

SOIL CHEMICAL POLLUTION REFLECTED IN GROUNDWATER ELECTRICAL PROPERTIES

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Abstract. In a test area, bulk electrical conductivity was determined by *in-situ* measurements. Investigations were made by inducing very small electrical “eddy” currents in the ground and measuring the magnetic field generated by these currents. The quad-phase results were used to analyze the soil electrical conductivity variations. Based on these observations, small depth boreholes were projected and executed. Water and soil samples were extracted and analyzed in laboratory conditions. Evaluating the total hydrocarbons (THP) results, it was noticed that the high values correspond to the area characterized by low quad-phase values. In all investigated domain a high content of chlorides was detected, affecting the detectability of petroleum hydrocarbons residual pollution. By integrated evaluation of all results it was concluded that the high conductivity area observed by electromagnetic measurements is closest to the pollution source. As we move away from the source, groundwater conductivity decreases (less chloride content) but hydrocarbons presence was still at a high rate. Therefore, by analyzing ground water electrical properties salty water pollution sites can be evaluated, while hydrocarbons pollution needs to be assessed either by measuring bulk soil conductivity or by specific chemical analyses.

Key words: electromagnetic measurements, oil pollution, salt water pollution, groundwater electrical conductivity, hydrocarbons pollution.

1. INTRODUCTION

Soil affected by stock or fund pollutants can be evaluated by specific chemical and physical analyses. In many cases there is the need to transfer the chemicals (absorbed by the solid matrix or trapped in soil micro or macro pores) from solids into aqueous phase, in order to be able to evaluate their presence and concentration.

In this study, two types of pollutants with different electrical effects were analyzed: salty water and petroleum hydrocarbons. According to their electrical properties, the salty water is classified as a very good electrical conductor while crude oil and most of the petroleum derivative products (fuels, oils), in normal conditions, behave as electrical insulators.

Oil products are also characterized by a low degradation rate, in natural conditions [1]. Therefore, unrecovered pollutant constitutes a continuous source of contamination, affecting human health and plant growth by induced changes into soil and groundwater quality.

Cases of oil and salt pollution are frequently encountered in areas with hydrocarbons extraction wells or refinery activities, but the major preoccupation when assessing the environmental problem in such cases consisted in delineating the petroleum hydrocarbons polluted zones, even though soil degradation by salinization decreases soil fertility and in specific climatic conditions may lead to desertification [2].

Oil pollution cases were worldwide reported at loading docks, along transportation pathways [3], near refinery storages facilities or petroleum waste depository facilities [3, 4, 5]. Affected areas were studied using direct investigation methods (laboratory analyses) [6], or indirect methods (geophysical measurements) [4, 5, 7], but best results were noticed when both types of methods were applied [8, 9].

1.1. OIL POLLUTION

Frequently crude oil is framed into light-non-aqueous-phase-liquids class (LNAPL), having a density lower than the water. In cases of severe soil pollution, three pollution phases can be observed:

- Separate Hydrocarbons Phases – pollutant in a liquid form (observed sometimes as a floating layer or lenses of hydrocarbons on top of ground water) or trapped in porous space of vadose zone (adsorption or retained by capillary forces).
- Dissolved Phase Hydrocarbons (PDH) – resulted from hydrocarbons soluble fractions.
- Volatile Phase Hydrocarbons (PVH) – formed by oil volatile compounds.

If surface spills occur, hydrocarbons start a lateral migration and a downward movement. Immobile residual organic compounds will be encountered along contaminant migration paths.

The contaminant migration in soil is influenced by many factors. Most important are the source activity (continuous, discontinuous, accidentally), the volume of the contaminant solution discharged into the medium, its properties (soluble and volatile compounds, viscosity) but also the medium characteristics such as soil texture, its porosity and permeability, soil water-holding capacity and terrain slope.

For evaluating an area affected by oil spills, typical analyses employed on soil and water samples are the total sum of benzene, toluene, ethylbenzene and xylenes (BTEX) or total hydrocarbons (TPH).

1.2. SALT WATER POLLUTION

The presence into the environment of salty solutions can be a result of natural phenomena [10], like saltwater intrusion into coastal aquifer along fractured zones [11] or water dissolution of salty rocks [12, 13]. Frequently is also observed as a result of anthropogenic activities such as waste water discharge (its salt content being influenced by the specific utilization of the water into the industrial or domestic activities), distribution of salts on roads (as defrosting technique) [2] or as a consequence of human induced changes into hydrogeological natural equilibrium (over pumping of groundwater into coastal aquifers) [14, 15, 16].

Considering the content of Cl^- ions, water is classified as fresh water (<150 mg/l), brackish water (150–10,000 mg/l) or saltwater (>10,000 mg/l) [16]. Salts content have a big influence over the water electrical properties. If not measured directly, the water conductivity can be estimated by considering the ions concentration and ions mobility. As the ions mobility is controlled by the temperature, for comparing the data measured in field with laboratory determinations (standard expressed as 20°C or 25°C), the temperature correction should be applied.

2. ELECTRICAL PROPERTIES OF CRUDE OIL AND WATER

Both types of contaminants (petroleum products and salty water) offered distinct results when submitted to electrical tests. Crude oil is practically an insulator as resulted in normal laboratory conditions of determination [9] and also in nature, in geological traps [17]. Same electrical behavior was observed by the authors for derivate products such as diesel, gas and kerosene measured using resistivimeters with output current in the range of 1 mA–2 A. Resistivity values for the oil processed products may vary, as special electrical conductive additives are mixed in order to reduce ignition risk caused by the static discharge during transportation and utilization.

Water electrical properties are extremely variable. The admissible limit (in Romanian national standards: 458/2002; 311/2004) of electrical conductivity for fresh water is 2500 $\mu\text{S}/\text{cm}$ at 20°C. The groundwater (non-treated for consumption) samples analyzed by the authors, collected from different aquifers within Romania (predominantly phreatic aquifers), are in the range of 320–1500 $\mu\text{S}/\text{cm}$. Phreatic aquifer electrical properties have seasonal variations, due to the recharge with rainwater or melted snow.

Different electrical values were obtained from low mineralized mountain springs (resistive, 65 $\mu\text{S}/\text{cm}$) and highly mineralized springs samples (highest value of conductivity encountered in our samples was 7100 $\mu\text{S}/\text{cm}$).

Therefore, comparing with electrical response of petroleum contaminants, water that can be found into the soil and rocks may be always considered as electrically conductive.

3. ELECTRICAL CONDUCTIVITY OF POLLUTED SOIL

The soil/rock is a multiphase system. The major component is the solid phase, formed by minerals, predominantly present in a crystalized form. In porous space, gaseous and liquid phases are present. All of them give contributions to the bulk electrical conductivity of soil/rock.

Considering the capacity of conducting electrical current, minerals are classified as good conductors (metals), semiconductors or dielectrics [18]. If the soil/rock is dry, the electrical conductivity depends on the mineral type, the nature of chemical bound and soil/rock texture. As most soil/rock forming minerals are poor electrical conductors, in porous and wet soil/rocks the bulk conductivity is highly influenced by the electrical properties of the fluids. Pore fluids acts as electrolytes, the non-conductive mineral grains having a smaller contribution on the soil/rock electrical conductivity [18].

According to Archie's Law (1942) [19], the effective resistivity of rock formations is given by an empirical formula which considers the rock porosity, the volume of the pores which contains water and the water itself resistivity. The proportionality between the resistivity of the saturated rock with non-conducting matrix and the electrical resistivity of the brine has been first establish for saturated "clear" rocks:

$$\rho_m = \rho_w \cdot F,$$

where ρ_m is the measured electrical resistivity, ρ_w is the specific electrical resistivity of pore water and F the formation factor. The formation factor combines the material properties which influence the electrical current flow [16].

Archie's equation was a starting point for analyzing the cases when rocks are partially water saturated and that a part of the porous space might contain a resistive fluid (crude oil). A great variety of models which relates the dependence of the rock conductivity on pore fluid content and its electrical properties, porosity and rock structure, were developed [18], for the so-called cases of only one conducting component (pore water).

In case of solid matrix material with conducting minerals the equations developed for the "clean rocks" (clay and shale free) are no longer valid. For quantification of the "double-layer or interface conductivity" observed in presence

of clay minerals and for relating the cation exchange capacity observed for solid components to the measured electrical conductivity, numerous models were built. A review of the theories and results are given in [16, 18].

Experiments to observe the influence of contaminants on the electrical conductivity showed that the presence of contaminants as methanol, benzene, perchlorethylene [20, 21] will decrease the bulk electrical conductivity of affected rock. The results of laboratory experiments on oil products were confirmed by in-field measurements [9].

4. CASE STUDY

Direct and indirect investigation methods were applied in an area contaminated both by oil and salty water, located in Prahova County, Romania (Fig. 1). Two panels were selected for detailed geophysical investigation. In Panel I there is no evidence of pollution at the earth surface. Panel II was chosen for comparison, as in this area oil pollution is partially visible at the earth surface.

Based on geophysical results, small depth boreholes were projected and executed in Fig. 1, Panel I. Water and soil samples were extracted and analyzed in laboratory conditions.

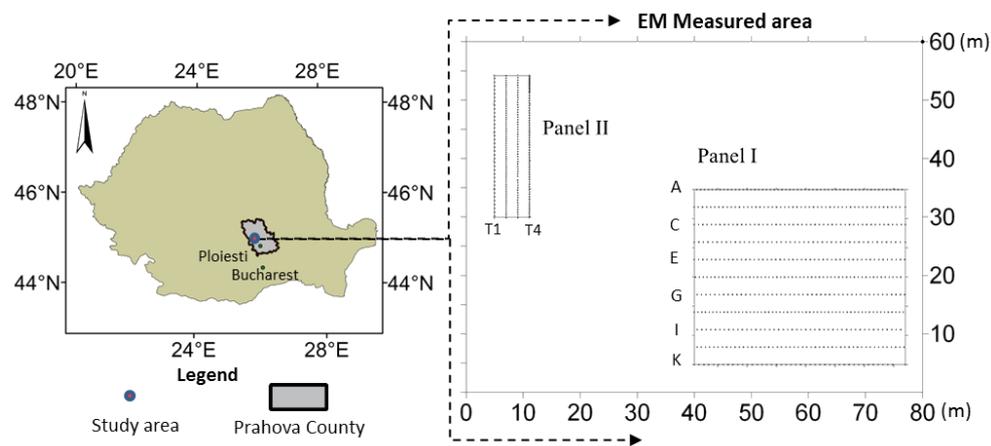


Fig. 1 – Location of study area and electromagnetic (EM) measured profiles:
Panel I – profiles code: A-K) and Panel II – profiles code T1-T4.

The source of soil and groundwater contamination is represented by a natural oil accumulation. The anticline structure which hosts the hydrocarbons is intruded by a salt diapir [22, 23], the latter being the main cause for the high chloride content of groundwater.

Even the oil accumulation has been exploited a few decades ago, the pressure maintained by the residual oil associated gases determines an upward displacement of oil, brine and groundwater. The ascension of fluids is made mainly through the vertical faults created by the salt diapir movements up to the surface

Following such tectonic paths, the groundwater is enriched with salts along the sections where exists a direct contact with the geological salt body.

4.1. GEOPHYSICAL METHOD

Methods which imply natural Earth fields or artificial fields for underground *in-situ* investigation are known as geophysical methods.

For measuring *in-situ* electrical conductivity of soil (ECa), a portable electromagnetic induction instrument was used. An electromagnetic (EM) transmitter located at the rear of the instrument outputs a time-varying electrical current into a small coil. The current generates an oscillating magnetic field which propagates in the air and in solid Earth. In case of a homogenous ground, a slighty reduction in wave amplitude will be observed for the one which propagates through the soil. In inhomogeneous soils, when the waves encounters a conductive material, the magnetic component of the incident EM wave induces small alternating electrical currents in the conductor body. The eddy currents will generate a secondary electromagnetic field wich is of interest for evaluating soil properties.

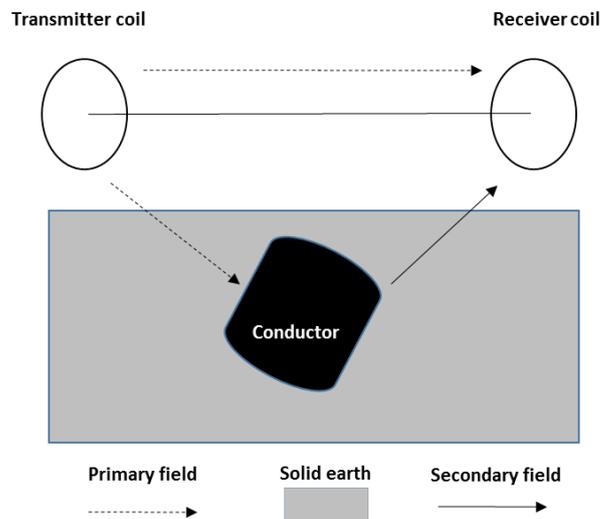


Fig. 2 – Sketch of shallow electromagnetic induction for field measurements.

The instrument receiver coil will therefore get the energy from two sources: the strong primary field generated by the transmitter and the much smaller

secondary fields given by the eddy currents (Fig. 2). To facilitate this measurement an internally generated signal is used to cancel or “null” the large primary signal so that it does not overload the electronic circuit [24]. Under the technical constrains incorporated into the design of the used EM instrument known as “operating at low values of induction” the magnitude of the secondary field is directly proportional to the soil conductivity [25]:

$$EC_{\alpha} = \frac{4}{\omega\mu_0 d^2} \frac{H_S^Q}{H_P}$$

H_S^Q is the measured in-quadrature component of the secondary magnetic field at the receiver coil to the primary H_P magnetic field at the transmitter coil, d is the spaces between the dipoles, μ_0 is the permeability of free space and the angular frequency $\omega = 2\pi f$, where f is frequency.

The recorded field is electronically split into the in-phase (I/P) and out-of-phase/quadrature (Q/P) component (90° phase shift with respect to the primary field) (Fig. 2).

The quadrature-component output (Q/P) gives information upon the soil electrical conductivity (expressed in mS/m). The ECa obtained is a weighted average across the investigated soil thickness. The in-phase (I/P) component of the secondary EM field (expressed in parts per thousands) can be used to evaluate the presence in soil of buried metals with magnetic properties. This type of materials can also give high electrical conductivity values, therefore is essentially to know their presence in soil for avoiding misinterpretation of ECa data. Buried magnetic pipes and cables are seldom recognizable by the specific dipolar susceptibility anomaly (I/P) (Fig. 3).

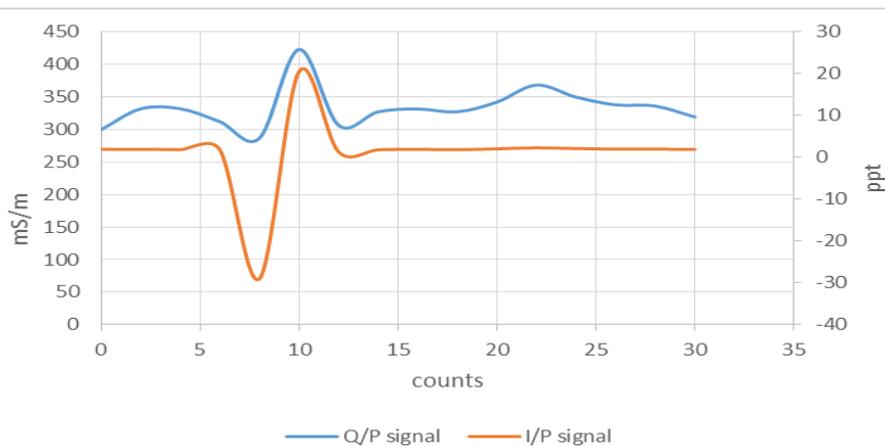


Fig. 3 – Electrical conductivity (mS/m) and magnetic susceptibility (ppt) anomalies recorded by EM field equipment over buried metallic cables.

The investigation depth is function of the coils spacing and orientation (vertical or horizontal dipole). For this investigation case the distance between the receiver and transmitter coil was of 1m and the vertical dipole mode was chosen. In the vertical dipole mode the relative sensitivity to near surface material is very low (being zero right at the surface); the sensitivity increases with depth, having a maximum at about 0.4 meters and decreasing slowly thereafter [24].

Before starting the measurements, the instrument was calibrated in a nearby area characterized by magnetic calm, following a specific protocol at ground level and at 1.5 m in the air.

Considering the operation frequency (14.6 kHz), for this particular configuration and soil composition, the maximum depth of investigation was approximatively 1.2 m. It is estimated that any conductive body located below this depth will not produce a recognizable EM anomaly.

EM investigations were performed in quasi-continuous measuring mode, on parallel profiles. Results were distributed considering the total length of the profiles and a constant acquisition velocity for each 5 m interval. In Panel I, eleven profiles (marked in Fig. 1 – codes A-K), placed at 3 meters distance were measured with a data coverage ranging from 139 points (profile D) to 163 points (profile A). Panel II was covered by measurements executed on profiles placed at 2m distance (marked in Fig. 1 – codes T1-T4), with a minimum data points of 170 (Profile T1) and maximum of 196 (Profile T4). Differences in data point density are due to natural obstacles sometimes encountered along the profiles.

4.2. DIRECT METHODS

Due to the double contamination problem (hydrocarbons and salts), direct methods applied can be divided in two categories considering the objectives:

- methods for assessing the hydrocarbon pollution, consisting in TPH determination of solid probes (soil) and electrical resistivity of oil pollutant (in the liquid form);
- methods for evaluation the salt pollution, consisting in chloride determination of solid substances and electrical conductivity measurements of water samples.

Borehole position (Fig. 4) was chosen after consulting the resulted maps, based on data from in-situ measurements (indirect investigation methods).

At short time after the boreholes were executed, groundwater accumulated into the resulted voids. The accumulation rate was different, and in one location free phase hydrocarbons filled completely the void. From each location water samples were extracted using a special 1.5 m pipette. As it was expected to exist a layering in water salinity, chemical analyses were performed using 1.2 liters of water, from each borehole location.

Soil samples were extracted at different depths from the 2 m length boreholes. For obtaining the chlorides and electrical conductivity of soil in laboratory condition, samples were dried and grind to 0.2 mm, and then mixed with demineralized water (electrical conductivity of 6 $\mu\text{S}/\text{cm}$ at 20 °C), respecting the proportion of 1:5.

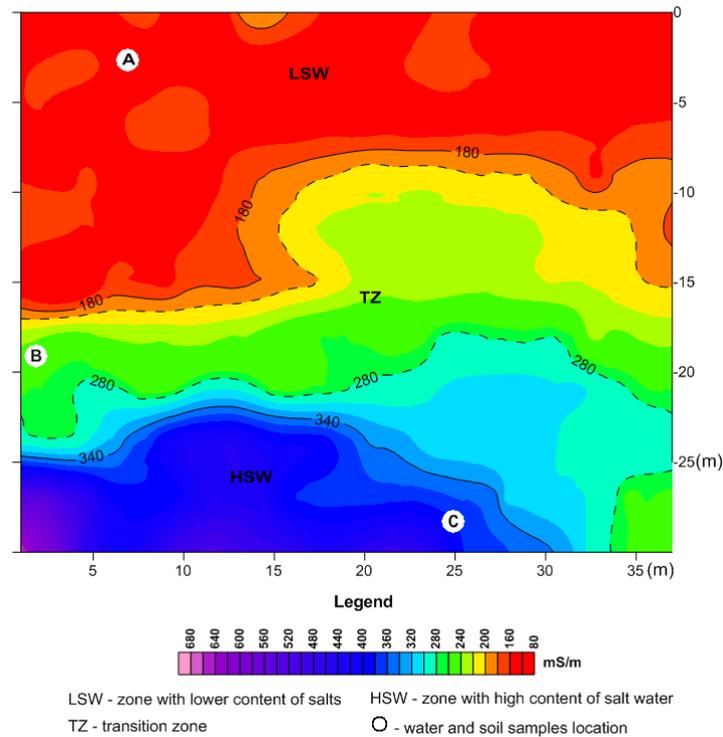


Fig. 4 (Panel I) – Electrical conductivity map obtained from EM measurements.

4.3. RESULTS AND DATA INTERPRETATION

In-situ investigation datasets were used for obtaining ECa graphs of variation or maps for the measured areas. A local coordinate system was used for each sector, in order to interpolate the bulk electrical conductivity results.

As it can be seen in Fig. 4, electrical conductivity shows a wide range of variability (80 – 665 mS/m). According to these results, the area was divided in 3 zones:

- highly polluted zone (HSW), characterized by electrical conductivity values higher than 280 mS/m;
- zone with low salt content (LSW), characterized by electrical conductivity values lower than 180 mS/m;

– transition zone (TZ) for the conductivity values in the range of 180–280 mS/m.

Each zone was further investigated by analyzing soil samples from different depths (0.5m, 1.0m, 1.5 and 2.0m) and later by water samples (code probes locations A, B, C). Results of TPH and chlorides from solid substance samples are shown in Table 1.

Table 1

Results of TPH and Chlorides chemical analyses for soil samples extracted in time of boreholes execution

Sample location	Soil sample code	Depth (cm)	TPH (mg/kg)	average TPH (mg/kg)	Chlorides (mg/kg)
A	AD1	50	22,3	134,37	80,34
	AD2	100	23,5		
	AD3	200	357,31		
B	BD1	50	519,67	4494,48	2595,37
	BD2	150	6232,78		
	BD3	200	6731		
C	CD1	50	392,11	5946,89	3948,75
	CD2	100	6016,31		
	CD3	150	7800,44		
	CD4	200	9578,68		

In all locations it was observed the tendency of increased TPH content with depth (Fig. 5), the highest level of petroleum contamination being observed in the area of the borehole located in the HSW zone. This result leads to the conclusion that both contaminants migrated into this area from the same source.

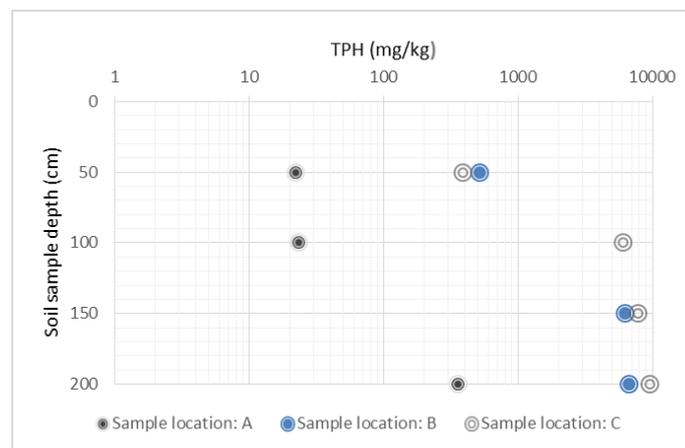


Fig. 5 – TPH variation with depth in soil samples from boreholes located in HSW zone (CS), TZ (BS) and LSW (AS).

Water samples, extracted from the boreholes, despite their relative close location, showed great variability in terms of electrical conductivity. The lowest values were obtained from borehole position code A ($500 \mu\text{S}/\text{cm}$) while values for the borehole position code C overpassed the value of $20000 \mu\text{S}/\text{cm}$. Total dissolved solids range of variation was between 321 ppm at pH value of 6.24 (borehole position code A) to 11000 ppm at pH value of 6.84 (borehole position code C).

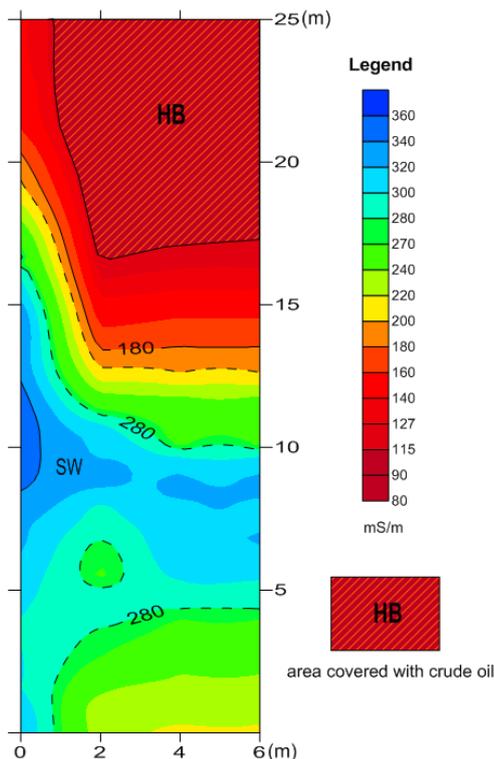


Fig. 6 (Panel II) – Electrical conductivity map obtained from EM measurements.

Same protocol of indirect investigation was applied for the second studied area (Panel II), where hydrocarbon pollution was observed at the earth surface. The EM sensors were kept at 0.30 cm above earth surface during measurements. Low conductivity values (80–95 mS/m) were observed on profile sectors where free phase hydrocarbons were visible. It was noticed that the ECa values were increasing as the hydrocarbon thickness diminished (Fig. 6). Despite the lower ECa values observed on the Panel II (maximum value of 360 mS/m), the entire area was also affected by salty water pollution. Both contaminants came from the same source (buried source), but due to the higher viscosity of the oil, its migration through the soil is slower and therefore the affected area is restrained.

5. CONCLUSIONS

For the investigated depth a three phase system was considered, the porous space being filled with water, oil and gases (air or VHP). The amount of each component contributed to the total bulk conductivity measured by means of EM field method.

Due to the high electrical conductivity of the soil, caused by salty solutions present in the soil porous spaces, the indirect investigation measurements were less sensitive to the presence of residual hydrocarbons (Panel I) and even to the mobile fractions of hydrocarbons (Panel II).

However, the maps resulted from the EM investigation were in accordance with results from the direct methods. High ECa values correspond to high chloride content determined from the solid substance, high electrical conductivity of water sample and high TDS values.

In areas where crude oil was observed at the earth surface, the EM equipment recorded lower ECa. Considering that this specific pollutant is an electrical non-conductor, from the theoretical models it was estimated to obtain much lower ECa values. This effect is a consequence of the petroleum hydrocarbon disposal inside the porous space. The oil is the second wetting fluid in this three phase systems, the first wetting fluid being the high electrical conductive groundwater. Soil grains were wetted by thin film of electrolyte, creating fast pathways for the electrical current flow.

Following this experiment we can conclude that a decrease in electrical conductivity is expected to be encountered in fields polluted with petroleum products. If the soil is humid and have high salts content (which can be transferred by dissolution into aqueous phase) the areas polluted with hydrocarbons may be underestimated.

Electromagnetic measurements showed a great potential in rapidly locating areas affected by petroleum products pollution or by salty water. Results can be used for creating sections and maps which allow a better understanding of the environmental problem and even the location of the pollution sources.

Detailed investigations (based on samples) can benefit of the zonation resulted by using non-invasive indirect methods, limiting the number of samples and therefore the costs.

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