Master Thesis

Biodegradable dust suppressants using biomass-based polymeric materials

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Biodegradable dust suppressants using biomass-based polymeric materials

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ABSTRACT

Either natural or anthropogenic fugitive dust causes a serious hazard to the environment and human health, in many countries and areas all around the world. In this study, development of biodegradable dust suppressants and their environmental impacts were evaluated Biodegradable dust suppressants were synthesized using various biomass-based polymeric materials such as crude glycerol (a by-product of biodiesel manufacturing), biodiesel, palm oil, cooking oil, seaweed mixtures, wakame (Undaria pinnatifida), and red algae. The results of wind tunnel tests with Korean standard sand demonstrated that spraying diluted mixtures of crude glycerol and biomass materials can significantly reduce the generation of dust. The optimal molar mixing ratio of crude glycerol and the biomass materials was 1:1, and the optimal dilution concentration was determined to be 100 times for the mixture of crude glycerol and biodiesel, palm oil, and cooking oil and 50 times for the mixture of crude glycerol with a seaweed mixture, wakame, and red algae. The suppression ability was 83.4%, 60.4%, 99.5%, and 98.1% for the mixtures of glycerol with soybean oil, palm oil, wakame, and red algae, respectively. The mixtures of glycerol plus wakame or red algae were the most efficient suppressants; they also have substantial biodegradability. Our results suggest that the mixture of crude glycerol with the various oils or the seaweeds may be a promising option to develop non-toxic biodegradable dust suppressants.

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1. INTRODUCTION

Fugitive dust is one of the serious problems among environmental issues which can arise from the mechanical disturbance of soils that injects fine particles into air. Atmospheric particulate matter contains various metals, depending on natural or anthropogenic factors, which are harmful to the human body. These suspended particles have been shown to constitute a large fraction of PM10 (particles with aerodynamic diameters less than 10 micrometers) in many urban and non-urban areas [1], [2]. These fine dust sources were generated from a variety sources such as roads, construction sites, combustion processes, and yellow dust (incoming from China and Mongolia), and might cause harms to humans in the short or long term. In particular, a significant impact in the case of the fugitive dust generated from construction sites nearby residential living environment, it is necessary to control these fine dust [3]. A 2008 study found that PM10 generated by erosion of road pavement by studded tires provoked an inflammatory responses in cells as potent as the response caused by diesel particles [4]. Severe health effects include a significant reduction in life expectancy of the average population by a several months, which is linked to long-term exposure to moderate concentrations of PM10 [5].

Various methods have been employed to suppress the dust such as street washing with water, street sweeping and using chemicals as suppressants. Street washing with water has been considered by several studies as a method able to reduce the mobility of dust load deposited on street surfaces [6]. However, watering, has a short restraint time and limits the ability of mud to adhere to the wheels of the vehicle. It also has the disadvantage of the secondary pollution arising from the spent water. One study reviewed more than 60 commercial suppressants in the categories of salts, asphalt/petroleum emulsions, tree resin and organic emulsions, lignin sulfonates, polymers, fibers and mulches [7]. According to Chang, 2003 [8] and Kirk, 2018 [9] the experiments using general dust reducer showed superior efficiency in dust suppression over a longer period than when water used. Salts and Brines are the most common type of dust suppressant used. In 1991, 75-80% of all dust suppressants used were chloride salts and salt brine products, 5-10% were lignin sulfonates, and 10-15% were petroleum-based products [10]. Calcium chloride ($CaCl_2$) and magnesium chloride ($MgCl_2$) are the major products in this category. The major known effects of salt in the environment relate to its capacity of moving easily with water through soils [11]. In the area near the application of salts, there have been negative impacts to the growth of fruit trees [12], and alterations in the plant nutrition due to increases in the osmotic pressure of soils [13]. Chloride concentrations as low as 40 mg/L have been found to be toxic to trout, and concentrations up to 10,000 mg/L have been found to be toxic to other fish species [14], [15]. High levels of lignin sulfonate in water bodies have high coloring effects, increase biochemical oxygen demand, reduce biological activity, and retard growth in fish [15], [16], [17], [18]. The environmental impact of suppressants depend on composition, application rates, and interactions with other environmental components. Potential environmental impacts include: surface and groundwater quality deterioration; soil contamination; toxicity to soil and water biota; toxicity to humans during and after application; air pollution; accumulation in soils; changes in hydrologic characteristics of the soils; and impacts on native flora and fauna populations [7].

Therefore, it is imperative to develop a new dust suppressant that can be biodegraded in the soil and that is not hazardous to human health or the environment when blown in the wind or absorbed into a body of water. It has been hypothesized that the combination of high viscous glycerol and readily biodegradable oily compounds may perform well as long lasting, high performance suppressants. Therefore, this study aims to develop a biodegradable dust suppressant with low toxicity, to evaluate various potential dust suppressants, and to propose promising candidate products for commercialization and mass production. By products of biodiesel production (as well as biodiesel ingredients themselves), marine biomass, and commercial vegetable oils were selected for the synthesis of suppressants. The optimal mixing ratio was determined, and the suppression ability was evaluated via wind tunnel tests. Considering biodegradability, the most effective suppressants were determined.

2. MATERIALS AND METHODS

2.1 Crude glycerol

Crude glycerol is the by-product of biodiesel production and it has very low value because of the impurities contained. However, the utilization of the glycerol became an urgent need, as the demand and production of biodiesel grow continuously [19]. The previous studies report indicating that crude glycerol is effective in some dust suppression applications [20]. Besides, there are some reports crude glycerol can be digested by microbial bioconversion [21], [22]. In the scope of this study, evaluated for dust suppressant performance when applied alone as aqueous solutions and in combinations with biomass-based polymers.

2.2 Materials

Potassium permanganate ($KMnO_4$, 99%) was purchased from Daejung Chemical (Gyeonggi, South Korea). Sodium oxalate ($Na_2C_2O_4$, 99%) and sulfuric acid (H_2SO_4, 95%) were purchased from OCI (Seoul, South Korea). Silver sulfate (Ag_2SO_4, 99.5%) was provided by Kojima Chemicals (Aichi, Japan). The PolySeed® BOD seed inoculum capsules, contains a minimum of 100 mg of specialized microbial cultures were purchased from Hach (Loveland, CO, USA). The raw materials crude glycerol, biodiesel, and palm oil were supplied through JC Chemical, a company that develops and produces alternative fuels such as biodiesel to be added to diesel in Ulsan, South Korea. Between various types of cooking oil, most commonly used soybean oil was purchased from Ottogi Co., Ltd (Gyeonggi, South Korea). We selected biodiesel due to its property that it has low viscosity, biodegradable unlike fossil fuel and has no toxicity. The same reasons apply for crude glycerol and palm oil. Molar mass of crude glycerol, palm oil, biodiesel, and soybean oil is 92, 256, 270, and 874 g/mol, respectively [23]. "Seaweed" was the washed out mixture of many kind of seaweeds and obtained from East Sea beach (Ulsan, South Korea). "Seaweed (wakame)" is the specie of edible seaweed, a type of marine algae, and a sea vegetable and easily purchased from grocery stores in Ulsan, South Korea. “Red algae” powder used as an ingredient in desserts throughout Asia, substitute for gelatin and purchased from grocery stores in Ulsan, South Korea.

2.3 Synthesis of dust suppressant

Crude glycerol, states in a solid state at room temperature therefore it was heated in an oven at 120 °C for 30 min. To prepare 1 M solution, in 1 L volumetric flask, approximately 200 ml of deionized (DI) water and 92 g of pre-heated crude glycerol was introduced and vigorously stirred. After 1 L of solution was made by adding DI water to the volumetric line, the 1 M glycerol solution was stirred at 180 rpm for 1 h. The solution was mustard-yellowish color and completely miscible. Palm oil, biodiesel, and soybean oil state in the liquid state at room temperature, therefore, didn’t require pre-heating. The 0.5, 1, 2 and 3 M of palm oil solutions were prepared in the same method as the crude glycerol, according to their molar mass. For biodiesel, 0.5, 1, 2, and 3 M of solutions were prepared under the same methods as well. It was observed that the prepared solutions were immiscible, the oil and water components were divided into layers within 2 to 3 minutes. Therefore, 20 g of sodium dodecyl sulfate (SDS) surfactant was added and stirred at 180 rpm for 3 h to make the solutions completely miscible. Palm oil and biodiesel solutions were visibly very similar with milky white color and foamy. To find the optimum mixing ratio of suppressants, crude glycerol and palm oil (or biodiesel) were mixed with ratios ranging from 1:0.5 to 1:3 M. The samples were shaken at 180 rpm and sampled by time 15, 30, 45, 60, 90, 120 and 180 min. Taken samples show that palm oil concentration increase in mixture, solution began to appear solid and more foamy but pH amount 11.6 didn’t change significantly and mixtures of crude glycerol and biodiesel characteristics were identical. Therefore mixing ratio of palm oil or biodiesel with glycerol was most suitable for 1:1 M.

To prepare seaweed mixture and wakame solutions, the two types of seaweeds were dried at room temperature for 24 h. The seaweeds were ground and passed through a sieve (#4 mesh, ≤4.75 mm). Then, 50 g of ground seaweeds were added to 1 L DI water, and boiled for 4 to 5 h, with constant stirring at 180 rpm. The solutions became visibly slimy and clammy. After cooling at room temperature for 2 h, the solutions were diluted to 10 g/L with DI water. Because the synthesized solutions included suspended materials, they were filtered through a glass fiber filter (Type B) (Millipore, Burlington, MA, USA) and the filtrates were stored in a refrigerator. Both 2 kind of seaweeds were prepared same conditions. The red algae we obtained was powder state thus not required to pre-dry and grind it. To prepare the red algae solution, 10 g of the powder was added to 1 L of DI water. The mixture was heated at 100°C with constant stirring (180 rpm) for 10 min, and cooled to room temperature. After filtration with the glass fiber filter, the algae solution was stored for further experiments.

2.4 Wind tunnel tests

A wind tunnel experiment was designed to measure whether the dust suppressant candidate samples could sufficiently suppress scattering dust generation. A laboratory-scale tunnel test model was designed (Fig. 1). According to the Big Spring testing model suggested by Fryeat [24], a rectangular prism-tunnel was made, using a wooden table with size of 1.7 m(length)*0.7 m(height)*0.9 m(width) as a structural frame and covered sides with a vinyl sheet, except for 2 ends. A commercial drum-type fan (Hans Electronics, Busan, South Korea) was installed as the blower and places one end of the tunnel. The performances of the fan are as follows, compact and wind sizes is 0.93 m(length)*0.93 m(height)*0.33 m(width) and 0.76 m/76 m, respectively. Revolutions per minute (RPM) of fan is 1085, where air volume is 17100 m^3/h and have 0.54 horsepower. The wind speed was maintained at 7.5 m/s measured with an anemometer (Testo 410i, Lenzkirch, Germany). As a scattering dust material, we used Jumunjin sand, which has served as a standard and is the most commonly adopted sand in Korea. The Jumunjin sand was chosen because preliminary tests with fine particles 2 such as clay and silt did not show any significant difference between suppressants and water. Jumunjin sand is classified as a poorly graded sand (SP) with a specific gravity of 2.65 and mean grain size (D50) of 0.52 mm, where the uniformity coefficient (Cu) is 1.94 [25].

The tunnel test procedures were as follows. First, an aluminum tray (area 0.16 m2) was filled with the Korean standard sand and weighted. Then, sand-filled tray was placed in the tunnel, in front of the fan, and experiments were started by turning on the fan to mimic wind blowing. After a pre-determined period (15, 30, 45, 60, and 120 min), the weight of the remaining sand was recorded, which was calculated by subtracting the weight of the sand remaining in the tray from the initial weight of sand. To determine the effect of the suppressant, 1 kg/m^3 of suppressant was spread evenly on the sand. As a control experiment, identical experiments were conducted with and without water. Preliminary tests using DI water without suppressant showed no weight loss in the wind tunnel tests (without drying periods). When the suppressant solution-spread sands were dried at room temperature for 3 days, clear distinctions among the suppressants were observed in the wind tunnel tests. Since further drying time did not increase the effects, the 3-day drying step was inserted between suppressant adding and wind tunnel testing. All tests were conducted in duplicate.

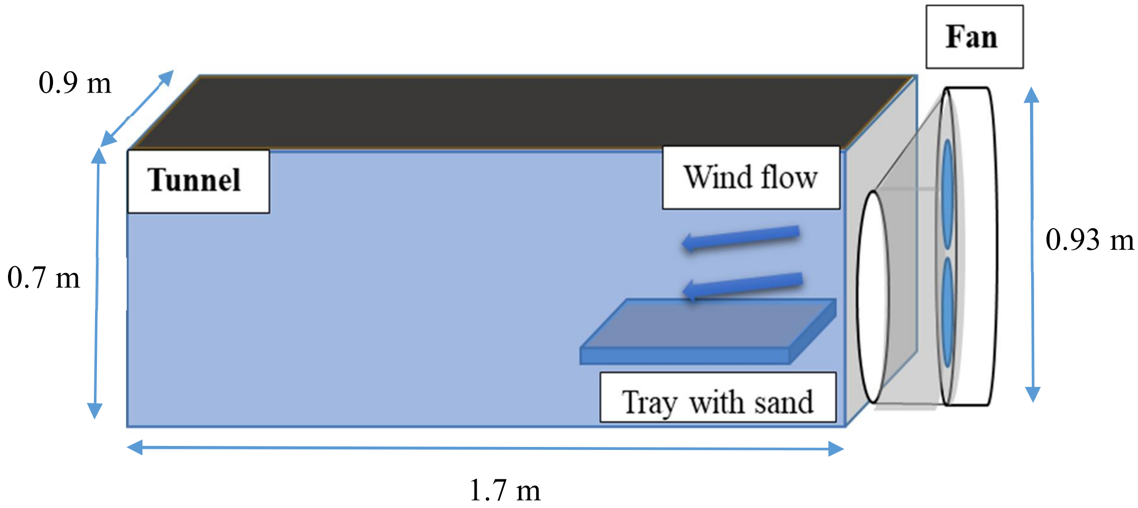

Fig. 1. Schematic graph of the wind tunnel test design. Air flow rate is 17100 m^3/h and wind speed is 7-7.5 m/s.

3. RESULTS AND DISCUSSION

3.1 Wind tunnel test results of control experiments

The control experiment results shown in Fig. 2. For the sand control experiment, after 15 min, over 71% of the sand mass was lost, and after 2 h more than 90% of the sand mass was lost. When DI water was sprayed, the suppression effect was evident at 1 h, with only 3.6% of sand mass loss. However, at 2 h, the water started to evaporate, and the loss of sand mass increased to 60.9%. For a 72 h drying time after DI water was applied 41.3% and 83.8% of the sand mass was lost in 15 min and 2 h, respectively. It was confirmed that the loss of sand mass was increasing as the water dries.

The control experiments were conducted using crude glycerol, palm oil, biodiesel, and soybean oil to ensure each of the materials could act as dust suppressants. The pre-prepared 1 M glycerol solution was diluted with DI water to 10 and 100 times and sprayed onto sand. After 72 h of natural drying, the results were as follows 0.1 M glycerol sprayed sample lost no sand mass but for 0.01 M of glycerol sprayed sand weight loss was 45.4% it indicated more than two times higher dust suppression ability than water alone (Fig. 3a). In a similar way, palm oil, biodiesel, and soybean oil were also diluted and applied onto the sand, and their suppression results are shown in Fig. 3b. All the 0.1 M diluted solutions had no loss of sand mass, and among the 0.01 M diluted solutions, soybean oil had the highest suppression ability (100% with 0.01 M soybean oil). All the solutions had a better suppression effect than DI water.

Two types of seaweed and red algae were used for the control experiment to ensure each of the materials could act as dust suppressants as well. Pre-prepared 10 g/L of the solutions were diluted by 10, 50, and 100 times and applied to the sand. The results of the wind tunnel tests showed that the seaweed solutions had a suppression effect. For the 10 times-diluted seaweed mixture and wakame samples, no loss of sand mass was detected. For instance, 50 times-diluted seaweed mixture, the loss of sand mass was 44.3% which was two times higher dust suppression ability than water. For the 50 and 100 times-diluted wakame samples, the loss of sand mass was 62.3% and 73.6%, respectively. Red algae spray showed lower suppression ability than 2 kinds of seaweeds but higher than water. The 10 times diluted red algae spray had 63.1% of sand mass loss, 50 and 100 times diluted sprays sand mass loss was 80.2% and 82.8%, respectively (Fig. 4). All of the materials have confirmed that better suppression ability than water.

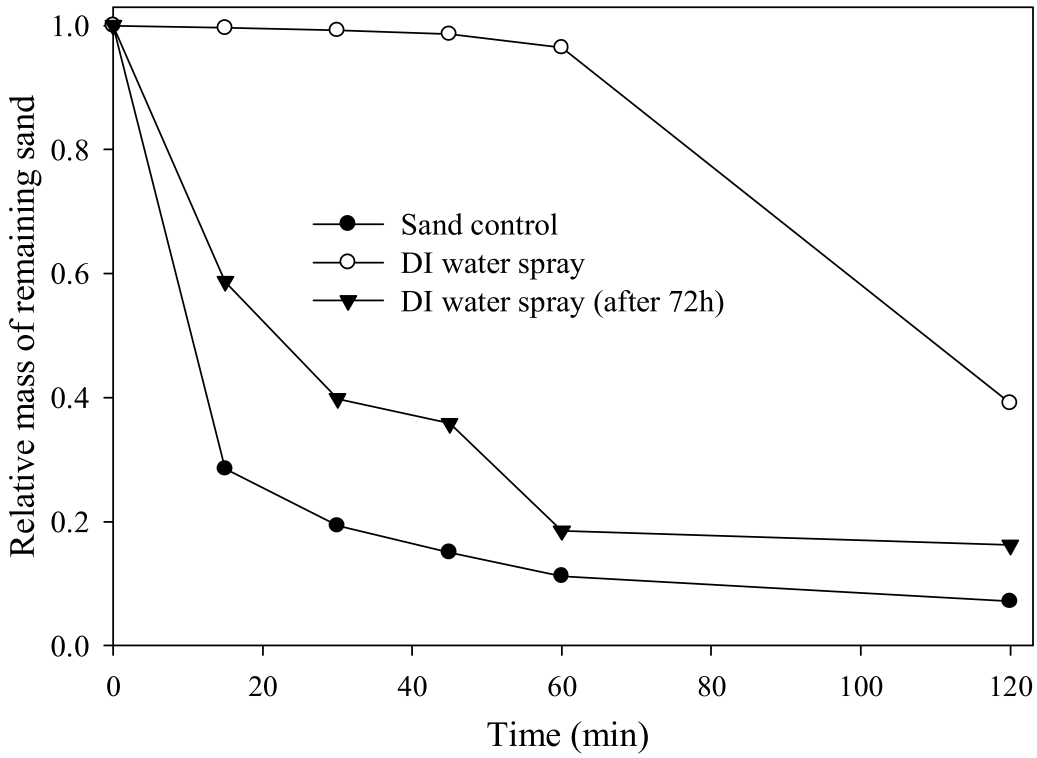

Fig. 8. Relative remaining sand mass of the standard Korean sand and DI water control.

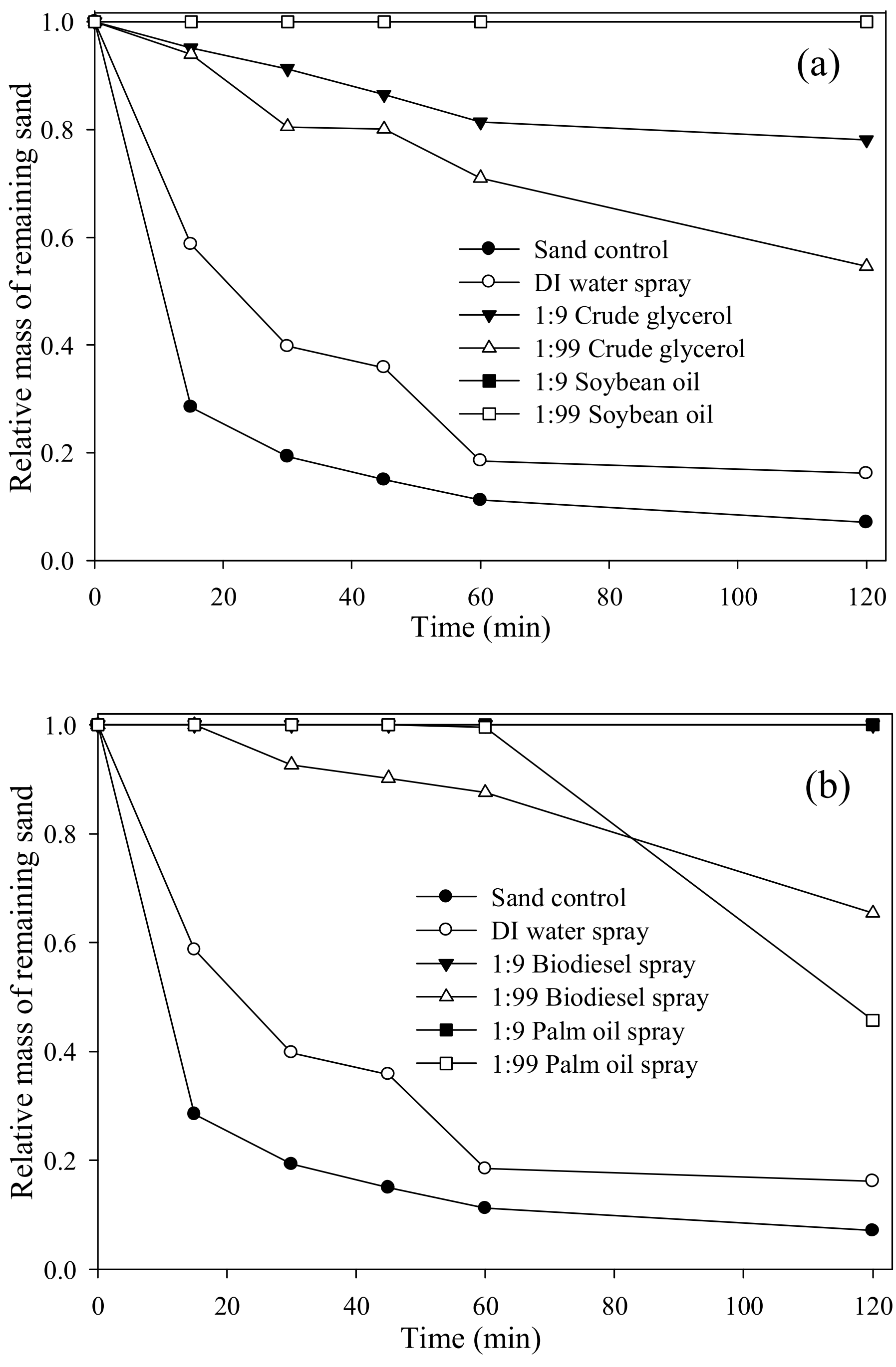

Fig. 3. Relative remaining sand mass of (a) crude glycerol control and (b) biodiesel and palm oil sprayed without glycerol.

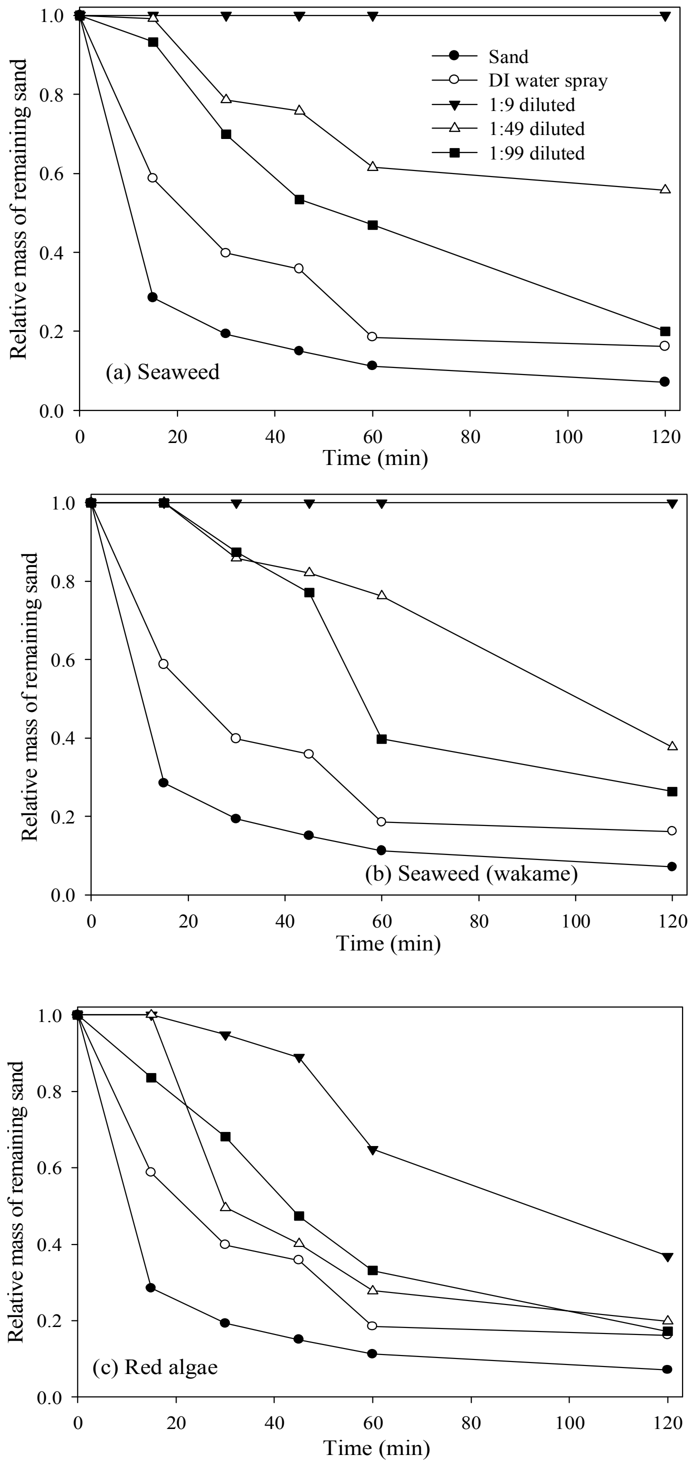

Fig. 4. Relative remaining sand mass of (a) seaweed, (b) seaweed (wakame) and (c) red algae sprayed without glycerol.

3.2 Wind tunnel test results of suppressant candidate experiments

Further, to obtain a suppressant that is more stable and has a higher suppression ability, we mixed the crude glycerol with other materials, in a molar ratio of 1: 1 and added 20 g/L surfactant as mentioned before. These candidates were diluted 10 (0.05 M) and 100 (0.005 M) times and the dust suppression efficiency was evaluated with wind tunnel test, results shown in Fig. 5. The 0.05 M of mixture of glycerol and palm oil, biodiesel, and soybean oil suppressants had no sand mass loss determined. For the 0.005 M mixtures of glycerol and palm oil, glycerol with biodiesel and glycerol with soybean oil sand mass loss was 39.6%, 49.3% and 16.6%, respectively. Adding palm oil and soybean to the glycerol enhanced the suppression ability of glycerol but for biodiesel did not had the same effect. Although the mixture of glycerol and soybean oil was most effective in suppression ability, we assumed it is not appropriate to food product and as a suppressant also the following experiments showed that this mixture has the lowest biodegradability. Therefore, glycerol and palm oil mixtures sand mass loss was lowest and further used as a baseline of suppression ability.

The 10 g/L solution of red algae and 2 kinds of seaweeds were prepared in advance and each were mixed with 1 M glycerol in a ratio of 1: 1 to prepare candidates for the dust suppressant. After that the solutions were diluted 10, 50 and 100 times and wind tunnel test was conducted. The 10 times diluted 3 types of mixtures had no mass loss determined. Experiment results showed that the 50 times diluted samples of glycerol and seaweed mixture sand mass loss was 34.8% and it means higher suppression ability than mixture of glycerol and palm oil. For mixture of glycerol and seaweed (wakame) sand mass loss was 0.5% and for glycerol and red algae mix relative sand mass loss was 1.9% where the suppression ability is 60 and 20 times higher than glycerol and palm oil mixture, respectively. Adding glycerol to the seaweed, seaweed (wakame) and red algae solutions showed the synergistic effect on suppression ability, sand mass loss was decreased by 21.4%, 99.1%, and 97.6% when included glycerol solution, respectively. However, 100 times diluted mixtures suppression ability was relatively low, the sand mass loss of mixtures of glycerol and seaweed was 65.9%, glycerol and seaweed (wakame) was 73.6% and glycerol with red algae was 73.4% (Fig. 6).

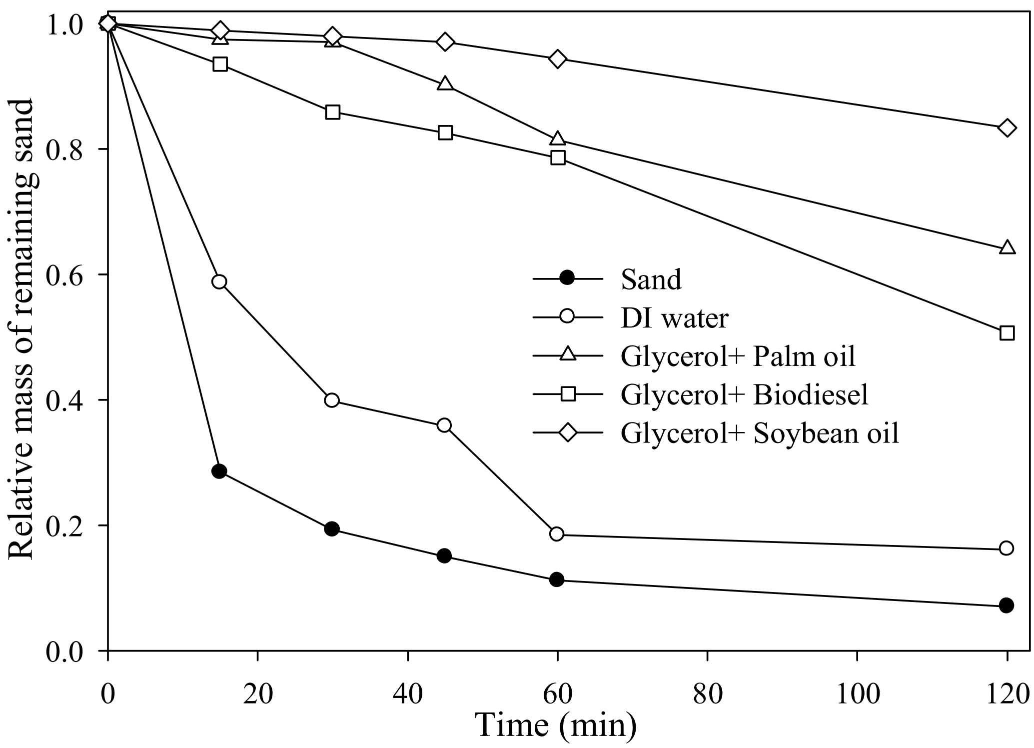

Fig. 5. Relative remaining sand mass when 0.005 M mixtures of glycerol and palm oil, glycerol with biodiesel, and glycerol with soybean oil used.

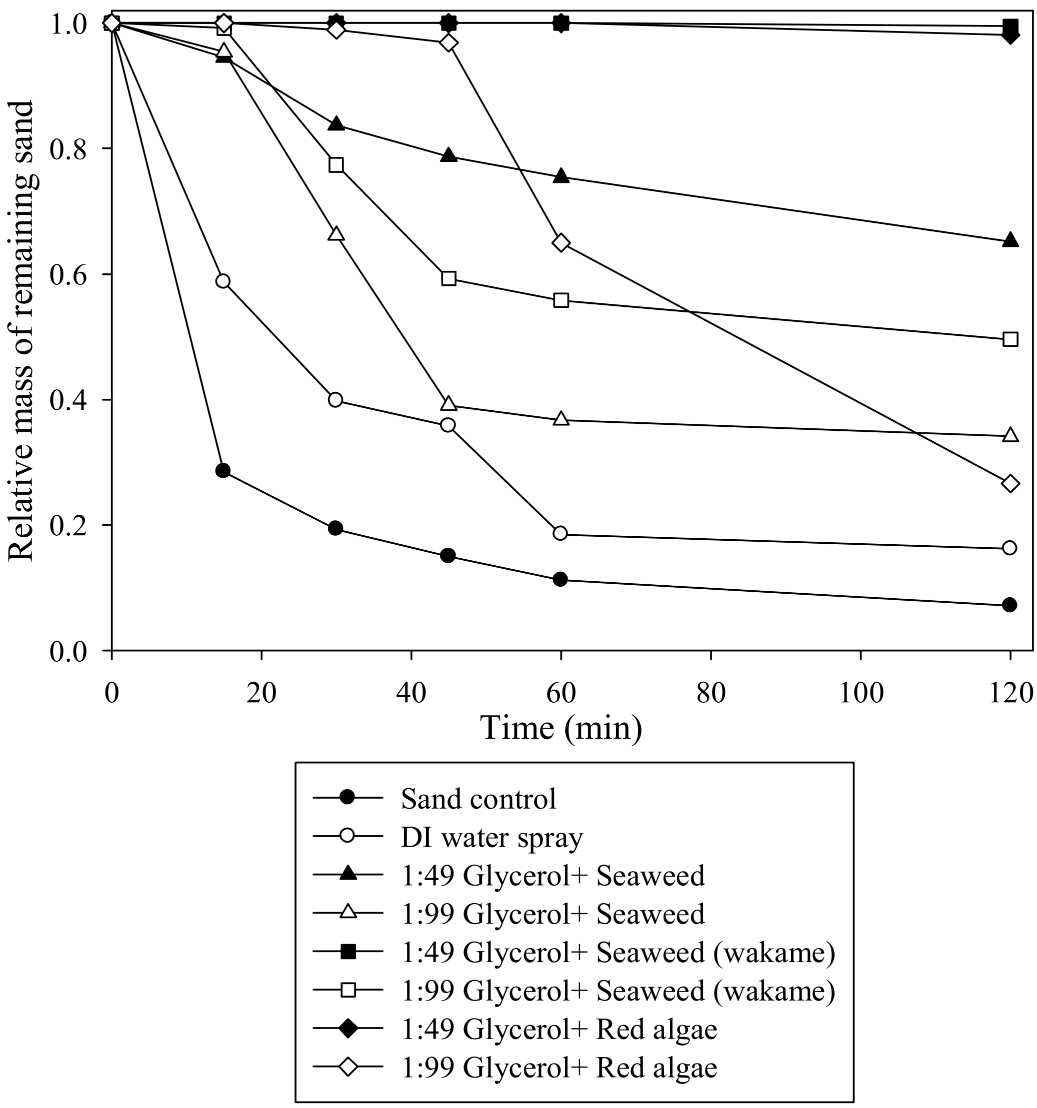

Fig. 6. Relative remaining sand mass when mixtures of glycerol and seaweed, glycerol with seaweed (wakame), and glycerol with red algae used.

3.3 Wind tunnel test results of long-lasting effect experiments

To determine the effect of drying time (between suppressor application and the wind tunnel test) on suppression ability, the drying time was increased from 3 days to 5, 7, and 10 days for the mixtures of glycerol plus palm oil, seaweed mixture, and wakame. As the drying time was extended, the ability to suppress the dust was reduced for all mixtures. After 7 days, the suppression ability of the mixture of glycerol plus palm oil was significantly reduced so it is near that of water (Fig. 7). After 10 days, its suppression effect was lower than that of DI water. The suppression ability of the other biomass-based candidates also decreased steadily. However, their suppression ability did not become less than that of water.

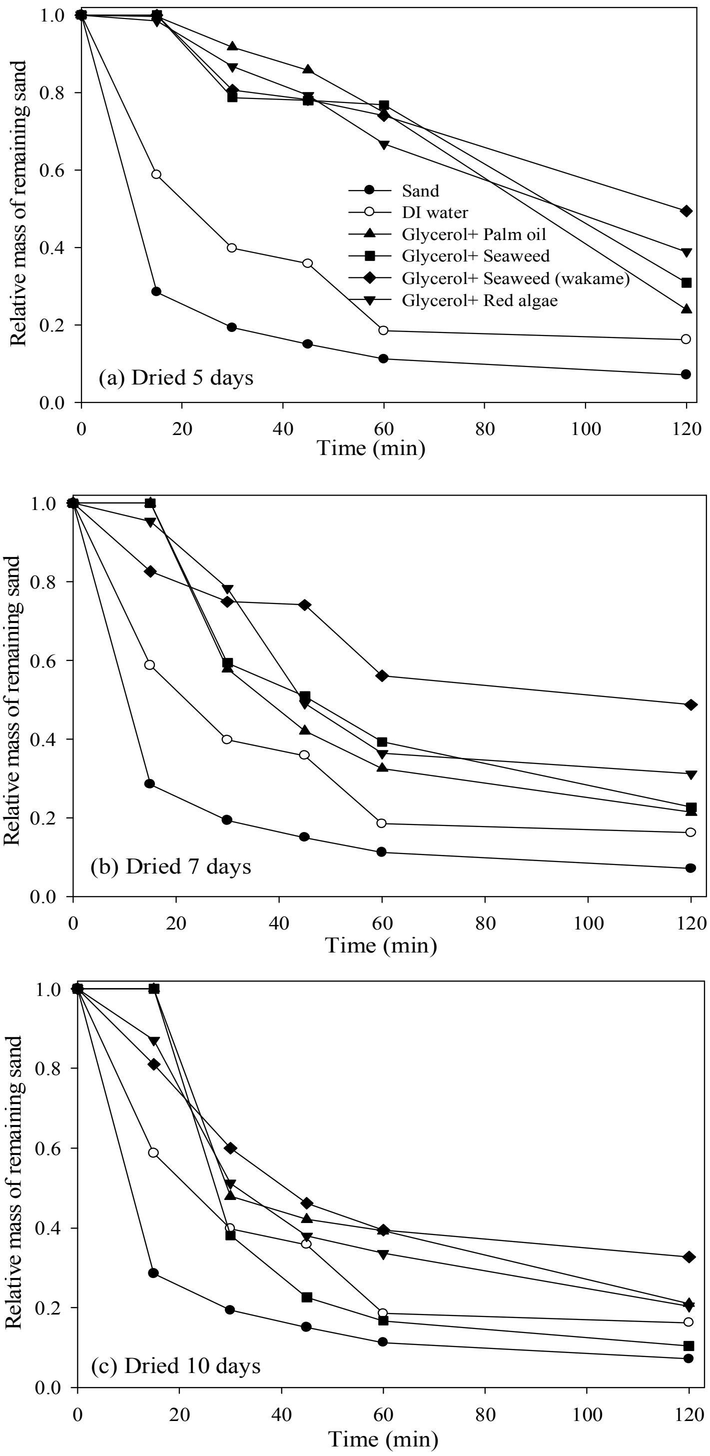

Fig. 7. Relative remaining sand mass when mixtures of glycerol and palm oil, glycerol with seaweed, glycerol with seaweed (wakame), and glycerol with red algae sprayed and dried for (a) 5 days, (b) 7 days, and (c) 10 days.

3.4 Environmental evaluation test results

To evaluate the environmental effects of the prepared dust suppressant samples we performed the BOD_5 and COD_{Mn} tests. The BOD and COD tests were carried out as the water pollution standard test method. The results shown in Table 1. The BOD result of surfactant used for suppressant was 1.97 mg/L. For 100 times diluted biodiesel solutions result was 21.7 mg/L similar with 50 times diluted seaweed, seaweed (wakame) and red algae solution results indicating possible biodegradability. Glycerol added suppressants palm oil, biodiesel and soybean oil results were 17, 21.1 and 0.6 mg/L, respectively. The mixture of glycerol and seaweed, seaweed (wakame) and red algae results were 34.2, 35.6 and 24.9 mg/L respectively. As expected marine-biomass derived materials BOD results were higher than biodiesel by-products. Glycerol added suppressant BOD results were slightly lowered due to of glycerol BOD result was relatively lower.

In Table 1. COD_{Mn} test results shown. The biodegradable surfactant used for suppressant (20 g/L) result was 21.2 mg/L. For 100 times, diluted glycerol, palm oil, biodiesel and soybean oil solutions results were 49.0, 36.4, 114.2 and 80.4 mg/L, respectively. For 50 times diluted seaweed, seaweed (wakame) and red algae solutions results were 33.0, 27.4 and 70.0 mg/L, respectively. Mixtures of glycerol and palm oil, glycerol with biodiesel and glycerol with soybean oil results were 48.8, 41.4 and 45.6 mg/L, respectively and mixtures of glycerol and seaweed, glycerol with seaweed (wakame) and glycerol with red algae results were 68.0, 43.4 and 58.6 mg/L respectively.

Although BOD_5 is not the ultimate result and COD_{Mn} is not oxidize all organic compounds with 100% efficiency, from the BOD/COD ratio we can assume biodegradability of the suppressants. The BOD/COD ratio shown in Table 1. As expected marine-biomass derived materials such as seaweed (wakame) as well as mixture of glycerol and seaweed (wakame) biodegradability highest. BOD/COD ratio indicate that adding glycerol was slightly lowering biodegradability of the solutions except interestingly for seaweed (wakame) and red algae. The seaweed (wakame) and red algae solutions BOD/COD ratios were increasing due to adding crude glycerol, from 0.78 to 0.82 and 0.36 to 0.43, respectively.

Table 1. Biodegradability of suppressants used in this study

Suppressants		$BOD5_{avg}$ (mg/L)	COD_{avg} (mg/L)	BOD/COD_{Mn} ratio
Surfactant	20 g/L	1.97± 0.03	21.2± 0.03	0.09
Glycerol	100 times diluted	6.78± 0.51	49.0± 1.0	0.14
Palm oil		18.72± 0.75	36.4± 0.4	0.51
Biodiesel		21.69± 0.78	114.2± 3.8	0.19
Soybean oil		10.74± 0.24	80.4± 1.6	0.13
Seaweed	50 times diluted	18.69± 0.12	33.0± 1.0	0.57
Seaweed (wakame)		21.48± 1.71	27.4± 0.6	0.78
Red algae		25.17± 0.06	70.0± 2.8	0.36
Glycerol+ Palm oil	100 times diluted	16.98± 2.07	48.8± 0.8	0.35
Glycerol+ Biodiesel		21.07± 0.94	41.4± 0.6	0.51
Glycerol+ Soybean oil		0.62± 0.29	45.6± 0.4	0.01
Glycerol+ Seaweed	50 times diluted	32.40± 0.03	68.0± 1.6	0.48
Glycerol+ Seaweed (wakame)		35.64± 0.63	43.4± 0.6	0.82
Glycerol+ Red algae		25.20± 0.33	58.6± 0.4	0.43

4. CONCLUSIONS

This study was conducted to investigate the following biomass-based polymeric materials as dust suppressants: crude glycerol, biodiesel, palm oil, soybean oil, a seaweed mixture, wakame, and red algae. Using crude glycerol as a base material, potential biodegradable suppressants were prepared and tested. All of the polymeric materials had better suppression ability than DI water. Under the given conditions, the 100 times-diluted mixture of glycerol plus palm oil showed a loss of sand mass of 36% in 2 h. The 50 times-diluted mixture of glycerol plus the seaweed mixture showed a loss of sand mass of 34.8% in 2 h. The 50 times-diluted mixtures of glycerol plus wakame and glycerol plus red algae showed the highest suppression ability, with 0.5% and 1.9% loss, respectively. According to the BOD5, values, the biodegradability of the suppressants using materials derived from marine biomass was higher than that of the suppressants synthesized with oil materials or byproducts from biodiesel production processes. Both the glycerol plus wakame and the glycerol plus red algae suppressants showed good suppression ability and biodegradability. Our results suggest that the mixtures of glycerol plus either wakame or red algae may be good biodegradable suppressants for fugitive dusts in urbanized and industrial areas.

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