

## Removal of Aliphatic Hydrocarbons from Gas Oil Contaminated Clay Soil via Soil Vapor Extraction

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### Abstract

In this research, the performance of soil vapor extraction (SVE) for the removal of aliphatic hydrocarbons from gas oil contaminated soil and the effect of soil type on this method is studied. To explore the effect of soil type in the removal of hydrocarbons, SVE tests were conducted on 3 types of soil: 1) fine sand, 2) fine sand with 20% of kaolinite clay and 3) fine sand with 40% of kaolinite clay. Three extraction periods of 8, 16 and 24 hours were used. The results have shown that the efficiency of this method exceeds 78% in the removal of all hydrocarbons from fine sand, while an increase in clay significantly decreases the efficiency. Accordingly, the efficiency of this method decreases by 53.1% and 54.6% in fine sand with 20% of clay and fine sand with 40% of clay respectively. Furthermore, the results show that the higher concentration of some heavy and medium hydrocarbon leads to the more increase in early hours of SVE process, which indicates the alteration of hydrocarbons into each other.

*Keywords:* Soil Vapor Extraction; Fine Sand; Clay; Aliphatic Hydrocarbons; Gas Oil.

### 1. Introduction

In recent years, human activities, especially in developing countries, increase the amount of environmental pollutants. This situation will be crucial as the contaminants penetrate through the soil and give rise to groundwater contamination [1, 2]. Polluting the ecosystem affects the cycle of nature and will have negative feedbacks for humans, animals, plants, and structures [3]. One kind of ecosystem pollution is polluting the soil. Examples of soil pollutants are oil products and their derivatives which can cause pollution during transport or nonstandard storage [4]. Volatile Organic Compounds, petroleum hydrocarbons containing Benzene, Toluene, Ethylbenzene, and Xylene, are among the contaminants which are a threat to the porous media, soil. Considering the type of soil, type of contaminant, location, soil physical and chemical characteristics and operating conditions are necessary for eliminating soil contamination [1].

Vapor extraction from soil is one method of soil remediation and by using it the concentration of the volatile substances in oil products which are absorbed by the non-saturated parts of the sand, can be reduced [5, 6]. Over the past 35 years, one of the conventional technologies used in the United States is Soil Vapor Extraction (SVE) [7]. Statistics show a 26% of the soil remediation, implementing SVE in the US. Soil Vapor Extraction is an efficient way of contaminant extraction from the soil in the vadose zones [8]. Using vacuum blowers, creating airflow, inducing the air flow through the soil matrix and collecting the extracted contaminated soil vapor are the basis of this soil remediation technology [9].

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The negative pressure caused by the vacuum leads to evaporation and separation of particles from the sand and the pollutants state changes from liquid to gas. Extraction pits remove all these vapors. The removed vapors are either released into the atmosphere after being filtered or injected into the soil again [9, 10].

The effectiveness of this technology in reducing the concentration of organic components (VOCs) and some semi-active organic components has been proven [11]. The benefits of SVE method compared to other methods are, (1) accessibility of Equipment (2) easy set-up (3) minimal interference with the site's other Functions (4) short filtering time approximately 6 months to 2 years (5) easily compatible and integrated with other remediation technology such as air inject and finally (6) cost efficiency. Perusing the literature, it can be found that the followings can affect the efficiency of the remediation process and time: operating conditions (e.g. soil temperature, airflow rate), contaminants' characteristics (e.g. vapor pressure and solubility) and soil characteristics (e.g. soil type, aggregation and water content, natural organic matter percentage) porosity and permeability, the pressure of polluting vapors, volatility of vapors, rate of passing air and the temperature of the systems.

The spread and arrangement of soil particles which affect its permeability are very influential in the extraction of vapors [12]. The more permeable a soil, the easier air passes through it and the higher the efficiency of the SVE method [13, 14]. For example, if the clay proportion is higher in the soil, the rate of removing will be reduced [15]. The kind of soil also affects the treatment rate for some reasons. Firstly the capacity for superficial absorption in different soils is different. In small particle soils, the surface of the soil is more significant compared to large particle soils making the removal of contamination more difficult. The permeability of different soils also differs. A soil with high permeability enables air to pass through nearly all the possible routes in the particular soil structure, evaporating pollutants. In small particle soils (soils with low permeability) air can only pass through a limited amount of routes which is due to its low permeability [16, 17]. In a comparison between parameters which affect the efficiency of SVE, the kind soil proved more important than the kind of pollutant [18].

Numerous studies have been carried out to analyze the effective factors on the efficiency of vapor extraction. Examples of these factors for the SVE method are the kind of soil and the temperature of the system. The effect of soil temperature is to increase the vapor pressure of pollutant leading to an increase in the contaminant extraction using SVE method. In a study, it was observed that an increase of 20 degrees centigrade in temperature caused a 3-fold increase in the contaminant mobility [19].

The effect of airflow rate is to influence the transfer of contaminant dispersed mass to the gas phase, desorption of the contaminants from soil and its dissolution from the aqueous phase. In another study, it was concluded that in sandy soil with a water content of (0-4)%, contaminated with BTEX, the equilibrium in the mass transfer was achieved blowing the airflow rate of  $1.9 \text{ cm}^3\text{s}^{-1}$  and the effect of slow diffusion was not seen. This led to higher soil remediation compared to those blown with higher airflow rates ( $19 \text{ cm}^3\text{s}^{-1}$ ) [17].

Decreasing the availability of contaminants caused by dissolving in aqueous phase tendency and accordingly lowering the efficiency of soil vapor extraction is the influence of the presence of contaminants with high water solubility [20].

Reduction in the air-filled pores of the porous media, barrier action between the soil matrix and pollutants and dissolving the contaminants are the influence of soil water content (SWC). Reduction in soil porosity is observed in soils with high SWC, leading to difficult air movement through the soil porous media and consequently affecting the efficiency of SVE process. On the other hand, due to the competition between the contaminants and soil for adsorption, a positive impact of SWC occurs. Therefore, desorption of the contaminants and increasing the contaminants' mobility occur as a result of this competition and eventually increase the efficiency of SVE [17, 21].

Using laboratory experiment data after the screening, Sabour et al., developed a model for determining the amount of contaminant removal rate based on the different temperature and available data by reasonable measures. They validate the model with the help of an artificial neural network using response surface methodology. The correlation coefficient square was equal to 0.95 in the validation section by the neural network. In the original model, the coefficient was equal to 0.99. Besides, the contaminant removal formula for sandy soils was presented. The results show that the model can be utilized for prediction of removal rate of thermal enhancement in the relevant situations with the slight error [22].

Huang et al. presented a new analytical solution for the equations that describe radial gas phase transport of a contaminant to a well-used for extraction. This equations model rate-limited mass transfer, dispersion and advection of dissolved, separate phase, and sorbed contaminants into the gas phase. Using Laplace transform concerning time the model equations are analytically solved. The results show that the effect of gas phase molecular diffusion upon concentrations at the extraction well is relatively small, although the effect upon the distribution of concentrations in space is significant. This model can be useful for the design of SVE remediation strategies [23].

Yang conducted a laboratory research and used TMVOC numerical model for the BTEX migration occurred during an SVE process in low temperature. The simulation results were consistent with the empirical data obtained, and the three conclusions were obtained. (i) The removal rate of SVE from benzene, toluene, ethylbenzene, and o-xylene was

89.8%, 71.3%, 29.7% and 14.4%, respectively. (ii) In two extraction processes, the benzene and toluene masses in the NAPL gas-water phase are reduced by approximately 20%: 70%: 10%, with the highest reduction in the water phase. During the period between these two extraction processes, benzene and toluene migrated from the NAPL to the gas and sewage phases, and their fractions were approximately 30%: 70%. (iii) It has been proven that in low-temperature conditions,  $-10-5^{\circ}\text{C}$ , the ratio of SVE removal to benzene was higher than that of the four pollutants tested. It is therefore determined that TMVOC can provide scientific guidance for determining whether to optimize or end SVE operations in low-temperature conditions [24].

A new solution for a soil venting problem operating a partially penetrating, vertical well at a fixed pressure under leaking ground surface, uncovered and covered conditions was presented by Ching-Lin et al. The authors found that the solution can predict the distribution of different pressures from those given by a solution using a constant flux rate at a single screen well. The influence radius for SVE is strongly affected by vertical permeability, radial permeability, and the ratio between these two. The pressure of vacuum values was predicted by the proposed solution and compared to measurements at different observation wells and depths under venting experiments at the former SVE wells. The vacuum pressures were measured, and the gas flow rate around the wells was significantly lower than the predicted values. Therefore, the wells of this site were suspected to be positive skin zones [25].

The purpose of this study is to explore the effect of soil type in the removal of hydrocarbons, on three types of soil: 1) fine sand, 2) fine sand with 20% of kaolinite clay and 3) fine sand with 40% of kaolinite clay. Soils with different percentages of clay were used to examine the effect of clay percentage temperature on the efficiency of soil remediation via the SVE method.

## 2. Methods and Materials

Considering the fact that a laboratory method was chosen for this study it is necessary to state the quantitative and qualitative characteristics of the substances used. Soil, pollutants and also the specifications of the experiment and the way it is carried out to the final results were acquired.

### 2.1. Material

To analyse the effect of small soil particle on treatment, fine sand which has a lot of use in the collection of oil spillage will be used as the main soil in this study. Then using kaolinite clay [26], two new samples of soil are made so that the effect of having clay in the soil can also be analysed:

- Fine sand with 20% kaolinite clay
- Fine sand with 40% kaolinite clay

In mixing sand and clay, we use a fixed size container which is filled 20% and after that 40% of its volume with clay and the rest with air-dried sand. The kaolinite clay used was acquired from the Iran china sand company and the sand from the Iranian Oil product distribution company. According to experiments, the moisture for each of the items, sand, sand with 20% clay and finally 40% clay was consecutively 3%, 2.85% and 2.32%. The soil organic content was also consecutively 1.6%, 1.56%, and 1.32%.

One of the vastly used oil products in the world is gas oil which has been chosen as the pollutant in this study. Gas oil is acquired by refining crude oil, and in terms of usage as fuel is in second place to petrol. Due to the stability of the oil compounds which act as pollutants, the damage caused to human resources, water and eco-system are different and more complex. For example, according to studies, in an area where excessive amounts of gas oil are respired, it causes itching of the eyes, nose, throat and pulmonary disorders. Gas oil can be absorbed through the skin and long-term exposure can cause skin cancer. In the case of contact with eyes it causes mild sensitization, and swallowing gas oil can be very dangerous. Also, poisoning can occur if gas oil vapors enter the body through the mouth.

The gas oil used in this study was acquired on the Iranian oil products distribution company. The specifications of the hydrocarbons in this gas oil are shown in Figure 1.

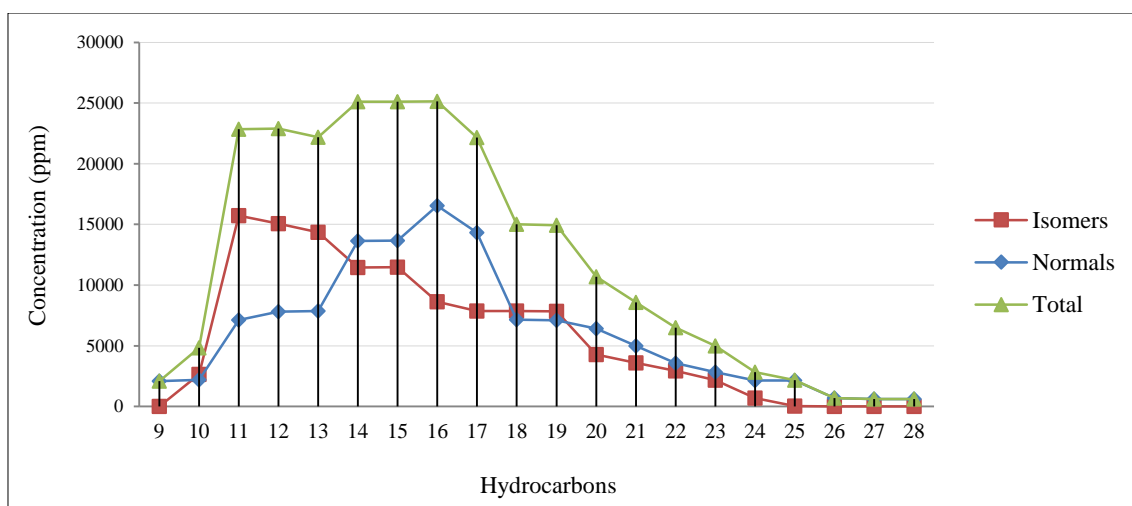


Figure 1. Existing aliphatic hydrocarbons in gas oil sample

2.2. Method

A pilot was made to analyze remediation on a laboratory scale using the vapor extraction method. The pilot consisted of seven parts that are shown in Figure 2.

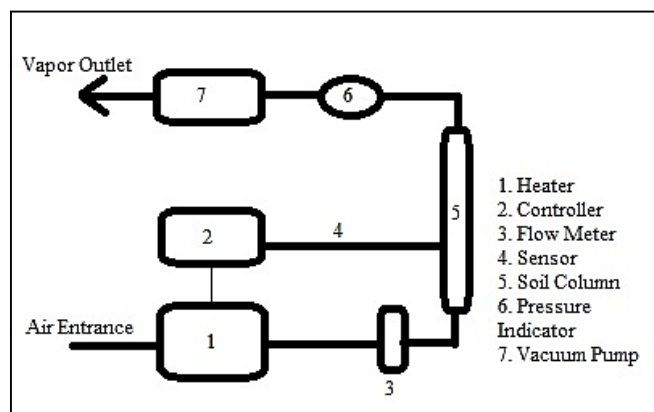


Figure 2. Schematic of vapors extraction pilot tests [16]

The soil column is made from Plexiglas and has a height of 30cm and a diameter of 4cm. The use of plexi glass to make to the column enables us to see the sand during the experiment. To extract vapors from the system, the Sparmax TC-63/vd model pump was used. There was a pressure indicator and an air filter installed on this pump and it had two exits for air. In all of the experiments, the flow in the system was 0.25 m<sup>3</sup> per hour. To manage and control the system's temperature, a heating device, and a digital temperature controlling device was used. This device consists of a heat producing element in a galvanized container and also a digital regulator which has a sensor positioned in the path of the airflow going into the soil column, which recognizes the temperature of the air going in and changes it to reach the desired temperature and keeps it at that point. This device works in such a way that when the temperature of the systems is not the defined range it turns the heating element off or on to keep it in the range desired. In all experiments the temperature was set at 30 degrees centigrade. For the purpose of keeping the conditions of the system the same in all the experiments, two pressure meters were installed, one on the soil column and another on the vacuum pump. Because of this whenever in an experiment the conditions changed from the rest of the experiments, it was stopped and repeated so that the final results wouldn't be affected by these changes. To control the temperature even further a thermometer was installed on the soil column to see whether the temperature before and after the column is the same or not. To control the flow of air a flow meter was put in place.

After all the essential material was provided, making the soil samples was started. For the first sample, sand was polluted with gas oil at a rate of 50000 ppm, for every 100 grams of sand 5 grams of gas oil was hand mixed. In the second sample (and third sample) where the sand was mixed with 20% (and 40%) of clay, first the 20% (and 40%) mixtures were made into small samples in a sufficient amount, then the polluting sequence was carried out the same as in the first sample.

To start the experiment, first the sand was placed in the column in three stages and in each stage it was beaten with 5 hits, then the lid was placed. Then the column was placed on its vertical stand, and the inlet and outlet pipes were linked

to their proper places. Then the desired temperature was set and at the same time, the vacuum pump, air inlet and temperature control system were turned on. With the experiment started, the pressure and temperature of the air flow were recorded at fixed time intervals. The chosen time for the experiments was 8, 16 and 24 hours. Using the SVE method, as time passed, the soil was cleaned and the concentration of the aliphatic hydrocarbons were reduced. At 8, 16 and 24 hours samples were taken from the sand in the column. The samples were kept in special containers and for determining the concentration of aliphatic hydrocarbons before and after applying the vapor extraction method, gas chromatography was carried out (GC). To use this technique (GC) on the specimens, around 2 grams of each sample was extracted using the standard method and was test according to the GC method. The chromatogram used was the HP 6890 GC /5973 model and it was equipped with an ionizing detection flame. The separating column was 3 meters tall and 0.32 mm in diameter. The temperature of the column and detector was 230 degrees centigrade. The flame producing gases were air (270 cm<sup>3</sup>/min) and hydrogen (30 cm<sup>3</sup>/min). The vector gas was nitrogen. Figure 3 shows the gas chromatography.



Figure 3. Gas chromatography

### 3. Results and discussion

In this section the results of the gas chromatography test from the initial samples after 8, 16 and 24 hours are shown in three groups of soil: 1) sand 2) sand with 20% clay 3) sand with 40% clay. At the end, the efficiency of elimination of the aliphatic hydrocarbons from the three kinds of soils was analyzed and compared.

#### 3.1. Fine sand

There are 20 differentiable hydrocarbons in the sand polluted with gas oil which consist of C-9 to C-28. The process of elimination for each of the normal hydrocarbons is shown in Figure 4.

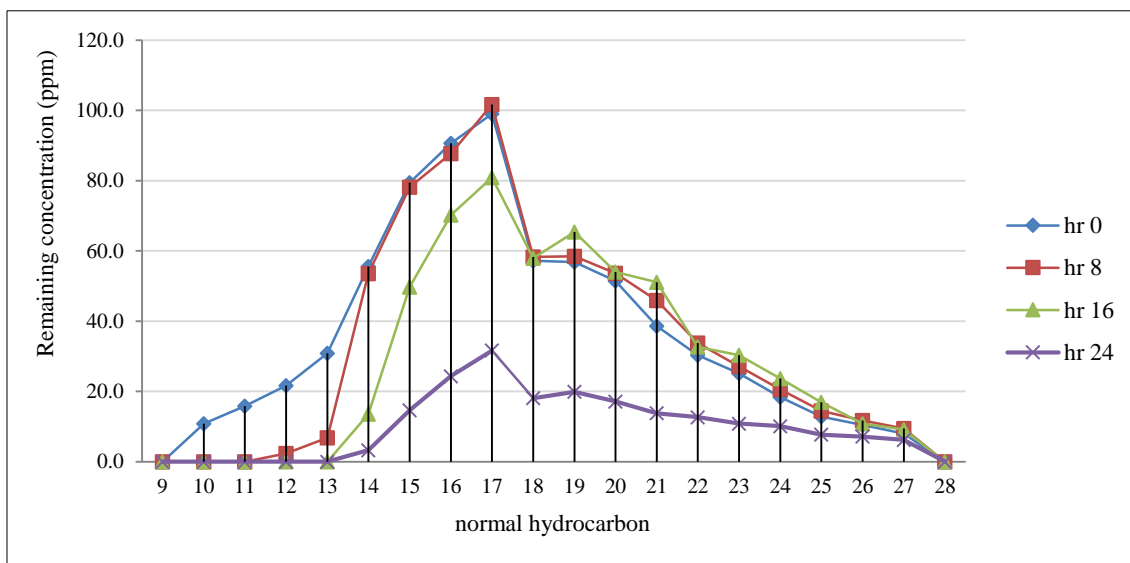


Figure 4. Changes of normal hydrocarbons concentrations in the fine sand

It should be mentioned that the concentration percentage of hydrocarbons are measured with respect to the most concentrated hydrocarbon at the beginning of the test. The efficiency of removal of hydrocarbons in the first 8 hours was 7%, after 16 hours it was 13% and after 24 hours it was 58%. The most efficient period was the one between 16 and 24 hours. In other words, if the cleansing procedure were to follow the same trend as in the first 16 hours, the efficiency would have been 20% but we can see the final 8 hours the efficiency is tripled approximately to what we expected. At the end, after 24 hours the cleansing rate was 78.4%. Figure 5 shows the changes of isomers concentration in fine sand.

As we can see in the diagram, the majority of the light hydrocarbons (C-9 – C-13) are cleaned very efficiently in the first 8 hours so we can conclude the best time to clean them is in this first 8 hours. Some of the heavy hydrocarbons have an additional sequence and then they are cleaned after 16 hours. The reason for this increase in the concentration of heavy hydrocarbons is the transformation of hydrocarbons to one another, especially the transformation of isomers to normal hydrocarbons [27, 28]. Figure 6 shows the changes for each of hydrocarbon isomers in the polluted sand as time passes.

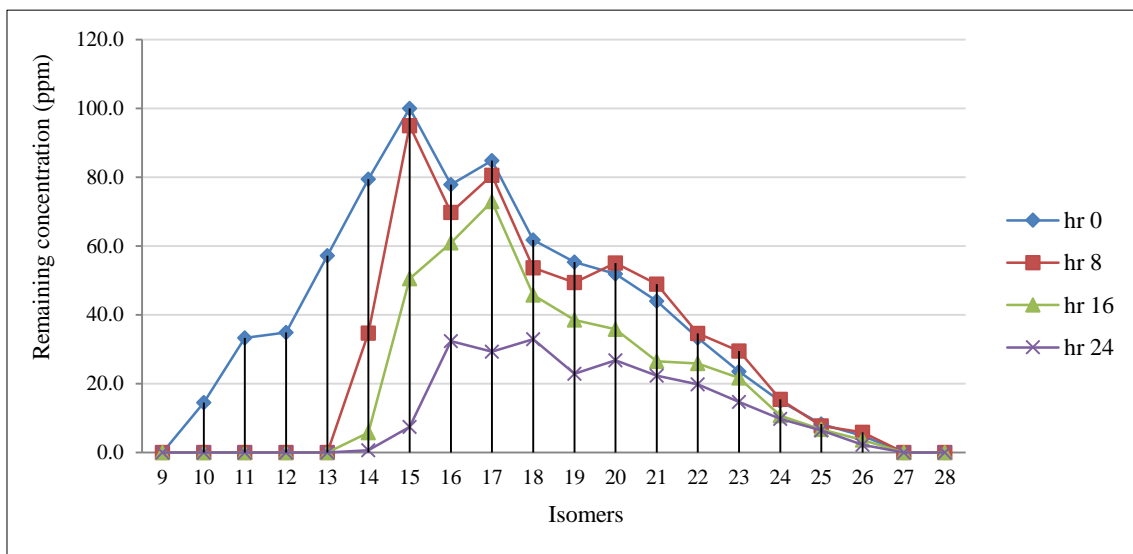


Figure 5. Changes of isomers concentrations in the fine sand

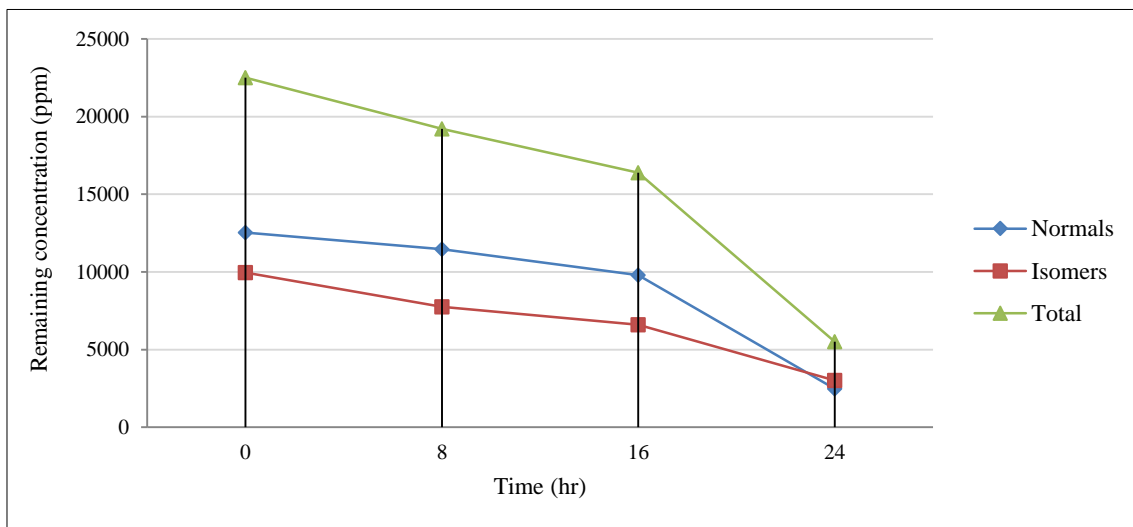


Figure 6. Comparing the processes of elimination of isomers, normal hydrocarbons and the total hydrocarbons

According to Figure 6, the overall changes for normal hydrocarbons are similar to isomers and naturally the process of cleaning soil from the total hydrocarbons (total of normal and isomers) will be similar to these two processes. The only difference is the speed of elimination of isomers compared to normal hydrocarbons in the first 8 hours. After 8 hours the percentage of efficiency for cleaning isomers is 14% more than normal hydrocarbons. On the other we can see that the concentration of some heavy hydrocarbons is increasing, this could be a reason to show that isomers transform into normal hydrocarbons. The percentage of concentration of remaining hydrocarbons in the sand after 24 hours in the temperatures 30 degrees centigrade is 21.56%.

### 3.2. Sand with 20% Clay

In this case the process of elimination is shown separately for each hydrocarbon in Figure 7.

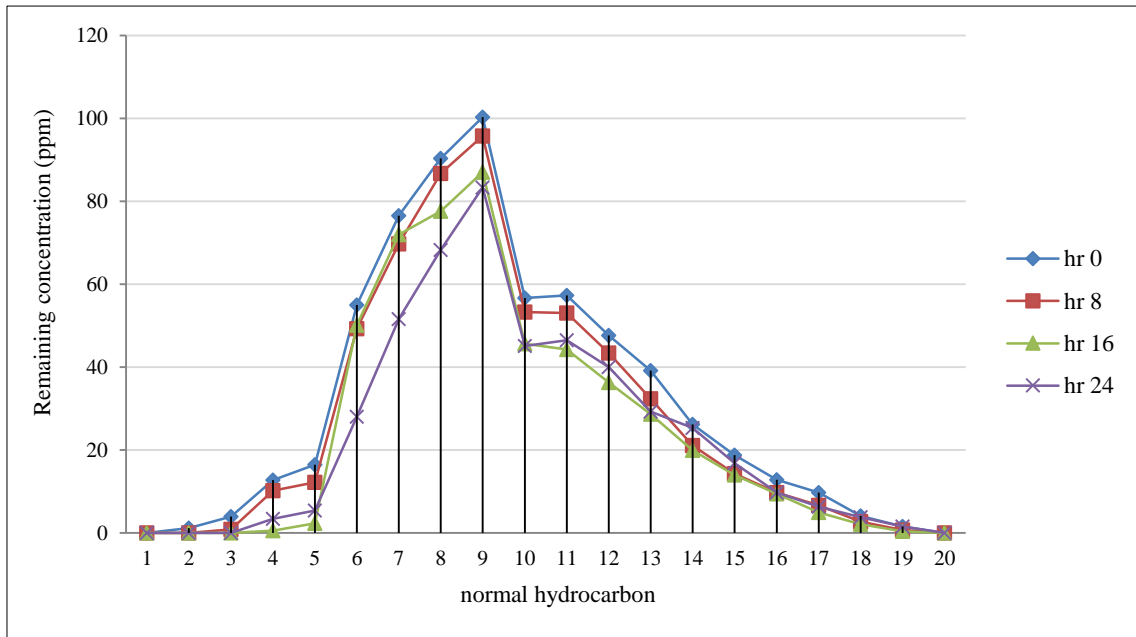


Figure 7. Changes of normal hydrocarbons concentrations in the fine sand with 20% of clay

Here the efficiency of elimination in the first 8 hours is 10%, in the second 8 hours is 9% and in the third, it is 6%. The existence of 20% clay causes a drop of 53.16% in the rate of cleaning compared to pure sand.

The main period of removal of heavy hydrocarbons from C-18 to C-28 and for light hydrocarbons, from C-9 to C-13 was in the first 16 hours. The removing of C-14 to C-18 hydrocarbons is largest in the last 8 hours. Figure 8 shows the changes in hydrocarbon isomers in the sand with 20% clay as time passes.

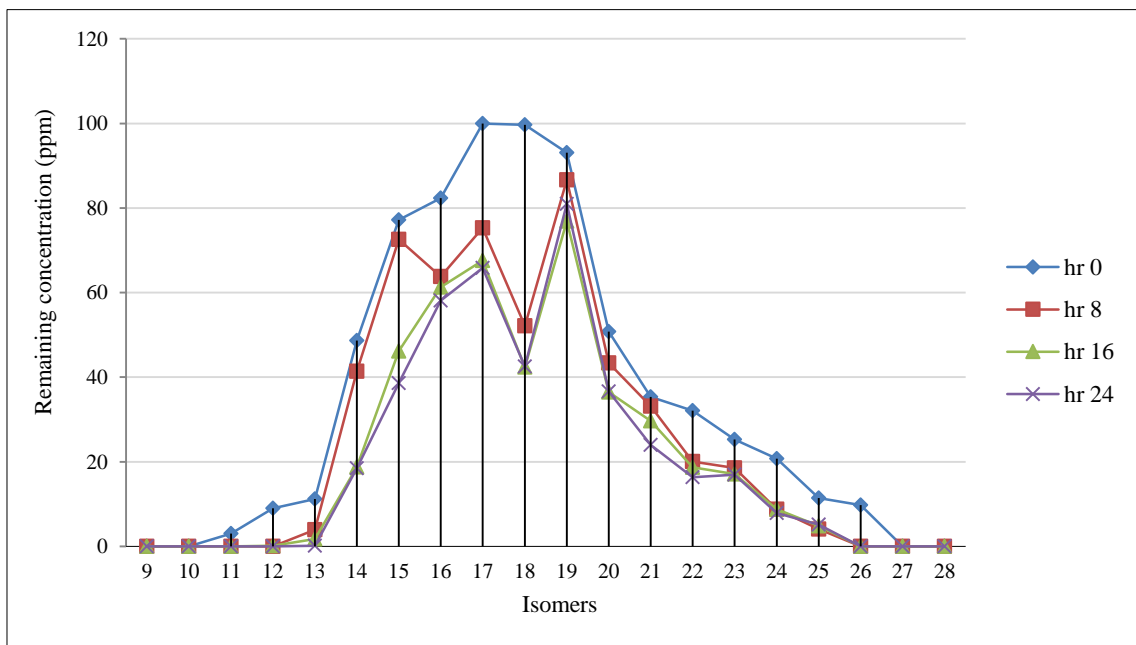
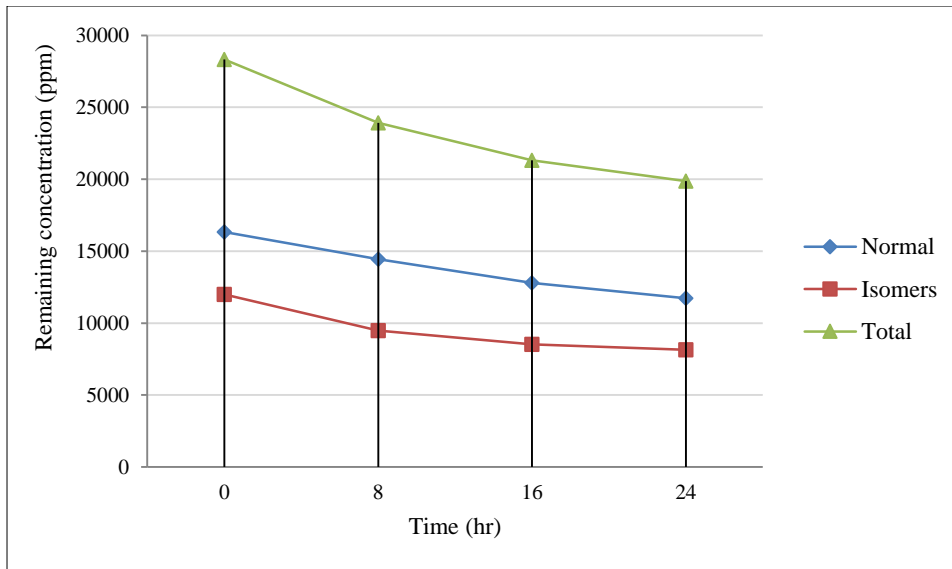


Figure 8. Changes of Isomers concentrations in the fine sand with 20% of clay

If we compare between the process of isomer and normal hydrocarbon removal and the removal of the total hydrocarbons from both these groups Figure 9 can be depicted.



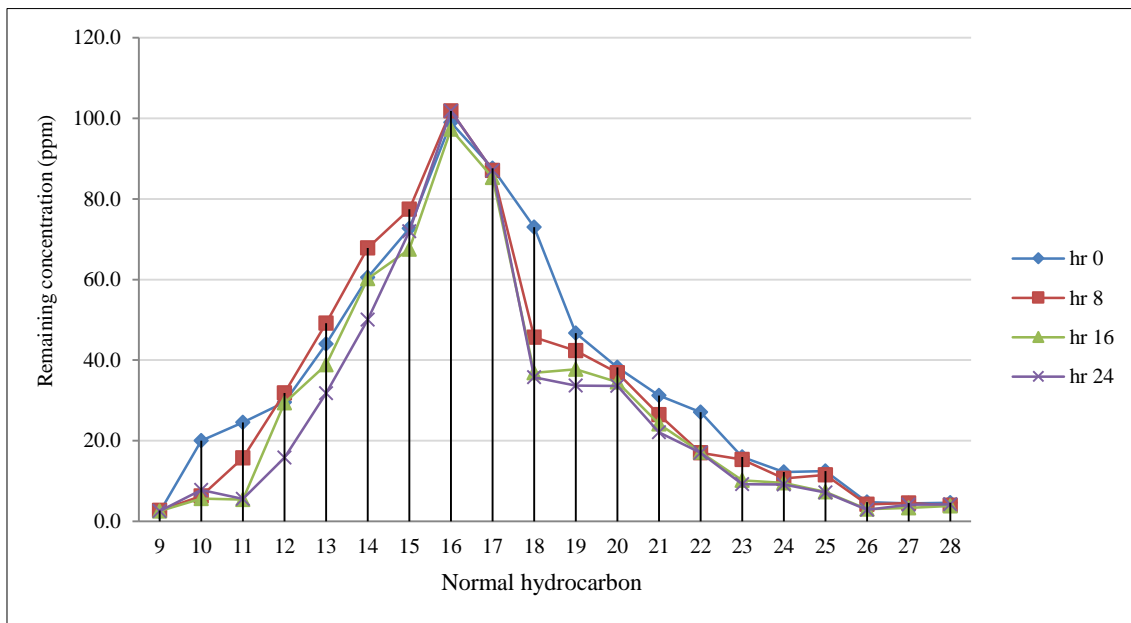


**Figure 9. Comparing the process of isomer and normal hydrocarbon removal and the removal of the total hydrocarbons**

The speed of removing for isomers compared to normal hydrocarbons in the first 8 hours is 12% more. The percentage of residual hydrocarbons in the sand with 20% clays after 24 hours at 30 degrees centigrade are, 74.76%.

**3.3. Sand with 40% Clay**

In this kind of soil, the process of elimination for each of the hydrocarbons is shown separately in Figure 10.



**Figure 10. Changes of normal hydrocarbons concentrations in the fine sand with 40% of clay**

If the percentage of clay is increased to 40% the process of cleaning in the first 8 hours becomes somewhat compromised and decreases to 2%. In the following, hours the process is similar to that of sand with 20% clay composition and continues with lower speed. In this soil, the rate of removing decreases 1.49% compared to the soil with 20% clay and stands at 23.75%.

This trend in the decrease of efficiency points out that when there is 20% clay in the sand, it is saturated with small grain soil and after this percentage, the drop in efficiency is very slight. Before the saturation of sand with small grains, any change in the amount of clay present would have a big impact on efficiency but after saturation adding clay will affect the cleaning rate in smaller scales. Figure 11 shows changes of isomer hydrocarbons in the sand with 40% clay with the passing of time.



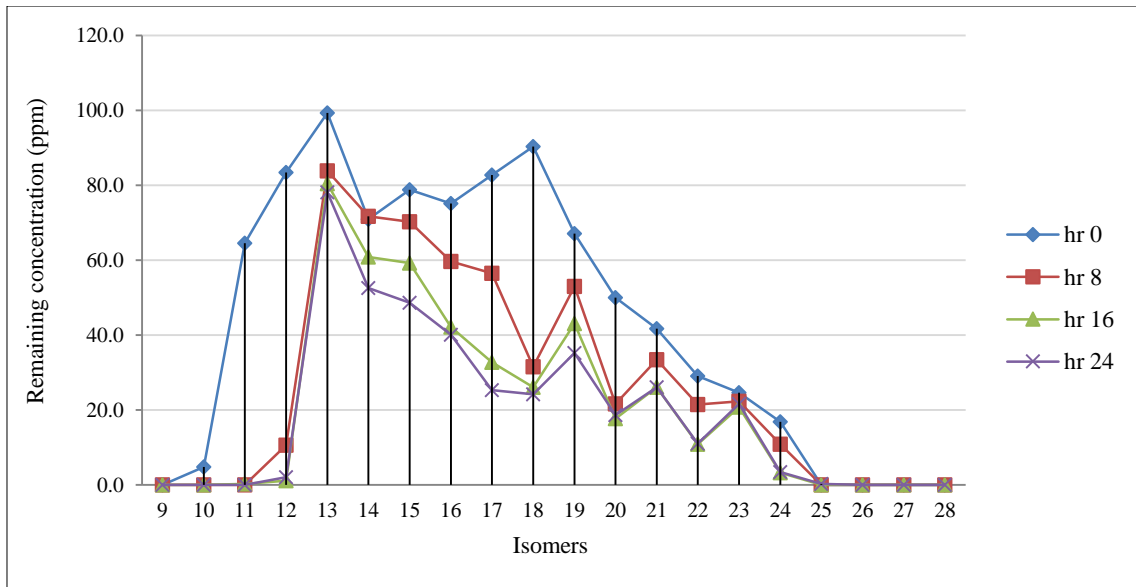


Figure 11. Changes of Isomers concentrations in the fine sand with 40% of clay with the passing of time

If we compare between the processes of elimination of normal hydrocarbons, isomers and the total hydrocarbons from both groups Figure 12 can be drawn as a result. The percentage of residual hydrocarbons in sand with 40% clay after 24 hours at 30 degrees centigrade is, 76.25%.

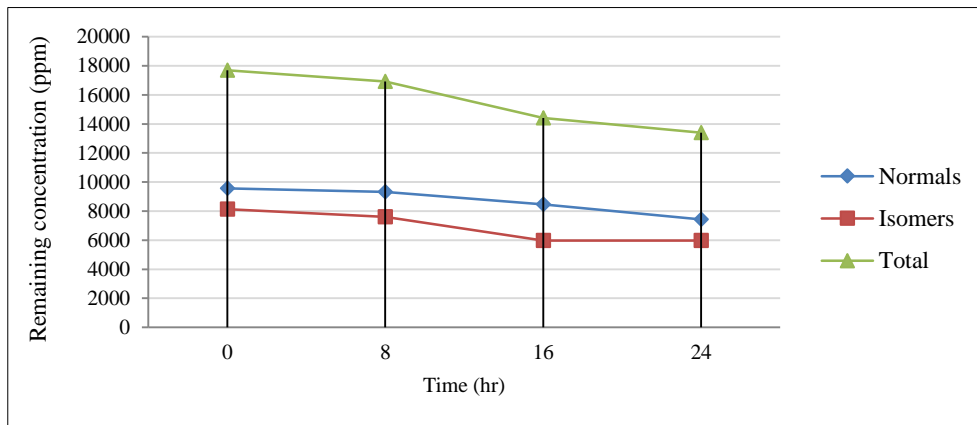


Figure 12. Processes of elimination comparison for normal hydrocarbons, isomers and the total hydrocarbons for sand sample with 40% of clay

### 3.4. Comparison of soils

By comparing the efficiency of cleaning of the hydrocarbons in pure sand, sand with 20% clay and sand with 40% clay at 30 degrees centigrade, Figure 13 was illustrated.

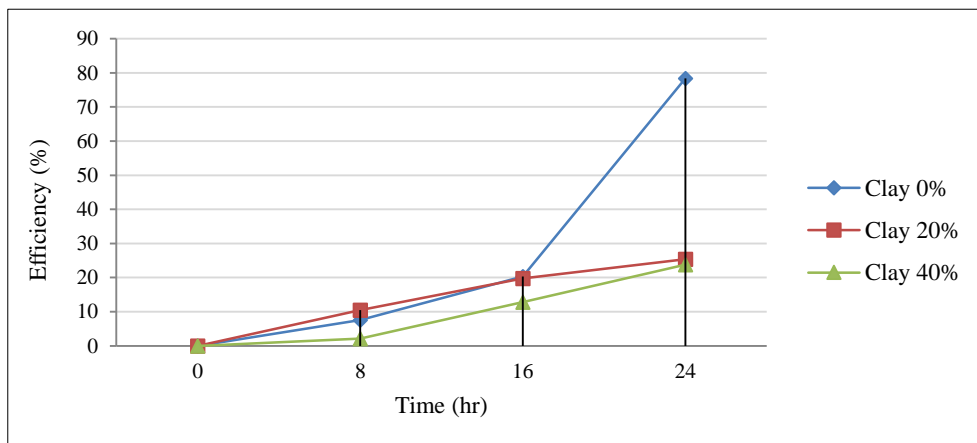


Figure 13. Efficiency comparison for 3 types of soils

## 4. Conclusions

Soil Vapor Extraction is recognized as an efficient method for removal of VOC's in the unsaturated polluted soil. In this study by analysing the SVE method in soil with variable amounts of clay the following results were deduced;

1. When there is no clay in the sand the efficiency of cleansing after 24 hours stood at 78.6%, the efficiency when there was 20% clay was reduced by 53.2% and when there was 40% clay it was reduced by 54.7%.
2. By dividing the hydrocarbons into three groups, light hydrocarbons from C-9 to C-11, medium hydrocarbons from C-12 to C-16 and heavy hydrocarbons from C-17 to C-28, the following results were obtained:
  - When there is no clay available, the concentration of heavy hydrocarbons after 8 hours increases and the concentration of the rest of the hydrocarbons decreases. But overall, the decrease is larger than the increase caused by the transformation of hydrocarbons to one another. After 24 hours the sand is noticeably cleaner. According to observations, the reason for the increase in heavy hydrocarbons is the transformation of hydrocarbons to one another especially the transformation of isomers to normal hydrocarbons. Also considering the fast cleaning of light hydrocarbons the best period to clean them is in the first 8 hours.
  - When there is 20% clay, all the hydrocarbons are cleaned to an average amount as the total amount of hydrocarbons cleaned in the first 8 hours, compared to when there is no clay in the sand where the hydrocarbons transform to one another, is higher. In this situation the amount of light hydrocarbons cleaned is more than the others. In this kind of soil the best period to clean light hydrocarbons using the SVE method seems to be 16 hours, and after this point the removal of this kind comes to halt.
  - When there is 40% clay, the speed of light hydrocarbon removal is faster than the other hydrocarbons. The medium hydrocarbons in the first 8 hours, experience a slight increase due to transformations and after that to the end of the 24-hour sequence have a decreasing process. The removal of heavy hydrocarbons follows a slow and steady rhythm.
3. The other interesting point is that when comparing the removal of the three kinds of hydrocarbons we see that with the increase of clay in the sand, the transformation process shifts from transforming to heavy hydrocarbons, to transforming into medium hydrocarbons.

The removal process for isomeric hydrocarbons for all three situations shows that this removal is faster in the first 8 hours compared to similar but normal hydrocarbons (when there is no clay, 14% faster, and with 20% clay it is 12% faster). In these 8 hours (initial) the concentration of some heavy hydrocarbons increases which emphasizes the transformation and breakdown of isomeric hydrocarbons into normal hydrocarbon and an increase in their concentration in the first 8 hours.

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