# 4. PETROLEUM PRODUCT CONTAMINATION INSIDE THE PERIMETER OF OIL TERMINAL SOUTH STORAGE AREA

## 4.1 PREVIOUS STUDIES ON THE ECOLOGICAL STATE OF OIL TERMINAL SOUTH STORAGE AREA

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The geoecological study presented in this chapter was carried out inside the perimeter of Oil Terminal South Storage Area located in the south-eastern part of Constanța City, South of the Constanța Sud and Faleza Sud quarters, between the Mangaliei Road to the West and the Constanța South Harbor to the East (Figure 4.1). South of the perimeter of the storage area the land slopes down gently towards Lake Agigea.

**Figure 4.1** *Location of the Oil Terminal South Storage Area*

The first investigations concerning the ecological state inside the perimeter of Oil Terminal South Storage Area were performed in 1995 by PROLIF S.A. as ordered by Oil Terminal Constanța. The conclusions of these geotechnical, hydrogeological and ecological studies (Spiridonică, 1995a; 1995b) referred to the geological structure and the ecological state inside the perimeter of the Oil Terminal South Storage Area before year 2000.

A total of 88 new 3”-diameter boreholes and 11 new $8^{5/8}$”-diameter boreholes were installed for the purpose of this study. The boreholes were placed both inside and outside the perimeter of the storage area. Sediment samples were collected from the boreholes, providing data on the lithology of the deposits (visual description and grain size analysis), the physical and mechanical characteristics of the sediments, as well as the water table in the unconfined aquifer. The level of contamination was evaluated based on the visual and olfactory description of sediments, as well as through sample analysis.
By investigating the causes for the rise of the water table inside the perimeter of the Oil Terminal South Storage Area, it was concluded that an inflow from the sewer line located outside the perimeter, from the drinking water pipeline or from the local irrigation system was occurring. For this reason, losses from the firewater pipeline network located in the eastern part of the Oil Terminal South Storage Area were considered. The damaged internal sewer network determined the continuous recharge of the water table.

The resulting potentiometric lines (Spiridonică, 1995b) indicated two groundwater flow directions (NW to SE and NE to SW), with hydraulic gradients of 1.6% and 2%, respectively. In the eastern part of the storage area the main groundwater flow direction is to the East towards the Black Sea.

The measurements emphasized the distribution of the zones of depth to the water table, from close to surface to more than 4 m, in the southern part of the storage area.

Three horizons were recognized inside the succession of sediments in the Oil Terminal South Storage Area (Spiridonică, 1995a; 1995b): permeable clayey silt on top, low-permeability silty clay in the middle and a basal red clay horizon.

The contamination with petroleum products in the eastern zone of the Oil Terminal South Storage Area and the discharge of petroleum contaminated water at the base of the seaside slope were discussed in a study carried out in 1995 (Spiridonică, 1995a). The investigations carried out emphasized the following main issues:

- Petroleum product losses determined the impregnation of sediments and the contamination of groundwater;
- The main groundwater flow direction, as well as the aquifer slope caused the movement of the petroleum product towards the base of the seaside slope;
- In some of the boreholes, free petroleum product was identified;
- Surface contamination was identified locally;
- Most of the contamination occurred in the past;
- The local contamination sources and water sources needed to be identified; interception and pumping of contaminated groundwater in a separator was recommended for remediation.

**Figure 4.2** *Location of the Study Boreholes Made in 2000 within the Perimeter of the Oil Terminal South Storage Area*

At the end of 2000, a sedimentological and hydrogeological study was carried out in the area of the Oil Terminal South Storage Area (Jipa et al., 2001), to evaluate the ecological state inside this perimeter. The data collected in this study were from boreholes performed by PROLIF S.A., Constanța (Figure 4.2).
By integrating the data from 1995 and 2000, the evaluation of the environmental state of the Oil Terminal South Storage Area was based on a relatively extended and reliable amount of information.

The results of the study are presented in this chapter.

## 4.2 LITHO-FACIAL SEQUENCE, THE GEOLOGICAL STRUCTURE AND THE WATER HEAD INSIDE OIL TERMINAL SOUTH STORAGE AREA

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### The Network of Boreholes

Because the study area was covered, to a great extent, by buildings, asphalt pathways, mounds etc., the investigation was performed on boreholes.
The boreholes were drilled manually in 2000, and they are of 3" and 8 $^{5/8}$" in diameter. 24 boreholes were drilled inside the perimeter of Oil Terminal South Storage Area (Boreholes f1/2 to f24/2), placed in the precincts of the big tanks. These boreholes had 3"diameters and depths between 5 and 7 meters. 6 additional boreholes (Boreholes f25/2 to f30/2), also of 3" diameter and 5 m depth, were installed on the seaside slope near the storage area between Gates VI and VII.
3 boreholes of 8$^{5/8}$"-diameter and 15 m depth (F1, F3 and F6) were placed in the agricultural area located South of Oil Terminal South Storage Area.
18 additional boreholes of 8$^{5/8}$"-diameter and 15 m depth were drilled inside the perimeter of Oil Terminal South Storage Area. These boreholes have 4” or 6” inside diameter metal casings.

Figure 4.2 shows the location of the boreholes installed in 2000.

**Sediment Types**

The sediments collected from the perimeter of the Oil Terminal South Storage Area were classified using the triangular grain-size diagram *sand/silt/clay,* by projecting the percentage values. Three main grain-size categories were distinguished: clayey silt, silty clay and clay.
The clayey silt can be divided in two categories, based on its content of sand:

- Brown or brown-reddish clayey silt, containing 1 - 2% sand, and
- Yellowish clayey silt, containing 2 - 4% sand.

For both categories, the silt content is 67-72% and the clay content is 26-29%.
Most of the samples analyzed belong to the category of silty clay. Three types of silty clay were distinguished:

- Brown or brown-yellowish silty clay, containing 1-2% sand, 51-63% silt and 35-46% clay, or 2-4% sand, 63-68% silt and 30-35% clay,
- Brick or brown-reddish silty clay, with sand content of 1-2%, but with 52-57% silt and 41-45% clay or 59-64% silt and 33-40% clay, and
- Yellow or yellow-grayish silty clay, containing 1-4% sand, 60-66% silt and 31-36% clay.

The third category of sediments consists of clays containing 1-1.5% sand, 47-48% silt and 50-59% clay.

**Litho-facial Sequences inside the Perimeter of the Oil Terminal South Storage Area**

In order to emphasize the types of litho-facial sequences, the lithological columns of the $8^{5/8}$"-diameter boreholes were examined to obtain thicker sequences, extended at the scale of the whole Pleistocene succession from the study area. The results of this sequence analysis were as follows (Figure 4.3):
Most of the lithological columns examined (around 48%) correspond to the *standard sequence* (the simplified model of the Pleistocene succession). The three lithological horizons were recognized: clayey silt (loessoid; 3-8 m thick), silty clay (prevailing paleo-soil; 2-4 m thick) and clay (mostly red clay; up to 6-7 m thick).
In some cases (Boreholes F2, F3, F4, F11, F11/2) the thickness of the silty clay horizon was significantly reduced, while the thickness of the loessoid horizon reached 10 m in Borehole F2.
In many cases (around 41%) we noticed an *atypical sequence*, the standard sequence being modified in the following way:
The clay was not present at the basal part;
The loessoid horizon was very thin, and
Red clay intercalations appeared in the silty clay horizon.
Around 11% of the examined lithological columns provided a *completely atypical sequence*, with a clear dominance of the silty clay horizon (Boreholes F7; F5/2), or with the combination of certain deviations from the standard sequence (red clay in the silty clay horizon and very low thickness of the loessoid horizon, etc.).
In conclusion, an attempted order the lithological succession (the simplified Pleistocene model) was found but with frequent deviations.

**Figure 4.3** *Pleistocene Lithology Sequences Emphasized within the Perimeter of the Oil Terminal South Storage Area- Comparison to the Standard Sequence*

## The Geological Structure of the Pleistocene Deposits

In order to determine the spatial distribution of the litho-facial sequences in the structure of the Pleistocene deposits, several lithological cross-sections were made, on the North – South and East – West directions, as well as a structural map at the base of the permeable horizons.

Data provided by the boreholes made in 2000 as well as data from previous studies (Spiridonică, 1995 a, b) were used in the geological cross-sections.

The analysis of the East – West cross-sections indicates the following (Figure 4.4):

- A general thickening tendency of the permeable horizons (the clayey silt and the silty clay horizons) from North to South, with rather significant thickness variations,
- An accentuated thinning of the upper horizon in the area of the tanks, which is the result of excavation activities during the construction of the storage area; in some sectors this horizon is completely absent (for example, on the cross-sections N-S3, N-S5);
- The base of the silty clay horizon is not uniform, reflecting the deposition of the red clay horizon on a paleo-relief.

The analysis of the North – South cross-sections indicates the following (Figure 4.5):

- In the central zone there is an uplift of the limits between the sequences; the eastern and western compartments are lower than the uplift;
- In the zone of the uplift, the upper horizon is very thin and even disappears. This thinning is partially due to excavations;

The thickness of the intermediate sequence (the silty clay horizon) was rather difficult to determine, because of the small number of boreholes, which crossed it completely especially in the eastern part of the storage area. Nevertheless, the thickening of the intermediate sequence is more accentuated in the central zone of the storage area compared to the northern and southern sectors.

It was sometimes difficult to trace the limit between the upper and middle horizons due to an alternation of coarser and finer layers (for example in Boreholes F7, F14, F15 and F17). The difficulty in separating the three horizons was also encountered in the analysis of the detailed lithological columns (both for the boreholes made in 2000 and 1995), which accompany each cross-section.

The lithological horizons are not homogeneous from the grain size point of view. Inside these horizons, there are lens of different grain size (coarser or finer) than the general background.

**Figure 4.4** *West – East Geological Cross-Section within the Perimeter of the Oil Terminal South Storage Area*

**Figure 4.5** *North – South Geological Cross-Section within the Perimeter of the Oil Terminal South Storage Area*

**Figure 4.6** *Map with Isolines of the Top Elevation of the Red Clay - Oil Terminal South Storage Area*

*(Points represent the locations of the boreholes used.)*

### Map with Isolines of the Top Elevation of the Red Clay

In order to emphasize the variation in thickness and the paleo-relief of the aquifer (from the surface to the first continuous level of red clay of significant thickness), a map with isolines at the top of the red clay was drawn. This map indicates the following (Figure 4.6):

- The upper part of the clay horizon is very nonuniform from a morphological point of view; a series of uplifts and depressions are emphasized, with developments in the order of hundreds of meters and altitude variations of 3 – 7 m;
- A general deepening tendency from North to South is observed; on this background, a ridge is emphasized in the southeastern zone, on the Northeast to Southwest direction, separating two lower zones with the same orientation;
- The central zone shows two sloping tendencies: one very accentuated towards the Southwest (paleo-valley) and another one, slightly emphasized, towards the Northeast (towards the sea).

The determination of the limits between the three horizons was dificult due to the slight variations in lithology (very similar grain sizes). Therefore, the geometry obtained for the sequences has a certain degree of uncertainty.

### Hydrostatic levels in the Oil Terminal South Storage Area

*Dan C. Jipa, Gicu Opreanu, Irina Dinu*

Inside the perimeter of Oil Terminal South Storage Area, the hydrostatic heads in the unconfined aquifer were measured in two periods: December 2000 /January 2001 and May 2003.

Table 4.1 presents the elevations of the water table in the aquifer.

The highest water table elevations are in the northwestern (Boreholes F15, F16), northern (F7/2) and southeastern (F9/2) parts of the Oil Terminal South Storage Area (Figure 4.7). By correlating these heads to the ground elevations and also to the water table elevations in the other sectors of the storage area, we can conclude that the water table inside the Oil Terminal South Storage Area is higher than expected. The water table is supplied both naturally (from precipitation) and artificially, by losses from water pipelines, as also noticed by Spiridonică, 1995b. Thus, in Boreholes F13/2 and F14/2, the water table is 3.63 m and 1.41 m deep, respectively. At the same time, North of the storage area, only few tens of meters away, the ground elevations are 6 – 8 m lower, with no groundwater discharge.

The conclusion is that the water table is exaggeratedly high, and this is due to an artificial recharge inside the storage area also sustained by the fact that, at the northern boundary, the sloping ground is usually wet, even when precipitations are much reduced.

**Figure 4.7** *Piezometric Map of the Unconfined Aquifer based on the Hydrostatic Heads Measured in September 2002- Oil Terminal South Storage Area*

Table 4.1
*Elevations of the Water Table in the Unconfined Aquifer in the Oil Terminal South Storage Area*

| Borehole | Date of the measurement | Water table elevation (m asl) | Date of the measurement | Water table elevation (m asl) |
|---|---|---|---|---|
| F1/2 | 12/07/2002 | 27.98 | 05/14/2003 | |
| F2/2 | | 29.09 | | 29.14 |
| F3/2 | | | | |
| F4/2 | | 22.36 | | 21.81 |
| F5/2 | | 29.41 | | 29.83 |
| F6/2 | | 26.60 | | |
| F7/2 | | 31.81 | | 31.98 |
| F8/2 | | 22.18 | | 21.92 |
| F9/2 | | 30.26 | | 30.31 |
| F10/2 | | 28.72 | | 28.52 |
| F11/2 | | 27.92 | | 27.99 |
| F12/2 | | 27.14 | | 27.92 |
| F13/2 | | 25.52 | | 25.93 |
| F14/2 | | 24.95 | | 25.05 |
| F15 | 01/19/2002 | 32.47 | | 32.70 |
| F16 | | 31.17 | | 31.65 |
| F17 | | 29.37 | | 29.39 |
| F18 | | | | 21.05 |

## 4.3 LITHO-FACIAL SEQUENCE, GEOLOGICAL STRUCTURE AND HYDROSTATIC HEADS EAST OF OIL TERMINAL SOUTH STORAGE AREA

*Dan C. Jipa, Gicu Opreanu*

**The Network of Boreholes**

During the assessment of the ecological state of the Oil Terminal South area, initiated by Oil Terminal Constanța, six 3”-diameter uncased boreholes were performed by Prolif Constanța, using hand sludge drilling. The data provided by these boreholes were used for the flow model in the zone of Oil Terminal South Storage Area.

Five boreholes reached 5 m in depth, while the sixth reached only 2.5 m in depth.
The boreholes were aligned on two East – West lines, located immediately eastward of the steep slope (Figure 5.2). The boreholes were numbered f25 to f30.

### Litho-Facial Sequence of the Deposits Intercepted by Boreholes East of Oil Terminal South Storage Area

The lithological columns made based on the information provided during the drilling of the boreholes show that the Pleistocene deposits from the central and eastern sectors of the perimeter located East of Oil Terminal South Storage Area consist mainly of very fine sediments (Figure 5.3).

The column of Borehole f27 consists of yellowish or brownish silty clays, with grey, greenish or reddish tints. A 30 cm thick intercalation of brownish clay with yellowish intercalation appears in the lower half of the column.

Boreholes f28, f29 and f30 are also crossing mainly silty clay, with clayey silt intercalations, 1.2 to 1.7 m thick.

The lithological column of Borehole f26 represents a clear clayey sequence, with two thin clayey silt intercalations.

Very fine sediments (mostly clay, but also silty clay) prevail in Borehole f25. The upper third of this lithological column consists of yellow-brownish clayey silt.

Carbonate concretions of milimetric dimensions (or less than 1 mm) are sometimes disseminated within the mass of clayey or silty sediments. Macropores are often observed in the sedimentary material collected during the drilling. At the upper part of the drilled column certain characteristics indicate that the quality of fill material is related to the significant construction works in this zone.

The dominance of clayey or prevailing clayey sediments show that the largest part of the sedimentary sequence crossed by the boreholes East of the Oil Terminal South Storage Area corresponds to the middle horizon, with prevailing paleo-soils of the standard lithological sequence (Figure 5.3). The clayey silt intercalations show that the upper part of the sedimentary columns (except in Borehole f26) may belong to the basal part of the clayey silt horizon.

### Geological Structure of the Pleistocene Deposits East of Oil Terminal South Storage Area

The geological cross-sections through the Pleistocene deposits East of the Oil Terminal South Storage Area followed the two borehole alignments (Figure 5.4).

The interpretation of the cross-sections raised some questions because the available lithological columns were not deep enough. For this reason, boreholes from inside Oil Terminal South Storage Area were also used.

The cross-sections were drawn using the hypothesis that the clayey silt intercalations would indicate the presence of the lower part of the clayey silt horizon.

According to the tendency observed in the zone of the Oil Terminal South Storage Area, the geological cross-sections emphasized the eastward deepening (towards the sea) of the Pleistocene deposits.

### Hydrostatic Heads in the Unconfined Aquifer East of the Oil Terminal South Storage Area

Table 5.1 shows the ground elevations and the water table elevations for the six boreholes.

These measurements emphasize the steep eastward decrease of the groundwater level in the unconfined aquifer, revealing a high hydraulic gradient.

This significant decrease of the water table elevation follows the steep ground slope from the vicinity of the eastern boundary of Oil Terminal South Storage Area and the sea shore.

## 4.4 FLOW MODEL OF THE UNCONFINED AQUIFER IN THE ZONE OF THE OIL TERMINAL SOUTH STORAGE AREA

*Irina Dinu*

The unconfined aquifer, located in Pleistocene loess, in the zone of the Oil Terminal South Storage Area, was modeled using Visual Modflow, which is based on the 3D finite difference method.

The represented domain is more extended eastward than the perimeter of the Oil Terminal South Storage Area. It is bounded by the Boreholes F15, F16, F17 West, F6, F3 and F1 South, f30 and f25 East and F13 and F14 North.

The domain was discretized in a grid with 64 rows and 88 columns of parallelipipedic cells with the horizontal dimensions 15 m x 15 m and a height equal to the thickness of the aquifer, determined based on lithological data provided both by the boreholes made in 2000, as well as older ones, made in 1995.

The boundary conditions are constant-heads, selected based on the hydrostatic heads measured in 2002 (Figure 4.7).

Thus the set constant-heads are the following:

- Along the northwestern boundary, increasing from 24.95 m at F14 to 32.47 m at F15, then decreasing from 32.47 m at F15 to 31.17 at F16;
- Along the western boundary, increasing from 31.17 m at F16 to 26.6 m at F6;
- Along the southern boundary, decreasing from 26.6 m at F6 to the minimum value of 21 m South of F18, then increasing from 21 m to 27.98 m at F1;
- Along the southeastern boundary, 28 m from F1 to a cell located South of F9, then decreasing from 28 m to 15 m at f30;
- Along the eastern boundary, increasing from 15 m at f30 to 11.5 m at f25;
- Along the northeastern boundary, increasing from 11.5 m at f25 to 25.52 m at F13, and
- Along the northern boundary, slightly decreasing from 25.52 m at F13 to 24.95 m at F14.

The recharge from effective infiltration was also introduced in the model. In this case, the recharge is mostly due to precipitation, but there are additional losses from the existing water pipeline network. An average value of 50 mm/year was introduced for the recharge from effective infiltration. Moreover, point sources, represented by injection rates, were introduced in the vicinity of the Boreholes F9 and f15, in order to calibrate the high hydrostatic heads. The injection rates were relatively low, of 0.08 l/s near F9 and 0.2 l/s near f15. These values were determined by the model calibration and may represent local losses from the water pipelines located in the eastern half of the Oil Terminal South Storage Area, a very plausible explanation for the locally increased values of the hydrostatic heads.

The model calibration was achieved by adjusting the values of hydraulic conductivity, until the calculated piezometric heads provided by the model were close enough to the

measured piezometric heads, at all the locations of the study boreholes, the head differences being less than 1 m.
The piezometric map provided by the model is presented in Figure 4.8 while the flow directions and the pathlines in the aquifer are shown on Figures 4.9 and 4.10. The aquifer discharges to the North, East and South, with consequences on the extent of the hydrocarbon contamination (Figure 4.9).
The values of hydraulic conductivity resulted from the calibration of the model (Figure 4.11) are between 0.01 and 6 m/day. These values are in the normal range for loess deposits. Higher values, between 3 and 6 m/day are present in the southern zone, in the vicinity of Boreholes f3 – f6, F8, F18. In this area, the measured hydrostatic heads have the lowest values within the perimeter of Oil Terminal South Storage Area.
After the calibration of the flow model, the water balance of the unconfined aquifer was calculated (Table 4.2).

Table 4.2
*Calculated Water Balance of the Unconfined Aquifer in the Zone of Oil Terminal South Storage Area*

| Recharge/discharge | Inflow ($m^3$/day) | Outflow ($m^3$/day) |
|---|---|---|
| Effective infiltration + point sources | + 176.25 | |
| Northwestern boundary | + 67.47 | - 77.90 |
| Western boundary | + 19.82 | |
| Southern boundary | | - 109.25 |
| Southeastern boundary | | - 17.95 |
| Eastern boundary | | - 9.35 |
| Northeastern boundary | | - 21.57 |
| Northern boundary | | - 27.52 |
| TOTAL | + 263.54 | - 263.54 |

The aquifer is mainly supplied by effective infiltration and point sources and, to a less extent, from the West. The northwestern boundary represents, partially, a recharge zone, on its southern half (between Boreholes F15 and F16), where the piezometric head reaches the maximum elevation in the study area, but on its northern half, it is a discharge zone (Figure 4.9). The main discharge zone of the aquifer is the southern boundary. The northeastern, eastern and southeastern boundaries are also discharge zones, but the sum of the outflows through these ones represents almost half of the outflow through the southern boundary. The aquifer also discharges through the northern boundary.
Therefore, dissolved pollutants may be discharged northward, eastward, southward and northwestward. This may lead to the further speading of the petroleum contamination. At the same time, the model emphasizes the influence of effective infiltration on the hydrostatic heads and therefore on the groundwater flow directions.
The main negative consequence of the hydrodynamic conditions in the zone of the Oil Terminal South Storage Area, also indicated by the available thickness measurements of the hydrocarbon layer accumulated on top of the water table, is the movement towards the Black Sea of the contamination.

**Figure 4.8** *Piezometric Map of the Unconfined Aquifer in the Oil Terminal South Storage Area, Resulted after the Calibration of the Model*

**Figure 4.9** *Groundwater Flow Directions in the Oil Terminal South Storage Area, Resulted after the Calibration of the Model*
**Figure 4.10** *Pathlines in the Unconfined Aquifer in the Oil Terminal South Storage Area, Resulted after the Calibration of the Model*
**Figure 4.11** *Distribution of the Hydraulic Conductivities (m/day) in the Oil Terminal South Storage Area, Resulted after the Calibration of the Model*

## 4.5 GEOELECTRICAL STUDY IN THE AREA OF THE LOADING RAMP OF OIL TERMINAL SOUTH STORAGE AREA

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The complex physical and chemical processes occurring in aquifers contaminated with various pollutants cause the appearance of zones of various electrical resistivity values. The contamination can be delineated using adequate electrometric devices, for non-distructive surface geoelectric measurements. In this case, a vertical electric sounding (VES) device was used.
In order to determine zones contaminated with petroleum products, a 680 m long profile was chosen, parallel to the loading ramp of the Oil Terminal South platform.

This profile is along Boreholes F12, F11 and F9:
- Borehole F9/38.48, with the hydrostatic head at a 8.22 m depth, corresponding to VES o18;
- Borehole F11/36.49, with the hydrostatic head at a 8.57 m depth, corresponding to VES o11; in this borehole the measured thickness of the free hydrocarbon layer was 16 cm in 2001 and 12 cm in 2003, and
- Borehole F12/36.34, with the hydrostatic head at a 9.2 m depth, corresponding to the VES o1; in this borehole the measured thickness of the free hydrocarbon layer was 32 cm in 2001 and 40 cm in 2003.

The application of the VES method is based on the apparent resistivity. In our case study, where the aquifer is in loess formations, with pores impregnated with more or less mineralized solutions, an ionic (or electrolitic) conductance occurs. The apparent resistivity (which is the inverse of the electric conductance) is mainly influenced by the ion concentration and less by the chemical composition of the dissolved elements.
The background is a macroscopic loess, with an apparent resistivity between 30 and 54 Ohmm, in which zones with petroleum product infiltrations can be distinguished by measurable geoelectrical resistivity variation (less than 20 Ohmm).
The vertical electric sounding measurements emphasized three geoelectric regimes, each being related to a certain type of lithological structure affected by infiltration of complex petroleum products – light non-aqueous phase liquids (LNAPLs) and dense non-aqueous phase liquids DNAPLs – in the infiltrated water.
This fact determined the change in the natural resistivity of the structure and, therefore:
- The dry clayey silts and silty clays (macroscopic loess) are emphasized in the geoelectric cross-section by values over 20 Ohmm;
- The moist clayey silts and silty clays (macroscopic loess) constitute the level where the petroleum products are located; the 20 Ohmm value limits the depth and extension along the flow direction of the contamination;

- The abnormal values under 20 Ohmm (20 - 10 Ohmm) contour the limit between loess and the basal brown/red clay, while on this background, there are zones under 10 Ohmm, of intense contamination.

The zones of minimum resistivity, between 10 and 20 Ohmm and located at 8 – 9 m deep (in the upper part of the aquifer) represent the geoelectric effect of the macroscopic loess impregnated with petroleum products. The width of this strip is controlled by the fluctuations of the water table.

The geoelectric cross-section 1-1', in the zone of the loading ramp of the Oil Terminal South Storage Area, includes both geological (Figure 4.12) and contamination information (Figure 4.13).

**Figure 4.12** *Geoelectric Interpretative Cross-Section 1-1', in Geological Terms, in the Zone of the Loading Ramp of the Oil Terminal South Storage Area*

**Figure 4.13** *Geoelectric Interpretative Cross-section 1-1', in Contamination Terms, in the Zone of the Loading Ramp of the Oil Terminal South Storage Area*

## 4.6 DATA ON THE HYDROCARBON CONTAMINATION OF THE SOIL, SEDIMENTS AND GROUNDWATER INSIDE THE PERIMETER OF OIL TERMINAL SOUTH STORAGE AREA

### Identification of Possible Infiltrations of Hydrocarbons

*Gicu Opreanu*

Considering to the main groundwater flow directions emphasized by the study, the investigations were carried out East and South of the perimeter of the storage area.

The zone of the seaside slope, East of Oil Terminal South Storage Area was well known for the presence of groundwater contaminated with hydrocarbons on the ground surface. The seaside slope was close to the Harbor; therefore contaminated groundwater could possibly reach the sea.

An evaluation of the presence of contamination was also carried out South of Oil Terminal South Storage Area. These lands were used for agriculture, and no signs of surface contamination were noticed. The ground sloped down southward. In the vicinity of the village of Lazu, about 2000 meters South of the storage area, there was a very low, swampy area, covered by tenths of square meters of water and lacustrine vegetation (mainly reed). The water did not have a visible petroleum film.

### Contamination of Soil/Sediments with Hydrocarbons inside Oil Terminal South Storage Area

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The impregnation of sediments with petroleum products was observed by Spiridonică (1995a, 1995b) in the zone of the Oil Terminal South Storage Area, based on data provided by more than one hundred boreholes. Additional data on the contamination of sediments with hydrocarbons were provided five years later in a new study.

The contaminated intervals in the sediments inside the perimeter of Oil Terminal South Storage Area were identified and contoured based on the field observations during the drilling of the boreholes and sample analysis (Figure 4.14).

Table 4.3 shows the degree of UV luminescence determined on groundwater samples from the Oil Terminal South Storage Area.

Based on these data, three directions of groundwater contamination by hydrocarbons resulted:

- zone of Boreholes F11/2, F12/2, F13/2, f27/2, eastward and northeastward;
- zone of Borehole f7/2, southward and southwestward;
- zone of Borehole f16/2, southward and southeastward.

**Figure 4.14** *Distribution of the Pleistocene Soil/sediment Intervals Contaminated by Oil Products in the Oil Terminal South Storage Area, Year 2000*

Table 4.3

*UV Luminescence Degree of Samples*

| Sample No. | Borehole No. | UV luminescence |
|---|---|---|
| 1 | f1/2 | * |
| 2 | f2/2 | * |
| 3 | f3/2 | *** |
| 4 | f7/2 | **** |
| 5 | f8/2 | ** |
| 6 | f9/2 | * |
| 7 | f10/2 | * |
| 8 | f11/2 | * |
| 9 | f15/2 | ** |
| 10 | f16/2 | **** |
| 11 | f17/2 | * |
| 12 | f18/2 | * |
| 13 | f19/2 | * |
| 14 | f20/2 | ** |
| 15 | f21/2 | ** |
| 16 | f24/2 | * |
| 17 | f27/2 | **** |
| 18 | f30/2 | * |
| 19 | F1/2 | ** |
| 20 | F2/2 | ** |
| 21 | F4/2 | – |
| 22 | F5/2 | * |
| 23 | F7/2 | – |
| 24 | F8/2 | ** |
| 25 | F9/2 | * |
| 26 | F10/2 | * |
| 27 | F11/2 | *** |
| 28 | F12/2 | **** |
| 29 | F13/2 | **** |
| 30 | F14/2 | – |
| * intensity degree of luminescence | | |
| – no luminescence | | |

## Free Product Contamination on the Water Table inside Oil Terminal South Storage Area

*Gicu Opreanu*

The measurements of the thickness of the free product layer accumulated on top of the water table were carried out at the end of the year 2000 in the cased boreholes, of $8^{5/8}$"-diameter, inside Oil Terminal South Storage Area. The same measurements were carried out in January 2001.
Table 4.4 shows these results.
A bailer-like device was used for measuring the thickness of the free product layer. The patent of this device belongs to Gicu Opreanu (from Geoecomar, Constanța). The precision of the device is +/-0.5 mm. Layers with a thickness under 0.5 mm were documented as *films* (Table 4.4, Figure 4.15).
Many measure points of the thickness of the free product layer emphasize low amounts of petroleum product accumulated on top of the water table, denoted as "*film*". However, the presence of petroleum product in these points is uncertain.
In the northeastern sector of the Oil Terminal South Storage Area, important amounts of petroleum products accumulated on top of the water table were emphasized (23%). The measured thickness of the free product layer is between 10 and 40 cm (Table 4.4).
The distribution of the thickness of the free product layer on top of the water table expresses the state of contamination at the moment when the measurements were made. The results show that the most significant contamination occurs in the northeastern sector of the Oil Terminal South Storage Area, where the loading ramp is located.

**Figure 4.15** *Distribution of the Thickness of the Petroleum Product Layer Accumulated on Top of the Water Table in the Oil Terminal South Storage Area, in 2001 and 2003*

Table 4.4

*Thickness of the Free Petroleum Product Layer Accumulated on top of the Water Table inside the Perimeter of Oil Terminal South Storage Area*

| Borehole No. | Date | Thickness of the free petroleum layer (cm) | Date | Thickness of the free petroleum layer (cm) |
|---|---|---|---|---|
| F1 | December 7, 2001 | film | May 14, 2003 | |
| F2 | | film | | |
| F3 | | | | film |
| F4 | | film | | |
| F5 | | 0.05 | | |
| F6 | | film | | |
| F7 | | film | | |
| F8 | | 0.1 | | film |
| F9 | | film | | |
| F10 | | film | | film |
| F11 | | 16 | | 12 |
| F12 | | 32 | | 40 |
| F13 | | 2 | | film |
| F14 | | 0.05 | | |
| F15 | January 19, 2002 | 10 | | 1.5 |
| F16 | | | | |
| F17 | | | | |
| F18 | | | | |
| | | | | |
| M1 | | | | |
| M2 | | | | 10 |
| M3 | | | | 30 |
| M5 | | | | film |
| M6 | | | | |
| M7 | | | | |
| M8 | | | | |
| M9 | | | | 2.5 |
| M10 | | | | 13.5 |
| M11 | | | | 11 |
| M12 | | | | 1.5 |

## Contamination of the Unconfined Aquifer with Dissolved Hydrocarbons inside Oil Terminal South Storage Area

*Rodica Popescu, Consuela Milu*

Table 4.3 above includes a summary of the data on the hydrocarbon contamination level in groundwater samples from 30 boreholes.

Based on the intensity of luminescence (directly proportional to the level of contamination), it was possible to draw a map with isolines that allows us to appreciate the movement of the contamination plume (Figure 4.16).

Inside the area investigated, two other zones with high infiltration degree can be contoured. These zones are related to the tanks located close to the Boreholes f3/2 (R34), f7/2 (R30) and f16/2 (R38).

The luminescence isolines indicate three main directions for the spreading of the contamination:

- From the zone of Boreholes F11/2, F13/2, F12/2 and f27/2 towards East (where springs are located) and Northeast;
- From the zone of Borehole f7/2 towards South and Southwest;
- From the zone of Borehole f16/2 towards South and Southeast.

While the first zone is on the main groundwater flow direction, the other two contaminated zones are smaller and probably supplied by leaks in the neighboring tanks.

**Figure 4.16** *Relative Degree of Dissolved Hydrocarbon Contamination Based on UV Luminescence in the Oil Terminal South Storage Area*
**Figure 4.17** *Depth of the Sedimentary Intervals Contaminated with Petroleum Products in the Oil Terminal South Storage Area*
**Figure 4.18** *Distribution of Contamination of the Unconfined Aquifer in the Oil Terminal South Storage Area, with Respect to the Main Groundwater Flow Directions.*
*(a – contamination by dissolved hydrocarbons; b – hydrocarbons floating on top of the water table; c – main groundwater flow directions)*
**Figure 4.19** *Influence of the Pleistocene Paleo-Relief on the Groundwater Flow in the Oil Terminal South Storage Area*

## Groundwater samples from Oil Terminal South Storage Area analyzed by the gas – chromatography method

*Consuela Milu*

*Gas-Chromatography Analysis*

The samples analyzed by gas-chromatography (soil and water) were chemically treated with solvent (chloroform, petroleum ether or dimethyl-ethyl-ketone) and evaporated after a while down to a specified volume. Then the samples were injected into the column at a temperature corresponding to the method for petroleum products identification.

The location of the groundwater samples is shown on Figure 4.20. Based on the gas-chromatography results, it was possible to specify, for the first time, the types of contaminants in the Oil Terminal South Storage Area. Thus, the presence of the typical compounds was emphasized: diesel, crude oil and fuel oil. Pollution of groundwater over the standard limits (2000 mg/l, as established for the limit of intervention, by the Order 756/November 3, 1997) was determined for all the samples. The investigations by the gas-chromatography method indicated that, for the Oil Terminal South Storage Area, contamination by petroleum products is highest in the eastern zone and, locally, on smaller areas, in the south-western and south-eastern zones of the perimeter.
This conclusion is in agreement to the results of the other types of geoecological investigations.

**Figure 4.20** *Location of the Groundwater Samples Used in GC Analysis inside Oil Terminal South Storage Area 2000-2001*
**Figure 4.21** *F1 Chromatogram*
**Figure 4.22** *f9 Chromatogram*
**Figure 4.23** *F10 Chromatogram*
**Figure 4.24** *f15 Chromatogram*
**Figure 4.25** *F13 Chromatogram*

According to the F1, f9 and F10 chromatograms (Figures 4.21-4.23), a low quantity of petroleum product (less than 0.05 mg/L) is present in these samples. However, in f15 the petroleum product level is 0.0456% a medium level of contamination (Figure 4.24), and in F13 the level is 0.0873%, therefore a substantial level of contamination.

Table 4.5

*Groundwater Sample Results from GC Analysis inside Oil Terminal South Storage Area*

| **Constituents** | **Unit** | **Results** | | | | |
|---|---|---|---|---|---|---|
| TPH | mg/l | **F1** | **f9** | **f10** | **f15** | **F13** |
| | | < 0.05 | < 0.05 | < 0.05 | 1.5314 | 158.19 |
| C 10-C 14 | % | < 5 | < 5 | < 5 | 0.0609 | 50 |
| C 14-C 20 | % | < 5 | < 5 | < 5 | 1.3618 | 45 |
| C 20-C 26 | % | < 5 | < 5 | < 5 | 0.1087 | 5 |
| C 26-C 34 | % | < 5 | < 5 | < 5 | < 0.05 | < 0.05 |
| C 34-C 40 | % | < 5 | < 5 | < 5 | < 0.05 | < 0.05 |

According to the results above (Table 4.5), the contamination is relatively low for most Oil Terminal South Storage Area, except the NE part of the facility, where the contamination is substantial.

## 4.7 INTERPRETATION OF DATA ON HYDROCARBON CONTAMINATION OF THE PLEISTOCENE SEDIMENTS INSIDE THE PERIMETER OF OIL TERMINAL SOUTH STORAGE AREA

*Dan C. Jipa, Rodica Popescu, Gicu Opreanu, Irina Dinu, Consuela Milu, Marius Albu*

Petroleum product contamination behaves differently in the three lithological horizons identified in the sediment column in the zone of Oil Terminal South Storage Area. The distribution of the contaminated sedimentary units has the following main aspects (Figure 4.17):

- *Contamination located only in the upper clayey silt horizon*

Petroleum product contamination is present in the upper horizon as expected, taking into account the high porosity of this type of sediment; contamination is frequent in the other lithological horizons of the Pleistocene sediments.

- *Contamination in the silty clay horizon (with paleo-soils)*

Contaminated intervals often extend from the upper loessoid horizon to the one with paleo-soils. There are situations when the contamination is located only in silty clay (Figure 4.17).

- *Contamination in the basal red clay*

This situation is observed in several boreholes (F8, F9, f9/2, f5/2). Very high content of silt in the red clay (sometimes 40%) facilitated the petroleum product impregnations in this lithological horizon.

- *Two or three contamination intervals in the sedimentary column of a borehole*

In most of the cases there is only one interval of contaminated sediments in the sedimentary column of most boreholes. However, Figure 4.17 shows some lithological columns with two or even three contamination intervals.

- *Upper limit of the contamination*

The upper limits of the contamination intervals were identified from close to the surface down to almost a 10 m depth.

### *Distribution of the Contaminated Sedimentary Intervals*

The thickness and depth of the upper limit of the contaminated sediments were measured. The thickness of the contaminated sediments identified in the boreholes in Oil Terminal South Storage Area varied between 0.4 m (f200) and 10.1 m (F5/2). Almost 90% of the contaminated sedimentary units are 1 to 5 m thick. Almost half of the sedimentary units impregnated with petroleum products are 1 to 3 m thick.
The contaminated intervals are located at various depths. The depths of the upper limit of the contaminated intervals vary between 0 m (at the ground surface) and 9.5 m (F14/2).

### *Uncontaminated Sediments*

The delineation of the contamination was based on data from the boreholes that did not indicate the presence of petroleum products.

The southern and western limits of the contaminated zone were possible to be contoured according to borehole data (Figure 4.17). The delineation line surrounds most of the tanks from the southern and western parts of the Oil Terminal South Storage Area.

In the southern part of the Oil Terminal South Storage Area, inside the zone of deep contamination of the Pleistocene deposits, a sector with no contaminated sediments was identified.

*Free Petroleum Product Contamination*

The distribution of the thickness values of the free petroleum product accumulated on top of the water table shows a suggestive image of the dynamics of the contamination process that occurs inside the perimeter of the Oil Terminal South Storage Area (Figure 4.18, Table 4.4) summarized as follows:

- In most of the boreholes there were very low quantities of petroleum products accumulated on the water table, denoted as "*film*"; therefore the measurement is not very precise, so that the presence of free product is not completely certain in these points;
- In the northeastern sector of the perimeter of the Oil Terminal South Storage Area, important quantities of free product accumulated on the water table were identified, the thickness of the layer varying between 1.5 and 40 cm.

It is important to mention that the distribution of the thickness values of the free product accumulated on the water table in the Oil Terminal South Storage Area is particular to the moment when the measurements were taken. The results show that in the northeastern part of the storage area regular accidental losses are very important.

*Dissolved Hydrocarbon Contamination*

For the quick detection of the presence of dissolved hydrocarbons in groundwater, the samples were tested in order to establish the intensity of luminescence in ultraviolet radiations. The intensity of the luminescence is directly proportional to the amount of hydrocarbons dissolved in water. Four relative degrees of intensity of luminescence were distinguished. Using this relative intensity scale, water samples collected from 30 boreholes from the Oil Terminal South Storage Area were analyzed. For 24% of the samples, the analyses indicated relatively high contents of dissolved hydrocarbons in the zone of the Oil Terminal South Storage Area.

The resulting data may be summarized as follows:

- Only 10% of the analyzed samples were non-detect;
- Around 43% of the water samples had low hydrocarbon content;
- Around 23% of the water samples had medium hydrocarbon contents, and
- The contamination level of the water is high in 24% of the analyzed samples.

Groundwater contamination by hydrocarbon is high in three restricted zones (Figure 4.18). The most important zone of contamination was contoured towards the eastern boundary of the Oil Terminal South Storage Area (Boreholes F11/2, F13/2, f27/2 and in the zone of the springs, on the seaside slope). Two other zones, of lower extent, were identified in the western (Borehole f7/2) and in the southeastern (Borehole f16/2) parts of the storage area.

Therefore, there was a generally reduced level of groundwater contamination associated with the most part of the perimeter of Oil Terminal South Storage Area. Significant groundwater contamination was found especially in the eastern part of the

storage area and, to a lesser extent, in the western and southeastern parts of the storage area.
According to the distribution of the intensity degree of the UV luminescence, the relatively high contents of dissolved hydrocarbons were concentrated in the as follows inside Oil Terminal South Storage Area:

- The most important zone appears in the Northeast of the Oil Terminal South Storage Area, overlying the zone where the thickest layers or free product floating on the water table were emphasized;
- A restricted zone was contoured in the southeastern corner of the investigated perimeter, around only one borehole location, and
- A relatively restricted zone, in the western part of the perimeter.

## Migration of Hydrocarbon Contamination in Sediments and Groundwater inside and from Oil Terminal South Storage Area

*Dan C. Jipa, Marius Albu, Irina Dinu*

Contamination of sediments and groundwater due to accidental infiltrations is mainly controlled by the groundwater flow. The petroleum products floating on top of the water table are carried away, while they also penetrate in water and in sediments. The fluctuations of the water table accentuate the contamination of sediments.
The groundwater flow model of the unconfined aquifer in the Oil Terminal South Storage Area emphasized two main groundwater flow directions, to the East and to the South.
The main groundwater flow direction towards the East may constitute the most serious threat associated with the contamination at Oil Terminal South Storage Area. The contaminated groundwater is discharged, through springs, in the eastern vicinity of the Oil Terminal South Storage Area. Here, the thickness of the free product layer is the highest. This indicates that very serious the accidental discharges of petroleum products have been occurring in the northeastern zone of the storage area.
The main groundwater flow towards South is associated with a much lower presence of petroleum products on top of the water table.
The flow model showed that, in the northern part of the perimeter of Oil Terminal South, there is a minor tendency of groundwater flow to the North. The natural relief conditions (the existence of a relatively steep slope) favor an eventual occurrence of contaminants on the seaside slope.
The only fairly precise delineation of the area contaminated with petroleum products has been accomplished in the western part of the Oil Terminal South perimeter (Figure 4.18). All the tanks located in the western sector of the Oil Terminal South Storage Area are inside the contaminated zone, possibly except for the tanks R30-R34.
The downward migration of the petroleum products represents an important aspect of all the cases of subsurface contamination. For this area, the basal red clay could be the impermeable bed. However, the impermeable characteristics or the red clay are not clearly established, taking into account its high silt content, as well as the presence of hydrocarbon infiltrations in these sediments. The available data concerning this important aspect in the investigated area is currently insufficient.

## Geological and Hydrogeological Factors in the Zone of the Oil Terminal South Storage Area

*Dan C. Jipa, Marius Albu, Irina Dinu, Gicu Opreanu*

The geology or more specifically the lithology of the contaminated deposits is the most significant factor in the movement of petroleum products, expressed by means of two physical properties of the sediments from the subsurface of the affected zone:

- Porosity, which is the ratio between the pore space and the total volume of sediment, and
- Permeability or the ease with which a fluid cad diffuse through a particular material.

The lithological succession emphasized by the boreholes in the zone of the Oil Terminal South Storage Area shows that the liquid phase of the oil contaminants has the best migration conditions in the upper part of the Quaternary deposits (the upper loessoid or clayey silt horizon). The migration conditions of the contaminants are more restrictive in the middle horizon (silty clay), while the basal red clays are considered virtually impervious for the contaminants. These are the prevailing conditions inside the investigated perimeter. However, local variations appear, mostly due to the existence of intercalations with different grain size deposits compared to the prevailing deposits. The alternation of deposits with differing grain size also induces lateral migration of petroleum products, enlarging the contaminated zone selectively, at various levels.

In the basal horizon, the red clays contain high amounts of silt (up to 40 – 45%), which lead to an increase in permeability, and to the presence of intervals of contamination in the red clays.

The piezometric map of the aquifer and the flow model suggest that the main groundwater flow directions are to the South, Northeast and Southeast (Figure 4.9). The model also shows the importance of the inflow by effective infiltration, especially due to losses from the water pipelines. Unlike in the area of Oil Terminal North, groundwater flow in the area of Oil Terminal South is controlled by the spatial variation of recharge. Therefore, dissolved pollutants are discharged towards the seaside slope, possibly reaching the Black Sea.

The Pre-Quaternary paleo-relief represents another geological factor with a significant influence on the distribution of the oil contaminants. The term "Pre-Quaternary paleo-relief" refers to the irregularities of the bottom of the lower horizon that constitutes the existent relief before the deposition of permeable sediments, which form the unconfined aquifer.

On a local scale, in the zone of Oil Terminal South Storage Area, the paleo-relief consists of a surface with a prevailing southward lowering, representing the passage from the northern hilly paleo-zone towards Agigea Valley. Because of this important paleo-morphologic character, the Quaternary sediments from the perimeter of the Oil Terminal South Storage Area are gently lowering southward. Moreover, their thickness increases towards South, determining the main characteristics of the geological structure in this zone.

The migration process of the petroleum contaminants is determined mainly by the fact that, inside the investigated perimeter, the water table decreases southward.

On the surface of the southward slope, there are irregularities that represent morphological elements less representative of the pre-Quaternary paleo-relief. The studies carried out inside the perimeter of Oil Terminal South Storage Area (Figure

4.19) emphasized the existence of an oblong ridge, approximately on the North to the South direction. Local-scale ridges and valleys are forming on the main ridge. The morphological uplift determined second order relief slopes. Among these, the most obvious one is an eastward slope-down. This slope seems to represent the paleo-morphologic element that generated an eastern lateral component of the groundwater flow. Therefore, a part of the petroleum products penetrating the sediments are carried away by groundwater.

Nevertheless, the minor paleo-morphological elements (third order) induced more local groundwater flow (and contamination) directions.

## 4.8 CONCLUSIONS ON THE HYDROCARBON CONTAMINATION IN THE ZONE OF THE OIL TERMINAL SOUTH STORAGE AREA

*Dan C. Jipa, Irina Dinu, Rodica Popescu, Consuela Milu, Gicu Opreanu,Doru Lutac, Nicolae Prodan*

The borehole data revealed the existence of sedimentary intervals contaminated with hydrocarbons.

In most of the boreholes, only one interval of contaminated sediments was found. In the whole perimeter, the vertical distribution of the contaminated sediments varies considerably; hydrocarbon contamination appears both in the upper part of the Pleistocene sedimentary succession and in deeper intervals. The upper limit of the contaminated units is located at depths varying between 0 m (ground surface) and 10 m. The areas with surface soil contamination are concentrated in few sectors, the most important ones being located in the western and north-eastern parts of Oil Terminal South Storage Area (Figure 4.18). We mention that several boreholes intercepted two or even three levels of contaminated sediments (Figure 4.17).

The thickness of the contaminated intervals is rather high, varying between 0.4 m and 10.1 m. Almost 90% of the contaminated intervals are 1 to 5 m thick.

Hydrocarbon contamination is present in all the types of sediments identified in the boreholes from Oil Terminal South Storage Area. As expected, the level of contamination is higher in coarser sediments; these ones are represented both by clayey silt and silty clay (paleo-soils). The red clay sediments, located at the bottom of the Pleistocene deposits, also contain contaminated intervals (Figure 4.17).

The contamination process of the sediments is influenced by the fluctuations of the water table caused by the variation in the amount of precipitation (and, therefore, infiltration) and by leaks in the water pipelines. In most of the cases, the water table is located inside the contaminated sedimentary intervals.

The main groundwater flow direction determines the movement of the contaminants to the East and their discharge at the base of the cliff. The groundwater flow model inside Oil Terminal South Storage Area emphasized the influence of the recharge on the piezometric heads and therefore on the main groundwater flow directions. The model suggests that the high water table elevations in the southeastern sector are due to losses from the water pipelines.

The pre-Quaternary paleo-relief also leads to the main groundwater flow direction to the East. It consists of an oblong ridge, approximately North – South orientated and an eastward slope down, which causes the groundwater flow to the East (Figure 4.20).

The variation in the thickness of the hydrocarbon layer on top of the water table (free product layer) confirms the influence of the groundwater flow on the transport of contaminants. Thus, the highest thickness of the free layer appears in the northeastern sector of the storage area. At the same time, the most significant zone of dissolved hydrocarbons is towards the eastern boundary of Oil Terminal South Storage Area.

In conclusion, the studies carried out in the zone of Oil Terminal South Storage Area show that the contamination is considerable, affecting the entire series of Quaternary deposits and represents a significant threat for this sector of the Romanian Black Sea Coast.