STUDY OF OIL POLLUTION IN MEXICO WITH RESISTIVITY SOUNDING

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Abstract

This study was performed by EcoExel company for PEMEX. IMP group consulted application of resistivity sounding. The pollution in this place is known for a long time and some remediation was already performed. The purpose of present stage has to estimate an ecological state of ground and to plan areas, amount and sequence for the next remediation.

Resistivity sounding (VES) here was applied first, before only drilling and chemical sampling was used. The area is occupied by buildings and industrial constructions and is covered by concrete 20 cm thick, and the studies were performed along the internal factory roads. The high level of geologic and electromagnetic noise caused some problems in VES field application and data analysis. To decrease geological and EM noise the special field technology and data processing for two-sided pole-dipole array was applied.

VES profiles' system has allowed seeing investigated objects both in plan and depth. From several wells the groundwater conductivity was estimated, that allowed to calculate theoretically the rocks' (sand and clay) resistivities. In oil-polluted places low resistivity anomalies were found, because of bacterial oil degradation. The comparison of theoretical calculations of rocks resistivities with VES results has allowed performing correctly polluted places mapping.

Oil pollution was found at two levels: near GWL and below in water saturation zone (on 4 - 10 m depth). Oil pollution anomalies were localized in several areas of linear outlines bound with fault zones. The oil pollution is placed in clays, not in sands. This fact was confirmed by drilling and chemical analysis.

The block structure of territory built on VES data, has allowed explaining numerous chemical analyses, and detecting pollution differences in some blocks, resulted probably from different sources and of different age.

Introduction

Oil pollution is exhibited in a field of resistivity method as a zone of low resistivity already after several weeks (3-4) or months (1-4) after the leakage, - according to different publications. At the same time there is a variety of opinions about polluted zone resistivity, from very high values (Olhoeft, 1992) up to very low ones (Sauck, 1998). Now it is more evident, that resistive zone appears in the case of fresh pollution, while conductive zone is the feature of mature pollution (after several months or years). The first publications with the information that the zone of oil pollution has the low resistivity appeared rather recently (Sauck and McNeil, 1994; Modin, Shevnin et al., 1997). The process of formation of oil pollution zone was described with many details about variations of physical characteristics and chemical reactions in the article of Sauck (1998). According to Sauck (1998), the source of the low resistivity is leachate from the acid environment created by the intense bacterial action on the residual hydrocarbons near the base of the vadose zone. This zone is produced by a high TDS leachate, which is aperiodically flushed down from the volume of intimately mixed hydrocarbon, water, oxygen and soil where microbial activity is a maximum. This leachate is a result of acidification of the heterogeneous free / residual product levels by organic and carbonic acids and is produced by the leaching and etching of the native mineral grains and grain coatings. Aperiodic infiltration or recharge events flush out the accumulated pore waters, which are high in TDS, and dump them into the underlying aquifer. There it may persist as a zone with a vertical extent of less than 0.6 or 1 m, or it may diffuse downward in a continuous gradient, approaching background water quality 3 to 8 m deeper. The pore waters from the base of the vadose zone should prove to be surprisingly saline, especially in climates where there may be a long time between recharge events.

The choice of a geophysical method for some object investigation is based on the physical properties' contrast, experience of the method application and accessibility of instrumentation. Our choice of VES method is based on a wide experience of its successful usage for oil pollution study (Modin, Shevnin et al., 1997, Geoecological inspection... 1999). There are three main questions in oil-pollution mapping with resistivity method. These are: 1. Changes in rock properties resulted from oil pollution. 2. Field problems and measuring technology. 3. Problem of separation of polluted and non-polluted areas.

Sauck (1998) and his colleagues from WMU (Atekwana et al., 2001) studied oil pollution in sand deposits. Modin, Shevnin et al., (1997) studied oil pollutions in sandy-clayish and carbonate rocks. In these publications were described some changes in rock's properties, resulted from oil pollution:
1. Increase of oil transforming bacteria (at 10^2 times).
2. Increase of groundwater conductivity due to increase of TDS (up to 5 times).
3. Change of oil density (up to slightly great value then that of water) and its movement below GWL.
4. Diminish of dissolved oxygen (up to 10 times).
5. Appearing surfactants produced by bacteria diminish surface tension of water (up to 2.5 times) and dimension of oil particles (emulsification). More of oil surface appears accessible for bacteria.
6. Vertical changes of GWL position cause vertical smearing of LNAPL.
7. Appearing organic and non-organic acids in polluted zone arises karstic processes, increases porosity and cavernosity of carbonates and decreases of load-carrying ability of carbonate grounds.

VES technology

Resistivity sounding (VES) in Mexico was performed as tomographic survey or the total electrical sounding - TES (Modin, Shevnin et al., 1997), because we proposed high geological noise at the territory of oil refining factory. This technology developed in Moscow state university (MSU) is very effective for obtaining geologically reliable results, since allows canceling geologic noise (resulted from near-surface inhomogeneities - NSI), placed near measuring (P-effect) and current (C-effect) electrodes).

In each separate case the TES field technology aims to investigate a depth interval from h_{min} up to h_{max}. Base model for TES is represented on fig.1. The horizontal size of any object to be visible should be approximately equal of its depth. The step on profile for detailed study of such object should be 2-5 times smaller of its size. The deeper

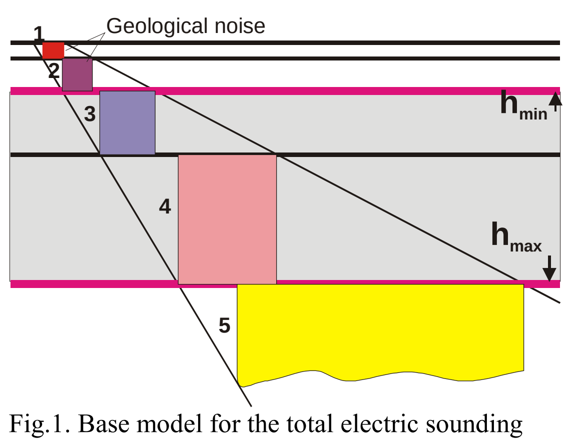


Fig.1. Base model for the total electric sounding

objects (5 in fig.1) are not visible due to restricted penetration depth. Other objects at the depth smaller than h_{min} (1-2 in fig.1) with proportionally small dimensions - may influence and the more noticeably, the smaller their depth is, because these are closer to the points of the current and potential electrodes position. These objects are considered as geological noise. VES curves distortions, caused by such objects may be divided into two types: caused by objects near potential electrodes (or P-effect) and near current electrodes (or C-effect). Deep inhomogeneities size in the case of our interest determines the step between VES on profile and the depth of study determines maximal AO distance. On the contrary, NSI are usually out of our interest and their size is so small that it's impossible to investigate them in details and select VES distance step on the base of their size. But it is not possible to ignore the existence of NSI. The best way in this case consists in minimizing the number of electrodes hits in NSI's and making their influence more regular and recognizable. That may be done by AO distance growth with linear step equal to the step between VES. In this case current electrodes from different VES will hit in the same points and NSI influence became regular. The linear step of AO distance growth is needed not for sounding, but for clearer influence of NSI (C-effect). On the same reason it is better to use array with one moving current electrode (AMN) instead of two.

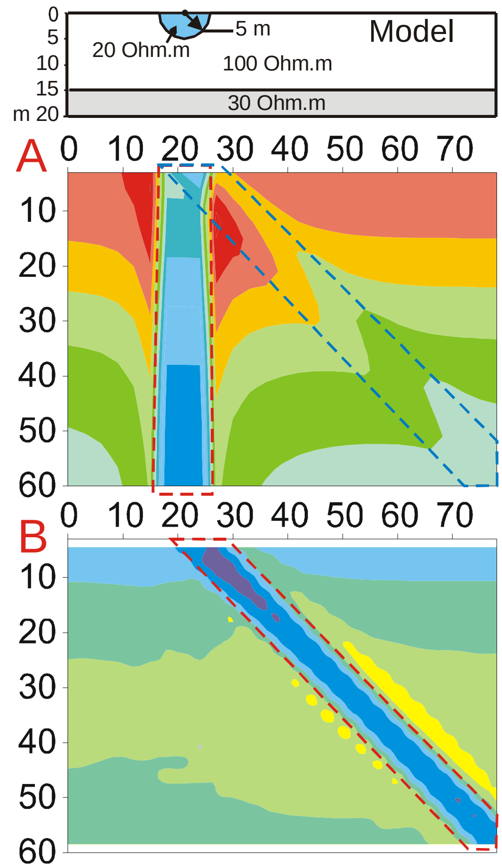


Fig. 2. P and C effects on pseudosections ρ_a (A) and its V-transformation (B) from one hemispherical NSI

So, TES field technology includes next rules: 1. VES locations on the profile are regular with equal distances. 2. Sounding at each location is fulfilled with two-sided pole-dipole (AMN + MNB) array. 3. Step in current electrodes' spacing growth is constant (that is linear) and equal to a sounding step. In this case places for current electrodes grounding will be the same for all VES points on the profile. 4. Step on profile for h_{min}-h_{max} depth interval should be equal to h_{min}.

Data Processing

For data processing these field data (for AMN+MNB arrays) the Median algorithm developed by E. Pervago in MSU in 1994 - 1998 (Ritz, Robain, et al., 1999) was applied. The algorithm operation consists of three stages: separation of apparent resistivity pseudosections' matrix for AMN and MNB arrays on some components, separation of these components on regional and local (with low and high spatial frequencies), smoothing the first ones and removing others components and then restoring apparent resistivity field, cleaned from distortions, caused by NSI. At separation of apparent resistivity pseudosections' matrix for AMN and MNB arrays on components the first

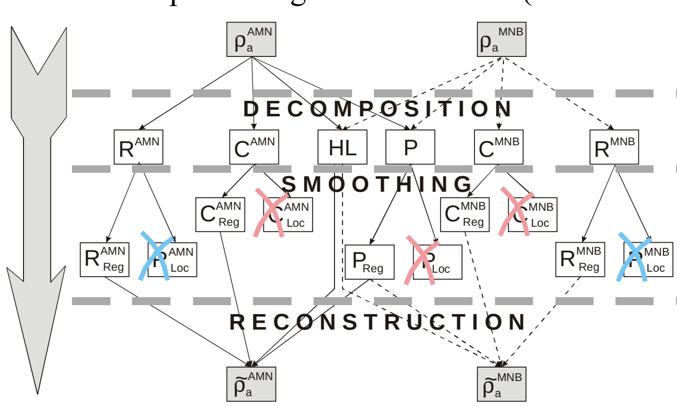


Fig.3. Scheme of Median program operation.

step is the horizontally-layered component removing. Then components, resulted from distortions effects from near-surface inhomogeneities are removed; separately distortions resulted from inhomogeneities near measuring electrodes (P effect) and then ones connected with current electrodes (C effect). The removal of these effects is based on VES modeling results for inhomogeneous media and theory of distortions caused by near-surface inhomogeneities, developed at MSU. Some explanations of algorithm operation were published in (Ritz, Robain, et al., 1999; Modin, Shevnin, et al., 1997). After removal above-mentioned components from the apparent resistivity matrix the only residual component (R) remains in matrix. It is also separated on a regional and local parts and local one is removed.

Dispersion Analysis

The wide experience of Median program usage in MSU for VES data processing, has shown, that P and C effects restrict the increasing of the survey accuracy. After removal of the geological noise the interpretation accuracy increases in 4-5 times. The role of random component usually is less than P and C effects at the use of standard Russian resistivity instruments. At relative instrumental accuracy 2-3%, the misfit error value at interpretation is about 8-12 %, and after removing geological noise decreases up to 2-3 %, i.e. practically up to instrumental accuracy.

VES data analysis of field data has shown, that more than 70 % of all VES curves are distorted. That means, that the distortions are practically inherent features of resistivity sounding.

Receiving the large volumes of VES field data and their interpretating we analyzed misfit errors of VES interpretation. The average value of misfit error (average RMS or $\sqrt{D}$, where D is dispersion) consists 8-12 % that seems rather high value and needs to be explained. The theory of geophysical interpretation (Tarkhov et al., 1982) says, that the total dispersion of geophysical field includes technological and geological dispersions, according to formula:

$$D_{total} = D_{technol} + D_{geol}$$

Dtotal can be determined as background dispersion of geophysical field (in area without strong anomalies). Indirectly it is possible to estimate this value on misfit error. Dtechnol can be estimated on control measurements, and, according to the instruction on resistivity method, RMS measuring error should not exceed 5 %. In practice measuring error depends on accuracy of measuring instrument (which does not exceed 2-3 %) and on accuracy of electrodes' arrangement (geometrical error, which does not exceed 1-1.5 %). Thus, the main factor of the differences between Dtotal and Dtechnol is geological noise Dgeol. Near-surface inhomogeneities (NSI) are the main source of geological noise, because these are close to sites of current excitation and electrical field registration. The influence of near-surface inhomogeneities is analogous to "broken glass" or wavered see surface, prevented from clear seeing deeper objects through them. In many cases the study of geologic noise can give the additional useful information on field technology and about position of separate NSI.

Fieldwork Problems

At the fieldwork at oil polluted place in Poza Rica, Veracruz, Mexico (fig.4) the instrument SYSCAL Kid (IRIS) was applied. At its usage two problems were found, which decreased a field data quality. At measurements of signals below 1 mV, the readings had very high errors and practically lost connection with geology (fig.5, for AO>35 m). To avoid this problem we tried to hold measured signals at a level above 1 mV and for that used MN lines as long as possible and changed MN regularly in the process of AO spacing growth,

Fig.4. Position of the study area

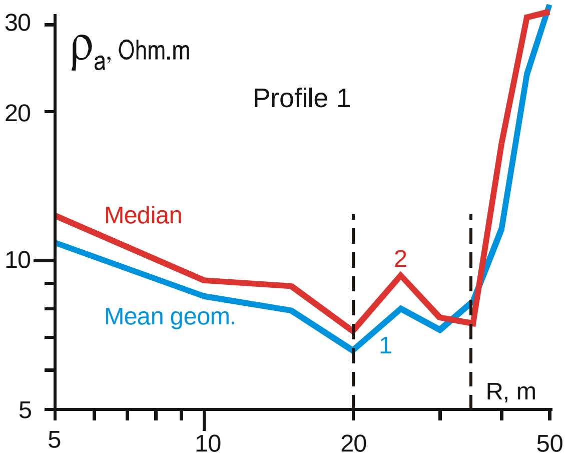


Fig. 5. Mean VES curves for profile 1, which show distortions of ρ_a values for weak signals at spacings more than 20 m and especially after 35 m.

trying to maintain AB and MN ratio close to Wenner array.

The second problem of fieldwork on industrial enterprise in city center was that the instrument appeared poor noise resistance. As a result the measuring accuracy was about 22-36%, instead of standard accuracy 5%, accepted in Russia. At low measuring accuracy C effect on VES data practically was not visible among the random noise. In such situation at data processing with the Median program the random noise was mainly filtrated (fig.6-7). The data quality after processing became noticeably better. The example of data processing results is displayed on fig.6 for profile 3 as dispersion graphs as function of spacing and on fig.7 as pseudo-cross-sections.

The total amount of sounding points was 180, measured along 7 profiles with two partial soundings AMN+MNB in each point (fig.8). Only the right part of profile 7 was measured with Schlumberger array.

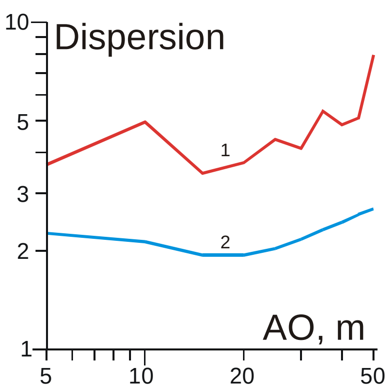


Fig.6. Graphs of dispersion for all VES on profile 3 before (1) and after (2) noise filtering

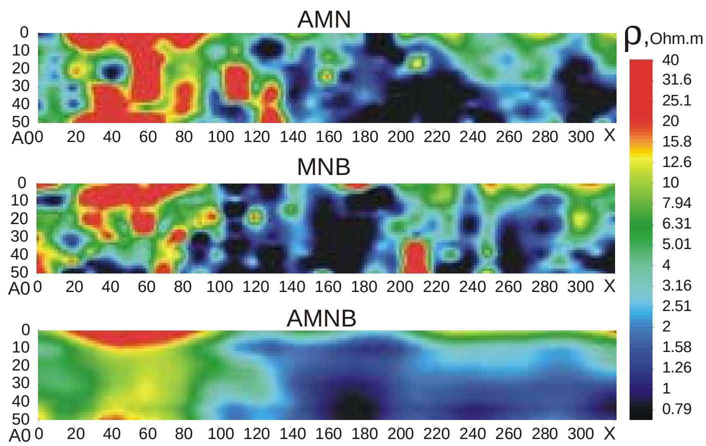


Fig.7. Pseudo-cross-sections for initial AMN, MNB arrays and AMNB array after noise filtering (Pr.3).

Median program shows the best results in the case of AMN+MNB array. Quality of filtering for Schlumberger array is lower. In fig. 9 the apparent resistivity pseudo-cross-section along profile 7 is shown. The initial part of the profile (up to point 210) was measured with AMN+MNB array and the rest part (to the right of location 210) was measured with AMNB array. In the last case the noise level (after data processing) has appeared much higher.

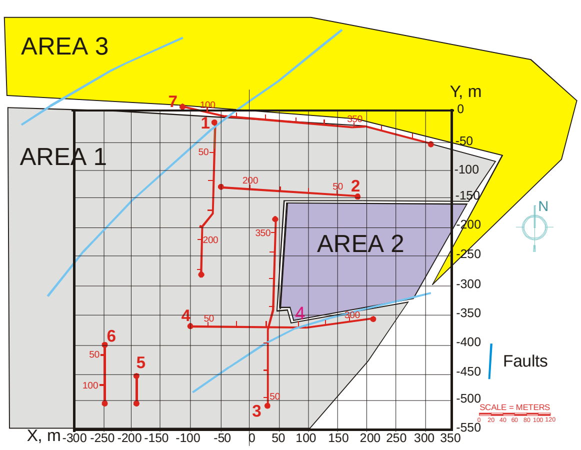


Fig.8. Scheme of the survey area with profiles.

Fieldwork results

VES data were displayed as pseudo-cross-sections

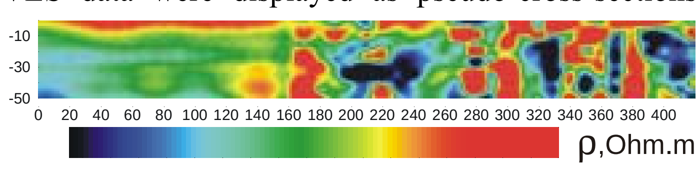


Fig. 9. Example of data processing for profile 7 with the left part, measured with AMN+MNB array and the right part, measured with AMNB array.

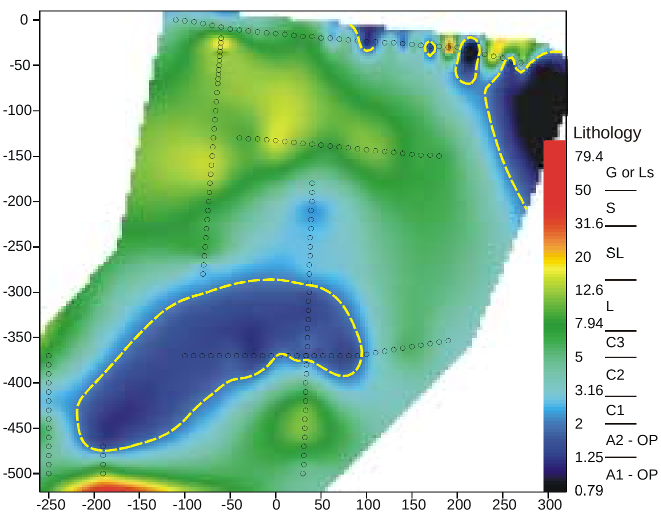


Fig.10. Apparent resistivity map for AB/2=15 m

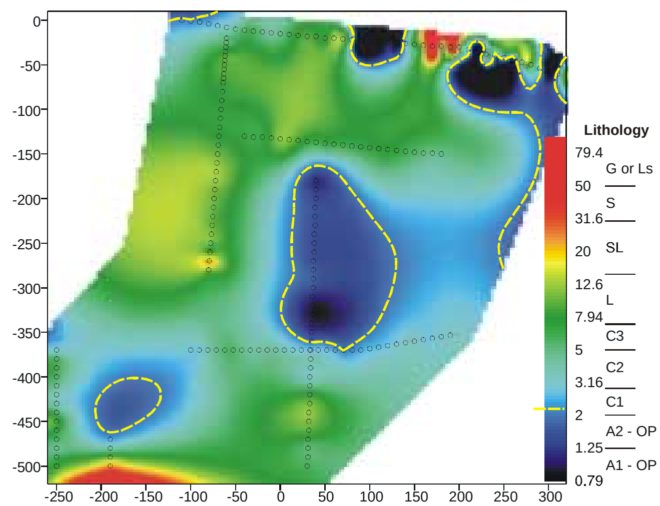


Fig.11. Apparent resistivity map for AB/2=35 m

along profiles (see fig.7, 9) and as apparent resistivity maps for different AO spacings (figs.10-11). VES sounding were performed at 10 electrode spacings from 5 up to 50 m with a step 5 m, and for each spacing the apparent resistivity map was built. Analysis of this data gives the first information about the study area. The greater is electrode spacing, the greater is depth, because there is some qualitative correspondence between electrode spacing and depth. For more accurate depth estimation the quantitative VES interpretation was performed. Nevertheless, on the set of maps for different electrode spacings several zones of abnormally low resistivity were mapped at depths. Some faults were also traced on apparent resistivity maps and then checked on VES cross-sections.

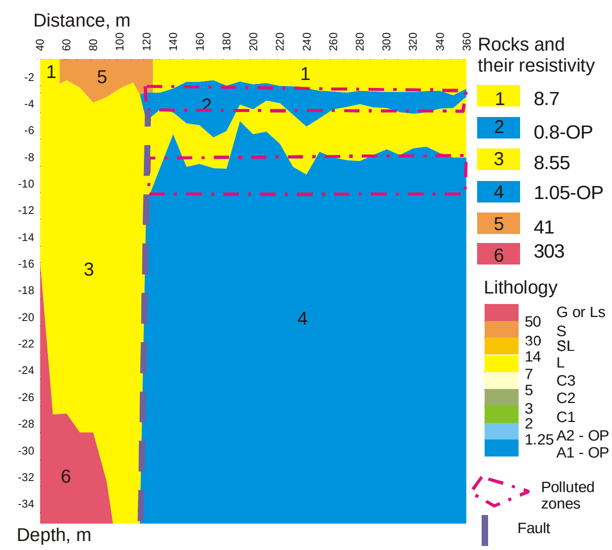


Fig.12. Geoelectrical cross-section for profile 3.

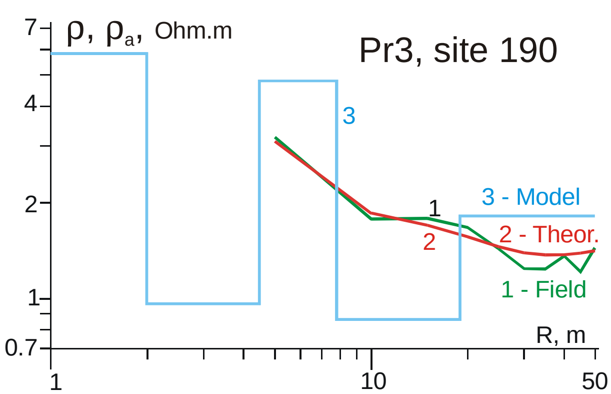


Fig.13. Field and theoretical VES curves and ρ(depth) model graph for the site 190 on profile 3.

Results for profile 3 (fig.12). On VES site 125 a fault was found due to noticeable difference of a medium structure and resistivity values. To the right of a fault there is the place of oil pollution (layer above GWL with resistivity 0.8 Ohm.m is detected) and layer on depth more than 7 m (1.05 Ohm.m), - the lower boundary of this layer was not definitely estimated.

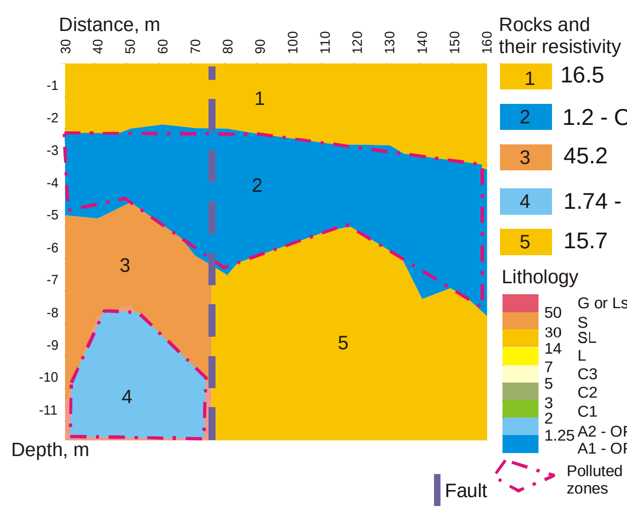


Fig.14. Geoelectrical cross-section for profile 6.

Typical example of VES curve with two conductive layers is presented on fig. 13. The conclusion about mature oil pollution here is based on the idea of bacterial oil degradation (Bailey et al., 1973, 1981; Dostalek, 1975) and low resistivity of polluted area. The borehole confirms both levels of oil pollution including depth interval from 7 up to 10 m with the maximum on 7 m. Downward movement of oil pollution below GWL is a result of oil density change after bacterial degradation, becoming slightly more than water density (Bailey et al., 1973, 1981, Dostalek, 1975).

Results for profile 6 (fig.14) are similar to profile 3. A fault and two conductive layers with oil pollution (close to GWL and on depth about 5 m) were detected, and the presence of oil pollution at two levels also was confirmed by drilling.

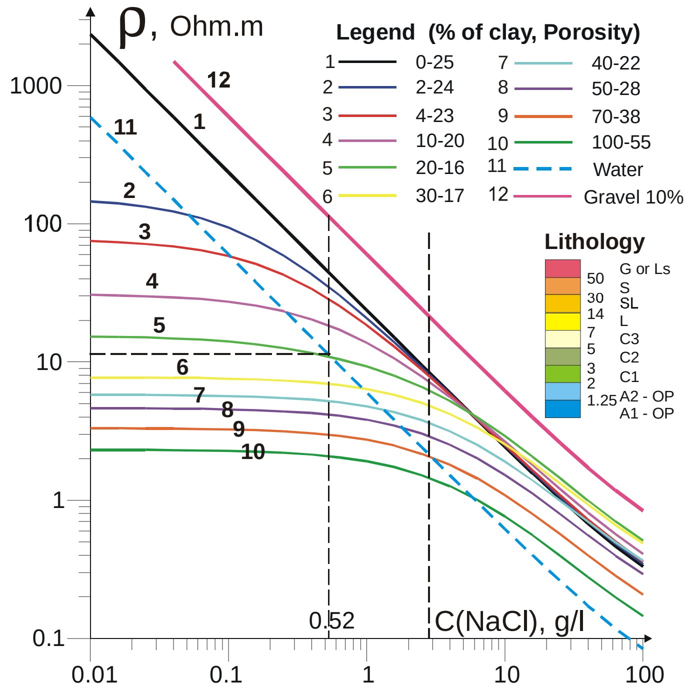


Fig. 15. Theoretical dependence of different rock's resistivity from porous water mineralization.

Theoretical Rocks' Resistivity Calculation

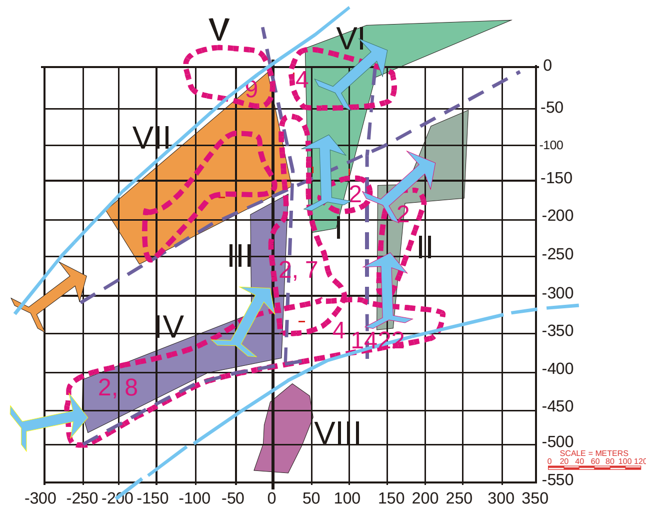
The problem of geological interpretation of VES data at oil-polluted places consists in separation of polluted and non-polluted rocks. For this problem's decision we used theoretical calculation of rocks resistivity based on water mineralization and rocks' lithology. This approach and software were developed by A.Ryjov (Ryjov, Sudoplatov, 1990, Ryjov, 1997, 2000, Ryjov and Shevnin, 2002). Program "Petrofiz" can calculate different rocks' resistivity on the base of an exact physical – chemical theory. Program works is several steps. 1. Calculation of water resistivity, taking into account types of ions (cations and anions) in solution, their mobility and concentration. 2. Account of conductivity in double electric layer (between water and solid phase), taking into account properties of solution and solid phase (Ionic Exchange Capacity - IOE). IOE is zero for sand, is maximal for clay and also depends on type of clay. 3. Calculation of rock resistivity taking into account rock porosity, pore size, humidity and clay content. Program can calculate resistivity for different positions of clay in pores – as corks in capillary paths and as thin layers at capillary wall. 4. All calculations are function of temperature.

In fig. 15 the results of theoretical calculations on A. Ryjov program - "Petrofiz" are shown. Clay, sand and some other rocks' resistivity is considered as function of water mineralization (for NaCl at 20°C). The dash line shows groundwater resistivity. It is interesting to note, that at high groundwater salinity the rock resistivity graphs are situated higher than water resistivity graph and practically in parallel to it. At smaller water salinity the resistivity graphs for clay – sand mixture are situated below resistivity of pore water. This case corresponds to the influence of double electric layer (DEL) in pores of clay. A vertical line, which passes across the water salinity value 0.52 g/l, shows water with resistivity 11 Ohm.m, measured on water samples taken from wells outside of a polluted zone. For this salinity the resistivity of clay and sand is in the range 2 - 47 Ohm.m. Practically in the area of study some resistivity values below 2 Ohm.m were measured. We consider these areas as oil polluted areas, in which the oil has been changed by bacterial biodegradation. Results of theoretical calculation were also used for preparing lithological legend, applied for VES models characterization (see the right part of fig.15, fig.12, 14). In this legend depending on resistivity interval the different rock names were used. C1 - C3 – are different clays (heavy, medium, light), L – loam (30% of clay), SL – sandy loam (10% of clay), S – sand, G or Ls – gravel or limestone with 10% of porosity. Two positions in the legend - - A1 and A2 are considered as definitely oil polluted rocks. This legend was used for geological interpretation of VES cross-sections (see fig.12, 14).

According to Atekwana, Cassidy et al., (2001) a groundwater resistivity (as a result of biodegradation) decreases about 5 times, that causes decreasing rock resistivity. A vertical line for water salinity C=2.8 g/l shows that in this case the clay resistivity decreases up to 1.5 Ohm.m. Practically we estimated resistivity values about 0.8 - 1 Ohm.m.

The main results of VES application.

Previous pollution model considered only pollution layer above GWL. VES showed that there was pollution below GWL until 10-15 m. Instead of horizontally layered model of pollution a new model appeared, with pollution zones position under the control of faults and mainly in clay. On fig.16 the zones of oil pollution on VES data are shown with red dashed lines. Comparison of block structure built on VES data with chemical analyses showed that each block has its own type of pollutant. This fact can be explained by different sources of pollution and/or by different time of pollution event. The position of oil pollution mainly in clay is very interesting fact, which is discussed below.



II - mineral spirits;III - kerosene;
IV - gasoline and diesel fuel; VII - unknown.

Fig.16. Scheme of oil pollution position and proposed groundwater movement. Different types of pollution are marked with different colors.

Position of oil in pores of clay

The oil enters saturated pores, displacing water and remaining in the center of a pore. As a result all remaining film of bound water appears completely in the area of a double electric layer (DEL), mainly in its diffuse part, which provides a surface conductance. Because of a surface conductance the clay filled with water and oil also remains electroconductive one. Moreover, the more is a contribution of a surface conductance, i.e. the fraction of pores occupied by DEL, the more conductive would the rock filled with water and oil. The surface conductance is absent in sands, and in this case the oil adding is only diminishes an electrical conductivity as insulator. Besides in sands also there is no evident bound water film (including an osmotic one), preventing from a sorption of oil immediately on the surface of rock particles. Therefore in sands the oil can wet mineral grains, whereas in clays the rock particles remain wetted by water.

Below groundwater level (GWL) all pores in clays are filled by bound (osmotic and adsorptive) water. Its movement in clayish rocks (and also migration of oil together with the water as a result of viscous friction) is possible only under the influence of different gradients:

- Salt content contrast, causing diffusive - osmotic water flow;
- Temperature contrast, causing heat and water transfer;
- Electric potential contrast, causing electro-osmotic water movement.

Basically, all these reasons are possible, since in clays these types of transfers could dominate above headwater filtration.

On the solid - fluid boundary operates a surface tension. Absorption of hydrocarbons in porous space of clays should occur enough rapidly due to a surface tension. The noticeable increase in surface-active agents' concentration in a polluted zone (Atekwana, Cassidy et al., 2001) probably promotes this.

Ions rather easily permeate in clay and their quantity in clays can be 50 times more than in electroneutral solution, as the DEL thickness frequently appears comparable with pores size in clays. And to extract ions from a pore space of clays is much more difficult, because the electric forces in pores of clays are very strong, and can be fulfilled only by washover of clay by water, executed on special technology.

The electric forces work as the pump, and a result is an oil transfer into clays due to diffusive - osmotic and electrokinetic mechanisms of secondary oil migration (Korolev et al., 1997).

Thus the water saturated rock containing oil drops in pores, is considered as a three-phase system with two interphase boundaries: mineral - water and water - oil. According to it, on each interphase boundary its own DEL is forming. These DEL patterns are different, as the oil is non-polar fluid with low electrical conductivity (about 0. 3 10^{-18} - 10^{-10} Ohm^{-1} cm^{-1}).

Threat of oil pollution situated in clays is that diverse dissoluble fractions or products of their disintegration originated from oil pollution could migrate in exploited underground waters by slow diffusion or the leaching.

Remediation of oil pollution in clay can be performed by imposing DC electric current, according to Korolev et al. (1997).

Conclusions

1. VES method allows studying zones of mature oil pollution with high certainty both in plan and depth.
2. Canceling geological and random noise in VES data has allowed increasing noticeably field data quality.
3. Joint analysis of field VES data and theoretical calculations of rocks' resistivity on the basis of groundwater resistivity or its salinity has allowed separating the contaminated and non-contaminated zones.
4. The maximal ecological effect is obtained as a result of integration of VES, chemical analysis and drilling data.
5. The presence of oil pollution was estimated as near to ground water level on 2 m depth, and on depth 4 - 10 m, concentrated mainly in clays. New model of polluted place includes faults and linearized polluted zones instead of a single uniform layer above GWL.
6. The concentration of oil pollution in clays, estimated as the fact in this fieldwork, requires additional studies of this phenomenon nature, study of oil pollution absorption's mechanism acting in clayish rocks, potential threat from such localization and developing remediation technology.

Acknowledgments

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