Oil contamination in mangrove ecosystems: impacts and rehabilitations

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Abstract. Mangrove ecosystem has multiple important ecological roles, like connecting nutrient flow from terrestrial ecosystem to marine ecosystem, nurturing place for several marine animals, home to many terrestrial animals, and prevention of erosion and controlling sedimentation process. Moreover, mangrove has many economic and social benefits for the community from the surrounding the areas. There are several factors which has potential impact both quantitatively and qualitatively upon the mangrove ecosystem. One of the influential factors is oil contamination. In this paper, it will be discussed further about oil contamination on mangrove ecosystem and efforts for overcoming its impacts.

Key Words: bioremediation, degradation, phytoremediation, remediation, surfactant.

Introduction. Mangrove is a combination from two words “grove” (English) and “mangue” (Portuguese). According to English, “mangrove” is defined as a plant community which is situated in the intertidal coastal area. This word is also understood as the individual plant of that foretold term. In short, mangrove forest can be defined as a forest type which grows on the intertidal coastal area (protected beach, lagoon, and delta). Meanwhile, mangrove ecosystem is the interconnection between organisms, abiotic, and its environmental surroundings inside the mangrove forest area. Hence, the scope of mangrove forest ecosystem can be detailed as: (1) plant species which only live inside mangrove forest area, (2) other associated plant species which can live in the mangrove forest area, (3) associated biota with mangrove ecosystem, (4) natural process which occurs throughout the mangrove forest ecosystem, and (5) open field or mud field limiting mangrove ecosystem with terrestrial and or marine ecosystem (Kusmana et al 2005).

The mangrove forest is highly influenced under intertidal process of nearby marine water body. It also in some places grows in the brackish water, or surrounding the delta of a river protected from sea waves. Therefore, this ecosystem should have to deal with many stressors like: salinity, sedimentations, and root aeration (Saenger 2002). Mangrove forest ecosystem is very unique for having a high level of soil salinity and frequently inundated. This condition leads to decrease the number of plant species which are able to adapt in this area. Several families are found in different places; it can be varied depended on the location and inundation frequency. In general, on the highly inundated area *Bruguiera* and *Xylocarpus* sp. are the dominant genus to be found, while *Bruguiera* sp. and *Lumnitzera* sp. are usually found on the rarely inundated area. In other case, where the inundation is highly influenced by the fresh water *Nypa fruticans*, *Oncosperma*, and *Cerbera* dominates (Kusmana et al 2005).

Mangrove forest ecosystem has a lot of economic benefits and ecological services. From the woods, woodchips, charcoals, and building materials (for home and for boat) can be utilized directly from wood harvesting. Other non-wood materials from mangrove forest are honey, fruit, antibiotics, medicines, food and feed materials, and salt. As the place for fish and shrimp spawning, mangrove ecosystem can enhance economic activity through fishery. Besides all of the benefits for human beings, mangrove ecosystem can protect the coastal area from erosion, gale, sea water intrusion, and also tsunami. It also rules the sedimentation process and it is a buffer ecosystem between terrestrial and marine environment (sea-grass, coral reefs, rivers, delta, estuary, etc.). Mangrove forest ecosystem is also able to stabilize the temperature disparity between night and daylight, thus micro-climate equilibrium becomes stable. As a scene view, mangrove ecosystem gives other aspect of human life, like recreation, spiritual nuance, and other social functions related to the culture of human community surrounding it (Saenger 2002; Kathiseran 2012).

Research on carbon sequestration shows mangrove ecosystem has a big role on carbon cycle (Lee et al 2014). Even though world mangrove forest area percentage is only around 0.5%, it can contribute to absorb carbon as much as 24 Tg annually. Carbon is sedimented or transferred in form of detritus or nutrient to the sea (Alongi 2014). Hence, ecological services of mangrove forest are very important to the global climate stability through CO_2 concentration stabilization in the atmosphere. Therefore, conservation efforts are conducted in many countries which have mangrove forest ecosystem.

Mangrove forest ecosystem is commonly found in the tropic area, between around 40° N and 40° S (Chapman 1975). However only a small portion of those areas has the greatest mangrove outspread, between 5° N and 5° S (Giri et al 2010). Mangrove distribution in general can be divided into two distinct areas: old world mangrove and new world mangrove. The old world mangrove consists of east African coastal area, India, South-East Asia, southeastern China, southern Japan, and many small islands on Pacific Oceans. Meanwhile, the new world mangrove covers much of east Africa coastal area, eastern of South America, Caribbean archipelago, and Gulf of Mexico, and also several places on Pacific Oceans like Galapagos (Chapman 1975; Kathiseran 2012).

According to Hamilton & Casey (2016) studies in 2014 there were 20 countries which possess mangrove forest ecosystem with minimum area of 900 km^2 (Table 1). Indonesia has the most mangrove area compared to the others. Almost a quarter of world mangrove forest area is in Indonesia. This percentage is decreased as much as 3 percent (around 1.100 km^2) from total world mangrove area on 2000.

Mangrove forest ecosystem has several inquiries for maintaining its existence. The structure, function, composition, biota distribution, and individual plants are depended on the external factors. Mangrove ecosystem is usually found on the plane coastal area, with low erosion and small wave. Mangrove forest also needs high sunlight intensity (3000-3800 kcal m^{-2} day^{-1}), average precipitation 1500-3000 mm per year, optimum temperature around 20-30°C. Other influencing factors are: sea level inundation frequency, wave, sea current, salinity, dissolved oxygen, soil sediment type, soil nutrients (N, P, K, Mg, Na). For soil nutrition, nitrogen and phosphorus always become the limiting factors (Kusmana et al 2005; Lovelock et al 2006). Several factors interact each other synergistically or not. Nitrogen compound assimilation inversely related to the salinity level. By increasing salinity level, the root and shoots biomass decrease, while hypocotyl biomass increases. This circumstance goes as the decrement of nitrogen absorbed by the plant (Shiau et al 2017).

Mangrove ecosystem area is very dynamic, influenced naturally and or from human. Each year, mangrove forest area coverage is decreased 1-2%. This number is as much as the rate of corral mortality and tropical rain forest deforestation (Duke et al 2007). Sedimentation from the river can enlarge the mangrove forest ecosystem area. This will affect the pattern of tide wave which acts as the control, positively shrinking the mangrove forest area. As the natural factors, the anthropogenic factors can be as a constructive (replantation of mangrove forest) or destructive (land conversion, and contamination by pollutions). Pendleton et al (2012) studies showed around 0.15-1.02

milliard ton CO_2 annually released due to the damage of coastal area (swamp, mangrove, and sea-grass). This rate is around 3-19% from total global deforestation.

Table 1

World mangrove forest area on 2014 (Hamilton & Casey 2016)

No.	*Country*	*Estimated mangrove ecosystem area (km^2)*	*Forest coverage in mangrove biome (km^2)*
1	Indonesia	23,324.29	44,038.77
2	Brazil	7,674.94	17,685.60
3	Malaysia	4,725.84	8,231.09
4	Papua New Guinea	4,172.29	5,677.16
5	Australia	3,316.21	3,251.24
6	Mexico	2,991.83	6,203.92
7	Myanmar	2,557.45	3,856.09
8	Nigeria	2,653.99	6,919.28
9	Venezuela	2,403.83	7,539.12
10	Philipina	2,064.24	2,089.24
11	Thailand	1,886.33	4,196.97
12	Bangladesh	1,772.98	2,314.56
13	Colombia	1,671.86	6,274.70
14	Cuba	1,633.46	2,454.10
15	United States of America	1,568.60	1,585.06
16	Panama	1,323.94	2,708.21
17	Mozambique	1,223.67	2,677.27
18	Cameroon	1,112.76	1,332.16
19	Gabon	1,082.11	3,882.95
20	Ecuador	935.74	1,945.27

Pollutant contamination affects ecosystem quality. One of the most destructive contaminant to mangrove forest ecosystem is contamination with heavy metals and oil. Heavy metals which are found generally in the sediments, potentially poisoning the biota. To the mangrove individual plant, heavy metal poisoning leads to several physiological diseases. Copper (Cu) poisoning can lead to potassium homeostatic trouble inside the cells. Excess of manganese (Mn) concentration leads to chlorosis, while lead (Pb) and cadmium (Cd) poisoning can decrease the photosynthetic rates (Kusmana et al 2005).

Phytoremediation is one of the alternative technologies for overtaking the heavy metal poisoning. Phytoremediation is a technique by using plants aimed to remedy the environment from chemical poisoning. This term is applied for the usage of vascular plant for decontamination process of inorganic and organic pollutants (Reichenauer & Germida 2008). The plant can accumulate the pollutants via root, trunk, and leaf and stored. This process is influenced by pH, exchange capacity factor of soil, dissolved oxygen, temperature, and plant secretion process (Cheng 2003). For oil contamination, it will be discussed in the later part.

Oil contamination on mangrove forest ecosystem. One of the damaging factor which can lead to total destruction of mangrove forest is the oil contamination. Oil contaminations can potentially devastating individual mangrove plant, both due to its toxicity and the impact to the mangrove physiology. In this case, oil as a petroleum, is a dark brown flammable liquid. The petroleum consists of many hydrocarbon compounds. During the oil refinery process the compounds are separated into several fractions which has specific characters, usually boiling point (Speight 2014). A lower boiling point is highly volatile. As the number of carbon atom increases, the boiling point also increases as well (Table 2). Each group consists of several groups of hydrocarbon compounds, alicyclic and aliphatic. The concentration of alicyclic can reach between 50-98% (Mangkoedihardjo 2005). The alicyclic compounds which can be found in the petroleum are BTEX (benzene, toluene, ethyl benzene, and xylene), and PAHs (polycyclic aromatic hydrocarbon, such as: naphthalene, anthracene, phenanthrene, pyrene, ect.).

Table 2

Fractions of petroleum oil and its boiling point (Eneh 2011)

Boiling point (°C)	*Carbon chain number*	*Product names*	*Usage*
<20	$C_1 - C_4$	Natural gas	Fuel, chemical synthesis (for chemical industry process)
0-30	$C_1 - C_4$	Natural gas (zymogene and rhigolene)	Liquid zymogene is used for ice manufacture, rhigolene is used for local anesthesia
20-90	$C_5 - C_7$	Petroleum ether	Solvent
30-150	$C_5 - C_{14}$	Naphtha	Solvent
70-90	$C_6 - C_{18}$	Petrol	Automobiles, dry-cleaning process
70-200	$C_6 - C_{10}$	Gasoline	Fuel
90-120	$C_7 - C_8$	Ligroin	Dry-cleaning
100-200	$C_5 - C_{10}$	Oil fuel	Fuel
120-160	$C_5 - C_{10}$	Benzene	Solvent (dry-cleaning, and colvent in paint)
150-300	C_{10} - C_{38}	Kerosine	Fuel
175-300	$C_{10} - C_{18}$	Kerosine (paraffin)	Jet fuel
200-300	$C_{12} - C_{18}$	Kerosine (paraffin)	Fuel (lamp and stoves)
>275	$C_{12} - C_{20}$	Gas Oil	Diesel fuel
>300	$C_{18} - C_{38}$	Heavy Oil	Diesel fuel
300-400	$C_{15} - C_{25}$ $C_{20} - C_{24}$	Diesel Oil Lubricant	Locomotive fuel Lubricant, wax, shoe polish
>400	$C_{21} - C_{30}$	Paraffin wax	Various
Non-volatile	C_{20} <	Lubricant, dll.	Various
Solid residue	C_{40} <	Asphalt, bitumen, vaseline, pitch	Lubricant, asphalt, paint, vanishing, ect

Oil spill in mangrove forest ecosystem occurs after leakage of oil substance. Many anthropogenic activities, like oil exploration, highly contribute to the higher concentration of oil substance. The oil leakage in nature can be transformed via photo-oxidation. Several volatile compounds are evaporated. During this process, many compounds are reacted and decomposed into CO_2 gas (Dutta & Harayama 2000), while other remained compounds spread through the wind or sea current. If the compounds are transported via sea current, mangrove forest ecosystem could be in danger. As the oil spill comes into mangrove ecosystem, the oil will be adsorbed at the sediment, roots or trunk of mangrove plants (Kusmana et al 2005).

Oil spill can cause serious damage to the mangrove ecosystem. In a huge volume of oil spill, the plants will die very quickly in vast area. The leaves will be defoliated then the plant dies. Other related biotas to mangrove ecosystem can migrate to another unpolluted area or die as the mangrove plants die. In another case, if the oil spill impact is in sub-lethal concentration, plants dying process will slowly occur. Defoliation happens, followed by decreasing of plant growth, disappearing of forest canopy, and rearrangement of plant composition. Oil residue can last longer in the mangrove sediment. After 10 years of oil spill, mangrove ecosystem can be fully recovered through recolonialization or perished (Lewis 1983; Mastaller 1996).

Impact of oil spill to the marine biotas varies. A study case in Ixtoc-1, Mexico, showed a decrement of fish biodiversity. Fluctuation of hydrocarboclastic microbes showed the impact of oil spill to the indigenous microbe population. Bentics, like shellfish, tended to be hardly recovered (Soto et al 2014). Meanwhile, physiologically mangrove plant's root failed to transport water to the other plants part. Tansel et al (2015) study about oil impact to the roots of *Rhizophora mangle* showed water transporting failure happened in xylem. Oil substances are adsorbed to the surface of epidermis and pneumatophores. Later it caused a decrement of water flux exponentially, from epiderm to the cortex tissue. Water flux failure also happened as the sample was introduced with dispersant, however the water flux was relatively higher compared to the former result.

Oil spill substance varies from one to another location. It depends on the composition of aliphatic and aromatic compounds inside. Conventionally, HPLC (high performance liquid chromatography) or GC-MS (gas chromatography-mass spectrophotometry) are used for analyzing the sample (Fernandez-Valera et al 2010; Yin et al 2015). The PAHs (polycyclic aromatic hydrocarbons) are frequently used as marker compound of oil spill. PAHs in the environment tend to be calcinated onto the sediment or other surface areas. PAHs are toxic and potentially also disrupt many metabolism pathways (Viñas et al 2009; Wang et al 2016). Therefore, it is important to monitor the concentration of PAH as an oil spill occurs.

FAO report (2007) showed that the oil spill is one of the destructive factors which influence the quantity and quality of mangrove ecosystem. A case of oil spill on mangrove ecosystem happened in Bahía las Minas, Panama in 1986. Although the residues of oil spill had been cleaned up, the long impact is still observed until today. Mangrove regeneration is affected many years after the oil spill event. This oil residue causes systemic disturbance for mangrove regeneration process (NOAA 2014). Other case of oil spill on mangrove ecosystem happened in Nigeria (Kadafa 2012). It was due to oil exploration around the mangrove forest. This oil spill event decreases biodiversity of mangrove forest ecosystem significantly. Earlier studies predicted the oil spill during the leakage for 50 years was around 9-13 million barrels, as much as 50 times of Exxon-Valdez oil spill estimated volume.

Several areas of mangrove forest ecosystem in Indonesia are reported to be affected by oil spill. Mangrove forest in di Tambak Lekok is polluted with oil compounds, such as BTEX (benzene-toluene-ethylbenzene-xylene) inside the sediment (Indawan & Ahmadi 2008). A study case in Kepulauan Seribu, Jakarta showed a threat of oil spill had impacted mangrove plant seeds in 4 islands (Saputra 2011). Dori (2016) reported that the mangrove ecosystem in Tarakan City, North Kalimantan Province, had been polluted with oil. Several hydrocarbon compounds are found throughout the sediment and roots of plants, such as: styrene, 2,4,6-trimethyl xylene, phenanthrene, and naphthalene. Aromatic compounds were highly found on mangrove plant roots and lower part of sediment which has fine texture.

As in 2012 it was estimated around 1.8 million hectares of mangrove forest had been damaged in the world. This number is equal to 58% of total mangrove forest area in Indonesia. Many people, government, and NGO made a concern upon this issue for decreasing the rate of mangrove forest damage. Therefore, Indonesian Government during Susilo Bambang Yudhoyono presidency (2010-2012), had made 21 Commitments. One of the issues was about enhancement of social-economic coastal society through mangrove conservation. From 2010 until 2013 45 thousand hectares were prepared for mangrove planting (Priyasidharta 2012).

Overcoming the impact of oil spill in mangrove forest ecosystem. If the oil spill is not cleaned soon, mangrove ecosystem is at high risk of danger. This even can potentially depopulate this ecosystem entirely. Adding the factor of high sedimentation process in mangrove ecosystem, mangrove recolonialization process can be disrupted. From several cases for the remediation process, several approaches have been conducted: addition of chemical substances or through biological process. TPH (total petroleum hydrocarbon), production of CO_2 and NH_3 gases are the common method to measure the degradation process (Chikere et al 2011).

One of the obstacles for remediation process is the property of oil substance. Oil is hardly soluble in water. Surfactant is the best chemical compound for dissolving oil into water. Surfactant acts as the bridge between oil and water via hydrophobic and hydrophilic interaction. Several oil spill cases had used synthetic surfactants for remediation process. Deepwater Horizon oil spill in 2010 had made 4.1 million barrels oil spilled to the Gulf of Mexico. For overcoming this event, as much as 2 million gallons of dispersants were used: Correxit 9500A and Correxit 9527A (Graham et al 2016). Other oil spill cases used surfactants for dispersing oil into water were in: Europe, Alaska, and Japan (Kleidienst et al 2015).

Using the surfactant aims for dispersing oil molecules into the water body. As the molecules dispersed, it is potentially converted by the aid of many aquatic marine microorganisms around the affected area. Other mechanism beside dispersion is making the oil substances are easily photo-oxidated, since the surfactant can promote faster photo-oxidation process on oil substances (Fu et al 2016). Therefore, controlling the oil pollution can be accelerated. Several commonly applied surfactants during the oil spill events are Corexit, Omni-Clean, or Dispersit. Each dispersant consists of several anionic, cationic, or non-ionic surfactants.

However, using the surfactant is under critics. The main critic to this action is, is it safe to pour a lot of surfactant's volume into the body of water? As the previous statement, the existence of dispersant can dilute many oil compounds into water. Therefore, many toxic compounds can be easily diluted into aqueous phase, like PAH. This process can lead into biomagnification and bioaccumulation into higher trophic level of organism. There are many proofs that PAH is absorbed into plankton in a fairly high level (Viñas et al 2009; Pampanin & Sydnes 2013; Kleidienst et al 2015). Though the food chain and food web, a huge amount of this dangerous compound can be accumulated to the highest trophic of food pyramid (Almeda et al 2013; Prince 2015). In this case, human beings are very vulnerable.

Considering the potential adverse impact of using surfactants, many other researches are conducted for a more ecofriendly surfactant. One of the candidates is the biosurfactant, a kind of surfactant which is produced by microbes. This surfactant is relatively safe, because it easily degrades. It also has lower toxicity level and a better dissolving power compared to the synthetic ionic surfactant. One of the biosurfactant which is investigated frequently is the rhamnolipid. The rhamnolipid is produced by microbes of *Pseudomonas* (Soberón-Chávez 2005; Xia et al 2012). However, extracting the biosurfactant from the microbes is relatively expensive. Moreover, the rhamnolipid is only produced by *Pseudomonas aeruginosa* which is identified as potential pathogen microbe. Using this microbe type in the environment in vast scale is feared to give very backwards and adverse effect to other aquatic biotas (Varjani & Upasani 2017). For overcoming this obstacle, other kind of dispersant is created, like polymer based compound or composites. Yang et al (2014) developed a xerogel lignin-based surfactant, while Pi et al (2016) developed a new dispersant using silicon nanoparticle and natural polymer of xanthan gum. Gong et al (2016) developed a modified non-ionic dodecanol-based surfactant. Another approach is by harvesting rhamolipid from other microbes via gene modification, thanks to genetic engineering (Reis et al 2011; Varjani & Upasani 2017).

Another method for remediation is by adding some microbes to the oil-polluted water body. This technique is called bioaugmentation. Therefore, several researches were conducted for searching the microbes that can degrade several targets of oil compounds. Several species are able to degrade oil compound in a broad spectrum, it means it can degrades many kinds of aliphatic and aromatic compounds in oil spill (Broojimans et al 2009; Das & Chandran 2011). Febria et al (2011) had found several bacteria (*Pseudomonas* sp. and *Burkholderia* sp.) from Indonesia which had the ability to degrade oil, especially pyrene, a group type of PAH compound. Both bacteria had been characterized their 16S RNA sequence. However, there are two things that we have to point. First, using a large number of bacteria for degrading oil spill has a risk. We do not know whether it can impact the native microbe population, or it has several unknown adverse impacts. Hence, it will create imbalanced microbe population which can have negative impact to the ecosystem. Second, in several cases adding some other microbes were not positively correlated with the success of oil spill degradation process (Thompson et al 2005).

Another approach is by adding some nutrients to the oil-polluted environment. This technique is called biostimulation. The additional nutrients are the nutrients which are needed by the microbes for its growth and for the metabolic process. In the oil-polluted soil, nitrogen (N) and phosphorus (P) tend to be a limiting element. Moreover, both elements are also the limiting elements for mangrove plants to grow (Lovelock et al 2006; Scharler et al 2015). Therefore, adding fertilizer (Walworth et al 2007; Chorom et al 2010; Hamzah et al 2014) or manure (Charlena 2010) can be applied for helping the indigenous organism for degrading the oil contamination. The microbes then can grow well since it is able to use oil contamination as the carbon source. As time goes, the oil contamination can be cleaned. However, adding the mentioned nutrients should be noticed, since it can also enhance eutrophication or algae booming (Fuhrer 2014).

Phytoremediation is potentially applied as the technique for bioremediation of oil contamination in mangrove forest ecosystem. Phytoremediation is a method of using whole plants as bioremediation agent. Constantly, mangrove forest ecosystem accumulates many kinds of pollutants through sedimentation process, so pollutant concentration is decreased in the nearby water body. Pollutant is restrained in the sediment and naturally degraded by the microbes associated with the mangrove plant roots. Mangrove roots support oxygen to be dissolved, so it helps the nearby microbes to degrade the oil. As the vegetation zone is getting wider, the rhizosphere zone which is rich of oxygen content, can promote oil degradation (Kusumastuti 2009). Phytoremediation can be applied to with toxic metal absorption. Several mechanisms are proposed: (1) through direct taking of pollutant, and accumulated inside plants' organs, (2) transpiration of volatile compounds, (3) stimulation of indigenous microbes for actively degrading the pollutant, (4) enhancing pollutant mineralization process becoming the non-toxic substance (Burken & Schnoor 1997).

Several researches had been conducted for screening the potency of mangrove plant to assist the oil degradation. Several species are known to have these properties, like: *R. mangle* (Morerira et al 2011) and *Avicennia schaueriana* (Moreira et al 2016). However, there is a fundamental disadvantage for using phytoremediation as the sole method. Before applying phytoremediation, we have to know the range of oil tolerance from each species, beside phytoremediation is only feasible to be applied in the low level oil contamination. It cannot be applied in the cases of major oil contamination (Gomes 2012).

Conclusions. Due to the complexity of mangrove forest ecosystem, we must consider several processes for conducting the remediation from oil contamination, and this action should be concerned by many related stakeholders, like local government and nearby community impacted from oil contamination. No one method can be applied 100% without risk. The approaches have many advantages and disadvantages, however they are complementary each other. It needs a previous examination to measure the oil spill impact through laboratory test or in situ measurement. As the level of contamination has been measured, those remediation processes can be applied, depending on the field situation.

Dispersion process and isolation of contaminated area is the crucial aspect. We do not want the contamination area getting wider. Therefore, a manual cleaning process should be conducted by adding surfactants as the auxiliary element. Bioaugmentation and biostimulation processes can be applied sometimes after the introductory of surfactant. The surfactant and nutrients concentration should be monitored, so the adverse effect cannot be manifested easily. After the oil contamination decreases rapidly, mangrove recolonialization should be induced, considering to the impacted area. A model of handling process should be investigated later so, the integrated remediation process can support the restoration process, hence it can accelerate the remediation process. The communities surrounding mangrove area can participate actively in the remediation process, during oil contamination. Government policies before, during, and after the spill event is absolutely required for strengthening the stakeholders' roles.

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