

Article

# Removal of Petroleum Hydrocarbons from Brackish Water by Natural and Modified Sorbents

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**Abstract:** Crude oil and petroleum products made from it are increasingly being extracted and consumed worldwide as an important energy source. During necessary transportation, e.g., by tanker, an oil spill might occur, which leads to water pollution by oil. One of the methods of cleaning up oil spills is to use sorbents, preferably made from natural materials. This study evaluates the remediation efficiency of brackish water polluted with crude oil, marine diesel oil (MDO) and lubricating oil. The experiment was performed with three different sorbents (straw, straw modified with methoxytrimethylsilanes (MTMS) and wood chip shavings) and without them. The evaporation loss and the dissolved and sorbed fractions of oil were measured by gas chromatography (GC) to evaluate remediation efficiency. Hydrophobization made the natural sorbents buoyant for the duration of the experiment, with only a slight increase in the maximum sorption capacity. The sorbents increased the evaporation of the oils and also of the water, reduced the proportion of the oil dissolved in water and retained the sorbed proportion for the lubricating oil and partly for the MDO, to such an extent that it could not be extracted entirely even after a 60-min extraction time.

**Keywords:** oil pollution; oil spill clean-up; sorbent; sorbent modification; sorption capacity; MTMS treatment; evaporation rate; THC analysis



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## 1. Introduction

As an essential energy source worldwide, crude oil and its products are increasingly produced and consumed [1]. Crude oil is not found everywhere on earth and has to be transported from its sources to consumers. In the process, transport accidents occur, such as on a tanker ship, which then causes water pollution by oil [2].

Crude oil is a complex mix [3,4], and it is toxic to fauna and flora [5–7]. The main components of crude oil and its products are non-polar water-insoluble hydrocarbons [8]. Since crude oil and its products float on the surface as a separate phase, it permanently acts as a reservoir for the toxic components. The toxicity of oil spills decreases as the concentration of the light fractions decreases. Still, at the same time, the toxicity of the aromatic compounds increases as the relative concentration of these compounds increases.

The horizontal and vertical distribution of oil in water, such as marine or freshwater, is shown in some literature [9–12], but only as a static profile. A temporal distribution is only available for very few studies [13,14], although with the assumption that oil ageing or weathering takes place over time [15,16]. Some overviews with detailed descriptions of the analytical methods are available on the characterization of petroleum hydrocarbon profiles [17–19].

One of the many ways of eliminating oil pollution from water is using different kinds of sorbents [20–22]. At present, a lot of attention is paid to researching natural

biodegradable sorbents, some of which are even recycled from waste [21]. Many different types of sorbents are listed in the literature: corn stalks [23], cotton [24], milkweed floss [25], curaua fibers [26], kapok [27], nettle fiber [28], orange peel waste [29], palm fibers [30], pineapple waste [31], sugarcane bagasse [32], walnut shell media [33], etc. [34,35]. However, the biggest disadvantage of natural sorption materials is that their specificity for oil in water is rather low. Due to their hydrophilicity, they absorb water even after the previous contact with oil and then sink. Hydrophobization considerably increases the specificity of oil, as demonstrated in the literature for various materials [36–39]. MTMS could be used as a modifier, resulting in excellent hydrophobicity and operational stability [40]. Hydrophobicity also leads to an increase in buoyancy, although the specific gravity does not change much: the specific gravity of wood is about  $1500 \text{ kg m}^{-3}$  [41], and that of straw is  $1.1 \text{ g cm}^{-3}$  [42]. The entrapment of air achieves buoyancy through hydrophobization [43].

There are many publications on the effectiveness of sorbents. Still, to the best of the authors' knowledge, none of them describes the evolution of oil distribution on and in water and in the presence of sorbents over a longer period of time. The aim of this study was to evaluate the remediation efficiency of oil-contaminated Baltic Sea water by applying natural sorbents with and without hydrophobization for a longer period of time. To allow comparability with commercial sorbents [2,44], natural sorbents have to be made permanently buoyant by hydrophobization while maintaining high specificity and a sorption capacity for oil.

## 2. Materials and Methods

### 2.1. Chemicals and Materials

For the extraction, n-hexane and n-heptane (Honeywell/Riedel-de-Haën, Cromasolv for HPLC  $\geq 97\%$ , Seelze, Germany), anhydrous sodium sulphate (Sigma-Aldrich, puriss 99.0–100.5%), aluminum oxide (Sigma-Aldrich, puriss) and acetone (Honeywell,  $\geq 99.5\%$ ) were used.

Two natural sorbents were used in this study: oat straw from a farm near Klaipeda (Lithuania) and wood shavings from a wood processing plant in Klaipeda. The straw was chopped into equal-sized pieces with a 1.0 to 1.5 cm length before the experiment.

For hydrophobization of the sorbents, MTMS (98%; Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany) was used.

The water was taken from the Baltic Sea next to Karkle [ $55^{\circ}48'33'' \text{ N}$ ,  $21^{\circ}4'38'' \text{ E}$ ] (Lithuania). The salinity of the Baltic Sea water used for the study was 5.76‰, and the pH was 8.39.

Petroleum products used for the experiment include the following:

- Crude oil (SC "ORLEN Lietuva", Mazeikiai, Lithuania): black-brown colored; measured density ( $T = 20^{\circ}\text{C}$ ) is  $867 \text{ kg m}^{-3}$ ; viscosity ( $T = 25^{\circ}\text{C}$ ) is  $0.0097 \text{ Pa s}$ ;
- Lubricant oil PEMCO iDrive 105 SAE 15w-40 (JSC "SCT Lubricants", Klaipeda, Lithuania): measured density ( $T = 20^{\circ}\text{C}$ ) is  $877 \text{ kg m}^{-3}$  and viscosity ( $T = 25^{\circ}\text{C}$ ) is  $0.0038 \text{ Pa s}$ ;
- Marine diesel oil (MDO) (SC "ORLEN Lietuva", Mazeikiai, Lithuania): measured density ( $T = 20^{\circ}\text{C}$ ) is  $852 \text{ kg m}^{-3}$  and viscosity ( $T = 25^{\circ}\text{C}$ ) is  $0.0024 \text{ Pa s}$ .

For the GC, standard mixtures of n-alkanes (Shimadzu) and n-hexane (Honeywell/Riedel-de-Haën, Cromasolv for HPLC  $\geq 97\%$ ) were used.

### 2.2. Hydrophobisation of the Sorbents and Determining of Their Bulk Densities

1 g MTMS per g sorbent was filled in a 50 mL crystallizing dish, which was put to the bottom of a 3 L beaker. MTMS was chosen because its chemical vapor injection into the porous material is easy to apply. The crystallization dish was covered with a plastic mesh to separate it from the samples. The sorbents were placed on the plastic mesh. The top of the beaker was covered with aluminum foil and placed into an oven at  $70^{\circ}\text{C}$  for 12 h.

The bulk density of the two sorbents with and without MTMS treatment was determined as described in [10].

### 2.3. Determination of Maximum Sorption Capacity

Determination of the maximum sorption capacity was performed according to [20].

Before the experiment, sorbent samples were weighed. Then, samples were immersed in 250 mL of an oil (crude oil/marine diesel oil/lubricating oil) and left for 2 min. Further samples were taken out, drained for 1 min, and weighed again. The maximum sorption capacity of the sorbents was calculated using Formula (1):

$$Q_s = \frac{m_w - m_d}{m_d} \quad (1)$$

$Q_s$  is the sorption capacity, g;  $m_d$  is the weight of sorbent before sorption, g;  $m_w$  is the weight of the sorbent after sorption, g.

### 2.4. Determination of the Evaporation of the Different Oils

The evaporation loss of pure oil was determined in the presence of the sorbent at room temperature (25 °C). To determine the evaporation loss of pure oil, 2 g each of MDO, lubricant oil and crude oil were placed in a Petri dish and weighed after 1, 7 and 14 days. To determine the evaporation loss of oil in the presence of the sorbent materials, 1 g of each sorbent material was added to the Petri dish and weighed again after 1, 7 and 14 days.

The evaporation of oil products was calculated using Formula (2):

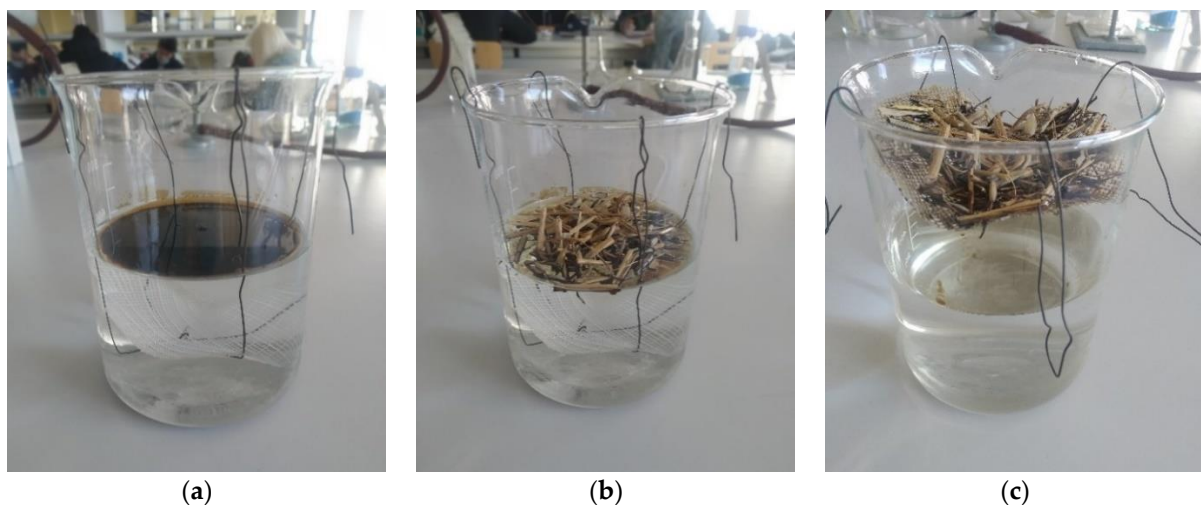
$$E_t = \frac{m_x - m_0}{m_{oil}} \cdot 100\% \quad (2)$$

$E_t$  is the evaporation at the relevant time (t), %;  $m_0$  is the weight of the sample before evaporation in g;  $m_x$  is the weight of the sample after x day, g;  $m_{oil}$  is the weight of added oil product, g.

### 2.5. Determination of the Distribution of Total Petroleum Hydrocarbons in the Water and Sorbent

The total hydrocarbons (THC) content in water and sorbents was measured to investigate the distribution of oils between sorbents and sea water.

A beaker with a total volume of 1000 mL was filled with 500 mL of Baltic Sea water. A plastic mesh was fitted with iron wire, weighed and fixed in a beaker as shown in Figure 1.



**Figure 1.** Analysis of THC in the aquatic system: (a) sea water with crude oil film; (b) sea water with crude oil and sorbent (straw) on it; (c) view of the water draining process after the experiment.

1 mL of oil and 2 g of sorbent were placed on the water surface (Figure 1). After an exposure time of 1, 7 or 14 days, the sorbent was lifted out of the water with the mesh, allowed to drain for 15 min and then weighed. All analyses were carried out with and without under the same conditions with three replicates.

The amount of crude oil/MDO/lubricant oil extracted from the water was determined according to [45] and from the sorbents was determined according to [46].

The water was extracted with 50 mL n-hexane by stirring in a magnetic stirrer at 500 rpm for 30 min and separated in the separating funnel. The mesh was washed with hexane and the wash solution was added to the extracted hydrocarbons from the sorbent. The sorbent was extracted with 100 mL of a mixture of solvents: heptane and acetone in ratio 1:2 in a sealable 250 mL Duran glass bottle extracted on a shaker at 120 rpm for 60 min. The hexane extracts of the water phase and the sorbent were purified separately over a short column of 3 g alumina oxide for water extracts and 6 g for sorbent extracts. Beforehand, they were applied to a filter in a funnel with 10 g of sodium sulphate on the column and dried.

The respective purified phases were transferred to labelled glass vials with rubber crimp caps for GC analysis, diluting by a factor of 20 with n-hexane for sorbent extracts.

GC analysis was carried out on a GC-2010 Plus gas chromatograph, equipped with an automatic liquid sampler and a flame ionization detector (FID) operated at 350 °C. Helium (purity > 99.999%) was used as the carrier gas at a flow rate of 30 mL min<sup>-1</sup>. Chromatographic separation was achieved on a Rxi-1MS 20 m, 0.18 mm ID, 0.18 µm film thickness capillary column, coated with cross bond 100% dimethylpolysiloxane polymer (Restek, Bellefonte, USA). The gas chromatograph oven program was kept at 77 °C for 2.5 min, then was increased at 200 °C/min to 350 °C and held for 5 min. A constant temperature splitless injection technique (at 350 °C) was used for injections of 1 µL. Instrument control and data evaluation were conducted with the LabSolution software.

Three series of calibration standard solutions with concentrations of 0.01, 0.05, 0.1, 0.5 and 1.0 mg mL<sup>-1</sup> were made of crude oil, MDO and lubricating oil. 1.0 mg mL<sup>-1</sup> standard solution was made by accurately weighing 100 mg of oil product and dissolving it in 100 mL of n-hexane. Other standard solutions were made by the dilution method.

The total peak area was measured in the range bounded by the n-decane (C<sub>10</sub>H<sub>22</sub>) and n-tetracontane (C<sub>40</sub>H<sub>82</sub>) standards.

The THC content in the water was calculated as follows [45]:

$$C_w = (C_{ex} \cdot V_{ex} \cdot 1000) / V_w \quad (3)$$

$C_w$  is THC concentration in water sample, mg L<sup>-1</sup>;  $C_{ex}$ —THC concentration in water extract, mg mL<sup>-1</sup>;  $V_{ex}$ —volume of extract, mL;  $V_w$ —volume of water sample, mL.

THC content in sorbent was calculated as follows [46]:

$$C_s = (C_{ex} \cdot V_{ex} \cdot F) / M_s \quad (4)$$

$C_s$  is THC concentration in sorbent sample, g g<sup>-1</sup>;  $C_{ex}$ —THC concentration in extract, mg mL<sup>-1</sup>;  $V_{ex}$ —volume of extract, mL;  $F$ —dilution factor;  $M_s$ —mass of sorbent, mg.

The crude oil/MDO/lubricating oil content of the sorbent was calculated according to [47]:

$$w_h = \rho \cdot \frac{V_h}{m} \cdot \frac{100}{w_s} \quad (5)$$

$w_h$  is the crude oil/MDO/lubricating oil mass of the sorbent, mg kg<sup>-1</sup> dry mass;  $\rho$  is the crude oil/MDO/lubricating oil mass concentration of the extract calculated from the calibration function, mg L<sup>-1</sup>;  $V_h$  is the volume of the n-hexane extract, mL;  $m$  is the mass of the sample taken for analysis, g;  $w_s$  is the dry mass content of the sorbent, % (mass fraction).

The sorption measurements for the crude oil/MDO/lubricating oil were carried out in three replicates for each type of sorbent under the same conditions.

### 3. Results and Discussion

The physical properties of straw are described in [48]: it has a porosity of about 96% and a skeletal density of about  $868 \text{ kg m}^{-3}$ . The maximum sorption capacity is reached after approx. 10 min, whereby approx. 80% of the maximum capacity is reached within less than 2 min. The maximum sorption capacity of straw for distilled water is about  $5.179 \text{ g g}^{-1}$ , whereas for Baltic Sea water it is  $3.766 \text{ g g}^{-1}$  [10].

Since unmodified wood shavings sink in water after a short time, they were not considered further. To make the results comparable in the following, the same conditions should be ensured during the experiments. This includes that the sorbent material floats on the surface during the test period. Since straw also sinks slowly, it was hydrophobized with MTMS.

#### 3.1. Maximum Sorption Capacity of the Sorbents

The maximum sorption capacity of crude oil, marine diesel and lubricating oil was determined for the three selected sorbents—straw, MTMS hydrophobized straw and MTMS hydrophobized wood shavings. The effectiveness of MTMS hydrophobization of the sorbents can be evaluated from the increase in bulk density (Table 1).

**Table 1.** Bulk density of the straw and wood shavings before and after MTMS treatment.

	Straw	Wood Shavings
Without MTMS treatment	$0.032 \text{ g cm}^{-3}$	$0.027 \text{ g cm}^{-3}$
With MTMS treatment	$0.056 \text{ g cm}^{-3}$	$0.030 \text{ g cm}^{-3}$

Since the morphologies of the straw or wood shavings did not appear to change during the treatment, the increase in density must have been due to increased weight due to methylsilanization.

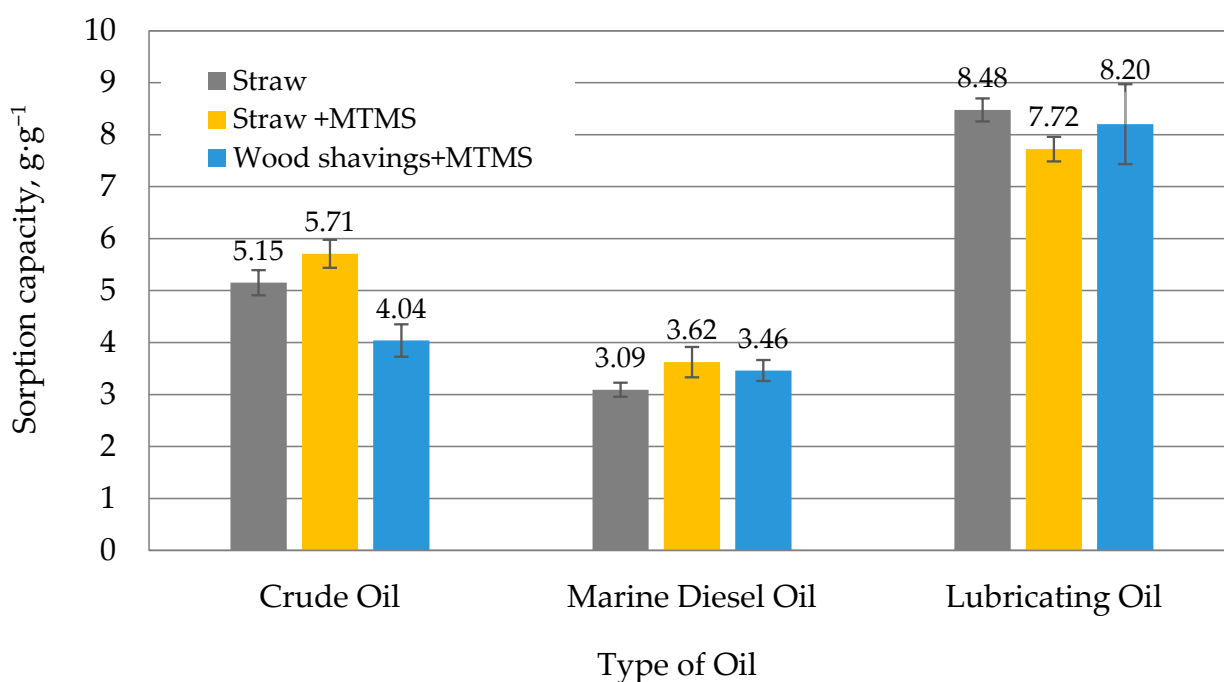
In the crude oil sorption tests, the maximum sorption capacity value ranged from  $4.04 \text{ g g}^{-1}$  using wood shavings with MTMS to  $5.71 \text{ g g}^{-1}$  using straw with MTMS. For marine diesel oil, the maximum sorption capacity ranged from  $3.09 \text{ g g}^{-1}$  when using straw to  $3.62 \text{ g g}^{-1}$  when using straw with MTMS.

These results agree with the results of [10], in which the maximum sorption capacity of straw for crude oil and diesel fuel was determined to be  $5.13 \text{ g g}^{-1}$  and  $3.86 \text{ g g}^{-1}$ , respectively.

The highest values of maximum sorption capacity were observed in the lubricating oil sorption test. The maximum sorption capacity ranged from  $7.72 \text{ g g}^{-1}$  using straw with MTMS to  $8.48 \text{ g g}^{-1}$  using straw (Figure 2).

MTMS treatment of straw caused a slight increase in the sorption capacity of crude oil by 10.9% and of diesel by 17.1%. Conversely, MTMS treatment of straw resulted in an 8.9% decrease in the sorption of lubricating oil. A possible reason for the somewhat lower maximum sorption capacity of the modified straw for lubricating oil could be the additives of the lubricating oil used [49]. Polymethacrylates form interfacial films in oils to surfaces [50]. At the same time, the polyisobutylene binds the lubricating oil [51]. In the case of the modified straw, both additives can penetrate into the pores and at least partially clog them [52–54]. This means that the sorption of the modified straw is reduced during the measurement period.

In terms of maximum sorption capacity, MTMS-hydrophobized straw was better for crude oil sorption ( $5.71 \text{ g g}^{-1}$ ) or diesel sorption ( $3.62 \text{ g g}^{-1}$ ). In the case of lubricating oil, natural straw showed the highest sorption capacity value of  $8.48 \text{ g g}^{-1}$  compared to MTMS-modified straw and wood shavings. The differences in the maximum sorption capacity of straw and wood shavings might be due to the different morphology. However, the maximum sorption capacity values of the MTMS-treated wood shavings do not differ very strongly from the straw with and without MTMS treatment.



**Figure 2.** Comparison of the maximum sorption capacity of straw with and without MTMS treatment and MTMS treated wood shavings for crude oil, MDO and lubricating oil.

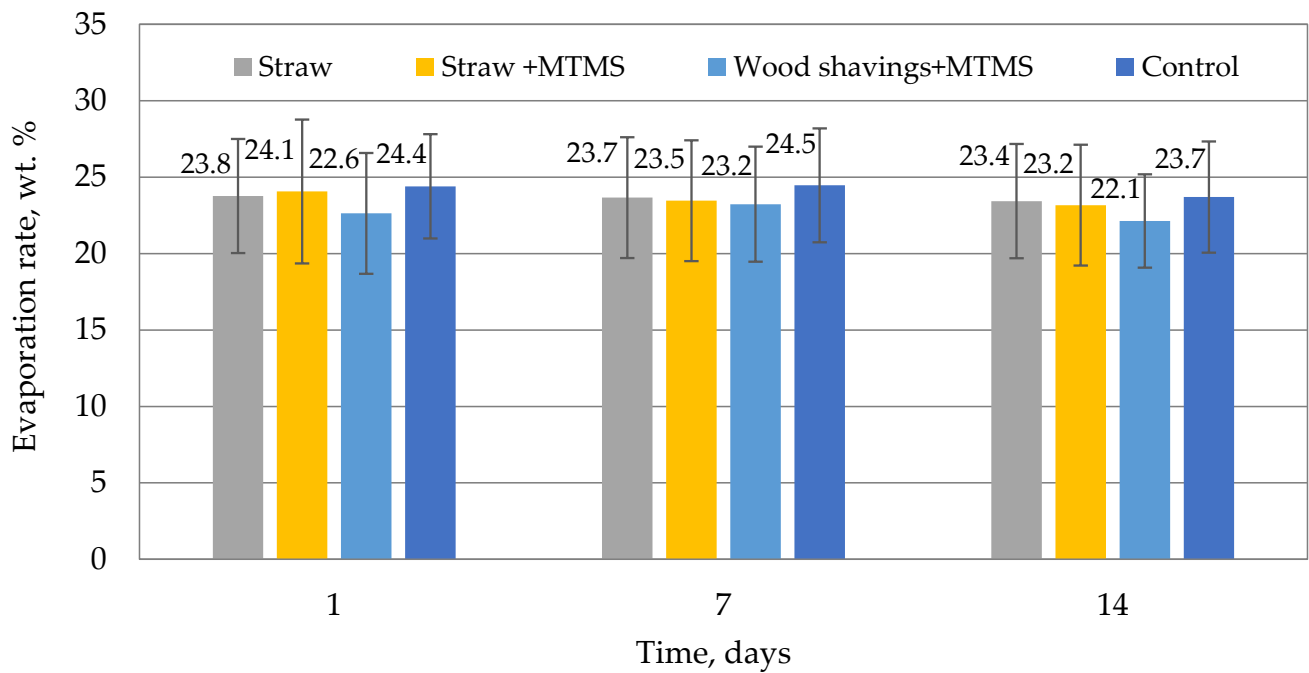
A similar slight increase in sorption capacity using the same procedure as in this study was found by [55]. They studied jute fibers treated with octadecyltrichlorosilane to increase hydrophobicity. The results of the experiment showed that the maximum sorption capacity of untreated jute fibers for crude oil, lubricating oil and MDO reached  $7.33 \text{ g g}^{-1}$ ,  $9.53 \text{ g g}^{-1}$  and  $8.38 \text{ g g}^{-1}$ , respectively. After modification by octadecyltrichlorosilane, the sorption capacity increased slightly by 1.1% and reached  $7.41 \text{ g g}^{-1}$ ,  $10.29 \text{ g g}^{-1}$  and  $8.48 \text{ g g}^{-1}$ , respectively [55]. Although the jute fibers, as seen from the SEM images in [55], have capillaries and have been hydrophobized, they hardly sorb oil. This could be explained in relation to the relatively short time period (that matches with the time scale of non-naturally grown sorbents such as PP nonwovens or aerogels) for determining the maximum sorption capacity of a few minutes by the effect described in [43], when the air is held in such structures. Then no fluid can enter, especially if its viscosity is increased. This effect would also explain the significantly increased buoyancy on the water surface. Natural sorbents first sink partially in water because of their density and through wetting with water, and, after water penetration, they finally sink entirely. Hydrophobization completely avoids this behavior. The hydrophobization also seems to prevent the subsequent penetration of water when the sorbent material has been wetted with oil and allows the investigation of the long-term effect of the sorbent material as a function of ageing oil. When oil enters marine waters, it is exposed to a number of factors that cause it to undergo various changes. These include mechanical movement (dispersion, advection), turbulent mixing (dispersion, diffusion, sedimentation) and physico-chemical factors (evaporation, photo-oxidation, dissolution, absorption, biodegradation, etc.) [15].

### 3.2. Evaporation Rates of Oils from Surface of Sorbents

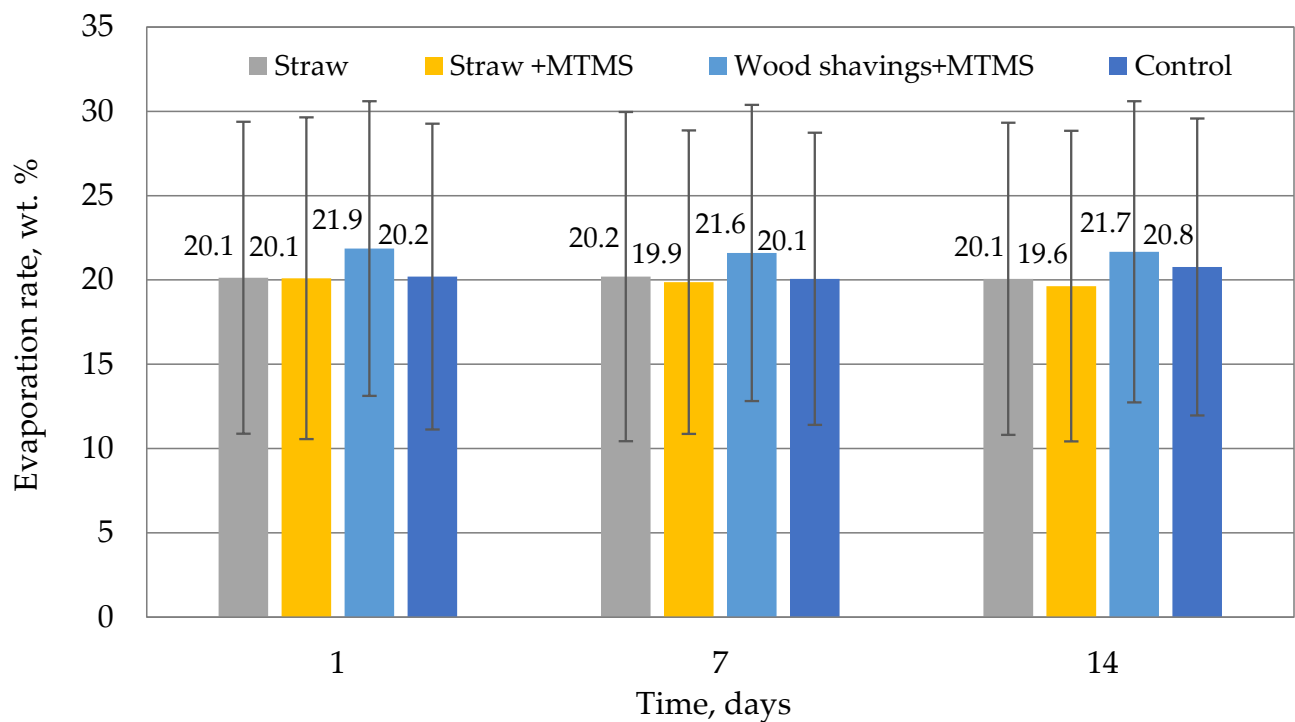
Evaporation is a very important process of weathering oil spills. In a few days following a spill, the volume of light crude oils can be reduced by up to 75%, medium crude oils—by up to 40% and heavy or residual oils can only lose about 5% [56].

In our experiment, it was important to investigate the oil-retention capacity of sorbents and oil products' evaporation rates from the surface of sorbents.

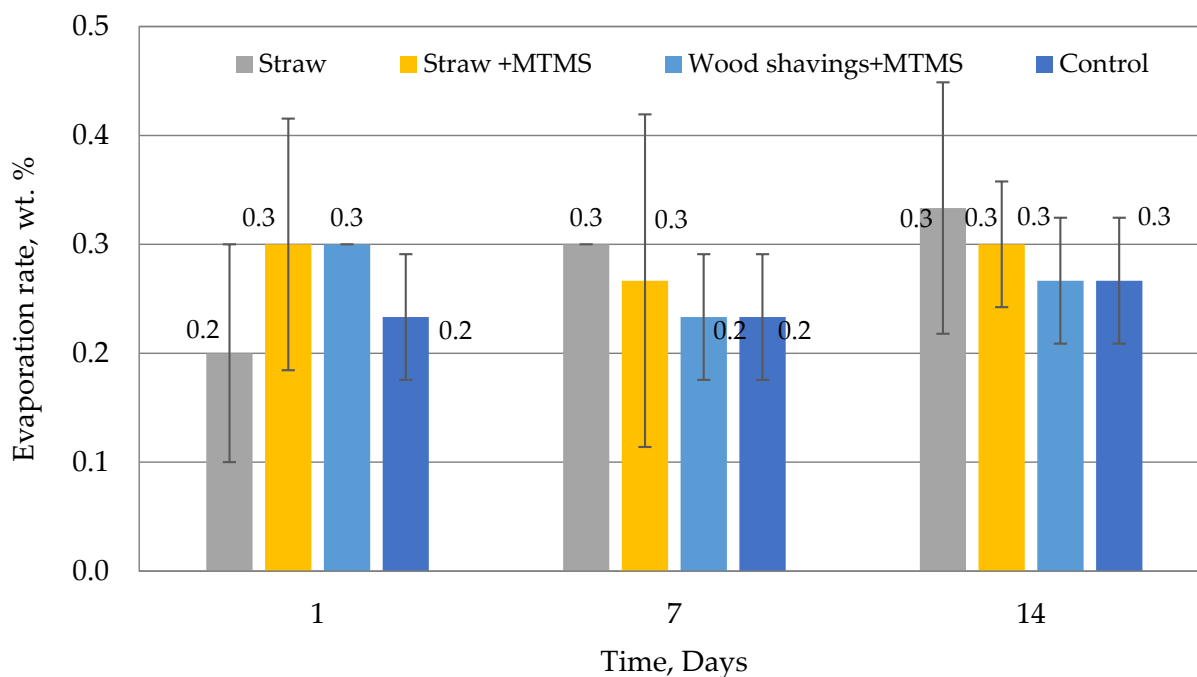
The dynamic oil evaporation of straw, straw + MTMS and wood shavings + MTMS are shown in Figures 3–5.



**Figure 3.** Evaporation rates of crude oil from the surface of the sorbents. Evaporation rate of crude oil on Baltic Sea water after 1, 7 and 14 days in the presence of straw with and without MTMS treatment and MTMS treated wood shavings compared with control samples.



**Figure 4.** Evaporation rates of MDO from the surface of sorbents. Evaporation rate of marine diesel oil on Baltic Sea water after 1, 7 and 14 days in the presence of straw with and without MTMS treatment and MTMS treated wood shavings compared with control samples.



**Figure 5.** Evaporation rates of lubricating oil from the surface of the sorbents. Evaporation rate of lubricating oil on Baltic Sea water after 1, 7 and 14 days in the presence of straw with and without MTMS treatment and MTMS treated wood shavings compared with control samples.

The amount of evaporated crude oil from the surface of sorbents was similar in 1, 7 and 14 days and varied from 22% for wood shavings to 23% for straw. It was similar to control samples, where 24% of crude oil evaporated (Figure 3).

The amount of evaporated MDO was similar and varied from 19.6% for the straw to 21.9% for wood shavings in 1, 7 and 14 days (Figure 4). It was similar to control samples, where 20.4% of the MDO evaporated. The total amount of evaporated oils was similar, being on average 23.4% for crude oil and 20.7% for MDO.

Evaporation of lubricating oil varied between 0.2 and 0.3% in 1, 7 and 14 days and did not depend on sorbents. This was similar in control samples, where 0.3% of lubricating oil evaporated (Figure 5).

Overall, the total amount of evaporated oil products from day 1 to 14 hardly depended on the type of sorbent. Furthermore, in all three cases, the evaporable components of the oils seem to have already evaporated within the first day, because the proportion of evaporated oil was always the same as on day 1, even on days 7 and 14. The differences in the evaporation of the various oils can probably be explained by their different compositions. Crude oil is a multicomponent mixture and therefore contains easily evaporable substances even under ambient conditions [18]. According to [56], within a few days of an oil spill, due to evaporation, the volume of light crude oils can be reduced by up to 75%, medium crude oils by up to 40% and heavy or residual oils by only about 5%. The vapor pressure of crude oil depends on its composition and varies between 5 and 40 kPa at 15 °C. However, MDO is also a mixture of various distillates from crude oil; depending on the specification, it still contains proportions of substances with a measurable vapor pressure at ambient conditions. The vapor pressure of diesel oils is lower than that of crude oil and ranges from 0.05 to 10 kPa. The vapor pressure of lubricating oil is even lower and does not exceed 0.0003 kPa [57].

These results are consistent with other studies that showed that, depending on the composition of the spill, about 50% of the petroleum hydrocarbons can vaporize within the first 12 h after the oil spill [16].

### Evaporation of Water

During the study, it was observed that water evaporated from the beakers containing sorbent. No water evaporated from the beakers, which contained only water and oil (no sorbent). Since this experimental setup is a simulation of an oil spill on the Baltic Sea, and the amount of evaporated water is relatively small, water loss was not quantified.

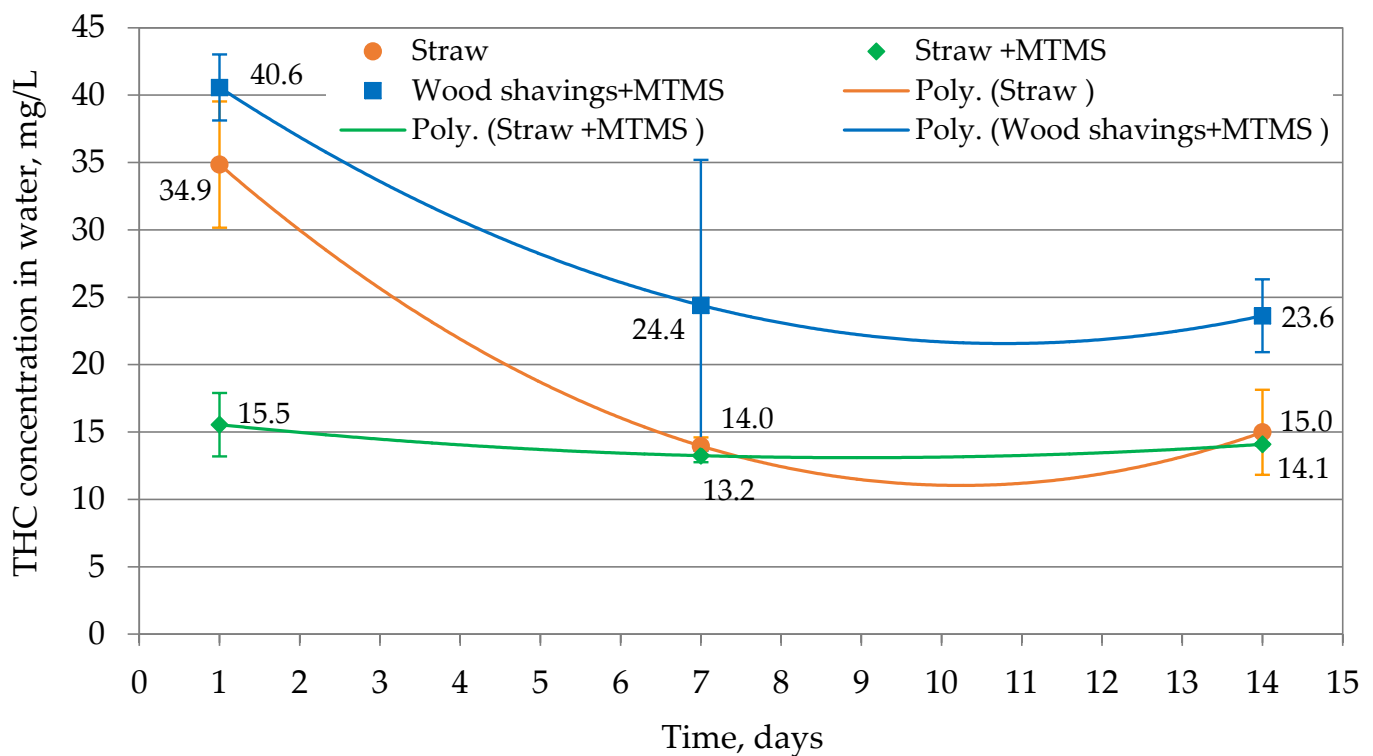
### 3.3. Oil Retention Capacity of Sorbents

A further part of our experiment in selecting suitable natural sorbents for oil spill clean-up was to evaluate the oil-retention capacity of sorbents based on the dynamic allocation of oil products between sorbents and sea water within 14 days.

#### 3.3.1. Crude Oil Dissolved in Water

Several investigations after the Exxon Valdez oil spill reported that approximately 20–30% of the spilled oil dispersed or dissolved into the upper water column in weeks after the spill [58]. Therefore, THC in the water was also determined in this study.

In the case of simulating crude oil spill clean-up, the THC concentration in the sea water decreased during 14 days from 40.6 to 23.6 mg L<sup>-1</sup> using wood shavings + MTMS; from 34.9 to 15.0 mg L<sup>-1</sup> using straw and from 15.5 to 14.1 mg L<sup>-1</sup> using straw + MTMS sorbents (Figure 6).



**Figure 6.** Dynamic of dissolving crude oil in sea water. THC concentration of crude oil in water in the presence of the sorbents: straw with and without MTMS treatment and MTMS-treated wood shavings after 1, 7 and 14 days.

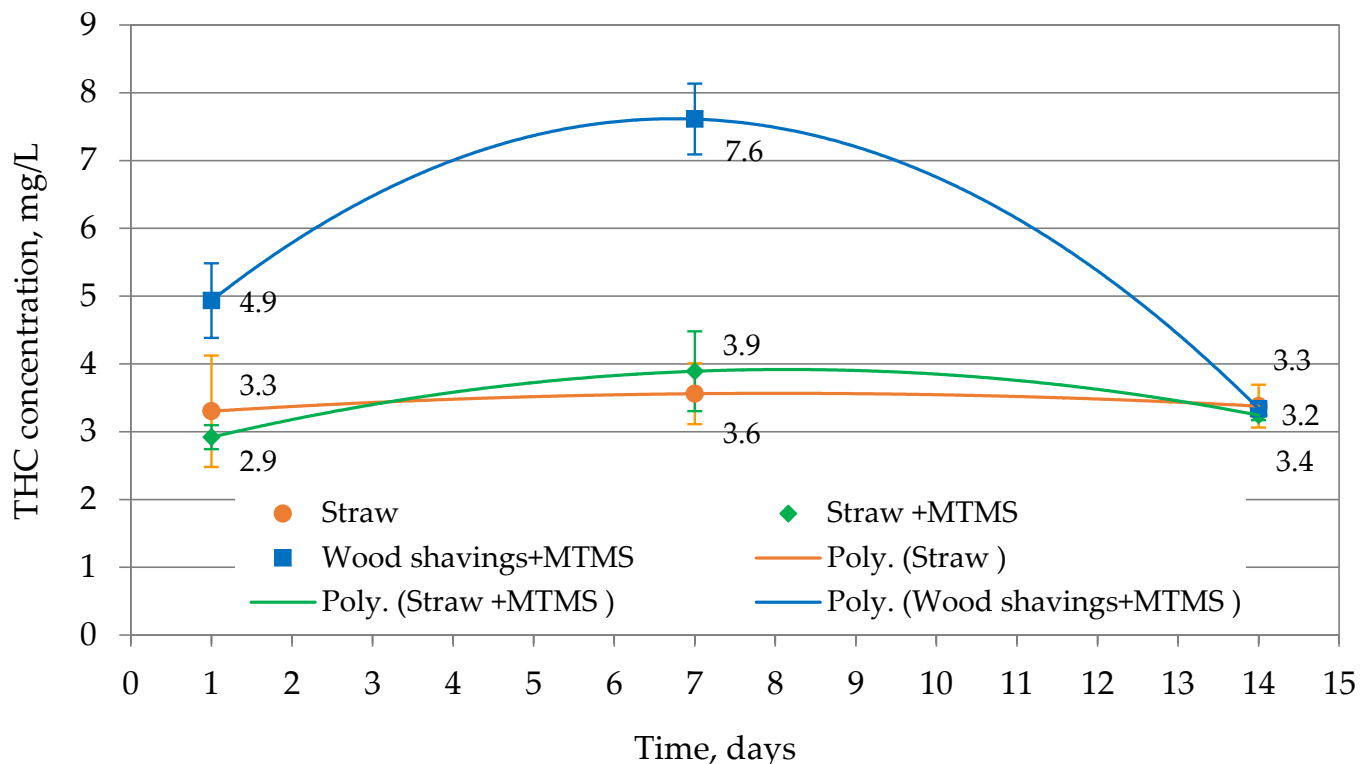
The maximum amount of THC in sea water (40.6 mg L<sup>-1</sup>) was determined for wood shavings + MTMS on the first day of the experiment. The minimum amount of THC in the sea water—13.2 mg L<sup>-1</sup> was determined for straw + MTMS on seventh day of the experiment. The water-soluble fraction of crude oil varied from 3.1% to 9.6% of spilled crude oil amount using wood shavings + MTMS and straw + MTMS as sorbents, respectively.

Our results are in line with research where it was noted that approximately 0.7–10% of the spilled crude oil dispersed into the water column within the first few days following the Hebei Spirit oil spill [59].

### 3.3.2. Marine Diesel Oil Dissolved in Water

Concerning simulating MDO spill clean-up, the THC concentration in the sea water increased on the 7th day and decreased on the 14th day of the experiment for all types of used sorbents.

THC concentration varied from 3.3 to 3.6 mg L<sup>-1</sup> using straw; from 2.9 to 3.9 mg L<sup>-1</sup> using straw + MTMS and from 7.6 to 3.3 mg L<sup>-1</sup> using wood shavings + MTMS (Figure 7).



**Figure 7.** Dynamic of dissolving marine diesel oil in sea water. THC concentration of marine diesel oil in water in the presence of the sorbents: straw with and without MTMS treatment and MTMS-treated wood shavings after 1, 7 and 14 days.

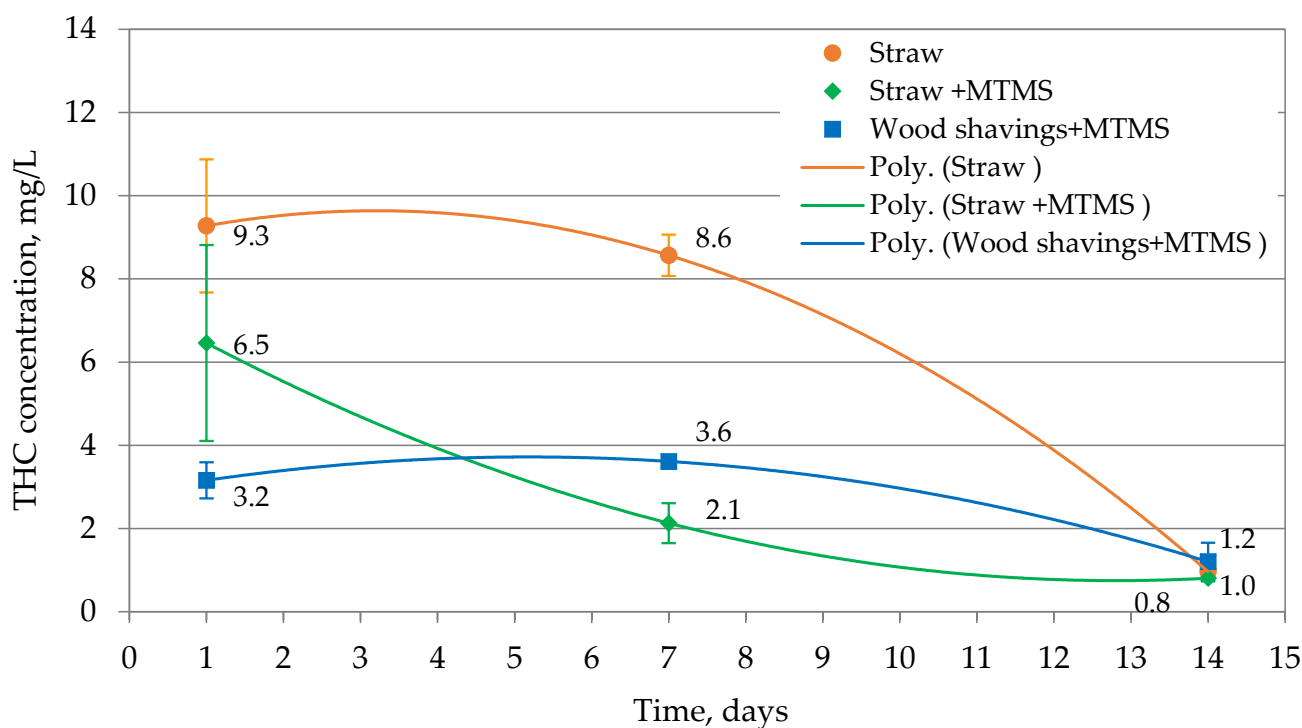
The lowest amount of dissolved MDO in sea water 2.9 mg L<sup>-1</sup> was determined on the first day for the use of straw + MTMS. The maximum amount of dissolved MDO in sea water 7.6 mg L<sup>-1</sup> was observed using wood shavings. The amount of water-soluble fraction of MDO increased by 74.3% and 94.8% using hydrophobized wood shavings in comparison with straw on the 7th day of the experiment.

The highest and lowest content of the water-soluble fraction of diesel consisted of 0.9% and 0.3% of the amount of spilled diesel, respectively, which was ten times lower than crude oil.

### 3.3.3. Lubricating Oil Dissolved in Water

Within simulating the lubricating oil spill clean-up, the THC concentration in the sea water decreased during the studied period for straw and straw + MTMS.

THC concentration decreased from 9.3 to 1.0 mg L<sup>-1</sup> using straw and from 6.5 to 0.8 mg L<sup>-1</sup> using straw + MTMS. In the case of using wood shavings + MTMS sorbent, the THC concentration in the sea water reached the highest value of 3.6 mg L<sup>-1</sup> on 7th day and the lowest value of 1.2 mg L<sup>-1</sup> on the 14th day (Figure 8).



**Figure 8.** Dynamic of dissolving lubricating oil in sea water. THC concentration of lubricating oil in water in the presence of the sorbents: straw with and without MTMS treatment and MTMS-treated wood shavings after 1, 7 and 14 days.

The highest amount of dissolved lubricant in sea water of  $9.3 \text{ mg L}^{-1}$  was determined on the first day of the experiment using a straw, which consisted of 1.1% of the spilled lubricating oil. The lowest content of water-soluble fraction of lubricating oil in sea water of  $0.8 \text{ mg L}^{-1}$  was observed on the fourteenth day using straw + MTMS; it consisted of 0.1% of spilled oil.

During this study, the THC content in the sea water depended on the composition of the oil products. Eshagberi and Ofotokun [60] found that the total petroleum hydrocarbons content in the water-soluble fractions of crude oil is higher at  $14.12 \text{ mg L}^{-1}$  than in diesel at  $6.34 \text{ mg L}^{-1}$ . The higher content of lightweight hydrocarbons in spilled oil produces a higher concentration of THC in sea water, which is in line with a number of spilled oil behavior studies [15].

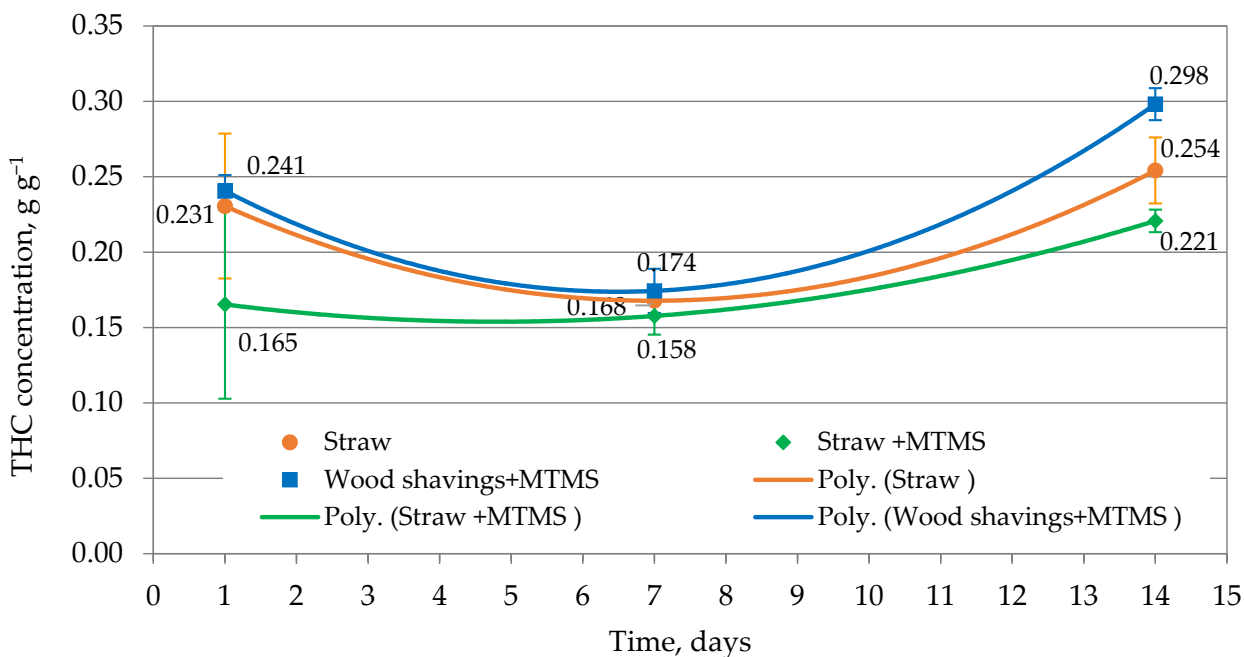
The highest concentration of THC in sea water of  $40.6 \text{ mg L}^{-1}$  was measured for crude oil, the lowest concentration of  $0.8 \text{ mg L}^{-1}$  for lubricating oil.

### 3.4. Crude Oil and Oil Products Retention in Sorbents

#### 3.4.1. Crude Oil Retention in Sorbents

As shown in Figure 9, wood shavings + MTMS were characterized by an insignificantly higher sorption capacity— $0.298 \text{ g g}^{-1}$ , compared with straw— $0.254 \text{ g g}^{-1}$  and straw + MTMS— $0.221 \text{ g g}^{-1}$ . While analyzing the results of the crude oil retention in the sorbents, maximum sorption capacity was observed on the fourteenth day of the experiment for all types of sorbents.

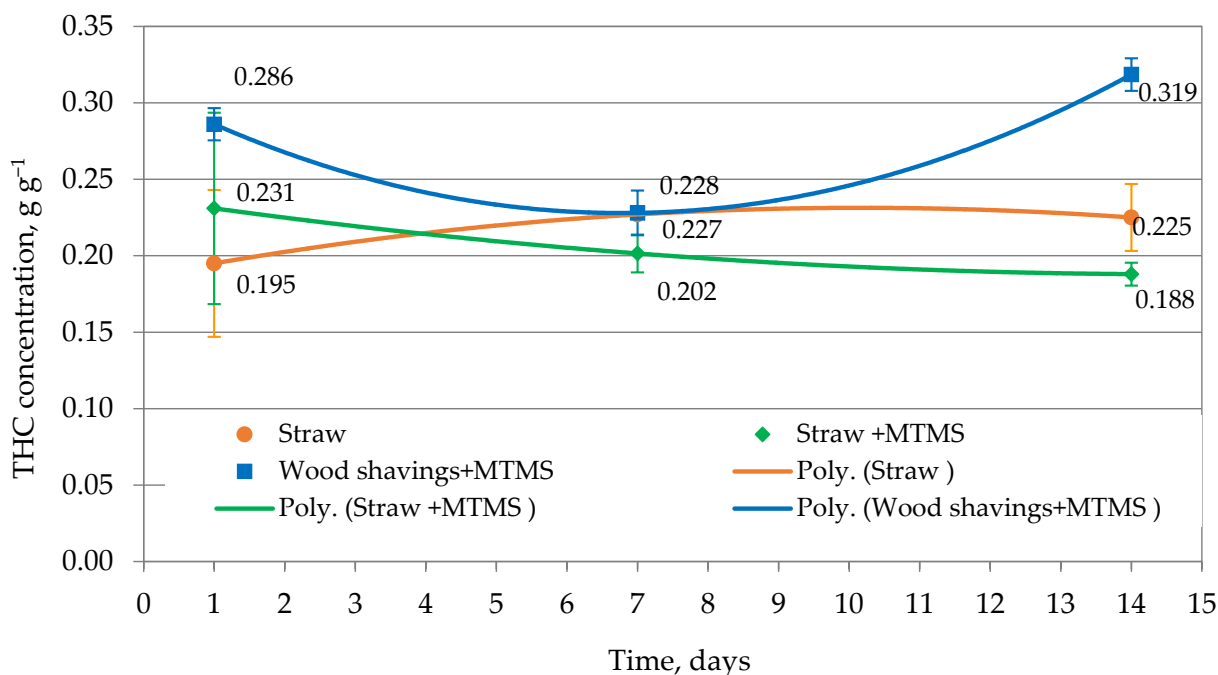
The dynamics of the crude oil retention in all types of sorbents demonstrated increasing sorption capacities from the first to the fourteenth day of the experiment for wood shavings + MTMS, straw and straw + MTMS by 22.1%, 9.9% and 33.9%, respectively.



**Figure 9.** Dynamic of crude oil retention in sorbents. THC concentration of crude oil in the sorbent straw with and without MTMS treatment and MTMS-treated wood shavings after 1, 7 and 14 days.

### 3.4.2. Marine Diesel Oil Retention in Sorbents

During the marine diesel oil sorption capacity investigation, the maximum of 0.319 g g<sup>-1</sup> was observed for wood shavings + MTMS. In contrast, straw + MTMS demonstrated a minimum sorption capacity—0.188 g g<sup>-1</sup> on the fourteenth day of the experiment (Figure 10).



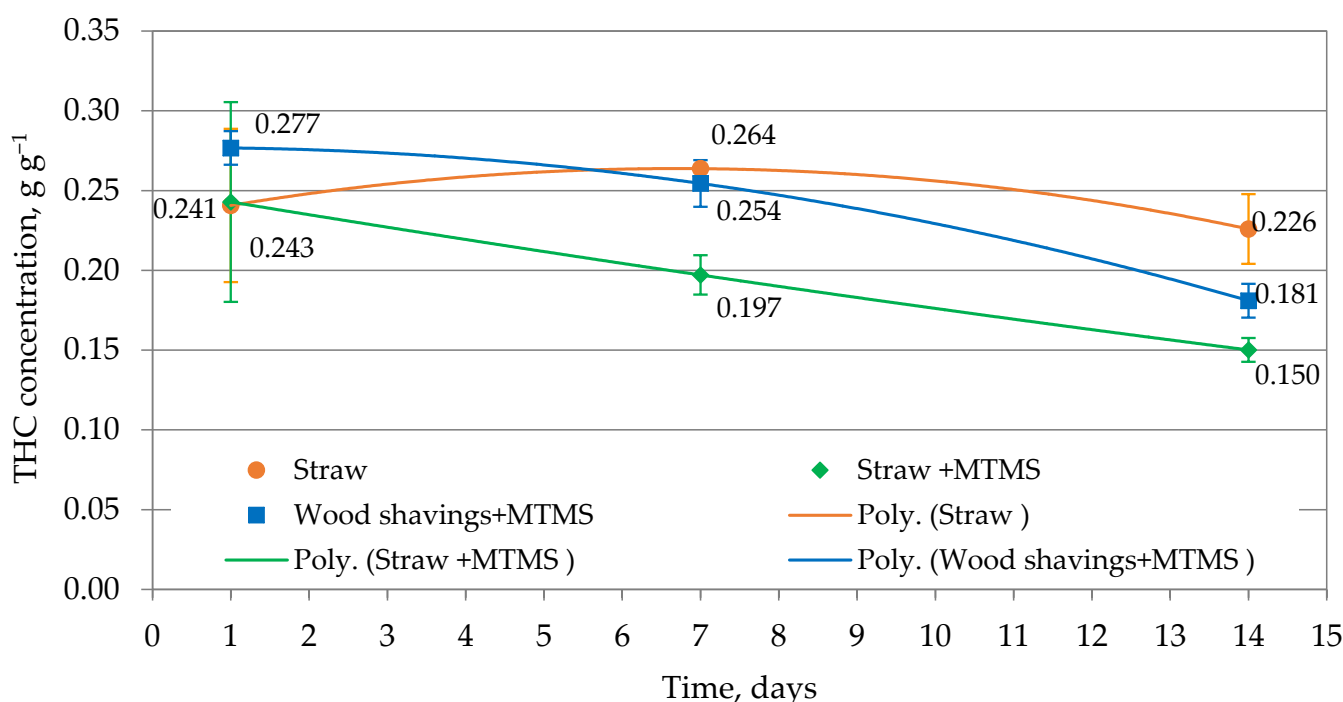
**Figure 10.** Dynamic of MDO retention in sorbents. THC concentration of MDO in the sorbent straw with and without MTMS treatment and MTMS-treated wood shavings after 1, 7 and 14 days.

Concerning the results of the MDO retention in the sorbents, it was noted that the sorption capacities of wood shavings + MTMS increased from the 1st to 14th day by 11.5%.

A possible explanation for the decrease in sorption capacity after 7 days could be the complex internal morphology typical of natural materials. The MDO is absorbed by the wood shavings and is not measurable within the extraction time. However, in the course of the 14 days, the characteristic of wood shavings emerges that it has absorbed moisture to a greater extent and drains the previously sorbed MDO out of the wood shavings again. Presumably, the hydrophobization with MTMS was not so complete that even the innermost structures were modified.

### 3.4.3. Lubricating Oil Retention in Sorbents

During the research on lubricating oil retention in sorbents, it was noted that the sorption capacity of the wood shavings + MTMS and the straw + MTMS sorbents decreased from the first to the fourteenth day with the maximum values of  $0.277 \text{ g g}^{-1}$  and  $0.243 \text{ g g}^{-1}$ , respectively. The maximum sorption capacity of the straw sorbent— $0.264 \text{ g g}^{-1}$  was observed on the seventh day (Figure 11).



**Figure 11.** Dynamic of lubricating oil retention in sorbents. THC concentration of lubricating oil in the sorbent straw with and without MTMS treatment and MTMS-treated wood shavings after 1, 7 and 14 days.

The dynamic of lubricating oil retention in all the types of sorbents demonstrated that sorption capacities decreased from the first to the fourteenth day of experiment for the wood shavings + MTMS, straw and straw + MTMS by 34.6%, 6.2% and 38.3%, respectively.

During the experiment, it was found that straw and wood shavings modified by MTMS demonstrated a sustainable capacity to retain adsorbed oil products within fourteen days in calm and stable conditions.

Hydrophobized wood shavings demonstrated a higher oil retention capacity for low density oil products, such as crude oil and MDO. The crude oil retention capacity of wood shavings + MTMS exceeded the capacity of straw and straw + MTMS by 17.3% and 34.8%, respectively. The MDO retention capacity of wood shavings + MTMS exceeded the capacity of straw and straw + MTMS by 41.8% and 69.7%, respectively.

In the case of lubricating oil, the natural straw showed a higher oil retention capacity than MTMS-modified straw and wood shavings by 50.7% and 24.9%, respectively.

In this study, MTMS treatment of straw not only improved the hydrophobicity of the sorbent, but even resulted in a decrease in the extractable amount of lubricating oil from the sorbent, which is shown as a decrease in sorption capacity by day fourteen of 13.0%, 16.4%, and 33.6% in the case of crude oil, MDO, and lubricating oil, respectively, in Figure 11. A possible explanation, similar to that described in Section 3.1, could be that the additives of the lubricating oil used may have clogged the pores of the hydrophobized straw during the measurement period and thus hindered extraction of the lubricating oil. This is also supported by the observation that the lubricating oil retention in the non-hydrophobized straw was about the same during the entire measurement period.

### 3.5. Balance of the Recovery of the Different Types of Oil

If the individual yields of the extractions of the individual experiments on evaporation, oil concentration in the water and oil content in the sorbent for an oil and the respective test durations are added, the values listed in the following Tables 2–4 are obtained.

**Table 2.** Recovery of different kinds of oil concerning the different sorbents on day 1.

	Straw	Straw + MTMS	Wood Shavings + MTMS
Crude oil	71.7 ± 13.5%	57.9 ± 17.5%	72.8 ± 6.1%
MDO	59.3 ± 9.9%	66.4 ± 11.1%	79.3 ± 12.1%
Lubricant oil	48.5 ± 7.2%	49.2 ± 6.0%	55.9 ± 4.0%

**Table 3.** Recovery of different kinds of oil concerning the different sorbents on day 7.

	Straw	Straw + MTMS	Wood Shavings + MTMS
Crude oil	58.0 ± 4.4%	55.8 ± 6.5%	59.2 ± 7.3%
MDO	65.8 ± 14.8%	60.5 ± 12.2%	67.6 ± 9.4%
Lubricant oil	53.5 ± 0.4%	39.8 ± 3.0%	51.2 ± 2.0%

**Table 4.** Recovery of different kinds of oil concerning the different sorbents on day 14.

	Straw	Straw + MTMS	Wood Shavings + MTMS
Crude oil	75.0 ± 8.3%	68.1 ± 5.4%	82.9 ± 5.4%
MDO	65.3 ± 16.9%	57.4 ± 15.2%	85.7 ± 10.7%
Lubricant oil	45.6 ± 7.2%	30.3 ± 7.6%	36.6 ± 10.8%

To estimate the influence of the sorbents, the following Table 5 shows the recoveries of the respective oils on the water without sorbents in % based on the amount of oil used.

**Table 5.** Recovery of different kinds of oil from water surface and body after the experiment time periods day 1, 7 and 14.

	Day 1	Day 7	Day 14
Crude oil	61.9 ± 10.2%	59.3 ± 0.8%	68.4 ± 19.6%
MDO	76.9 ± 9.2%	70.1 ± 10.4%	65.4 ± 2.6%
Lubricant oil	83.8 ± 29.6%	51.4 ± 7.6%	62.0 ± 12.7%

The values in Table 5 show the process-related losses due to extraction and subsequent cleaning.

As can be seen from the comparison of the recoveries for day 1 in Table 5 with Table 2, the sorbents have a large influence on the recovery value of the various oils, especially in the case of lubricating oil. In the case of crude oil, the sorbents on day 1 even seem to increase the recovery somewhat. In the case of MDO, this is only the case for wood shavings treated with MTMS, whereas for straw-based sorbents the recovery is reduced.

On days 7 (Table 3) and 14 (Table 4), the sorbents had a similar effect on the recovery amount of the different oils, with the recovery amount decreasing with increasing test

duration, especially for lubricating oil. This could be due to the lubricating oil being adsorbed by the natural sorbents in the deep, internal pores. If this is the case, the extraction time of 60 min is probably not sufficient.

Finally, it can be stated that the recovery of crude oil and MDO is on average 70%, and for lubricating oil is on average 50%. These values are still within an acceptable range as reported by [18].

#### 4. Conclusions

Hydrophobization with MTMS improved the buoyancy of both straw and wood shavings, enabling a long-term study over 14 days on the sorption behavior of crude oil, MDO and lubricating oil in water. This was not done before for sorption materials other than commercially available ones. The increase of sorption capacity for crude oil and MDO in engineered sorbents by hydrophobization with MTMS cannot be transferred to natural sorbents in this study. MTMS treatment of straw resulted in a moderate reduction of the sorption capacity of crude oil by 11%, diesel by 17% and lubricating oil by 9%. In terms of maximum sorption capacity, straw hydrophobized with MTMS was better for the sorption of crude oil and diesel. However, an unexpectedly high sorption capacity was determined for lubricating oil, with about  $8 \text{ g g}^{-1}$  for straw and wood shavings.

The evaporation rates of crude oil and MDO are not affected by the used sorbents.

Both natural straw and MTMS-modified straw and wood shavings can hold the adsorbed oil products in water under calm and stable conditions within 14 days. However, the extractable oil amounts for lubricating oil and partially for MDO are lower than the values for the comparative samples of water and oil layer without sorbent. It can be concluded that the extraction time of 60 min, which is common in the literature, was probably too short in this study for the latter oils. Presumably, the oils fully penetrated the network of empty cell walls during the course of the experiment and would now require a similarly long time to flow out or wash out for extraction.

It is intended that the sorbents investigated in this study will be used as carrier materials for oil-degrading microbes.

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