

The preliminary study of oil removal using lemon peel waste

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Abstract

While the discovery of oil contributes a lot towards a country's economy and technological development, it is also the cause for oil pollution. As such, this study proposes to use lemon peel waste as a low-cost adsorbent to manage oil pollution. For the untreated adsorbent, the lemon peels were cut into small pieces and dried under sunlight for 48 hours. Then, it was further dried in an oven for 24 hours and ground into powder. For the treated adsorbent, the lemon peels were soaked in 0.5 M of sodium hydroxide (NaOH) solution. The adsorbent was used to adsorb different types of oil (diesel oil, lubricant oil, waste vegetable oil) and in different types of water (ocean water, lake water, tap water) with different amounts of adsorbent which is 0.2 g, 0.4 g, 0.6 g, 0.8 g, and 1.0 g for adsorbent dosage experiment. While for types of water experiment, a ratio for volume of water and oil of 3:1, and constant mass adsorbent was used. The result showed that untreated adsorbent can adsorb higher amount of oils than treated adsorbent. The oil that could be easily adsorbed using lemon peels adsorbent is diesel oil with 89.91% adsorption. For the types of water, the result changes according to different types of water and oil used. It was found that the higher the mass adsorbent, the lower the percentage of oil removal. The highest percentage of diesel oil removed in ocean water is 81.68%. While the removal of lubricant oil and waste vegetable oil in lake water is 66.6% and 72.13%, respectively. Scanning Electron Microscopy (SEM) shows that treated lemon peels had small pores compared to untreated lemon peel waste. This study demonstrated and proposed that the lemon peel waste has a good potential in low-cost oil waste removal.

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1.0 Introduction

Oil is the world's most important fuel and underpins our high standard of living. It provides modern convenience and freedom of movement and is crucial to the functioning of the modern transport system. The uses of oil as an energy resource is very popular, and it is already being commercialized around the world. According to the frontline, world's oil consumption will increase from 76 million barrels per day in the year 2000 to 119 million barrels per day in the year 2020 (Dutta et al., 2011). However, if oil is explored, transported, stored, and used too widely, chances of oil spillage are high hence, posing severe problem and damage to the environment. The oil spilled is not only messy but is also hazardous and will eventually threaten our lives (Tembhurkar et al., 2012).

Oil is already known as a hydrophobic element which is immiscible in water because it has a lower density than water. An oil spill is the release of a liquid petroleum hydrocarbon into the environment, especially the marine ecosystem, due to human activity, and is a form of pollution (Abdullah et al., 2016). Usually the problems are marine oil spills, where oil is released into the ocean. Otherwise, sometimes these oil spills can occur in the land. Spilled oil can also contaminate drinking water supplies. It can cause waterborne and diarrheal diseases to human. Therefore, it is important to remove oil spillage to maintain the environment and ecosystem.

Various methods for removing oil has been introduced including gravity separation, dissolved air flotation, chemical coagulation, filtration, membrane process, and biological process. All these technologies

are very expensive (Sathya et al., 2019). Hence, a simple experiment of adsorbent has been explored using fruit peel for oil removal purpose. Unwanted citrus fruit peel could be re-purposed into ultra-light sponges that can clean up oil spills, while also reducing food waste. Citrus is one of the most important fruit crops in the world. Production of citrus fruits has increased enormously in the last few decades, going from an average of 62 million tons a year in the period of 1987–1989 to about 100 million tons in the year 2010 (Köse & Bayraktar, 2018). Citrus fruits belong to six genera (*Fortunella*, *Eremocitrus*, *Clymenda*, *Poncirus*, *Microcitrus*, and *Citrus*), which are native to the tropical and sub-tropical regions of Asia, but the major commercial fruits belong to genus *Citrus*. The genus citrus includes several important fruits such as oranges, mandarins, lime, lemons, and grapefruits (Giwa et al., 2018). Lemon peel (LP) is one of the valuable agriculture biomass wastes, principally consisting of cellulose, hemicellulose, lignin and pectin substances because of the huge consumption of lemon throughout the world, a large amount of wet solid waste is produced.

Oil removal using lemon peel can be achieved using extraction, chemical coagulation and ultra-filtration, however, it incurs high cost. The adsorption method in removing oil spills have been considered a better way of controlling pollution and the natural adsorbents are preferred to be used. Adsorption is the process by which a solid adsorbent can attract a component from aqueous phase to its surface.

A simple tool adsorbent approach which is from fruit peel was used in removing oil. The use of natural organic adsorbents was widely used because of their greater adsorption capacities, eco-friendliness and cost effectiveness. Natural adsorbents are not only biodegradable when disposed, but more efficient than chemical adsorbent as they showed a greater adsorption capacity (Abdullah et al., 2016; Abdullah et al., 2019; Azlin et al., 2019).

In this study, there are two methods that has been proposed. One is adsorbent dosage which is the use of adsorption method between lemon peel adsorbent and oil, and the other one is the use of different types of water with the lemon peel adsorbent and the different types of oil. At the end of the experiment, the percentage of oil removal and adsorption capacity of oil in the treated and untreated adsorbent is recorded. Hence, the objective of this study is to produce a low-cost adsorbent from lemon peels waste for oil removal.

2.0 Methodology

2.1 Material

The chemicals used were 0.5 M of sodium hydroxide (NaOH) and 65% of nitric acid (HNO₃). The NaOH was used to treat the lemon peels while HNO₃ act as a neutraliser after the peels were treated by NaOH. Distilled water was used in the preparation of NaOH solution and cleaning of the lemon peels. Four types of waste oils were used for the experiment namely diesel, petrol, lubricant oil and used vegetable oil.

2.2 Methods

The absorbent was prepared in two conditions known as treated and untreated conditions. For untreated adsorbent, the lemon peel was collected then rinsed with tap water and dried under the sunlight for 48 hours to remove some of the moisture. Next, it was put in the oven for one hour at 70 °C to remove the residual moisture in the peels (Abdullah et al., 2016; Abdullah et al., 2019; Azlin et al., 2019). After that, it was ground by using into powder. Lastly, it was sieved using a machine to get the particle size of 250 µm before it is used for the testing of the removal of oil.

Similar preparation was done for the treated lemon peels adsorbent with few additional steps. After drying under sunlight, it was impregnated with 0.5 M NaOH solution for 48 hours. The peels were then rinsed with distilled water and dried again under the sunlight for 3 days. The dried lemon peels were soaked in a beaker containing 1000 ml of 0.5 M HNO₃ for about 30 minutes. This was done to neutralise the peels surface which could stop the NaOH reaction (Tembhurkar et al., 2012). Afterwards, the peels were rinsed with distilled water and extra pure water. Then, the peels were dried under the sunlight for 2 days, prior to drying inside the oven for 4 hours at 60°C to eliminate all moisture. It is then ground into powder and sieved to achieve 250 µm of particles in size.

For the effect of adsorbent dosage, the samples are labelled as S1–S15 for untreated adsorbent and S1T–S15T for treated adsorbent, according to its process conditions as shown in Table 1. The specified amount of untreated adsorbent was packed in a tea bag and placed inside a beaker. Following procedures by Lazim et al. (2015), a volume of 20 ml of diesel oil was poured into the beaker and left at room temperature for 25 minutes to allow the adsorption of oil.

Table 1. Samples notation according to process parameters for untreated lemon peel adsorbent

Sample	Mass of adsorbent (g)	Types of oil	Sample	Mass of adsorbent (g)	Types of oil
<i>Untreated Adsorbent</i>			<i>Treated Adsorbent</i>		
S1	0.2	Diesel	S1T	0.2	Diesel
S2	0.4	Diesel	S2T	0.4	Diesel
S3	0.6	Diesel	S3T	0.6	Diesel
S4	0.8	Diesel	S4T	0.8	Diesel
S5	1.0	Diesel	S5T	1.0	Diesel
S6	0.2	Lubricant	S6T	0.2	Lubricant
S7	0.4	Lubricant	S7T	0.4	Lubricant
S8	0.6	Lubricant	S8T	0.6	Lubricant
S9	0.8	Lubricant	S9T	0.8	Lubricant
S10	1.0	Lubricant	S10T	1.0	Lubricant
S11	0.2	Waste vegetable oil	S11T	0.2	Waste vegetable oil
S12	0.4	Waste vegetable oil	S12T	0.4	Waste vegetable oil
S13	0.6	Waste vegetable oil	S13T	0.6	Waste vegetable oil
S14	0.8	Waste vegetable oil	S14T	0.8	Waste vegetable oil
S15	1.0	Waste vegetable oil	S15T	1.0	Waste vegetable oil

The final mass of the adsorbent in tea bag was recorded. Next, the percentage of oil removal and the adsorption capacity were calculated using Eq. (1) adapted from Abdullah et al. (2016), Abdullah et al. (2019) and Azlin et al. (2019).

$$\% \text{ oil removal} = \left(\frac{W_o - W_i}{W_o} \right) \times 100\% \quad (1)$$

where: W_i is initial mass of adsorbent (g) and W_o is final mass of adsorbent (g)

These procedures were repeated for all samples with, all the samples were collected and further tested for Scanning Electron Microscopy (SEM) analysis.

The oil removal and adsorption capacity were also studied for different types of water which are lake water, tap water and ocean water. For this study, the adsorption of diesel, lubricant and waste vegetable oil were tested, while the ratio of water and oil, and the adsorbent mass were kept constant at 3:1 and 0.5 g, respectively.

3.0 Results and discussion

3.1 Effect of adsorbent dosage on percentage of oil removal

Fig. 1–3 show the effect of dosage on removal of diesel, lubricant and waste vegetable oil, respectively,

using untreated and treated lemon peels adsorbent. Overall, it can be seen that increasing mass adsorbent dosage resulted in lower percentage of oil removal. The adsorption capacity of all adsorbents decreased by increasing the adsorbent dosage.

Previous studies have shown that percentage of oil removal is directly proportional to adsorbent dosage (Tembhurkar et al., 2012). The greater the dosage of the adsorbent, the more effective the adsorption of oil because more surface area is available for the adsorption to occur.

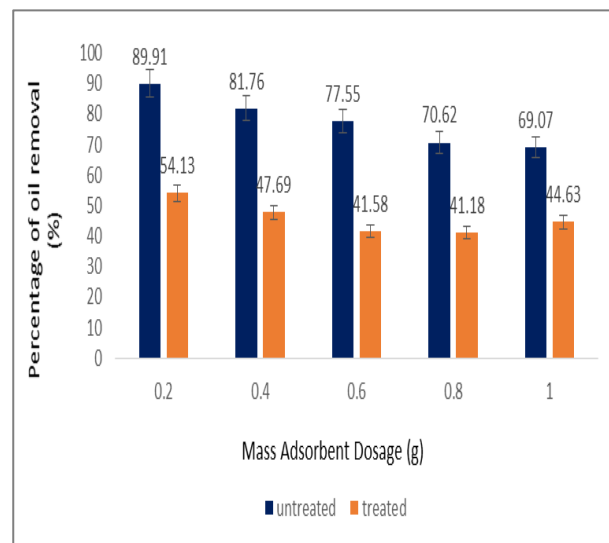


Fig. 1: Effect of adsorbent dosage on diesel oil removal using untreated and treated lemon peels adsorbent

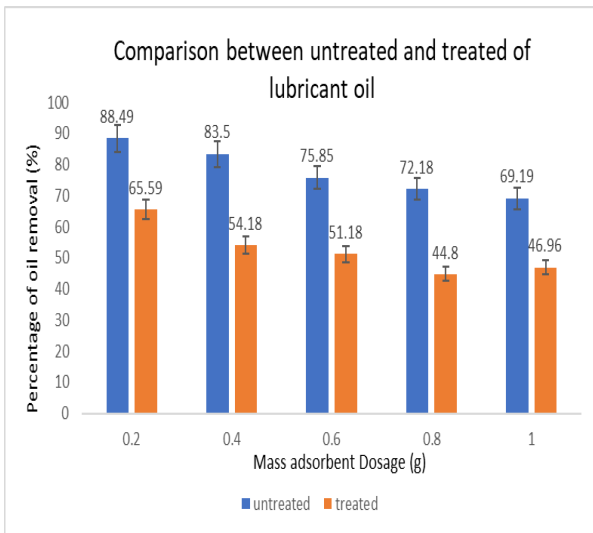


Fig. 2: Effect of adsorbent dosage on lubricant oil removal using untreated and treated lemon peels adsorbent

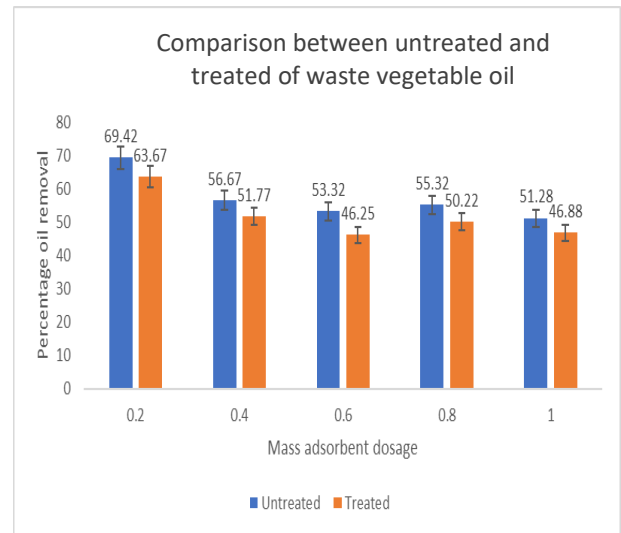


Fig. 3: Effect of adsorbent dosage on waste vegetable oil removal using untreated and treated lemon peels adsorbent

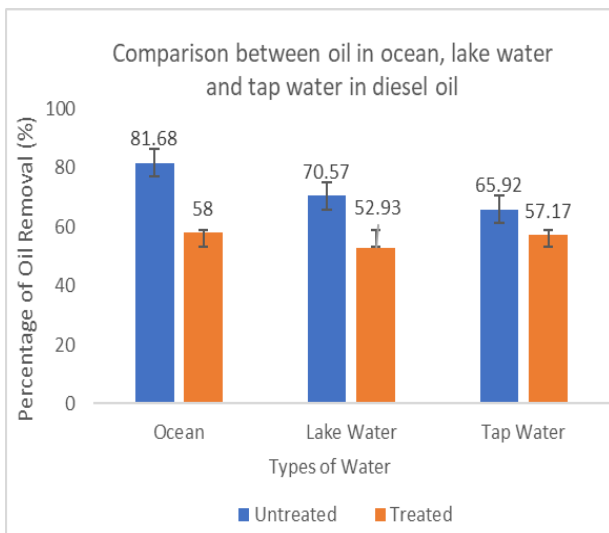


Fig. 4: Effect of types of water on diesel oil removal using untreated and treated lemon peels adsorbent

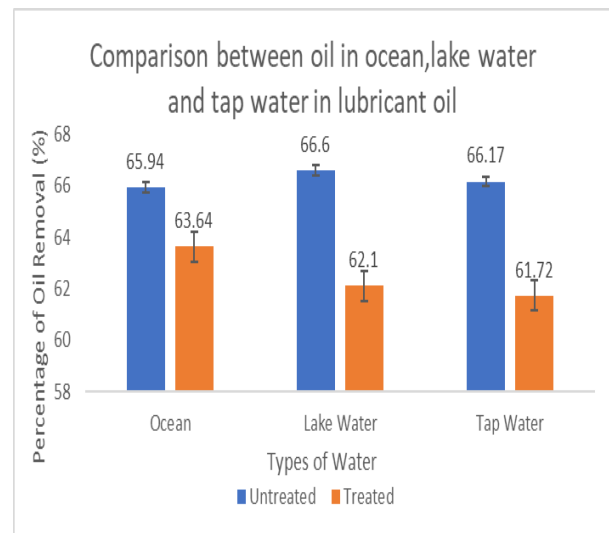


Fig. 5: Effect of types of water on lubricant oil removal using untreated and treated lemon peels adsorbent

Meanwhile, the percentage of oil removal using untreated adsorbent is higher than the treated adsorbent. From Fig. 1, the percentage of diesel oil removal for samples S1 and S1T are 89.91% and 54.13%, respectively. Samples S5 also shows higher percentage of oil removal (69.07%) than the treated adsorbent of sample S5T (44.63%).

From Fig. 2, it was seen that sample S6 was capable of removing 88.49% lubricant oil compared to the treated adsorbent of sample S6T (65.59%). Meanwhile, the percentage of oil removal for sample S10 and sample S10T decreased to 69.19% and 46.96%, respectively. This depicts that the higher the mass, the lower the percentage of oil removal. Moreover, from Fig.3, sample S11 of 0.2 g untreated adsorbent was able to remove 69.42% of oil, while sample S11T removed 63.67% of oil. Similar to the trend of diesel and

lubricant oil removal, the percentage of oil removal for sample S15 and sample S15T decreased to 51.28% and 46.88%, respectively.

This is because untreated adsorbent has larger and more surface area than the treated, thus adsorbs more oil and resulted in higher percentage of oil removal (Chaidir et al., 2015; Zhou et al., 2012). When the adsorbent is treated with acid and alkali, the particles in the adsorbent collide with each other, causing less surface area of the adsorbent and results to a less adsorption of the oil (Wan Ibrahim et al., 2013).

3.2 Effect of adsorbent dosage in different types of water towards oil removal

In this study, the experiment was carried out to observe the effect of adsorbent dosage on the oil adsorption in

different types of water (ocean, lake water, tap water) by using 0.5 g of untreated and treated lemon peels adsorbent. The volume ratio of water and oil used was 3:1. Fig. 4–6 show the percentage removal of diesel, lubricant and waste vegetable oil, respectively, in different types of water using untreated and treated lemon peels adsorbent.

In ocean water, the adsorbent removes diesel oil as much as 81.68% and 58.00% using untreated and treated adsorbent, respectively. Meanwhile, 70.57% and 52.93% oil were removed from the lake water using untreated and treated adsorbent, respectively. Lastly, diesel oil removal from the tap water shows the lowest percentage (65.92 %) using untreated adsorbent, while 57.17 % oil removal was recorded using the treated adsorbent. The highest percentage of diesel oil removal is in ocean water. The reason might be because of electrostatic interaction and salting out, the oil adsorption of the adsorbent becomes large and more hydrophobic (Wan Ibrahim et al., 2013).

The untreated adsorbent in lake water shows higher percentage of lubricant oil removal (66.6%) but for the treated, decreased to 62.11%. In the ocean water, untreated adsorbent was able to remove 65.94% of lubricant oil while treated adsorbent was able to remove 63.64%. The lubricant oil removal in the tap water using untreated and treated adsorbent was 66.17% and 61.72%, respectively. For lubricant oil, the untreated adsorbent removes more oil than treated adsorbent. Moreover, in this result in lubricant oil, lake water has the higher percentage of oil removal.

For waste vegetable oil, the percentage of oil removal for untreated adsorbent is almost equal to treated adsorbent. In the ocean water, the untreated adsorbent successfully removed as much as 66.94% of

oil while the treated was 60.18%. There was a high percentage of oil removal for untreated adsorbent in the lake water which is 72.12% and this decreased to 60.52% for the treated adsorbent. Meanwhile, in the tap water, treated adsorbent was able to remove 58.79% of oil, while the untreated was able to remove 69.60% of oil.

3.3 Scanning electron microscope (SEM)

The Scanning electron microscopy (SEM) was performed to observe the structure of lemon peels adsorbent. It uses a focused beam of high-energy electrons to generate a variety of signals at the surface of solid specimens. The SEM is routinely used to generate high-resolution images of shapes of objects (SEM) and to show spatial variations in chemical compositions (Wan Ibrahim et al., 2013). Fig. 7 shows the structure of lemon peels adsorbent before and after adsorption. Fig. 7 (a) and (b) are the structures before the adsorption of untreated and treated surface of lemon peels, respectively. The treated adsorbent has small pores and there is less surface area of pores than the untreated adsorbent. The surface area of treated

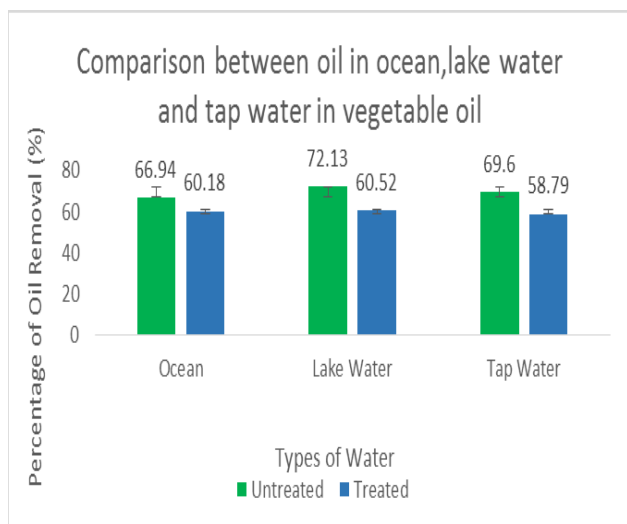


Fig. 6: The comparison types of water between untreated and treated adsorbent in waste vegetable oil.

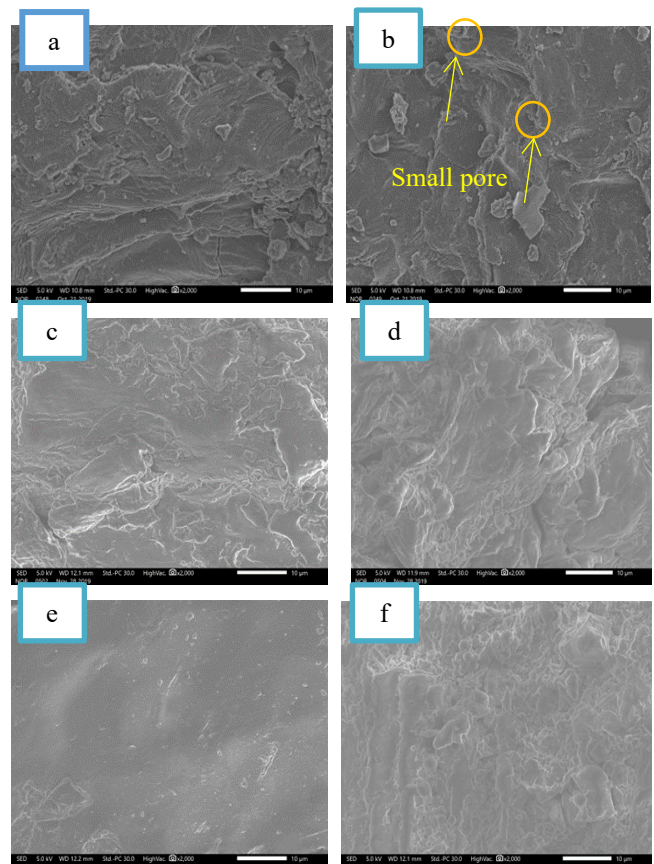


Fig. 7: SEM micrographs of (a) untreated adsorbent (b) treated adsorbent (c) untreated adsorbent after diesel adsorption (d) treated adsorbent after diesel adsorption (e) untreated adsorbent after diesel adsorption in lake water and (f) treated adsorbent after diesel adsorption in lake water.

adsorbent is larger, thus has small pores (Wan Ibrahim et al., 2013). The large pores in untreated adsorbent provide easier oil entrance into the internal parts and assist in the adsorption process. The larger the surface area of pores, the higher the percentage of oil removal.

Furthermore, the structures of lemon peels adsorbent after oil adsorption are shown in Fig. 7 (c), (d), (e) and (f). depicts the fibrous structure with the heterogeneous pores. However, a significant change in the structure of the lemon peels adsorbent is apparent after the oil adsorption (Tembhurkar et al., 2012). The structure appeared to have a more coated surface in Fig. 7 (d) and (f) with the oily substance. This is due to the oil molecules covering the substance adsorption whereas for Figure 7 (d) and (f) which has been treated with acid and alkali, it will easily be coated, and it has a small surface area (Chaidir et al., 2015). Therefore, the smaller the surface area, the lower the percentage of oil removal that is produced and the treated adsorbs less oil than the untreated adsorbent.

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4.0 Conclusions

The main objective of this study to produce a low-cost adsorbent Lemon Peel for cleaning oil spills. This study clearly proves that the process of removing oil using lemon peels waste as an adsorbing medium is efficient at laboratory scale. It can be concluded that the percentage of oil removal is directly proportional to the adsorbent dosage, but in effect to that, it is inversely proportional to the adsorption capacity. The results stated in the adsorbent dosage indicated that the higher the mass of adsorbent, the lower the percentage of oil removal in both untreated and treated adsorbent.

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