# ANNULAR FLUME EXPERIMENTS ON OIL - MINERAL AGGREGATES : AN OIL SPILL COUNTERMEASURE ON LOW ENERGY SHORELINES

Danielle Cloutier, Institut des Sciences de la Mer (ISMER), Université du Québec à Rimouski, Rimouski, Qc, Canada

Philip R. Hill, Geological Survey of Canada, Pacific Geoscience Centre, Sidney, B.C., Canada Carl L. Amos, University of Southmapton, Southampton Oceanographic Center, United Kingdom Kenneth Lee, Centre for Offshore Oil and Gas Environmental Research, Bedford Institute of Oceanography, Dartmouth, Nova Scotia

# ABSTRACT

The formation of oil mineral aggregates (OMA) has been identified as a significant process regulating the natural removal of stranded oil on low energy shorelines. This process involves the interaction of oil droplets with fines to form OMA which enhance dispersion rates of oil into the sea. Laboratory experiments were conducted in an annular flume to determine the current speed necessary to initiate the rupture of a stranded oil film of Hibernia crude oil (HCO). Suspended sediment concentrations (SSC)  $\leq$  600 mg L<sup>-1</sup> were used to investigate the effect of turbidity on the critical stress. The OMA formed were analyzed to determine the type and size of the aggregates corresponding to different environments.

# RÉSUMÉ

La formation d'agrégats argile-pétrole est reconnue comme processus favorisant l'auto-nettoyage des plages de faible énergie. Ce processus implique l'interaction des particules fines en suspension avec les gouttelettes de pétrole afin de former des agrégats dont les propriétés limitent l'adhésion de l'huile au substrat et facilitent leur dégradation dans le milieu. Des expériences réalisées en canal annulaire ont permis de déterminer une vitesse de courant critique permettant l'érosion du pétrole échoué. L'effet de la turbidité sur le stress critique a été évalué sous des concentrations de sédiments en suspension (SSC)  $\leq$  600 mg L<sup>-1</sup>. Les agrégats formés sous différentes conditions de vitesse de courant et de turbidité ont été analysés afin d'en déterminer le type et la taille.

# 1. INTRODUCTION

The formation of oil-mineral aggregates (OMA) through the interaction of fine mineral particles with oil stranded on shorelines following spills has been shown to be an effective natural cleaning process in most environments and particularly in low energy, sheltered shorelines where wave action and abrasion are negligible (Bragg and Owens 1994, 1995; Bragg and Yang, 1995; Michel et al., 1993; Lee et al., 1997). Oil-mineral-aggregate formation presents a highly positive potential in self-cleaning of spilled shorelines as OMA formation can result in enhanced removal of oil from the shore because OMA have overall properties that reduce residual oil coalescence at the sea surface and its adherence on solid substrates (Beslier et al., 1980; Gundlach and Reed, 1986). Moreover, small sedimentcovered oil droplets experience more rapid microbial degradation than "bulk free surface oil" and oil that has coalesced into large globules (Lee et al., 1997; Weise et al., 1997).

OMA have been generated easily in laboratory by shaking a variety of crude oils with seawater containing mineral particles (Lee et al., 1998). Predominantly, three types of OMA have been characterized by microscopy techniques : droplet, solid and flake aggregates (Lee and Stoffyn-Egli, 2001). Droplet OMA are composed of one or more droplets with mineral particles attached to their surface only. Solid OMA are nonspherical, often elongated aggregates (~ 250

 $\mu$ m) that exhibit particles within the oil boundaries. Flake OMA look like membranes, usually floating or neutrally buoyant, and can be several mm wide. Once formed OMA appear to be very stable structures.

The effectiveness of natural cleaning by OMA formation depends on a number of factors but is primarily related to the amount of wave energy reaching the shore. OMA formation requires wave energy to initiate 1) the rupture of the oil emulsion into small droplets; 2) sediment suspension into the water column; and 3) interaction of fine material with oil droplets. In low energy environments, the availability of fine sediment is usually not a limiting factor in the oil/sediment interaction process. It is more likely limited by the energy within the surf zone that is needed to break the oil emulsion into small droplets.

Studies have been conducted to show the formation and characteristics of OMA at various spill sites and yet quantitative evaluation on the physical parameters responsible for oil elimination is still lacking. This study was undertaken to examine the steady flow conditions under which a stranded oil film will be eroded. Hibernia crude oil (HCO) was used as the test oil. The effect of suspended sediment concentration (SSC) on the erosion process was examined. It is hypothesized that turbidity associated with suspended mineral fines will reduce the critical shear stress necessary to break the oil film and initiate the process of oilmineral aggregation due to the presence of a solidtransmitted shear stress. To test this hypothesis, a study with the Lab Carousel, a laboratory annular flume, was conducted. The characteristics (size and type) of OMA formed were analyzed under various current velocities and levels of SSC. They are essential parameters to examine as they will help determine the fate of dispersal aggregates.

#### 2. METHODS

#### 2.1 Lab Carousel

Lab Carousel is an annular laboratory flume designed to examine the erosion and settling of sediments under controlled, monitored conditions (Fig. 1). It consists of an acrylic annular trough, 2 meters in diameter, 0.15 m wide, and 0.45 m deep. The flume volume is 0.268 m<sup>3</sup> (to a depth of 0.30 m). The flow in Lab Carousel is induced by a top rotating lid equipped with eight paddles spaced equidistantly (0.78 m). The speed and acceleration of the paddles are controlled by a high performance microstepper driver & indexer which is controlled by a PC. Tangential and radial currents are measured using a Marsh McBirney® electromagnetic current meter positioned 0.15 m above the flume base. The relationship between motor speed (M), lid rotation ( $U_r$ ), and azimuthal current speed ( $U_v$ ) is :

$$U_r = 1.17 x 10^{-4} \text{M} \text{ (m s}^{-1})$$
 (1a)

and :

$$U_y = 0.574 U_r \text{ (m s}^{-1}) \text{ (1b)}$$

The friction velocity ( $U_*$ ) is calculated using equations derived from the Sea Carousel calibration, a benthic annular flume developed for field applications (Amos et al., 1992):

$$U_* = 0.0167 + 0.097 U_y \text{ (m s}^{-1})$$
 (2)

The critical bed shear stress ( $\tau_c$ ) is the force exerted on the bed per unit area above which erosion takes place ; it is determined from:

$$\tau_{\rm c} = \rho \ U_*^{\ 2} \ ({\sf Pa}) \tag{3}$$

where  $\rho$  is fluid density (1021 kg m<sup>-3</sup>).



Figure 1. The Lab Carousel filled with seawater and sediments.

### 2.2 Hibernia crude oil concentration monitoring

Oil erosion experiments were carried out using Hibernia crude oil artificially weathered by forced aeration for 24 hours. An oil slick was formed by stranding 200 g of oil on a section of the flume base. Natural sea water ( $13^{\circ}C$ , 30 ppm) from the Saint-Lawrence River pre-filtered on a 1 m thick sand bed was then introduced into the oiled flume. A Sea Tech fluorometer positioned at 0.15 m above the flume base was used to monitor oil content in the water column. Fluorometer calibration was performed using water samples collected from the middle sampling port mid-way through each applied current velocity. The total hydrocarbon content (dissolved plus particulate) was determined by measuring absorbance at a wavelength of 233 nm on a Beckman® DU 640 spectrophotometer using the same solvent as that used for extractions (CH<sub>2</sub>Cl<sub>2</sub>).

## 2.3 Suspended sediment concentration monitoring

The sediment used in this study was a commercially obtained red potters clay called PHB-non-grog and contained 85% clays and 15% silt and the mean size was 6  $\mu$ m. The mineral composition measured by X-Ray diffraction revealed that the sediments contained quartz, kaolinite, and illite. The sediments were devoid of organic matter. Sediments were deflocculated by ultrasounding prior to the introduction into the flume. Suspended sediment concentrations of 100, 200, 250, and 600 mg L<sup>-1</sup> were evaluated. Sediment slurries were introduced into the channel from the surface.

Turbidity in the flume was monitored using three optical backscatter sensors (OBS) (Downing et al., 1981; Conner and Visser, 1992) located in the inner wall of the annulus at heights of 0.03, 0.10 and 0.20 m above the flume base. These sensors determine the amount of backward scattering of infrared light caused by the fluid/sediment sample and hence monitor the temporal variation of turbidity in the flume. OBS calibrations were derived under well-

mixed conditions, using water samples collected from three sampling ports also located in the outer wall of the annulus, at the same heights as the OBS sensors.

# 2.4 Experimental design for OMA analysis

A video camera was focussed on a ruler placed on the flume wall. Images of the OMA formed under different conditions of turbidity and current speed were recorded immediately after the current was stopped. The video images were freeze-framed and image analysis was carried out using the PC-based analysis package Image Pro-Plus (IPP) from Media Cybernetics Inc.

# 3. RESULTS

3.1 Determination of a critical shear stress for the erosion of Hibernia crude oil

Two replicate experiments were carried out to determine the critical shear stress for oil erosion at  $13^{\circ}$ C in clear water (Fig. 2 and 3). The current speed was progressively increased with small step increments (0, 0.06, 0.14, 0.21, 0.27, 0.38, 0.41, 0.48, 0.55 m s<sup>-1</sup> for thirty minutes each) until erosion of the slick surface was observed. The current speed was held constant for one hour after oil erosion began, stopped for thirty minutes (to determine the buoyancy of oil droplets), and then further increased to 0.65, and 0.75 m s<sup>-1</sup> (Fig.3A). Periods of still water were again imposed between these last increments.

The results for the two replicate runs are presented in Figure 2 and 3. Figure 2, at an expanded scale, illustrates that some minor oil erosion took place during the early stages of the experiments ( $U_y$  < 0.55 m s<sup>-1</sup>). This was manifested by small increases in oil concentrations and the formation of an oil deposit around the flume wall at the level of the water surface, in the velocity range 0.06 <  $U_y$  < 0.27

m s<sup>-1</sup> (0.52 <  $\tau_{cl}$  > 1.88 Pa), for both runs. At these velocities, no visual deformation of the oil slick was observed and oil concentrations remained < 30 mg L<sup>-1</sup> (Fig. 2). This behavior is defined as Type I erosion.

At  $U_y$  = 0.55 m s<sup>-1</sup>, oil droplets detached from the flume base and concentrations increased to 39 and 26 mg L<sup>-1</sup> for

runs 1 and 2 respectively. This is defined as the beginning of type II erosion. The critical bed shear stress for Type II

erosion ( $\tau_{cll}$ ) was thus 5.0 Pa. After the current was stopped, the oil concentrations decreased and stabilized around 13 and 20 mg L<sup>-1</sup> suggesting that oil was present in the water column but in emulsion form (otherwise it would have floated to the surface).

At  $\,U_{_{\mathcal{V}}}$  = 0.65 m s  $^{-1}$  (T<sub>cll</sub> = 6.5 Pa), Type II oil erosion was

again evident, seen as a rapid increase in oil concentrations to values of 235 and 320 mg  $L^{-1}$  for the replicate runs (Fig.

3B). At  $U_y = 0.75 \text{ m s}^{-1}$  ( $\tau_{cll} = 8.2 \text{ Pa}$ ), oil continued eroding from the flume base, and concentrations reached approximately 450 and 535 mg L<sup>-1</sup>. During still water periods, the visible oil droplets in suspension floated rapidly to the surface but the oil content (C) remained in the range of 150 < C < 200, and 200 < C < 300 mg L<sup>-1</sup> suggesting increased oil dissolution rates under the more turbulent flow conditions.



Figure 2. Details of the first 6 hours of the experiments presented in Figure 3 but showing Type 1 and II erosion for runs 1 and 2.



Figure 3. Hibernia crude oil erosion under increasing current speed and shear stress (A). Run 1 and 2 represent clear seawater conditions (B).

## 3.2 Oil erosion under differing SSC and current speeds

Oil erosion was analyzed under SSC levels of 100, 200, 250, and of 600 mg L<sup>-1</sup>, and increasing current velocities (0.07, 0.14, 0.21, 0.27, 0.38, 0.41, 0.48, 0.55 m s<sup>-1</sup>). Each increment was held for thirty minutes (Fig. 4A), whereas the last was held for 1 hour. The current was then stopped for one hour between the last three velocity increments. The clear-water trend (run 1 in Fig. 3B) is used as a control. Sediment was introduced into the flume at the beginning of the first velocity increment.

The results showed similar trends for all levels of SSC. That is, above the erosion threshold, oil concentrations increase with increasing current velocities. Oil concentrations

increased sharply to values close to 210 and 510 mg L<sup>-1</sup> (Fig. 4B) for velocities of 0.65 and 0.75 m s<sup>-1</sup> respectively. Note that the 250 mg L<sup>-1</sup> curve is not complete (0 to 4 hours), but the trend is similar to that of the 200 mg  $\rm L^{-1}$ curve. For  $U_{v}$  = 0.65 and 0.75 m s<sup>-1</sup>, the oil concentrations were significantly lower for the 600 mg L<sup>-1</sup> run compared to the lower concentration runs. These results show that lower SSC were more efficient in eroding oil at a constant speed of 0.65 m s<sup>-1</sup>. For  $U_v \le$  0.55 m s<sup>-1</sup>, oil concentrations were more elevated at SSC's of 200 and 250 mg L<sup>-1</sup>, than for SSC of 0, 100, and 600 mg L<sup>-1</sup>.

During the settling period, oil concentrations decreased to values close to control values for all levels of SSC indicating that oil floated rapidly to the surface after the current was stopped. The oil concentration, however, remained higher for the control conditions (0 mg L<sup>-1</sup>) suggesting additional oil dissolution in the water column as a result of turbulent flow.



Figure 4. Erosion of HCO under increasing current speed and shear stress (A) and for different SSC (B).

3.3 Analysis of the OMA formed under different conditions of turbidity and current speed.

The size and type of the OMA formed under the more turbulent conditions (0.65 and 0.75 m s<sup>-1</sup>) of the previous set of experiments (Fig. 4B) were analyzed. Water samples collected from the middle port under each applied current speed were analyzed under light microscopic. The analysis revealed that most of the OMA formed were solid aggregates, that is, nonspherical elongated aggregate that exhibit particles within the oil boundaries.

The size of the OMA formed under turbulent conditions are presented in Figures 5A & B. The results show that the size of the solid OMA vary from 50 to 2100 µm for SSC = 250 mg  $L^{-1}$  (Fig. 5A), and from 10 to 700 for SSC = 600 mg  $L^{-1}$ (Fig. 5B). The bigger aggregates are observed immediately after the current is stopped. The results also show that under a SSC of 250 mg L<sup>-1</sup>, the size of the OMA tends to be more important under a current speed of 0.65 m s<sup>-1</sup> that for a higher current speed suggesting that more turbulent flow would limit the aggregate size. This effect is less evident under a SSC of 600 mg L<sup>-1</sup>.



Figure 5. The size of the solid OMA formed under current speeds of 0.65 and 0.75 m s<sup>-1</sup> and under SSC of 250 mg L<sup>-</sup> (A) and 600 mg  $L^{-1}$  (B).

00 00

5 6 7 8 9

OMA-VCR-0 65 m s<sup>-1</sup>

OMA-VCR-0 75 m s

Time after current stopped (min.sec)

#### 4. CONCLUSIONS

0i1

0

0

2

•

0

Two distinct types of oil erosion have been identified and are described as following : 1) Type I erosion caused by oil dissolution under an applied current without any visual disturbance of the oil slick surface, and 2) Type II erosion defined as visual erosion of the oil slick and entrainment of oil droplets. Type I erosion is important to consider as it may be linked to the onset of toxic responses while Type II erosion is important in oil/sediment aggregation processes that facilitate natural dispersion of stranded oil. A critical

stress of 5.0 Pa was necessary to initiate Type II oil erosion in clear seawater which was not influenced by turbidity. At velocities > 0.65 m s<sup>-1</sup>, a SSC of 200 mg L<sup>-1</sup>, was observed to produce the highest erosion rates. The bigger OMA sizes were also formed in that range of SSC. Therefore, this concentration is proposed as optimal for Hibernia crude oil erosion in warm seawater. A higher concentration of suspended sediment leads to significant suppression of turbulence that tends to diminish the rate of oil erosion.

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