

Modeling the Long Term Fate of Oil-Mineral-Aggregates (OMAs) in the Marine Environment and Assessment of Their Potential Risks

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ABSTRACT

Spilled oil can interact with suspended particles (mineral or organic) in marine environments and form aggregates, including oil-mineral-aggregates (OMAs). Some OMAs with densities greater than seawater can settle to the seabed and pose potential risks to benthic organisms. To understand the transport and fate of oil associated with OMAs and evaluate their potential risks, researchers recently developed an integrated hydrodynamic, fate/transport and risk assessment modeling system, and conducted several case studies. One of the limitations of these studies was the use of a conservative approach which neglected biodegradation processes. Although this approach is acceptable for the study of short term effects of settled oil, the long term effects cannot be studied. It is the objective of this paper to improve the existing modeling approach in order to simulate the long term fate/transport and potential risk of settled oil associated with OMAs. The improved approach used the DREAM model developed by SINTEF and a number of case studies under different combinations of oil, sediment type, wave and current conditions were conducted.

INTRODUCTION

The action of waves in a marine oil spill can break up the surface slick into micron-sized oil droplets. If the water column has a high load of suspended particles, they can interact with oil droplets to form oil-mineral-aggregates (OMAs). This natural process stabilizes dispersed oil droplets in the water column and enhances their biodegradation rate (Bragg and Owens, 1995; Le Floch et al., 2002; Owens and Lee, 2003; Owens et al., 2003; Page et al., 2000). Thus, impacts on sea birds and the probability of oil reaching shoreline environments are diminished.

The suspended mineral particles that constitute OMAs may have densities heavier than both crude oil and seawater. For example, Khelifa et al. (2008) has reported a lowest effective density of 25 kg/m³ for OMAs formed under laboratory conditions with several types of crude oil and natural sediments at different concentration levels. Therefore, there is a high probability that OMAs will transport residual oil to the seabed to adversely affect benthic organisms.

To evaluate the potential impacts of OMAs, mathematical models can be used to simulate the transport process of OMAs. Niu et al. (2009) has developed a particle tracking model and

conducted a case study for a hypothetical spill in the Gulf of St. Lawrence. The method has also been used by Niu et al. (2010a) in a probabilistic analysis to study the effects of uncertainty associated with OMA properties on predicted risk. Although the effect of waves has been studied, the model did not consider the important re-suspension process which may influence model accuracy. To overcome this limit, Niu et al. (2010b) integrated a fully three-dimensional hydrodynamic model with a fate and transport model and conducted a case study using hydrodynamic conditions of the Bristol Channel, U.K., which successfully demonstrated the capability of the model to simulate transport and to predict risk; however, the model was limited to specific hydrodynamic conditions. Hence, Niu et al. (2010c) conducted a series of simulations using a wide range of wave and current conditions.

It was found from these studies that although the transport process of OMAs and its risks to benthic organisms depend on several factors such as spill amount, sediment properties, water depth, and hydrodynamics, the risks from the various scenarios studied are generally low and not persistent. Nevertheless, this is not true for very weak currents. If the ambient current is weak, the OMAs tend to settle and concentrate near the spill site rather than disperse within a large area. In this case, the modeling approach used in previous studies becomes inadequate since the important process of biodegradation is not included. Our objective was to improve the existing modeling approach, conduct a series of simulations of the long term transport of OMAs, and to assess their potential risks.

METHODOLOGY

The new approach in this study takes into consideration the following fates for OMAs in the water column: 1) advection, 2) diffusion, 3) settling, 4) biodegradation of oil. The fates in sediment are 5) deposition of OMAs, 6) burial, 7) oxygen depletion, 8) biodegradation, and 9) re-suspension.

Governing equations for hydrodynamics (Sørensen et al, 2004, 2006) and fate process 1-3 and 5 (Reed and Hetland, 2002; Rye et al., 2006a, 2006b) have been described before in Niu et al. (2010b, 2010c). Process 4, the reduction of concentration due to biodegradation of the oil in the water column, is calculated by (Rye et al., 2006a) as

$$PEC_{water} = C_{discharge} \exp(-K_w t) / \text{DILUTION} \quad (1)$$

where PEC_{water} is the local concentration in water column, $C_{discharge}$ is the initial oil concentration, K_w is the first order biodegradation factor, t is time, and DILUTION is the dilution factor calculated from advection/diffusion processes.

The biodegradation process of oil associated with OMAs in sediment is modeled using diagenetic equations 2 and 3 (Rye et al., 2006b):

$$\frac{\partial C_1}{\partial t} = \frac{\partial}{\partial z} \left(D_B \frac{\partial C_1}{\partial z} - w C_1 \right) - [K_s C_1] \frac{O_2}{K_{O_2} + O_2} \quad (2)$$

where C_1 is the concentration of oil in sediment, D_B is a bioturbation coefficient, K_s is the biodegradation rate in sediment, K_{O_2} is the “Monod-type” saturation constant and O_2 is the free oxygen pore-water concentration, and

$$\frac{\partial O_2}{\partial t} = \frac{D_0}{\theta^2} \frac{\partial^2 O_2}{\partial z^2} - \frac{1-\phi}{\phi} [Y K_s C_1] \frac{O_2}{K_{O_2} + O_2} \quad (3)$$

where D_0 is the diffusion coefficient for oxygen in pore water, θ is tortuosity of sediment, ϕ is the porosity of the sediment, and γ is the Redfield number. The potential risk of settled oil on benthic organisms in terms of Hazard Quotient (HQ) was evaluated using the same method as Niu et al. (2009; 2010a, 2010b). HQ is the ratio of a contaminant's environmental concentration to a screening value selected for risk assessment of that substance (US EPA, 2009):

$$HQ = PEC / BC \quad (4)$$

where the PEC is the predicted environmental concentration in sediment of eight hydrocarbon groups (8-HCG), and the BC is the benchmark concentration (Table 1). If HQ is greater than 1.0, harmful effects can be expected; if HQ is less than 1.0, harmful effects are unlikely to occur. It should be noted that although oxygen depletion is considered in the computation of PEC , its effects is not included in the HQ calculation.

Table 1. Sediment benchmark for recommended petroleum fractions (Battelle, 2007) representing 8 hydrocarbon groups (8-HCG)

Hydrocarbon Fraction		K_{oc}	Final Chronic Value (FCV) ($\mu\text{g/L}$)	Sediment Benchmark (mg/kg oc)
Aliphatic	C ₅ -C ₈	7.24×10^3	218	1591
	C ₉ -C ₁₂	4.37×10^5	6.3	2722
	C ₁₃ -C ₁₈	1.10×10^8	0.05*	5543
	C ₁₉ -C ₃₆	8.32×10^{10}	0.0001*	9883
Aromatic	C ₆ -C ₈	4.47×10^2	1191	531
	C ₉ -C ₁₂	4.90×10^3	46.2	228
	C ₁₃ -C ₁₅	2.40×10^4	5.2	125
	C ₁₆ -C ₂₄	3.39×10^5	0.12*	40

* The fraction is not likely toxic because mean LC50 exceeds mean aqueous solubility;

CASE STUDIES

Niu et al (2010b, 2010c) conducted a series of simulations (15 cases) to study the transport of a hypothetical spill of 1000 tonnes South Louisiana crude oil in the Bristol Channel, U.K., under various hydrodynamic conditions. Two cases involved currents (Figure 1) at the spill site ranging from 0.1 to 1.30 m/s with a mean value of 0.65 m/s. As the simulated current for this site is relatively strong, a factor of 0.25, 0.5, and 0.75 was applied in the hypothetical studies (12 cases in Figure 2) to reduce the current magnitude and to represent more general environmental conditions. Therefore, the average current speeds (U) over the simulation period become 0.16, 0.33, and 0.49 m/s. The wave height for the studies ranged from 0 to 5 m. It can be seen from these studies that risk is present only for the case with very weak current (wave only) at the end of 9 days. Therefore, the long term simulation is important only under conditions of very weak current, and this situation was chosen for the long term fate and transport study.

The detailed conditions such as simulation domain, release amount, and hydrodynamics, of the weak current case have been described previously (Niu et al., 2010b). Unlike previous studies, the inclusion of the biodegradation process (equations 2-3) in the improved model required specification of the biodegradation rate. The rates of biodegradation vary greatly among the

various components of crude oils and petroleum products and depend on many environmental factors such as temperature, nutrient concentration, and oxygen content. Venosa et al. (1996) utilized hopane normalization in studying the kinetics of oil biodegradation and developed first-order biodegradation rate constants for resolvable alkanes and important two- and three-ring polycyclic aromatic hydrocarbon (PAH) groups. The method was also used by other researchers and the results of these kinetic studies done in the field and laboratory are summarized in Table 2. Khelifa et al. (2008) reported the properties of OMAs formed by South Louisiana oil with four types of sediment: Cook Inlet (CI), Columbia River Delta (CRD), Mississippi River Delta (MRD) and Standard Reference Material (SRM) under concentrations ranging from 25 to 300 mg/L. For a mid-range sediment concentration of 100 mg/L, the amount of oil that can be bonded by the sediments is 10%, 3%, 16% and 10% for CI, CRD, MRD and SRM, respectively. To study the effects of biodegradation on OMA transport, this study considered factors such as nutrient load and temperature. The effects of chemical dispersant on the size distribution of oil droplets and OMAs, and the amount of oil associated with OMAs, were also investigated.

Table 2. Summary of first order biodegradation rate constants from field studies (modified from Zhu et al., 2004).

Location	Shoreline Type	Oil Type	Treatment	First order biodegradation rate (day ⁻¹)		Reference
				Alkanes (nC ₁₀ -nC ₃₅)	PAHs (2-3 ring)	
1. Delaware	Sandy beach	Bonny light crude	Control Nutrient	0.026 0.056	0.021 0.031	Venosa et al. (1996)
2. Texas	Brackish wetland	Phase II: Arabian light crud	Control Nutrient	0.019 0.042-0.061	0.017 0.018-0.027	Simon et al. (1999)
		Phase III: Arabian medium crude	Control Nutrient	0.020 0.024	0.015 0.013	
3. Louisiana	Salt Marsh	South Louisiana crude	Control Nutrient	0.005 0.005	N/A	Shin et al. (1999)
4. Nova Scotia	Salt Marsh	Mesa light crude	Control Nutrient	0.020 0.026-0.039	0.010 0.011-0.013	Unpublished data owned by Fisheries and Oceans Canada
5. Laboratory	N/A	Prudhoe Bay crude (20 °C)	Control C9500 JD2000	0.17 0.23-0.66 0.21-0.35	0.13 0.19-0.218 0.15-0.233	Venosa and Holder (2007)
		Prudhoe Bay crude (20 °C)	Control C9500 JD2000	0.073 0.093-0.138 0.073-0.156	0.052 0.057-0.067 0.062-0.116	

To examine the effects of nutrients, the properties of OMAs formed by CI sediment and South Louisiana crude oil (Figure 3) was used given that the settleable amount of oil (10%) by CI sediment is about mid-range among the four sediments (Khelifa et al., 2008). Four scenarios using CI were studied: 1) no degradation, 2) low nutrients (control), and 3) high nutrients, to study the effect on the first order biodegradation rate coefficient. While using the same rate constant (control), an additional CI scenario (number 4) was simulated to study the effects of the sediment model (excluding equations 2 and 3). Since it is known that dispersant has the most

significant effect on OMAs formed with MRD sediment (Khelifa et al., 2008), we used the properties of MRD and South Louisiana crude oil OMAs (Figure 3) to examine temperature and dispersant effects. The five simulations performed for MRD were numbered thus: 5) no degradation, 6) no chemical dispersant at 5°C, 7) no chemical dispersant at 20°C, 8) with chemical dispersant at 5°C, and 9) with chemical dispersant at 20°C, to study the effects of temperature and chemical dispersant. Detailed conditions for the cases are listed in Table 2.

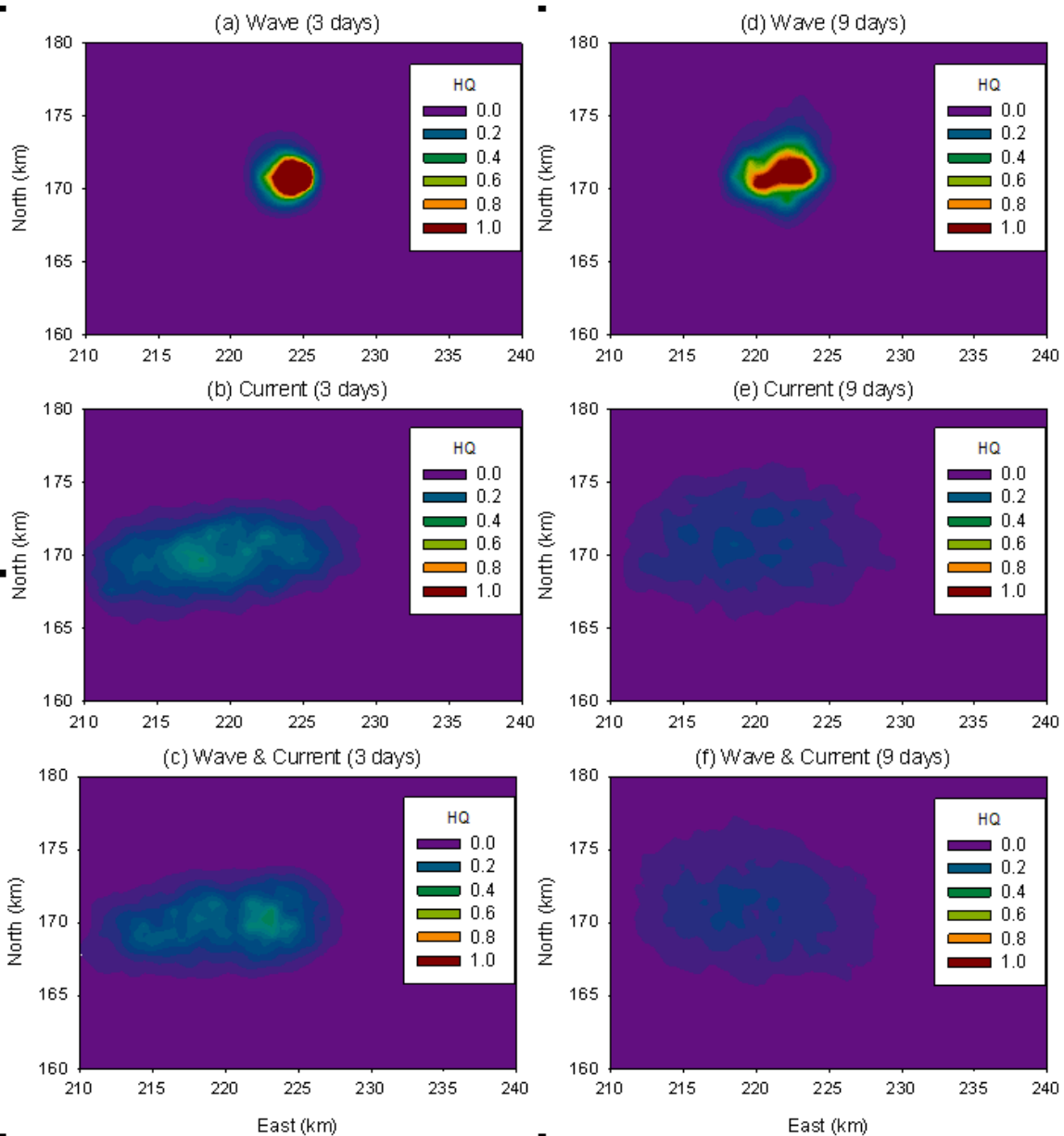


Figure 1 Risk in terms of *HQ* to benthic organisms at 3 days and 9 days based on simulated hydrodynamic conditions for Cook Inlet.

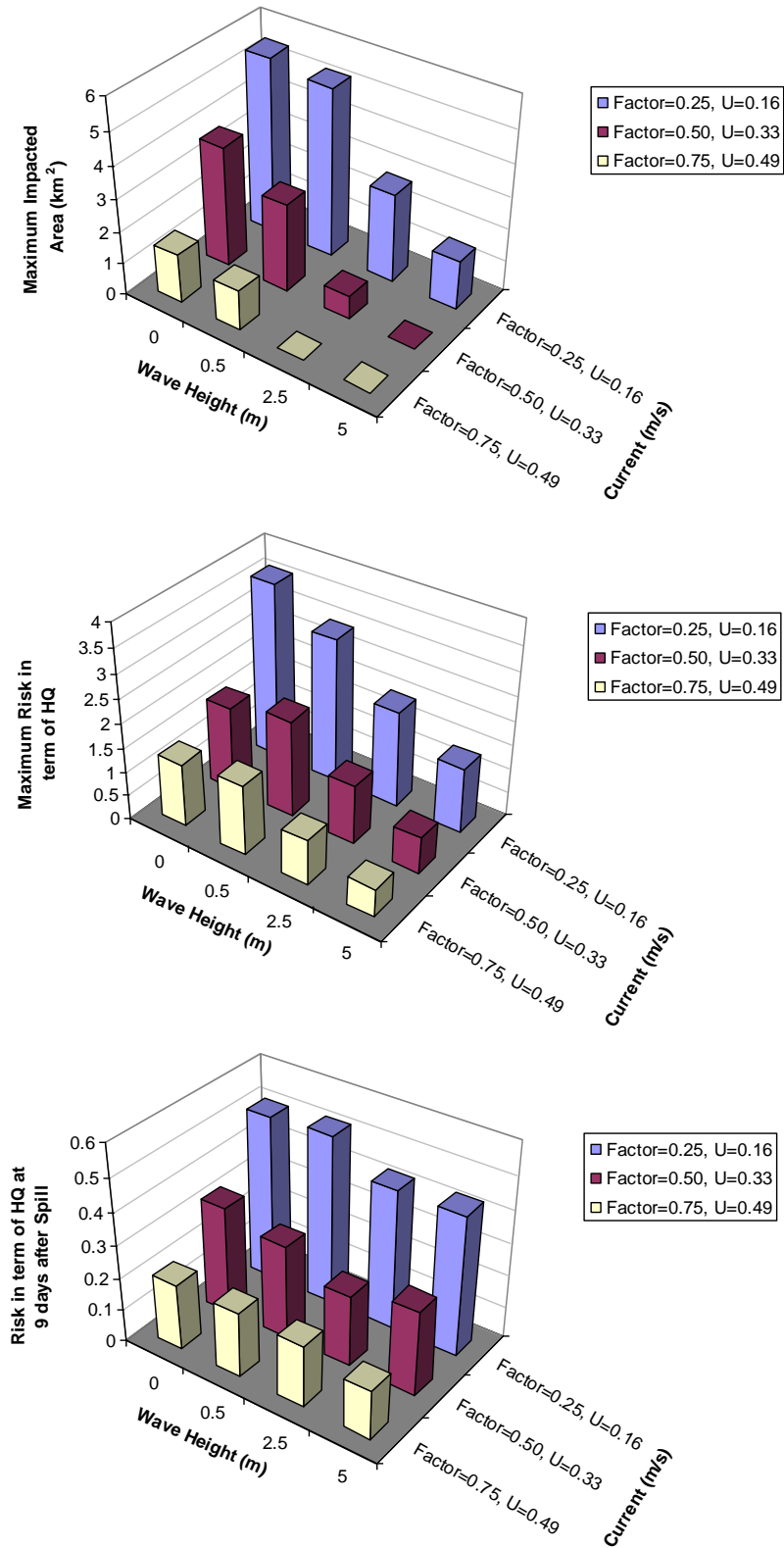


Figure 2 Maximum Risks and Impacted Area over the Simulation Period and Risks at 9 Days based on hypothetical hydrodynamic conditions.

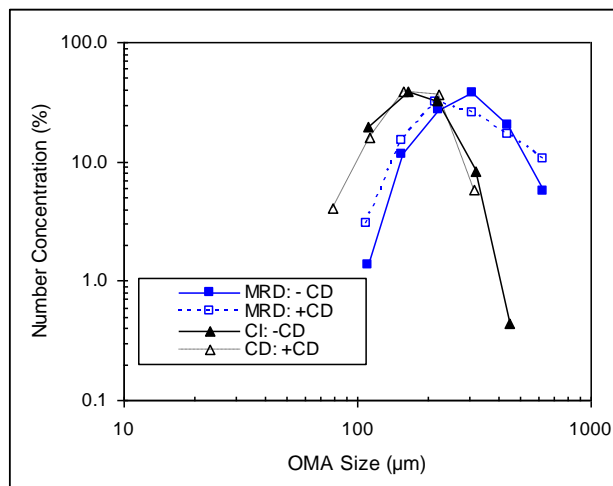


Figure 3 Particle size distribution for OMAs formed with CI and MD. Data source: Khelifa et al. (2008).

Table 3 Summary of case study parameters.

No.	OMA	K (day^{-1})		O_2 (mg/L)/ Depletion	Temperature	Chemical Dispersant
		Alkanes	PAHs			
1	CI	0.0000	0.0000	9/+	N/A	N/A
2	CI	0.0423	0.0356	9/+	N/A	N/A
3	CI	0.0757	0.0423	9/+	N/A	N/A
4	CI	0.0423	0.0356	9/-	N/A	N/A
5	MRD	0.0000	0.0000	9/+	N/A	-
6	MRD	0.073	0.052	9/+	5	-
7	MRD	0.17	0.13	9/+	20	-
8	MRD	0.115	0.059	9/+	5	+
9	MRD	0.363	0.198	9/+	20	+

+ included, - excluded

RESULTS AND DISCUSSIONS

The mass balances of oil, represented by 8 hydrocarbon groups (8-HCG), associated with OMA (CI) are plotted in Figure 4. For case 1, the OMAs rapidly settled from the water column to the sediment and the amount settled was about 98% at 10 days. Due to the exclusion of the degradation mechanism, the amount of the 8-HCG associated with OMAs in sediment changed little (97%) until the end of simulation for this case. With the inclusion of biodegradation (case 2), the amount of 8-HCG associated with OMAs in sediment was reduced to 92% and 5.9% was biodegraded at 10 days. After the first 10 days, the rate of loss due biodegradation becomes quite slow. The amount remaining in sediment at the end of the simulation was 88.8%. With higher nutrient concentration and increased biodegradation rate (case 3), the trend was similar to that of case 2 but with increased decay, such that at 10 days, the amount in sediment was 88% and the amount decayed was 9.7%. The amount remaining in sediment at the end of the simulation was 84.5%. The increase in decayed 8-HCG with higher nutrient concentrations is plotted in Figure 5 (left). It can be seen that the rate is much higher at the beginning (or in water column) than during the later stage (in sediment). This implies that the sediment model (equations 2 and 3) realizes a decrease in degradation as a result of oxygen depletion. This was confirmed by a

comparison of cases 2 and 4 (in which the sediment model was not used) in Figure 5 (right). Notice that the amount in the water column is almost the same which indicated that it was unaffected by the inclusion of the sediment model. However, if the sediment model was excluded, the degradation in sediment became much higher. The amount remaining in sediment for case 4 was 28% while the amount for case 2 was 88.8%.

The results using MRD sediment are depicted in Figure 6. A comparison of Figure 6 with Figure 4 shows that the deposition rates are much higher for MRD than CI. The time for 90% of the 8-HCG associated with OMA to be transported to the sediment was from 0.5 to 1.25 days for MRD, while the time required for CI was from 4 to 6 days. Rates for the cases with chemical dispersant were also higher than without dispersant. This was due to higher oil fractions being associated with the larger MRD sediment particles (based on laboratory measurements by Khelifa et al. 2008), but it may not be the same for OMAs generated from other oil or sediment types. Despite the deposition rate, the general trends of case 6 to 9 (MRD) are similar to that of case 2 and 3 (CI). Without chemical dispersant, the percentages of 8-HCG associated with OMAs in sediment at the end of simulation were 94% and 89% at 5 and 20°C respectively. The amounts decayed were 5.1% and 10% at 5 and 20°C respectively. The higher degradation rate at 20°C resulted a 5% decreasing of the total oil associated with OMAs in sediment. The amounts of 8-HCG decayed for the two cases with chemical dispersant at the end of simulation were 4.4% and 10.6% at 5 and 20°C respectively. Although the percentages in the presence of chemical dispersant were not significantly different from percentages without dispersant, the total amount of oil decayed or in sediment were different because oil associated with OMAs was 16% of the total spilled oil without chemical dispersant as opposed to 23% with it (Figure 7).

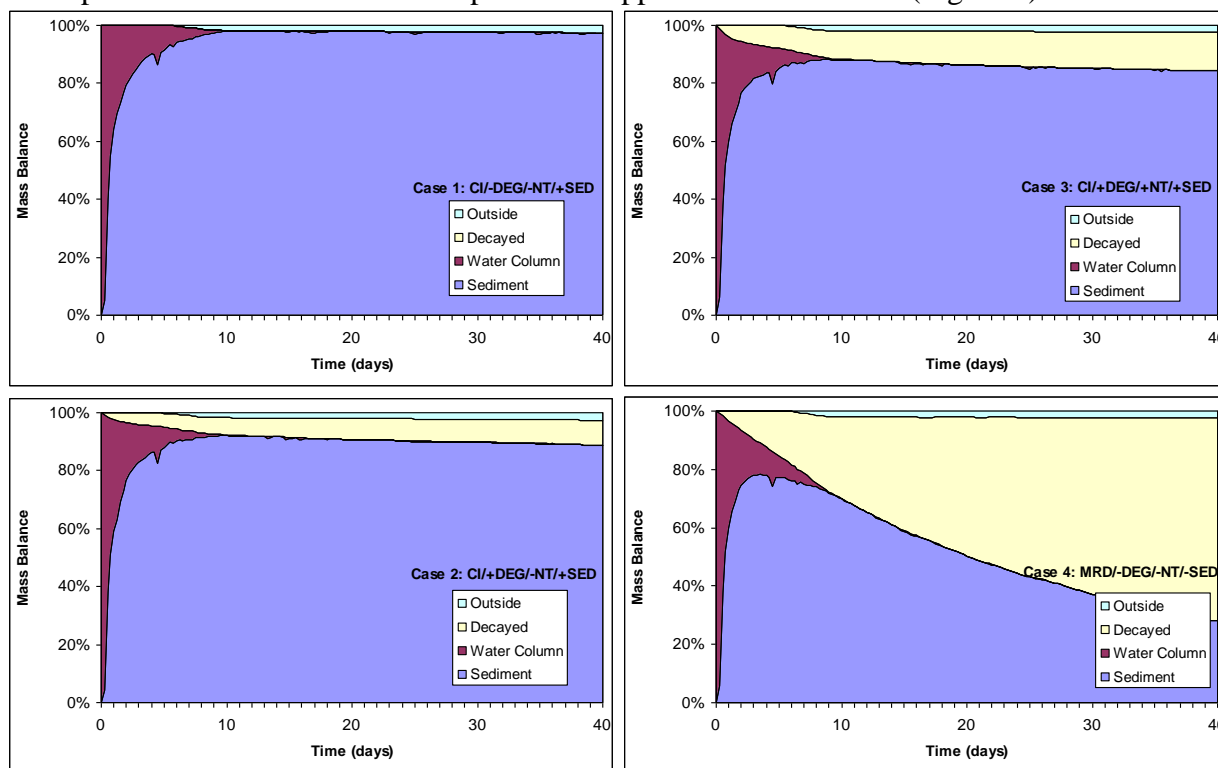


Figure 4 Mass balance for oil associated with OMA (CI). +DEG/-DEG is with/without degradation; +NT/-NT is high or low nutrient regime; +SED/-SED is with/without the use of sediment model.

The temperature effect for the case with chemical dispersant was more dramatic than the case without dispersant. The total amount of oil decayed due to increased temperature was 1.4% for cases with chemical dispersant while the amount was only 0.74% for those cases without dispersant. The effect of including the degradation process (cases 6 and 7 vs. case 5) can also be seen in Figure 7.

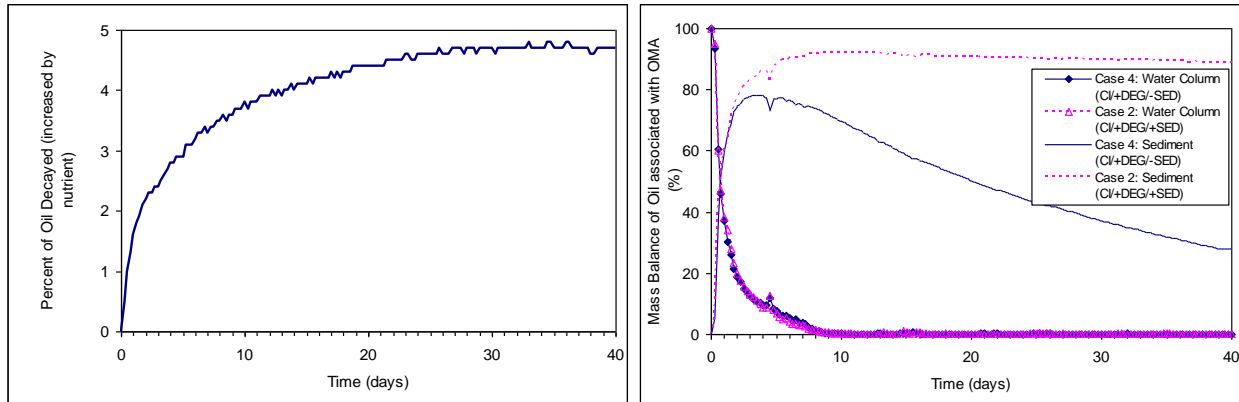


Figure 5 Effects on degradation of nutrients (case 2 vs case 3) (left) and inclusion of the sediment model (case 2 vs case 4) (right) in case 2.

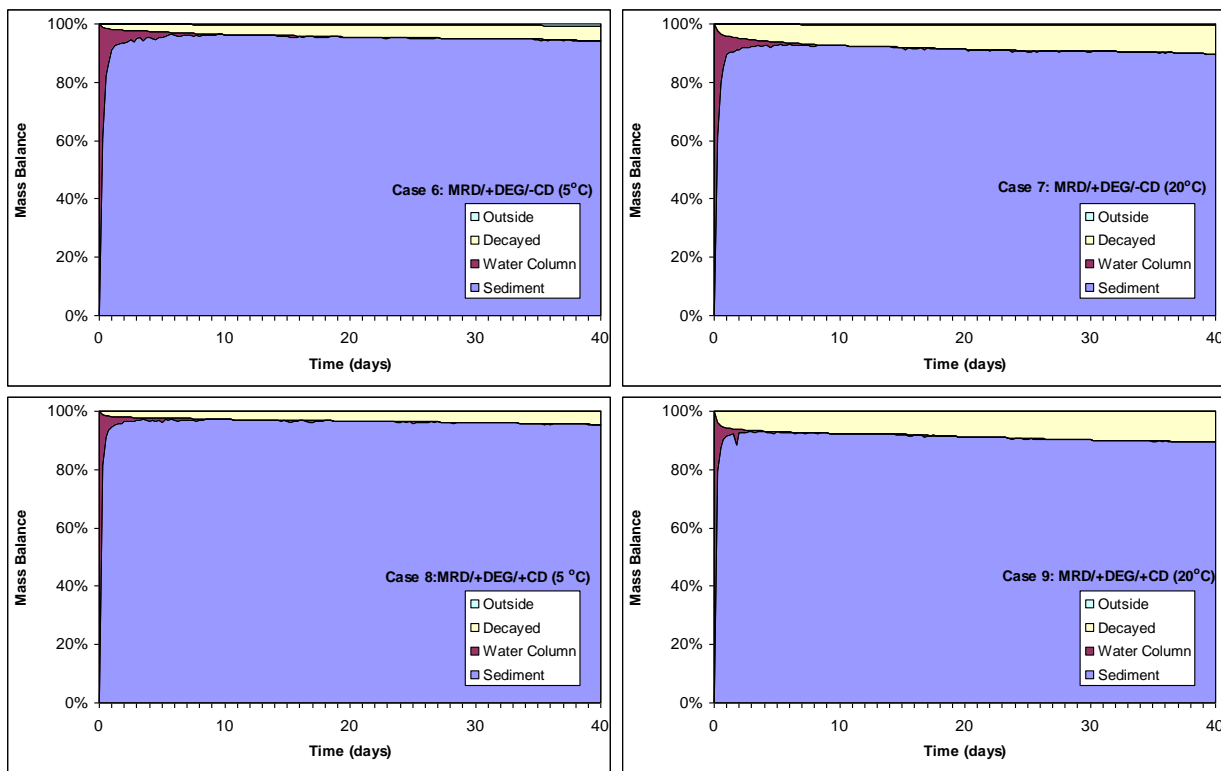


Figure 6 Mass balance for oil associated with OMA (MRD). +DEG/-DEG is with/without degradation; +CD/-CD is with or without chemical dispersant.

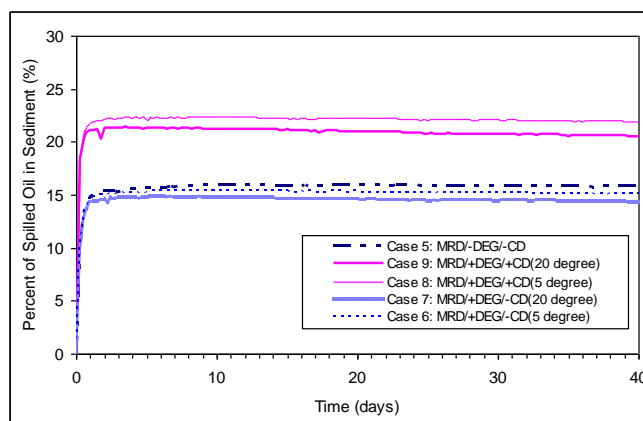


Figure 7 Percentage of spilled oil in sediment.

The risks in terms of impacted area for the cases 1 to 9 are plotted in Figure 8. Although biodegradation did reduce the oil in sediment, it is a relatively slow process (Figure 8). The figure also demonstrates the importance of including a sediment model in the study of degradation of oil in sediment. Failing to include the oxygen depletion process may cause underestimation of the concentration of oil in sediment. It should be noted that volatile components were not included here, although they were considered in previous studies (Niu et al., 2010b, 2010c).

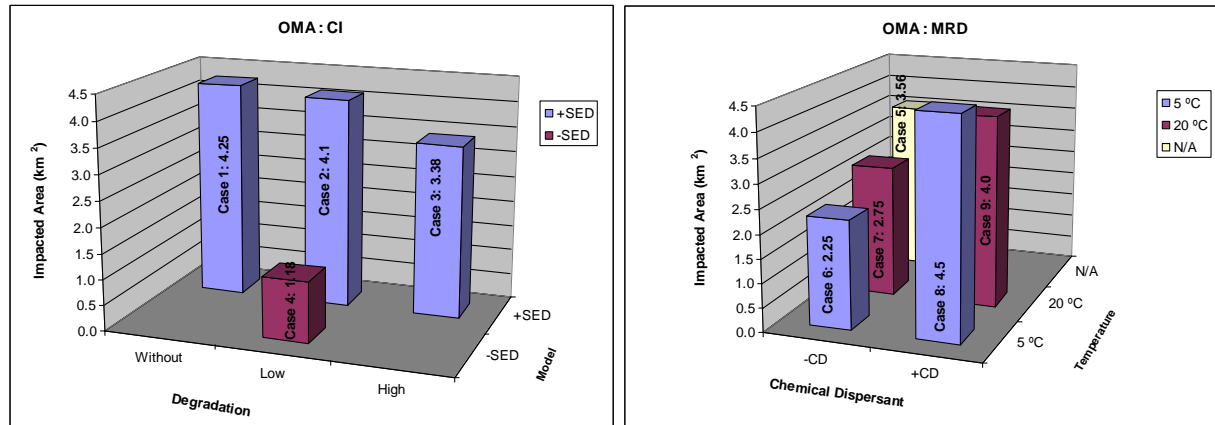


Figure 8 Impacted area at the end of simulation for the 9 cases. The temperature effect was not considered in Case 1 to Case 4.

CONCLUSIONS

Previous studies have shown that the formation of OMAs generally pose a relatively low risk to sediment organisms. However, their risks may increase in an environment with very weak currents. The methods used in previous studies are inadequate to simulate the fate of OMAs under such conditions due to the exclusion of the biodegradation process. An improvement of OMA modeling has been made in this study by the adoption of the diagenetic equations developed by Rye et al. (2006b). A total of 9 cases were studied using the improved approach to

simulate the transport of oil associated with OMAs in the water column and sediment. The result shows that the inclusion of biodegradation caused a reduction in the amount of 8-HCG in sediment (calculated by $(\text{Mass}_{+\text{DEG}} - \text{Mass}_{-\text{DEG}})/\text{Mass}_{-\text{DEG}}$) in the range of 8.9% (Case 2 versus Case 1) to 13.2% (Case 3 versus Case 1) for Cook Inlet depending on the nutrient load, which in turn affected the biodegradation rate. The comparison of cases with and without the sediment model (oxygen depletion) indicated the importance of using diagenetic equations to avoid underestimation of the concentration of oil in sediment. The effects of temperature and chemical dispersant were studied using MRD sediment and results showed that increased temperature helped to reduce the amount of oil in sediment as a result of increased biodegradation rate. Although the relative percentages of 8-HCG in sediment and decayed 8-HCG were not significantly different for cases with and without chemical dispersant, the absolute amount (percentage of total spilled oil) in sediment and decayed are higher for those cases with chemical dispersant.

It should be pointed out again that the transport process of OMAs and their potential risks depend on many factors, such as sediment properties, oil type, water depth, and hydrodynamic conditions. A higher potential level of combined oil and sediment (16%-23% for MRD compared with 10%-11% for CI based on Khelifa et al. (2008) experimental data) does not mean it poses higher risks (impacted area of 2.25 to 4.5 km² for MRD and 3.38 to 4.25 km² for CI). The method described in this paper provided a useful tool to evaluate OMA fate and transport on an individual basis.

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