

**Oil plume simulations: Tracking oil droplet size distribution and fluorescence within high-pressure release jets****R.N. Conmy<sup>1</sup>, B. Robinson<sup>2</sup>, T. King<sup>2</sup>, M. Boufadel<sup>3</sup>, S. Ryan<sup>2</sup>, C. McIntyre<sup>2</sup>, M. I. Abercrombie<sup>4</sup>, K. Lee<sup>2,5</sup>**

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**ABSTRACT**

Optical measurements have been used during oil spill response for more than three decades to determine oil presence in slicks and plumes. Oil surveillance approaches range from simple (human eyeball) to the sophisticated (sensors on AUVs, aircraft, satellites). *In situ* fluorometers and particle size analyzers were deployed during the Deepwater Horizon (DWH) Gulf of Mexico oil spill to track shallow and deep subsea plumes. Uncertainties regarding instrument specifications and capabilities during DWH necessitated performance testing of sensors exposed to simulated, dispersed oil plumes. Seventy-two wave tank experiments were conducted at the Bedford Institute of Oceanography. Simulated were oil releases with varying parameters such as oil release rate, oil temperature (reservoir temp ~ 80 °C), water temperature (<8 °C and >15 °C), oil type, dispersant type (Corexit 9500 and Finasol OSR52) and dispersant to oil ratio (DOR). Plumes of Alaskan North Slope Crude (ANS), South Louisiana Crude (SLC)

and IFO-120 oils were tracked using *in situ* fluorescence, droplet size distribution (DSD), total petroleum hydrocarbons (TPH) and benzene-toluene-ethylbenzene-xylene (BTEX). For the lighter SLC, bimodal droplet size with mean diameter  $< 70 \mu\text{m}$  was achieved for 1:20 and 1:100 DOR, regardless of water temperature. Similarly, the medium ANS crude exhibited mean droplet diameter  $< 70 \mu\text{m}$ , but was bimodal only for the 1:20 treatment. Bimodal distribution was not achieved with the heavy IFO, but droplet  $< 70 \mu\text{m}$  were observed for 1:20 warm waters, indicating poor dispersibility of the high viscosity oil even for jet releases. Results offer valuable information on the behavior and dispersibility of oils over a range of viscosity, DOR and environmental conditions. Findings have implications for fate and transport models, where DSD, chemistry and fluorescence are all impacted by release variables. This research was supported by the Bureau of Safety and Environmental Enforcement.

## INTRODUCTION

The 2010 *Deepwater Horizon* (DWH) oil spill highlighted the pressing need for a better understanding of the interaction of chemical dispersants and crude oil at ocean depth. Early in the blowout release, partial emulsification of oil and heterogeneous slicks were observed as it rose to the surface from 1500-m depth (NOAA Technical Report, 2010). As a countermeasure, Sub-Sea Dispersant Injection (SSDI) was used, where dispersants were injected directly at the release point as a possible means to increase dispersion efficiency and to potentially reduce the amount of dispersant needed if applied at the air-sea interface (CRRC Report, 2010). Potential advantages of this application during a continuous oil release included the fact that the fresh (un-weathered) oil was considered well suited for dispersion, operators were able to inject the dispersant directly into the oil stream thereby maximizing dispersant/oil contact, sufficient control of DOR (Dispersant-to-Oil Ratio) could be maintained, injection may minimize the need

for surface application because of reduced oil surfacing, and optimized subsurface application would likely promote formation of smaller and stable oil droplets, enhancing biodegradation (Lee et al., 2009). The Unified Area Command (UAC) coordinated a large-scale environmental monitoring program to track and characterize dispersed subsurface oil plumes based on field data and transport modeling outputs for amount and location of oil. Oil Droplet Size Distribution (DSD) and fluorescence intensity from submersible sensors were used as an indication of Dispersion Effectiveness (DE) onboard vessels, where concentrations were monitored to evaluate oil dispersion (presence of small droplets  $\leq 70 \mu\text{m}$ ) based on previous studies for surface dispersant applications (Li et al., 2009a). Monitoring provided sound evidence of the presence of oil-bearing small particles both in surface waters and in the subsurface plume (NOAA Technical Report, 2010). SSDI correlated with a higher proportion of low molecular weight compounds in subsea waters compared to surface, suggesting that SSDI may have promoted the formation of small oil droplets in the deep sea and thus would have the potential for enhancing the weathering and dissolution of oil in the water column.

However, uncertainties still exist regarding the effectiveness of this application. For example, assumptions of the optimal DOR are based on empirical data mostly obtained from bench-scale experiments designed for testing at standard temperatures and pressures (STP). Such experiments do not take into account the effects of natural dispersion during high-pressure jet releases (i.e. deep water blowout) or the range of conditions on ocean floor, anywhere along a riser, or within the water column that could be significantly different. Hence, DOR for direct injection needs to be better understood. Although theoretical analyses and experiments suggest that oil jet breakup is insensitive to the absolute value of hydrostatic pressure for incompressible liquid-liquid systems (Masutani and Adams 2000), the effects of several ambient environmental

factors on SSDI, including high release pressure, high oil temperature, low water temperature, and the presence of methane and suspended sediments in the oil plume and/or surrounding water column remain to be clarified. Improved understanding on the influence of these factors and the interaction of oil and chemical dispersant under a range of turbulent regimes at depth is required for informed decision-making for future SSDI use.

For evaluating DE, standard laboratory tests are inherently limited in simulating operational performance under realistic environmental conditions in part due to space constraints that are critical for transport and dilution efficiency (NRC, 2005). Meso-scale tanks capable of generating waves and flume flows to simulate ocean conditions help to address this issue. One such tank facility at the Bedford Institute of Oceanography (BIO; Dartmouth, Nova Scotia) has been used previously to assess chemical dispersant effectiveness for various oil types as a function of energy dissipation rate and particle size distribution (Li et al. 2009b) and demonstrated that the effectiveness of a dispersant is strongly dependent on wave conditions, dispersant type, and oil type (Wickley-Olsen et al. 2007; Li et al. 2008; Lee et al. 2009). A strong correlation has been established between dispersion effectiveness and *in situ* DSD within the hydrodynamic regime, particularly energy dissipation rate, under a variety of wave conditions (Li et al. 2008; Li et al. 2009b), where flume mode can simulate the effects of underwater currents on dispersion and dilution of oil (Li et al. 2009a; Li et al. 2010). Experiments have also shown the reliability of fluorescence intensity as a proxy for oil concentration within oil dispersions (Conmy et al., 2014).

Presented here are results from a series of high pressure jet releases of oil within the BIO flume tank to assess the operational performance of SSDI. DSD and fluorescence intensity were used as a proxy for dispersion efficiency for treatments with (chemically-enhanced dispersion)

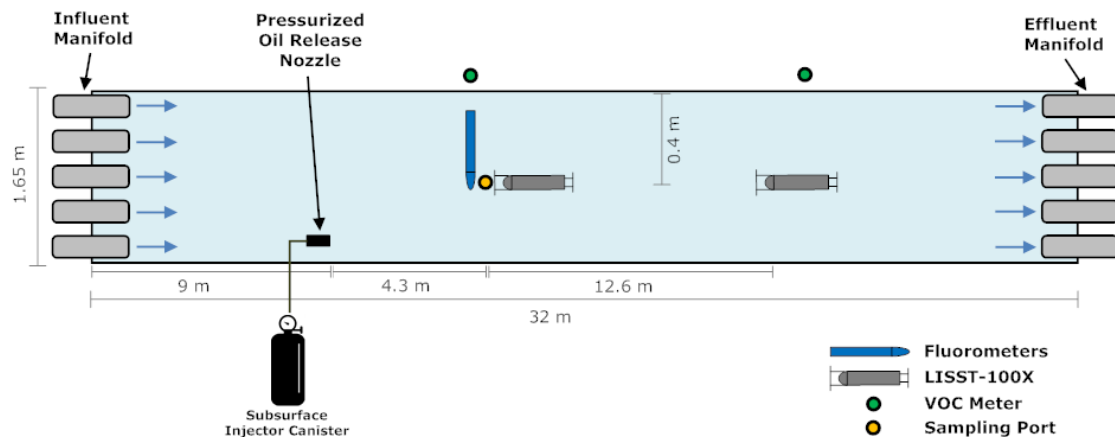
and without (physical dispersion) chemical dispersants. The objective of this work was to evaluate the effects of water temperature and dispersant-to-oil ratio on DE of three oils at high reservoir temperatures. Findings help (1) to determine the applicability of existing *in situ* monitoring technologies for SSDI and (2) inform efforts to integrate DSD into deep water blowout plume formation models that enable prediction of the dispersed oil droplets under high-flow subsurface release velocities. DWH spill monitoring program DSD and fluorescence data played a significant role in supporting emergency response operations, fate and transport modeling, and impact assessment. Findings from this study have implications for efforts in support of future deep-water spill preparedness.

## **METHODS**

**Flume Tank Description** - Oil dispersion experiments were conducted using Alaska North Slope crude oil (ANS), South Louisiana Crude (SLC) and Intermediate Fuel Oil 120 (IFO 120) with Corexit 9500 and Finasol OSR 52 chemical dispersants in the 32m long flow-through carbon steel flume tank (31,000 L) at BIO. Oils were chosen to provide a wide range of viscosity. Seawater was pulled from the Bedford Basin (50cm below surface) via two smaller centrifugal pumps (110 gpm), and filtered through high-flow polypropylene bag filters (5 and 25  $\mu\text{m}$ ) to a fiberglass holding tank (25,000 L) that supplies seawater for the system to ensure that a constant flow rate is maintained. Two high-flow centrifugal pumps (600 gpm) provided flow to generate horizontal water currents in the tank, where flow gauges on influent and effluent pipes were monitored and valve adjustments made to obtain a balanced flow rate throughout experiments. Water current velocities were measured in the tank using an ADV (Nortek Vectrino) to maintain consistent horizontal water current velocities (3.5 cm/s) at all measured depths. At the completion of experiments, oil absorbent pads removed oil from the water

surface. Effluent wastewater was filtered over layers of polypropylene PomPom Oil-Mops to remove insoluble oil from the water prior to discharging, where effluent samples were collected and PomPom's were changed if oil concentrations exceed the minimum guidelines (10 ppm) for wastewater discharge in Canada.

**Subsurface Oil Injection System** - A custom subsurface oil injection system was used to generate dispersed oil plumes in the tank (Figure 1), consisting of a 2 L stainless steel pressure vessel and a series of valves and pressure gauges, fastened to the outer wall of the tank. Inside the tank, a fitting connects the outer assembly to a nozzle (2.4 mm), which extends mid-width perpendicular to the tank wall (9 m downstream from the influent pipe) and is angled at the tip to direct the discharge plume downstream and use horizontal length of the tank to capture the plume movement. For each experiment, oil or oil/dispersant premix was added to the pressure vessel in order to reduce the influence of any additional confounding factor of mixing effectiveness. It should be noted that pre-mixing would expectedly enhance the dispersion compared to dispersant addition to an oil jet (Nedwed, 2015). Copper coil within the pressure vessel connects to a water bath to permit oil heating to 80°C, and then pressurized (40 psi for ANS and SLC; 60 psi for IFO 120) with compressed Nitrogen. A ball valve connected to the vessel controls the oil release through the subsurface nozzle into the tank. Release time and total volume (determined by mass) of oil injected are recorded. After each experiment, the subsurface injector system was cleaned by flushing with toluene, acetone and fresh water.



**Figure 1. Schematic diagram showing the location of the subsurface injector and *in situ* instrumentation submerged within the tank.**

**Submersible Sensor Deployment** - Sensors were mounted on an aluminum frame located 4.3 m from the oil release point with their pressure windows and at a depth of 0.4 m. The instruments were attached to a crosspiece support bar, so that they were all located the same distance downstream from the oil release point with the pressure window pointed directly towards the bottom of the tank. Data acquisition systems were used to control and collect data with real-time displays from the *in situ* fluorometers. A total of six *in situ* hydrocarbon fluorometers were evaluated during this study; Chelsea Technologies Group UV Aquatrackas - Refined and Crude models, Turner Designs Group Cyclops - Fine and Crude models, Sea Bird – Wet Labs Inc ECO-FLU, and GmbH Trios. Sampling rates and units of signal intensity varied for each instrument with one reading every 5 seconds for GmbH ( $\mu\text{g/L}$  PAH), every 3 seconds for Cyclops (auto-gain feature; mV), and every 1 second for the ECO and Aquatrackas ( $\mu\text{g/L}$  QSDE,  $\mu\text{g/L}$  perylene and  $\mu\text{g/L}$  carbazole, respectively). Oil droplet size was measured *in situ* using two LISST-100X (Laser *in-situ* Scattering and Transmissometry) particle size analyzers (Sequoia Scientific, Inc), measuring particle size from 2.5 – 500  $\mu\text{m}$ . The first LISST (upstream LISST) was located immediately after the fluorometer package support frame at a distance of 5.1 m from the oil release point and the second LISST (downstream LISST) was located at 16.9 m

from the oil release point and both at a depth of 0.4 m (Figure 1). Placement was informed by the numerical modeling efforts conducted previously in the tank to maximize oil droplet detection without saturating the instrument. Real-time data was provided by LISST-SOP data acquisition software (ver. 5) with a sample acquisition rate of one measurement every 3 seconds. Underwater video of oil droplets and the transport of the plume were captured using a GoPro Hero4 digital camera, as well as a Sony RX100 III digital camera with underwater housing.

**Submersible Sensor Data Processing** - LISST data files were processed using a statistically-based quality control script written using the R statistical package. This script identifies and removes “Over Range” samples (defined as 0  $\mu\text{L/L}$ ) and outliers. Outliers are defined as any reading that is greater than the moving mean (5 data points before and after the targeted time point) of the dataset multiplied by four times the standard deviation (over the same interval as the moving mean). Due to the potential for one or more extreme outliers to skew both the moving mean and standard deviation calculations for points around them, this outlier detection routine is run iteratively, excluding previously flagged points, until no more outliers are detected. The script then calculates the Total Particle Concentration (TPC), Volume Mean Diameter (VMD), and Particle Size Concentration (PSC). It then detects the plume curve (if present) and time-normalizes the data based on that location. Data are presented as Droplet Size Distribution (DSD). Plots presented include data 2 minutes before and 8 minutes after the start of the plume curve. Data from the Downstream LISST were normalized so that the plume began at  $t = 5$  min in order to visually convey that the plume was detected in the tank roughly 3 min after detection by the LISST further upstream near point of injection release. Similar to the LISST data, a script was used to detect outliers in the fluorometry data. Curve detection was performed and the data was time-normalized to include 2 minutes of data before, and 8 minutes of data after



the start of the plume curve. The baseline of the plume curve was calculated using data points observed in the first minute preceding the start of the curve and this baseline was subtracted from the data. Finally, factory calibration factors were applied to the data.

**Experimental Design** - Seawater temperature and salinity in the tank were recorded using a handheld probe (YSI Incorporated, Yellow Springs, OH). Prior to experiments, the tank was operated in recirculation mode (10 minutes) to allow current flow to stabilize. Five minutes prior to oil injection, data-logging on all instruments was started and background seawater samples were collected. After oil injection, the real-time readout of the fluorometer signal was monitored for the first spike in signal intensity (usually after 2 minutes), and bottle chemistry sample collection began. The experiments ran for 12 minutes and the tank was cleaned using Big Orange Degreaser, to prevent any potential contamination between experiments. Instrument windows were cleaned using disposable alcohol wipes. Water samples were returned to the lab and stored at 4°C.

**Discrete Chemistry Samples** - Water samples for chemical analysis were collected at various time points throughout the experiments. Three ¼” stainless steel tubes were attached to the aluminum fluorometer frame, so that the end of the tube was located at the same depth as the instrument pressure windows (0.4 m). These were attached via silicon tubing to a digital peristaltic pump with a 120 mL/min flow rate. Samples were analyzed for Total Petroleum Hydrocarbon (TPH) using a method developed by DFO in-house (Cole et al., 2007; King et al., 2015) for Gas Chromatograph (GC) - Flame Ionization Detector (FID) analysis. An eight point calibration was generated using standards prepared from the appropriate crude oil stock that was used to generate the TPH samples (e.g. ANS, IFO 120 and SLC). Peak quantification was performed using relative response factors. Samples were analyzed for benzene, toluene,

ethylbenzene and xylene (BTEX) using EPA Method 8240 (purge and trap), modified by running a GC - Mass Spectrometer (MS) in selected ion monitoring mode to include ethylbenzene. BTEX standards were prepared in 40 mL purge and trap vials; and samples and standards were analyzed using this method, along with sample blanks and duplicate samples. TPH and BTEX results are not shown here in detail but can be found in the EPA 600/F-16/250 Report (2016).

## RESULTS & DISCUSSION

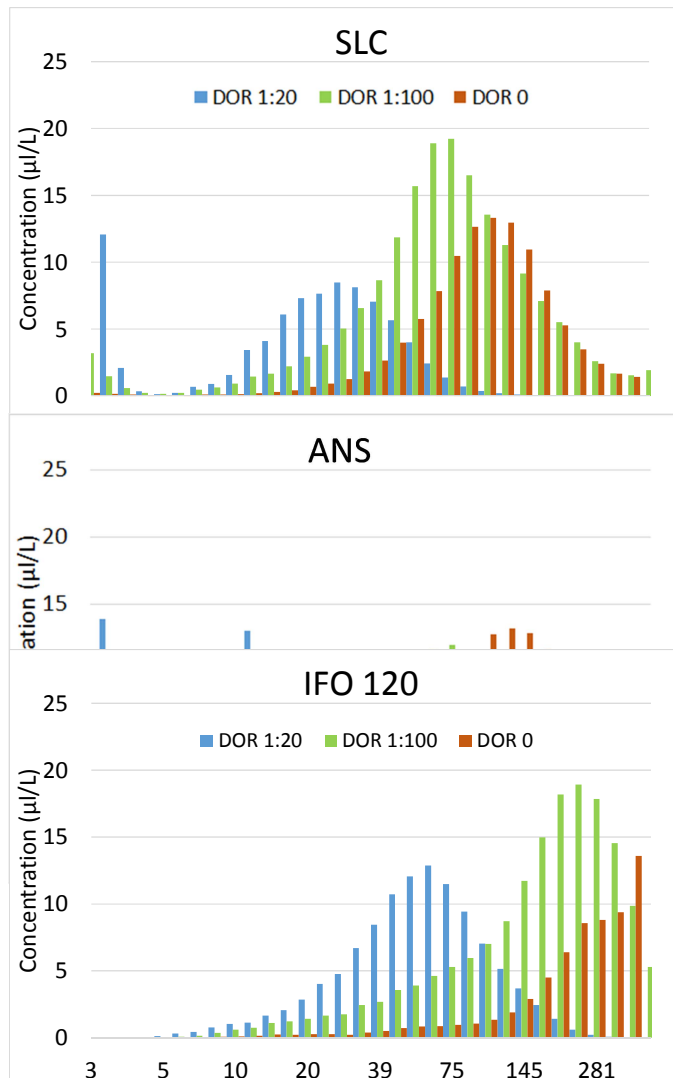
A total of 72 experiments were conducted in replicate, where water ranged in temperature from 4.9 - 20.3 °C, with 11 °C serving as the cut off between warm and cold water tests. Previous experiments within the BIO flume tank facility have assessed dispersion effectiveness as a function of energy dissipation rate and particle size distribution (Li et al. 2009a) and demonstrated that DE is strongly dependent on wave conditions, dispersant type, and oil type (Lee et al. 2009) and highly correlated with *in-situ* droplet size distribution (Li et al. 2008; Li et al. 2009a). Here, DE was evaluated via response monitoring tools (fluorescence and particle size analyzers) during high-pressure subsurface injection experiments.

**Dispersant-to-Oil Ratio (DOR) Effects**- Addition of either Corexit 9500 or Finasol OSR 52 chemical dispersants to Alaskan North Slope (ANS), IFO 120 and South Louisiana Crude (SLC) oils resulted in a decrease in the Volume Mean Diameter (VMD) and shifted the Droplet Size Distribution (DSD) to smaller droplets for warm water treatments (Figure 2). For ANS, dispersions created without chemical dispersants or Dispersant-to-oil ratio (DOR) = 1:100 yielded VMD > 70 µm and exhibited unimodal DSD. Dispersions created with DOR = 1:20 yielded VMD < 70 µm size range with a bimodal DSD. SLC oil was more dispersible compared to ANS for treatments with and without chemical dispersant. Bimodal distribution was observed during DOR = 1:20 and some DOR = 1:100 experiments indicating that the jet release of this

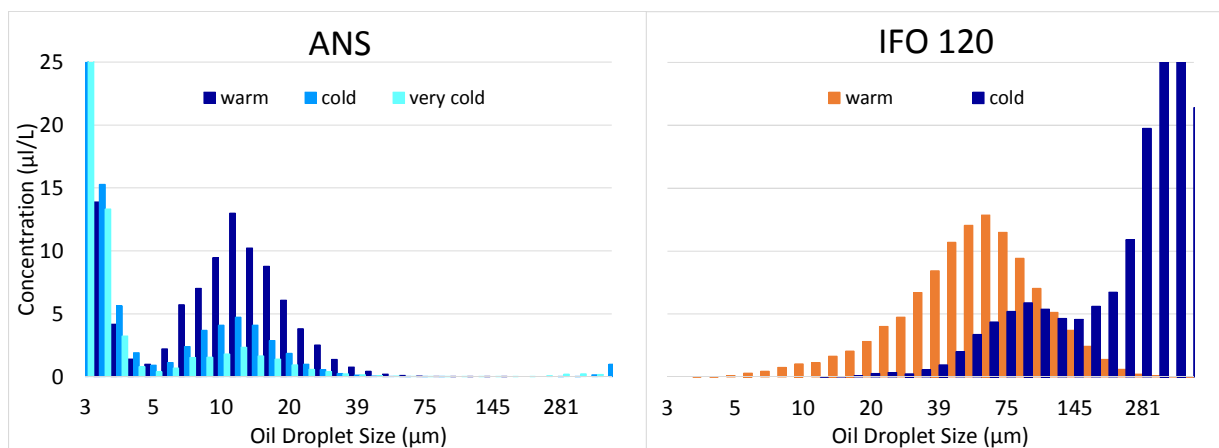
particular oil into warm water produced smaller droplets than the ANS. During the DWH spill, droplets  $\leq 70\mu\text{m}$  were considered small enough to remain in suspension and were monitored as a proxy for evaluating oil dispersion efficiency (NOAA Technical Report, 2010). This suggests that produced SLC droplets from a DOR = 1:20 or 1:100 dispersant injection, and ANS droplets at DOR = 1:20 would likely remain dispersed in the presence of mixing energy given the larger proportion of small droplet sizes observed. In general, Corexit 9500 produced slightly smaller droplets compared to Finasol OSR 52 for warm water treatments (11-20°C). The addition of Corexit 9500 or Finasol OSR 52 to IFO 120 during warm temperature experiments resulted in a shift in DSD and a decrease in VMD; however bimodal distribution was not achieved and even DOR = 1:20 did not yield VMD less than 70  $\mu\text{m}$  in most cases. This suggests that dispersant addition to this oil at warm temperatures would not yield droplet sizes that would likely remain in suspension.

**Water Temperature Effects** - For ANS, dispersion with  $< 70 \mu\text{m}$  droplet VMD was observed for the DOR = 1:20 treatments at both cold and warm water temperatures (Figure 3). Water temperature did not appear to influence the DSD or VMD for this lighter crude oil. However, a temperature effect was observed on the Total Particle Concentration (TPC), where lower temperatures coincided with fewer particles dispersed within the plume for a given volume of oil injected. This may be due to enhanced density differences between the oil and water at colder temperatures, thus quicker rise time of the produced droplets. For the addition of Corexit 9500 or Finasol OSR 52 to IFO 120 at cold water temperatures, smaller droplet sizes were not observed, where DOR = 1:20 remained above 200  $\mu\text{m}$ . This suggests that dispersant addition to this oil at cold temperatures would not yield droplet sizes that would likely remain in suspension.

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**Figure 2. LISST DSD for SLC, ANS and IFO 120 with Corexit 9500 at three DOR treatments in warm water (11-20°C). Histograms represent averages obtained at the 2 minute time stamp during each experiment.**



**Figure 3. LISST DSD for ANS and IFO 120 with Corexit 9500 at DOR = 1:20 and varying water temperature. ANS temperatures are 15, 9 and 6 °C for warm, cold and very cold classifications. IFO temperatures are 17 and 7°C.**

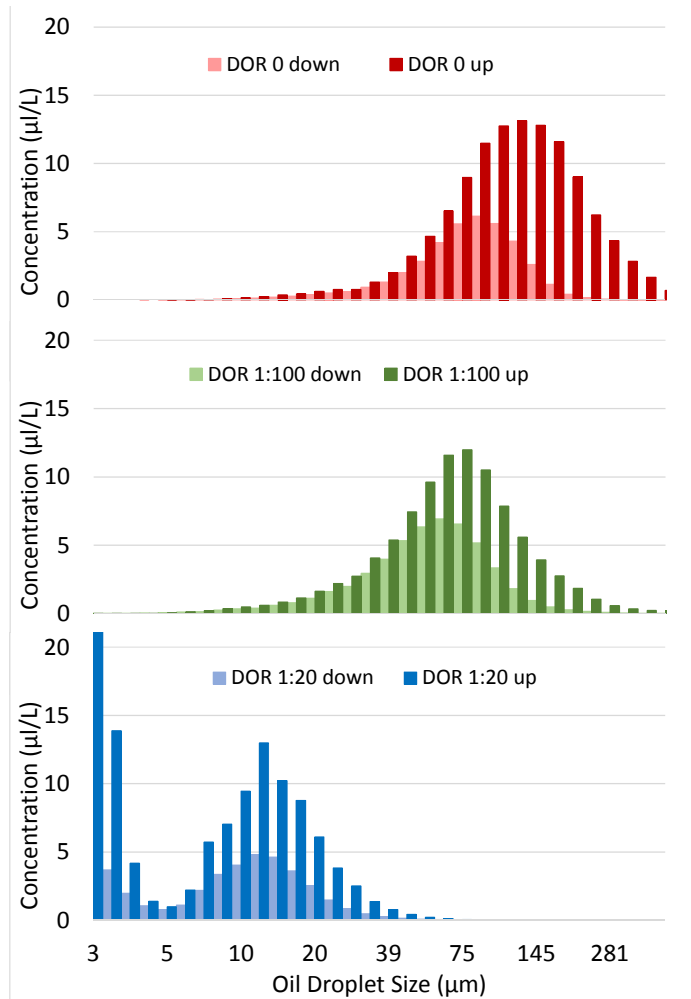
For experiments conducted at water temperatures  $< 5$  °C, the LISST particle size analyzer yielded unexpected DSD where even a unimodal distribution was not measured.

Chemistry and *in situ* fluorescence data indicate that the oil was in fact dispersed adequately. This suggests potential operational problems for this unit  $< 5$  °C, even though it is within the operating temperature (manufacturer manual). Additional testing of cold water temperature limits is recommended. Data collected for  $< 5$  °C temperatures are not included in this paper.

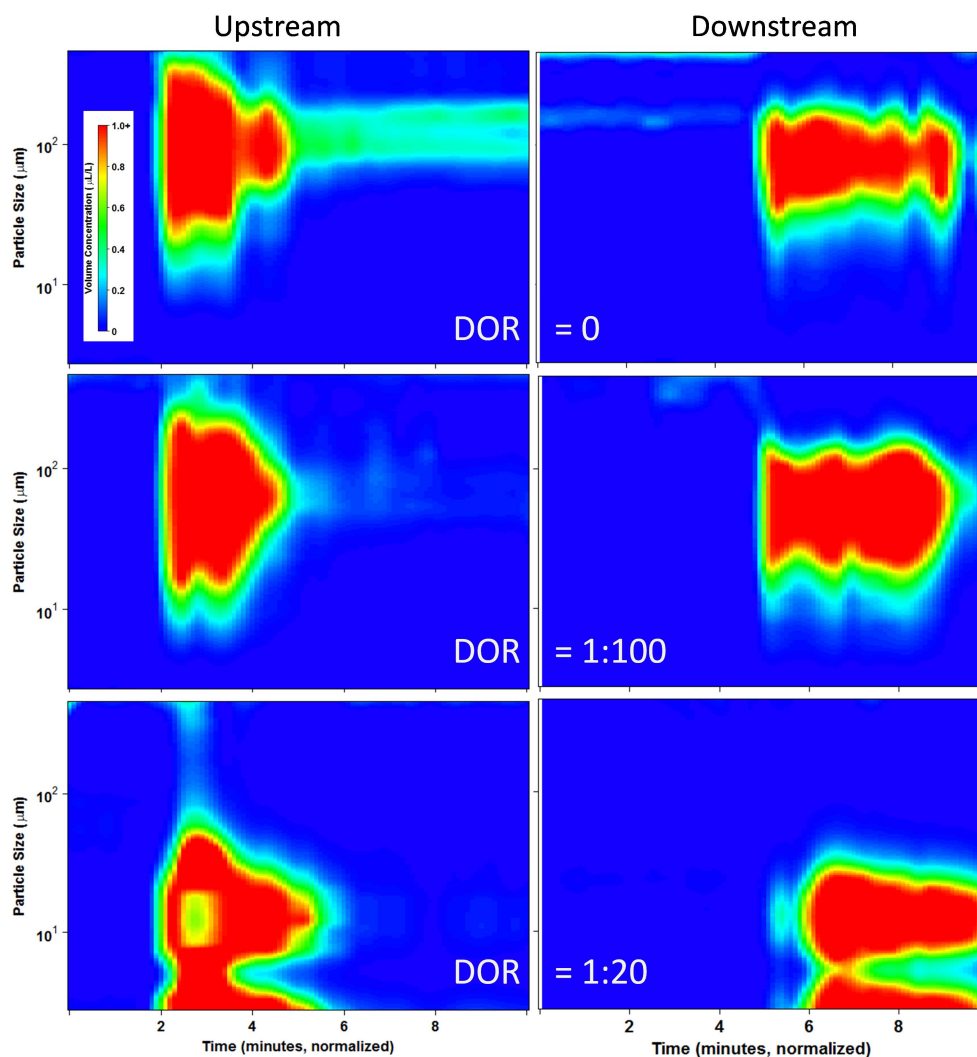
**Upstream and Downstream Particle Fractionation** - Histograms representing the particle concentration for a given size class from the upstream and downstream LISSTs suggest dilution and fractionation of particles within the plume. The downstream LISST positioned further from the jet release serves as an indication of plume evolution through the tank. For all oil and DOR treatments, dilution in particles (decrease in TPC) within the plume expectedly occurred. Also evident is a shift to smaller particles for all DOR treatments as the plume moves through the tank, where size fractionation was dependent on DOR treatment (Figure 4). Physical dispersion (DOR 0) treatments for lighter oils were found to have the largest shift in DSD and VMD between the upstream and downstream LISSTs suggesting the largest effect of particle fractionation in the plume. Where the decrease in TPC suggests plume dilution in the tank, the DSD shift to smaller particles suggests that within each experiment larger droplets were removed from the plume within 6 minutes of the oil release, most likely rising to the surface of the tank. The effect was least observed in the DOR 1:20 treatments where dilution occurred but the presence of smaller droplets (with lower rise times) at the point of release most likely didn't allow for fractionation. The effects of fractionation were observed in warm and cold treatments.

Time series particle size data allows for evaluating plume dynamics over the duration of the experiments. Figure 5 depicts time series contour plots of the plume, where colored contours represent the particle concentration (normalized to max value for comparison purposes) for given

droplet sizes through time. These plots allow for ascertaining how the DSD shifts over the duration of the release as the plume moves in the horizontal direction in the tank. For ANS, data suggest that the leading edge of the plume, as detected by the upstream LISST, exhibited a wide range in DSD during the first few minutes after release. As the plume advances down the tank, the DSD becomes narrower with a shift in the VMD for the DOR = 1:100 and 0 treatments. DSD remained similar for the DOR = 1:20 treatment. Warm and cold water treatments exhibit similar trends.



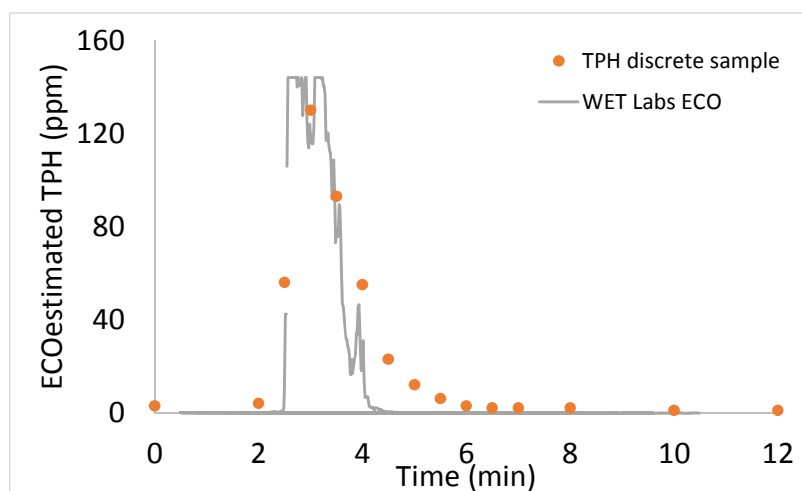
**Figure 4. Upstream and downstream LISST Droplet Size Distribution and particle concentration for ANS with Corexit 9500 during warm water treatments.**



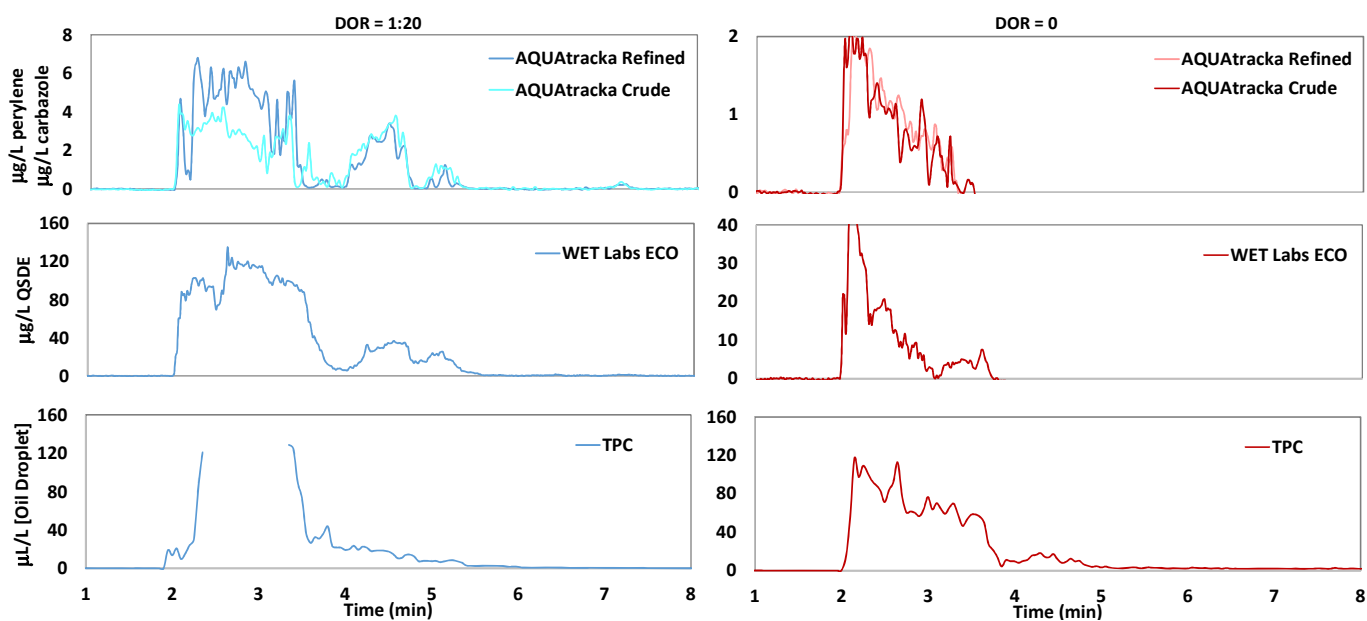
**Figure 5. Time series contours of droplet size distribution and particle concentration from the upstream (left panels) and downstream (right panels) LISSTs for ANS with Corexit 9500 during warm water treatments.**



**Plume Fluorescence Intensity** – In addition to the plume transport of particles (oil droplets), dissolved hydrocarbons were also measured via discrete samples and by proxy using fluorescence intensity. The reliability of *in situ* fluorescence measurements as a proxy for oil concentration within physically and chemically dispersed oil has been demonstrated previously within the tank (Conmy et al., 2014). Here, *in situ* fluorescence serves as a good proxy for oil concentration during the subsurface injection experiments. However, heterogeneity of the produced plumes and the short time scale of experiments (~10 min) led to difficulties in correlations between the plume particle size analyses and chemistry results. This is in part due to discrete samples representing 15 second averages as opposed to instantaneous measures given by fluorometers and particle size analyzers. Thus, although general trends were similar, exact matchups between *in situ* data streams and discrete samples proved to be challenging (Figure 6). Given the experimental design, fluorescence was found to be better suited for correlation with total particle concentration (Figure 7). Depicted are the *in situ* fluorescence and TPC data during a warm water experiment of ANS with (left) and without (right) Corexit 9500. When exposed to dispersant, fluorescence estimates of dissolved hydrocarbons was higher compared to oil alone treatments, and signal persisted for longer periods of time. Whereas a sharper decay in the fluorescence intensity and the TPH values were observed for the physically dispersed oil.



**Figure 6. Fluorescence intensity and TPH samples for a warm water treatment using SLC at DOR = 1:20.**



**Figure 7. Fluorescence intensity and TPC as a function of time for ANS warm water treatment.**

## CONCLUSIONS

*In situ* oil droplet size distribution, particle concentration and fluorescence data from this study are essential for better understanding dispersion effectiveness and for refining droplet formation and trajectory models, such as modified Weber Number technique (Johansen et al., 2013) and the VDROD-J model (Zhou et al., 2014). These results demonstrate that during high pressure releases, light crude oils such as SLC form droplets with VMD < 70  $\mu\text{m}$  and bimodal

DSD for 1:20 and 1:100 DOR regardless of water temperature. The effect was similar for the medium ANS crude oil, but bimodality was only observed for the 1:20 treatment. Conversely, Bimodal distribution was not achieved with the heavy IFO, but droplet  $< 70 \mu\text{m}$  were observed for 1:20 warm waters, suggesting poor dispersibility of the high viscosity oil even for jet releases. Droplet fractionation observed in the plumes was greatest for the lighter crude and demonstrates the separation that would occur as a plume rises and is advected horizontally in a water column. Thus, findings from this study have implications for predicting the behavior of oil and plume formation, decision-making pertaining to SSDI as a countermeasure, and assessing the transport and fate of slicks and subsurface plumes in support of emergency response operations during any future deep water spills.

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