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7 Production of carbon dioxide, methane and nitrous oxide gas emissions in biodegradation of waste oil sludge

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Abstract. Biodegradation Bioremediation of waste oil sludge through biodegradation using microorganisms still has an impact on the environment, because produces of gas emissions which causes to the global warming. The purpose of this study was to determine the production of carbon dioxide, methane, and nitrous oxide gas in biodegradation of waste oil sludge by using types of bioactivator from soil as the microbe inoculums sources was compost, mangrove and forest soil with adjusting the ratio of C:N:P of 360:60:1 to optimize the biodegradation condition. The results show that the addition of bioactivator on the treatment of waste oil sludge can increase the biodegradation process. The results show that the compost bioactivator treatment is 72 percent decrease in oil sludge, the forest soil bioactivator treatment of 58 percent and then the mangrove soil bioactivator of 46 percent. The observation of gas production obtained the highest concentration of carbon dioxide was produced on compost bioactivator treatment of 0.91 ppm on the 25 day, production of methane on compost bioactivator treatment was 0.66 ppm on the 25 day and production of nitrous oxide on the mangrove soil bioactivator treatment of 0.19 ppm on the 30 day; The decrease of hydrocarbon waste oil sludge and increased gas production are also associated with an increase in the population number of hydrocarbon degrading bacteria at all bioactivator treatments.

key words: gas emission, biodegradation, oil sludge

3 Introduction

One of the major problems faced by oil refineries is the safe disposal of oil sludge that is generated during processing of crude oil. Improper disposal of oil sludge leads to environmental pollution, particularly soil contamination and brings serious threat to the ground water. Many constituents of oil sludge are carcinogenic and potentially immunotoxicants [1]. Among many techniques used to decontaminate the affected sites, in situ bioremediation is the most widely used [2,3].

Bioremediation offers an alternative way that is not only effective in cost but also acceptable to the environment. Landfarming with using soil type is one of the advanced bioremediation methods that effective for contaminated soil of oil sludge. In this process, oil sludge from refinery waste is mixed with soil and subjected to enhance in situ bioremediation; inorganic fertilizer is applied to the site to



4 provide fixed nitrogen, and phosphate and pulverized limestone (CaCO_3) are added to raise the pH of the soil-oily sludge mixture about 7.8[4,5].

However, in the biodegradation process of oil sludge, it will produce several gases formed by the decomposition both soil total organic from plant material and the components of hydrocarbon contained in oil sludge. Gas emission that is produced in the biodegradation process to CO_2 , CH_4 and N_2O and many trace gases such as H_2 , H_2S , OCS and NO [6].

CH_4 and CO_2 are the dominant gases as the product of organic matters decomposition, while emission of N_2O depends on the form of N that is found in soil, whereas the other trace gases are only intermediates and are released into the atmosphere [7,8]. Gases emission of CO_2 , CH_4 and N_2O are included the main component that reduce the loss of outgoing infrared radiation [9].

Its Gases are relevant for atmospheric chemistry and climate that will lead to the higher temperatures at the earth surface leading to global warming [10,11].

Therefore, soil microbial activities are also the important source of the greenhouse gases [9, 12]. The third gases are interesting to be investigated in biodegradation process because they provide significant effect to the greenhouse effect.

2. Materials and Methods

2.1. Sampling.

There are three types of soil used as an inoculum source; soil forest; soil mangrove and compost. 1 The soil was air dried, crushed, and sieved (mesh size, 2mm) 2 and stored at 20°C in plastic bag. Previously, the soil with pH 6.8 to 7.0 and with $\text{Ca}(\text{OH})_2$ was used, oil sludge sample was collected from an open storage pit at the refinery at Unit Pertamina Indonesia, and it was stored in sealed plastic containers at a room temperature.

2.2. Characterization of soil and oil sludge

9 Physical and chemical properties of each sample of soil type was collected. 3 Soil samples were analyzed for organic carbon, nitrogen, available phosphorus and moisture level [13]. The analysis of organic carbon was also conducted on oil sludge. Determination of chemical properties was to optimize the biodegradation condition.

2.3. Biodegradation experiments

Biodegradation treatments soil were mixed with oil sludge ratio 1:1 based on its C organic content. Nitrogen and phosphorus content of the mixture were adjusted to reach a ratio of C:N:P as 360:60:1 by adding NH_4NH_3 and K_2HPO_4 mineral nutrient respectively. Water content of the mixture was maintained at 90% of water holding capacity. This treatments were carried out in microcosm landfarming system [2].

6 Microcosm was created by placing the biodegradation treatments in flakes and was temporarily sealed with rubber stopper that was equipped with a rubber septum and air samples. Each microcosm consisted of 75 g biodegradation treatment. These treatments were incubated at room temperature for thirty days and there were three replications for each treatment. The biodegradation treatments were as follows: Concentration of oil sludge, gas emission, and hydrocarbon degrading microbes were determined every five days.

2.4. Determination of oil sludge concentration

2 Determination of oil sludge concentration of each treatment biodegradation were extracted by the following methods: 10 g of biodegradation treatments was mixed with 7 g of 2 anhydrous Na_2SO_4 and extracted with diethyl ether in soxhlet extractor for 10 h. It was extracted and dried with an additional 0.1 g anhydrous Na_2SO_4 and transferred, with rinsing, to a tared vial. Evaporation was conducted at room temperature and the amount total of oil sludge hydrocarbon recovered was determined gravimetrically.

2.5. Gas emission analysis

Gas samples were collected using gas syringe. The samples were immediately injected into a gas chromatograph; CO₂, CH₄ and N₂O gas were detected with flame ionization detector. CH₄ and CO₂ were analyzed on an SRI GC (SRI Instruments, Torrance, Calif.), which was equipped with a flame ionization detector (FID) and a Hayesep D column (length, 2 m; 80/100 mesh). Helium was used as the carrier gas (flow rate, 20 ml · min⁻¹) and also synthetic air (20 ml · min⁻¹) and H₂ (20 ml · min⁻¹) were used as burning gas. The oven temperature was 80°C. Calibration was performed at each sampling occurrence by triplicate injection of 1,000 ppm of CH₄ in N₂ (Messer Griesheim). Standards were prepared by adding defined amounts to serum bottles of known volumes [7,10]. N₂O was analyzed with a gas chromatograph that was equipped with an electron capture detector and a Porapak Q-80/100 column (Supelco, Bellefonte, Pa). N₂O concentrations were expressed as the amount of N₂O in the gas phase plus the amount of N₂O [10].

2.6. Enumeration of microbial population

Enumeration of oil-degrading microbes used Bushnell-Hass agar (12 g KH₂PO₄, 1.0 g K₂HPO₄, 1.0 g NH₄NO₃, 0.2 g MgSO₄ · 7 H₂O, 0.05 g FeCl₃, 0.02 g CaCl₂, 20 g agar per liter). After steam sterilization, 5% of tween-80 was added into the medium of BH agar. Then, crude oil was mixed into BH agar and stirred by a magnetic stirrer. The medium of BH-oil agar was poured when it was cooled 50°C. Serial dilution of sample were prepared and spread onto BH-oil medium plates. After incubation at 30°C for 7 days, the colony was measured [14].

3. Results and Discussion

Determination of chemical properties on the soil sample including oil sludge was conducted to optimize the condition of biodegradation treatments, the results were analyzed in Table 1. such as; the content of nitrogen and phosphorus of the mixture was adjusted to reach a ratio C:N:P as 360:60:1 by adding NH₄NH₃ and KH₂PO₄ mineral nutrient respectively.

Table 1. Chemical properties of each soil types and oil sludge sample

Soil type	Organic carbon	Nitrogen	Available phosphorus
Soil of mangrove	31.9345	0.14	0.26
Compost	26.6766	0.08	0.45
Forest soil	34.6429	0.03	0.13
Oil sludge	62.8	0.067	0.19

From the analysis on total oil sludge hydrocarbon residue, it showed that all treatments had started to decrease slightly on day 5 where the highest was 14% at mangrove soil treatment. Then on day 10, it started experiencing significant concentration decrease especially at compost treatment compare with mangrove soil and forest soil treatment. The biodegradation process continued until the day 30 and remained on the compost treatment that showed a higher biodegradability indicated by the remaining oil sludge hydrocarbon residue of 28%. Then it was followed by mangrove treatment 42% and forest soil 63%, while on the control treatment there was a gradual decrease in the total hydrocarbon residue up to the 30th reaching 6% as shown in Figure 1.

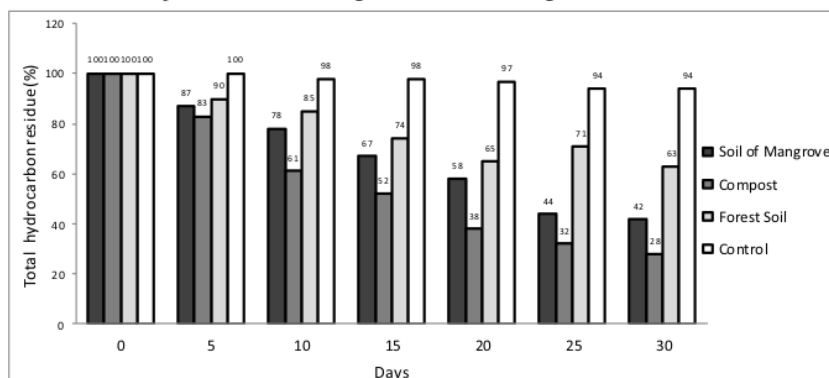


Figure 1. Total hydrocarbon residue in biodegradation of oil sludge with soil types are soil mangrove, compost and forest soil.

From the observation, it shows that on all treatments, biodegradation processes in oil sludge hydrocarbon occurred due to the presence of a number of microbes found in the soil type used as the inoculum source of degrading microbes [2]. The microbes present in this type of soil are easy to degrade the hydrocarbon oil sludge as petroleum hydrocarbon is similar to the molecular structure of the plant parts contained in the soil type. For example, hexane shares similarity with waxy substances in leaf, while aromatic hydrocarbon compound is similar to the lignin molecule structure in wood. This causes the microbes present in the soil type to be more enzymatically adaptive [15,16]. On the other hand, the loss of hydrocarbon residue in the control treatment occurred because abiotic loss process that is caused by environmental factors such as temperature, light or pressure that might cause hydrocarbon evaporation in oil sludge.

The addition of compost can give a great reduction of total hydrocarbon, followed by the addition of forest soil and mangrove soil. Production of CO₂ significantly increased at the 10th until 25th day after treatment. CH₄ gas was produced at the day and continuously increased up to the 30th day. While NO₂ gas that was treated especially with compost produced more gas than the other treatments that only produced gas at 20th day. Gas emission produced during biodegradation at forest soil of CO₂ was higher, which began at 10th day with 0.291 ppm and then decreased at 25th day. CH₄ was produced with high concentration at final incubation, but N₂O was lower than CH₄ as shown in Figure 2.

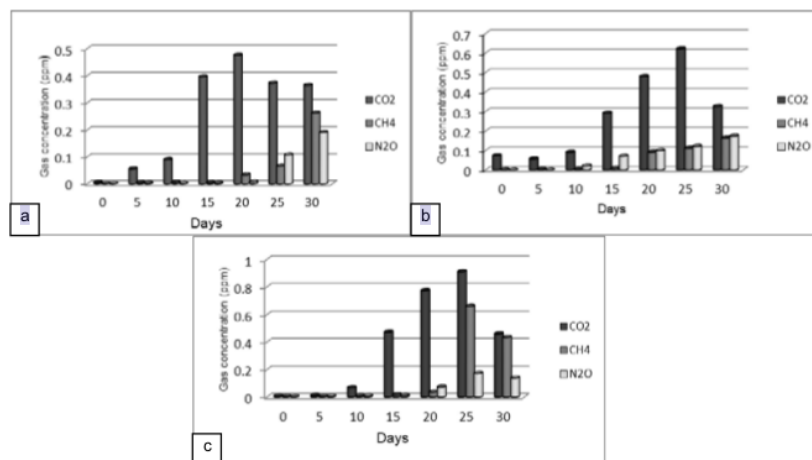


Figure 2. Production of CO₂, CH₄ and N₂O gas emissions in biodegradation treatment with (a) soil of mangrove. (b) forest soil; and (c) compost

The highest CO₂ gas was produced on the treatment of compost 0.91 ppm at the 25th day. The highest CH₄ gas on the compost treatment was 0.66 ppm at the 25th day and the highest N₂O was produced on the mangrove soil treatment was 0.19 at the 30th day. Observation of gas emission CO₂ of mangrove concentration that generated from 10th day to 25th day increased at 20th day. Even though the CO₂ was still the highest, by using compost treatment, CO₂ concentration increased at 15th day and N₂O increased at 30th day, so it was higher than mangrove soil and forest soil.

The process of hydrocarbon biodegradation of oil sludge is related to the growth of microbes and the resulting gases. The population of hydrocarbon degrading bacteria increased at 6th day after treatment. A significant increase in hydrocarbon degrading bacteria population was observed from the compost treatment up to the 30th day. While the treatment with mangrove soil and forest soil decreased at the 18th day as shown in Figure 3.

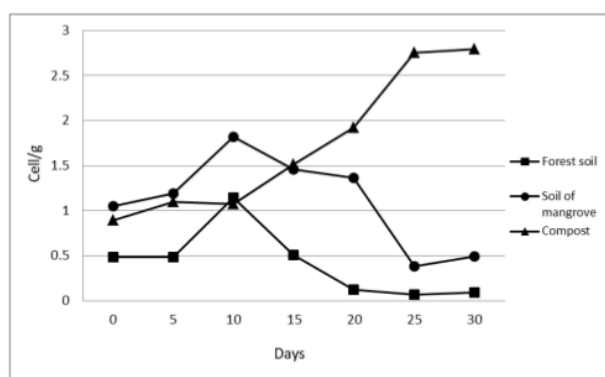


Figure 3. Number hydrocarbon degrading microbe of biodegradation treatment with forest soil, soil of mangrove and compost

The results of observation on all microbes that can degrade oil sludge hydrocarbon show that the increasing growth began at the 12 days in each treatment, except on forest soil that began to increase on the 6th day. It proves that microbe that can degrade oil sludge hydrocarbon need time to adapt with new substrate of oil sludge hydrocarbon. It also might occur due to the pressure to the microbe as the selection by the toxic component will decrease the population and diversity, so it gives the effect on diauxic growth of microbe [16,17].

Oil sludge is waste product of oil refinery activities that can badly affect the environment. Biodegradation is an effective method to solve this problem. In its application, soil is often being used as inoculum source. However, depending on vegetation growing on it, the content of organic material of each soil type has its own uniqueness. It affects the population and diversity of soil microbes in it [2,18]. The biodegradation process of oil sludge to produce CO₂, CH₄, N₂O and other gases emission is by decomposition both soil organic matter and hydrocarbon component contained in oil sludge [14].

Mineralization of organic component that comprises from inoculum source have been done by the microbe that can produce CO₂, CH₄, N₂O or any other gases. The gases produced are various depend on type of soil that is used as inoculum source. Different kind of soil contains unequal amount and type of microbe. In the other hand, they also include the compound of unequal organic composition. The simple organic molecules will crash sooner. Various inoculum source addition will affect the mineralization of oil sludge hydrocarbon [16]. Therefore, it will be seen from every type of various inoculum source addition and different gasses produced. The observation result showed that all treatments produced higher amount of CO₂. Here CH₄ and NO₂ gas were relatively in small amount, except in the second compost soil treatment in which these gases were shown in higher amount. It occurred because the compost soil contained a lot of microbe with many types. It also occurred on higher amount of nitrogen compound what was caused by higher level production. The higher level production of CH₄ gas was caused by anaerobic process that was made possible based on the compost soil texture itself.

The observation result on total residues of oil sludge hydrocarbon in each treatment of the added soil in oil sludge showed that the decreasing peak was at the 24th day of observation until the end stage of incubation especially on compost treatment. If it was closely related with gas production because it started on the 20th day. CO₂, CH₄ and NO₂ kept increasing during the observation. Even though it was not followed by the growth of microbe that could degrade oil sludge, the total amount of microbe still

increased relatively. The possibility of gas production also came from the reduction of soil organic sources added, especially NO_2 gas source from denitrification of soil organic nitrogen [19,20].

Therefore, in this study, the highest emission gas is from the soil of compost inoculum and then, forest soil and mangrove soil are still relatively lower in gas production. However, in the other hand the ability of compost degradation is higher.

4. Conclusions

During the hydrocarbon oil sludge biodegradation process, several gases were produced is CO_2 , CH_4 and N_2O . The increase in oil sludge biodegradation process was along with the increase in gas produced where CO_2 was the highest at compost treatment by 0.91 ppm, while CH_4 was 0.66 ppm on day 25, and N_2O was the highest on mangrove treatment by 0.19 ppm at day 30. The decrease hydrocarbon oil sludge and increase the amount of gases generated are also accompanied by an increase in the number of hydrocarbon degrading microbes at all treatments.

References

- [1] Mishra, S., J., Jyot, R.C., Kuhad, and B, Lal. 2001. *Appl. Environ. Microbiol* 67 (4):1675 - 1681.
- [2] Dibble, J.T. and R. Bartha, 1979. *Appl. Environ. Microbiol* 37 : 729 – 737.
- [3] Song, H. and R. Bartha, 1990. *Appl. Environ. Microbiol.* 56(3) : 646 – 655.
- [4] Eweis, J.B., S.J. Ergas, D.P.Y. Chang, and E.D. Schroeder, 1998. *Bioremediation Principles*. WCB, McGraw-Hill. Boston.
- [5] Efsun, E. F. Olcay, T. Şağban and H. S. Başkaya, 2016. *Turkish Journal of Agriculture and Forestry* 40: 631-641.
- [6] Shrestha M., W.R. Abraham, P.M. Shrestha, M. Noll and R. Conrad, 2008. *Environ Microbiol.* 10:400–412.
- [7] Hardy K.R. and G.M. King, 2001. *Appl. Environ. Microbiol.* 67 (8): 3671 – 3676.
- [8] Lori, B, B.Peter, B.Philippe, D. Edward and P.Tans, 2017. *Journal of Geophysical Research- Atmospheres* 7 (122): 4070–4083.
- [9] Trumbore S. 2006. *Global Change Biol.* 12:141–153.
- [10] Matthies, C., A. Griebhammer and M. Schittroth, 1999. *Appl. Environ. Microbiol.* 65 (8): 3599 – 3604.
- [11] Falkowski P.G, T. Fenchel and E.F. Delong, 2008. *Science* 320:1034–1039.
- [12] Wang C., D.Y.F. Lai, J. Sardans, W.Wang, C. Zeng and J. Peñuelas, 2017. *PLoS ONE* 12(1): e0169254. <https://doi.org/10.1371/journal.pone.0169254>.
- [13] Nelson P.W. and C.E. Sommers, 1996. Total C, organic C and organic matter. In: PAGE, A.L. (Ed.). *Methods of soil analysis. Part 2. Chemical methods*. Madison: SSSA. pp. 539-579.
- [14] Bagga J, M. Pandey and V. Pandey, 2015. *International Journal of Innovative Research in Science, Engineering and Technology*.4(10): 9993–10005.
- [15] Christos G., M.C. Joanna, and J.S. Liz, 2014. *J. Sci Food Agric.* 94(12): 2362–2371
- [16] Fahrudhin and A.Abdullah, 2015. *Asian Journal of Microbiology, Biotechnology and Environmental Sciences*17(2): 321-324.
- [17] Mamitha K.S., I. Grasian I and Albert H.S., 2013. *Bioinformation* 9 (3):150–157.
- [18] Langenheder S. and J.I. Prosser, 2008. *Environ Microbiol.* 10:2245–2256.
- [19] Brüggemann, N., P.Rosenkranz, H. Papen, and K. Butterbach-Bahl, 2003. *Geophys. Res. Abstr.*(5) 08463.
- [20] Thomsen, T.R., Y. Kong and P.H. Nielsen, 2007. *FEMS Microbiol Ecol.* 60:370–382.

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