature is inconsistent on the role of dispersant application in oil sedimentation, this process, which can also take place without use of dispersants, could be considered in the context of a NEBA analysis and evaluated along with other pathways and the above impact reduction criteria. The questions that should be considered are what is a concentration of hydrocarbons in the water column and in the benthic sediment, what is its chemical composition and toxicity, which species may it interact with, how long the exposure may be, what the impact is, and how quickly those populations can recover from this interaction. This can then be compared to the impacts of oil at the surface remaining untreated or treated with different response techniques. Considering any potential sedimentation of oil and associated environmental impacts in isolation from all possible oil pathways may result in a decision to protect benthic communities (especially if the presence of large volumes of fresh oil in benthic environments is mistakenly assumed) at the expense of marsh habitats, bird populations, or coastal communities that could be considerably more affected.

#### EFFECT OF SUBSEA DISPERSANTS INJECTION ON ATMOSPHERIC CONCENTRATIONS OF VOLATILE ORGANIC COMPOUNDS ABOVE WELL SITE

Protection of human health and stopping the oil from being released is always a priority in the unlikely event of the failure of all safety barriers resulting in a subsea oil blowout. Rapidly-deployable capping stacks capable of sealing off an oil well have been developed in

recent years. Industry has access to several providers of capping stacks that can be rapidly deployed globally. High concentrations of volatile organic compounds (VOCs) produced at the water surface by evaporation of surfacing oil may negatively affect deployment of this equipment, which requires direct vertical access to the well and the presence of response personnel on site for extended periods of time. Subsea dispersants use aims to prevent the oil released subsea from reaching the sea surface by dispersing it into the water column. This in turn reduces VOC concentrations near the release site resulting in greater health and safety protection for the response personnel and may serve as a critical enabler for crucial well control operations. Reduction of VOCs near the well site coinciding with periods of subsea dispersant injection was observed and anecdotally reported by the well control personnel involved in the Macondo oil spill response.

API initiated a project delivered by scientists from RPS ASA, AQA and Sage to evaluate the spatial and temporal characteristics of atmospheric concentrations of VOCs for a theoretical blowout event in 1400 m of water in the Gulf of Mexico, both with and without the use of subsea dispersant. The study was conducted in two phases and included an oceanographic and atmospheric assessment, evaluation of appropriate VOC thresholds of concern, oil fate and transport modeling, atmospheric dispersion modeling and post processing of model output to determine spatial and temporal statistics for human exposure to VOCs. The first phase involved evaluation of the appropriate thresholds of concern for the VOCs of interest (Benzene, Cyclohexane, Ethylbenzene, Heptane, n-Hexane, Toluene, Trimethylbenzenes, Xylene, and total VOCs) using an Industrial Hygiene data set available publicly from the Macondo oil spill response and other sources. Then an atmospheric dispersion tracer study with time varying meteorological forcing was used to evaluate the area affected by concentrations above the

thresholds and the maximum distance from a source to areas over thresholds. The results were analyzed to identify the correlation between meteorological conditions and metrics of atmospheric dispersion including area and distance within which concentrations exceed thresholds. The tracer study was performed using 5 years of meteorological forcing data to select specific periods of interest (7 consecutive days) with low (5<sup>th</sup> percentile), typical (50<sup>th</sup> percentile) and high (95<sup>th</sup> percentile) potential for dispersion of atmospheric concentrations of VOCs within a 2 km of the hypothetical well site.

The second phase of this project included oil fate and transport modeling coupled with atmospheric dispersion modeling of a single blowout release event for two response scenarios (with and without subsea injection) during each of the periods identified in Phase 1. Oil fate and transport modeling of a specific release scenario (single location, oil type, release rate, and Gas to Oil Ratio) was performed. Near field modeling with OilmapDeep (Spaulding et al., 2000; Crowley et al., 2014; Li et al. 2016) was used to determine the droplet size distribution and trap height of the blowout for initializing the far field model. Far field oil transport modeling with SIMAP (French McCay, 2015) was used to generate the spatial and time-varying flux from the water surface for input to the atmospheric dispersion modeling using SCIPUFF dispersion model (Sykes, 1996). SCIPUFF is a Lagrangian puff dispersion model that uses Gaussian puffs to represent a three-dimensional time-dependent concentration. The results were analyzed to develop the spatial and temporal patterns of atmospheric concentrations of VOCs within 2 km from the well site.

*/Placeholder for a one page of final results that will be available in the final draft of this paper. Phase 2 modeling is expected to be completed in January/*

## CONCLUSIONS

These and other API projects advanced our understanding of the scientific and environmental aspects of subsea dispersant use, expedite decision making and provide a solid basis for inclusion of this technique into contingency plans.

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