

DC RESISTIVITY METHOD APPLIED IN THE MONITORING OF DIESEL LEAKAGE IN A RAILWAY ACCIDENT IN SÃO MANUEL CITY, SÃO PAULO STATE (BRAZIL)

César Augusto Moreira¹, Lívia Portes Innocenti Helene² and Ariane Raissa Pinheiro Côrtes²

ABSTRACT. This paper presents an integrated analysis of geochemical and geophysical data in a contaminated site, currently under environmental monitoring, after a leakage of 240,000l of diesel in 1999. The main objective is to assess the presence of subsurface contaminants in residual and dissolved phases, using Electrical Resistivity Tomography (ERT), and to understand the effects of natural degradation of hydrocarbons and changes in electrical properties in soil and groundwater. The results allowed for the definition of two distinct resistivity patterns containing dissolved hydrocarbon phases, defined after historical chemical analyzes. The intermediate resistivity values (260 Ω .m and 511 Ω .m) are indicative of the coexistence of hydrocarbons in residual and dissolved phases. The higher resistivity values (>1000 Ω .m) reveal the prevalence of residual phases with incipient degradation, possibly due to high toxicity concentrations prevailing the effective action of biodegradation processes. The generation of 3D visualization models from 2D sections allows the use of the isosurface modeling technique and definition of a high resistivity body, consistent with conceptual models of hydrocarbons dispersion in the subsurface. The diagnostic of an area with potential accumulation of hydrocarbons in residual phase can subsidize the planning of remediation techniques to promote the complete decontamination of the area.

Keywords: contamination, hydrocarbons, residual phase, dissolved phase, electrical resistivity tomography.

RESUMO. Este trabalho realiza uma integração de dados geoquímicos e geofísicos obtidos numa área contaminada pelo vazamento de 240.000l de diesel, em 1999, e que atualmente está sob monitoramento ambiental. O principal objetivo é a verificação da presença de fases residual e dissolvida de contaminantes em subsuperfície, por meio da técnica de tomografia elétrica (ETR), para compreensão dos efeitos do processo de atenuação natural na degradação de hidrocarbonetos e alterações em propriedades elétricas do solo e águas subterrâneas. Os resultados permitiram a definição de dois padrões distintos de resistividade numa região com presença de fase dissolvida, definido a partir do histórico de análises químicas. Valores intermediários (260 Ω .m e 511 Ω .m) são indicativos da coexistência de hidrocarbonetos em fases residual e dissolvida. A região com elevados valores (>1000 Ω .m) indica a prevalência de fase residual com degradação incipiente, possivelmente sob concentrações altamente tóxicas para uma ação efetiva dos processos de biodegradação. A geração de modelos de visualização 3D a partir das seções 2D, possibilitaram o uso da técnica de modelagem de isosuperfície e definição de um corpo de alta resistividade, consistente com modelos conceituais de dispersão de hidrocarbonetos em subsuperfície. O diagnóstico de uma região com potencial acúmulo de fase residual de hidrocarbonetos pode subsidiar o planejamento de técnicas de remediação e propiciar a completa descontaminação da área.

Palavras-chave: contaminação, hidrocarbonetos, fase residual, fase dissolvida, tomografia elétrica.

¹ Universidade Estadual Paulista – UNESP, Applied Geology Department, Geoscience and Exact Science Institute, Rio Claro Campus, 24-A Avenue, 1515, P.O. Box 178, 13506-900 São Paulo, SP, Brazil. Phone: +55(19) 3526-9313 – E-mail: moreirac@rc.unesp.br

² Universidade Estadual Paulista – UNESP, Geoscience and Environmental Science Post-graduation Program, Geoscience and Exact Science Institute, Rio Claro Campus, 24-A Avenue, 1515, P.O. Box 178, 13506-900 São Paulo, SP, Brazil. Phone: +55(19) 3526-2819 – E-mails: liviapihelene@gmail.com; ariane.rpc@gmail.com

INTRODUCTION

Environmental contamination occurs as a result of lack of planning following the economic development and inadequate management of raw materials, products and hazardous waste at concentrations that, above the recommendation values, pose health risks to the quality of life and to the environment.

Groundwater contamination by organic chemicals is a recurring issue. Given the variability in terms of density and solubility in water, when reaching the soil and the groundwater, they can form plumes of contamination and turn this resource not potable for human consumption. The most common organic compounds capable of causing environmental impacts are chlorinated solvents and short chain hydrocarbons.

The origin of the contamination of soil and water by hydrocarbons usually occurs due to transport accidents, breakage of pipes or by improper storage. In the state of São Paulo, Brazil, contaminated areas rose from 225 to 4.572 in a ten-year period. The main groups of contaminants found in these areas are the aromatic solvents, represented by the BTEX compounds (benzene, toluene, ethylbenzene and xylenes), the aromatic polycyclic hydrocarbons (PAH), the metals and the halogenated solvents (CETESB, 2012).

For a long time, the contamination of soil and groundwater has been neglected by the difficulty of characterization. When in contact with the physical environment, the path and transport of organic contaminants can be modeled according to two stages. The first comprises all those that occur entirely above the groundwater level, in the vadose zone. The second considers the transport processes inside the aquifer, soluble/dissolved or insoluble/supernatant characterized by chemical analysis of water samples from monitoring wells (Newell et al., 1995).

Diagnostic techniques through direct methods such as physical and chemical analysis of samples are widely used in the diagnosis of these areas, given the possibility of quantifying the levels in terms of concentration. Although, the main disadvantage of this procedure is its punctual and little representative character in spatial terms, in a condition where the analyzed object is mobile and dynamic.

Particularly considering past contamination by hydrocarbons which occurred several years ago, content changes due to dispersion, dilution and degradation of the compounds by the action of various agents present in the geological environment are expected. The biodegradation of contaminants is one of the major mechanisms responsible for the consumption of contaminants with long periods of residence in the environment (USEPA, 1998). The efficiency of this process depends on a combination of factors such as the presence of electrical receptors (O_2 , SO_2^{-4} ,

Fe^{+3} , Mn^{+4} , NO^{-3}), appropriate conditions of pH, Eh, temperature and low levels of contamination (Ritmann, 2004; Adriano et al., 2004).

In many cases of contaminated sites with hydrocarbon there is a reduction on the concentration levels, independent of remedial action. The natural attenuation comprises a set of processes for the remediation in geological environments through the action of physical, chemical and biological processes. This process is especially important in the degradation of hydrocarbons, which may be converted by organisms found in nearly all environments (Mulligan & Yong, 2004).

Non-invasive geophysical investigation tools, have low cost and wide coverage in spatial and temporal terms, and have been used increasingly in recent years as diagnostic tools in the management of contaminated sites (CETESB, 2001). Electrical geophysical methods are particularly relevant in this type of research because of the frequent contrast of the physical properties between the contaminant and the uncontaminated soil or rock.

The degradation processes can cause some change in the electrical properties of the environment which allows the identification and monitoring of contaminant in soil and groundwater. The hydrocarbons have high resistivity in the free phase; however, factors such as the production of organic acids during the biodegradation cause a decrease in resistivity values. Several case studies involving electrical methods demonstrate the effectiveness, benefits and limitations when applied to the diagnosis and monitoring of contamination by hydrocarbons (Sauck, 2000; Batayneh, 2005; Moreira et al., 2006; Delgado-Rodríguez et al., 2006; Menezes et al., 2011; Porciúncula & Lima, 2012; Delgado-Rodríguez et al., 2014; Veloso et al., 2015).

In face of the delineation possibility of the contaminants in soil and groundwater, the geophysical methods allows the understanding of the evolution and persistence of hydrocarbon as a technique for monitoring the natural attenuation.

This work performs an analysis of geophysical and geochemical data from a benzene-contaminated area, currently under monitoring for natural attenuation processes, after a railway accident.

Behavior of hydrocarbons in soil and groundwater

Once the petroleum-derived hydrocarbons reach the ground, they can undergo evaporation or penetrate and reach the groundwater. When in contact with the groundwater, they are subject to dissolution or they remain in the residual free phase, named non-aqueous phase liquids (NAPL – Non-Aqueous Phase Liquids), which can fill the pores of the soil so that they become a continuous source for the contamination of the aquifer (Newell et al., 1995).

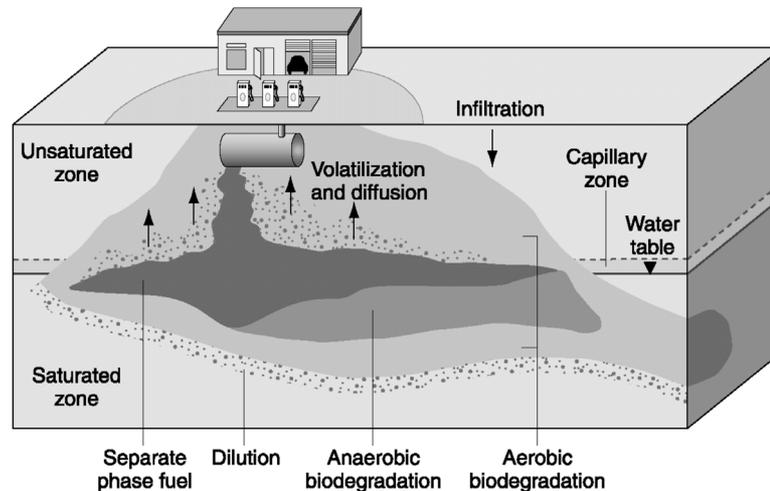


Figure 1 – Conceptual modeling of natural attenuation processes that affect the fate of hydrocarbons in aquifers (Bekins et al., 2001).

Reaching the aquifer, the NAPLs are divided according to their density relative to water in light phase LNAPL (Light Non-Aqueous Phase Liquids) represented by compounds BTEX (Benzene, Toluene, Ethylbenzene and Xylenes) and dense phase DNAPL (Dense Non-Aqueous Phase Liquids), consisting on the group of polyaromatic hydrocarbons (PAH). The NAPLs compounds have great potential for contamination in soils and groundwater.

Without human intervention and under favorable environmental conditions, organic compounds undergo a reduction in their mass, toxicity, volume and concentration through physical, chemical and biological processes. This process of natural attenuation occurs in the saturated and unsaturated zone. These processes include, besides biodegradation, mechanisms of dispersion, dilution, evaporation and adsorption (USEPA, 1998).

Dispersion and dilution of contaminants depend almost exclusively on the physical characteristics of the aquifer, such as hydraulic conductivity, which influences the transport of groundwater. This hydraulic conductivity, when high, increases the extent of contamination, but decreases the average concentration due to dilution (Fig. 1). Thus, at lower concentrations, microorganisms meet the most favorable environments for the performance of natural degradation (Wilson & Jones, 1993; Gan et al., 2009).

Volatilization of compounds is relatively low, because the surface area between the contaminant on residual or dissolved phase with the atmosphere is small, and are often neglected in diagnostics. Several studies shows that less than 5% of the dissolved phase of BTEX are converted into gas by volatilization (USEPA, 1998).

Other common destiny for contaminants are the pores of the soil matrix. Due to the hydrophobicity of the hydrocarbon molecules and to the mechanisms of sorption caused mainly by clay minerals, organic matter and iron and manganese oxides, these compounds tend to be trapped in these pores or in fractures of rocks at the surface (Thomas & Ward, 1989; Gan et al., 2009).

The affinity of the contaminant with the soil matrix may not be sufficient to isolate the contaminant from the permanent groundwater. The sorption does not remove the mass of contaminant, it only retards the migration, and depending on the desorption rate can contaminate groundwater again (Thomas & Ward, 1989; Wilson & Jones, 1993).

The main mechanism of natural attenuation of hydrocarbons in soil and groundwater is biodegradation, where microorganisms convert hydrocarbons to organic acids or promote the complete oxidation (mineralization). The generation of these acids in large amounts results in mineral dissolution and ion release to the saturated zone of the aquifer (Bennett & Siegel, 1987; Hiebert & Bennett, 1992; Moreira & Dourado, 2007).

This transformation is mainly limited by the availability of electron receptors and hydrocarbons that assume the role of electron donors to the degradation, and therefore need to be biochemically accessible to microorganisms. Besides, other parameters such as pH, temperature, salinity and redox potential are also important so that the biodegradation occurs (USEPA, 1998).

Under aerobic conditions, where dissolved oxygen is the electron acceptor, the biodegradation is more effective and allows complete conversion of hydrocarbons into non-toxic products such as CO_2 , CH_4 and H_2O .

METHODOLOGY

Location and Accident History

The area is located in the farm named Ribeirão do Paraíso, rural area of São Manuel city, São Paulo State, Brazil, specifically at the kilometer 296 of the railway, between the cities of Rubião Júnior and Bauru (Fig. 2).

In December 1999 a derailment and overturning of three wagons carrying oil fuel occurred between the Rubião Junior and Bauru cities. About 240,000 liters of diesel leaked, contaminating an area of approximately 30,000 m² (CETESB, 2006).

The first measures was the recovery and containment of the material retained on the surface. Large quantities of oil hit the groundwater and surface water, with contamination of the Igualdade creek, which supplies water to the São Manuel city (CETESB, 2006).

Probing and the installation of monitoring wells identified 0.3m to 1.0m sandy-clay soils, with presence of organic matter, superimposed over fine sandstones belonging to Adamantina Formation, Bauru Group, of the Paraná Sedimentary Basin.

Near the drainage, there are outcrops of Quaternary alluvial sediments consisting of medium to coarse quartz sand. The

hydrogeological data show that the level of water table is shallow, with a maximum depth of 2.9m in the higher topographic areas, and sub-outcropping near the drain. The permeability indicates a maximum hydraulic conductivity of $7,663 \times 10^{-4}$ cm/s and minimum of $5,783 \times 10^{-5}$ cm/s. The maximum calculated flow speed was 0.135m/d (CETESB, 2011).

The groundwater flow follows the local topography, from southeast to northwest, towards the Igualdade creek (Fig. 3).

The analytical results of the historical series of groundwater monitoring at the accident site indicate the presence of BTEX compounds (Benzene, Toluene, Ethylbenzene and Xylene), especially benzene with greater temporal persistence (Table 1).

RESULTS AND DISCUSSIONS

The geophysical data consisted of six lines of Electrical Resistivity Tomography (ERT), each 104m long, in a Wenner-Schlumberger arrangement with 2m spacing between electrodes, placed parallel to the railway with 12.5m spacing between lines (Fig. 1). The positioning of the lines considered the latest geochemical data available given by the monitoring of groundwater contamination as required by the environmental agency. These data

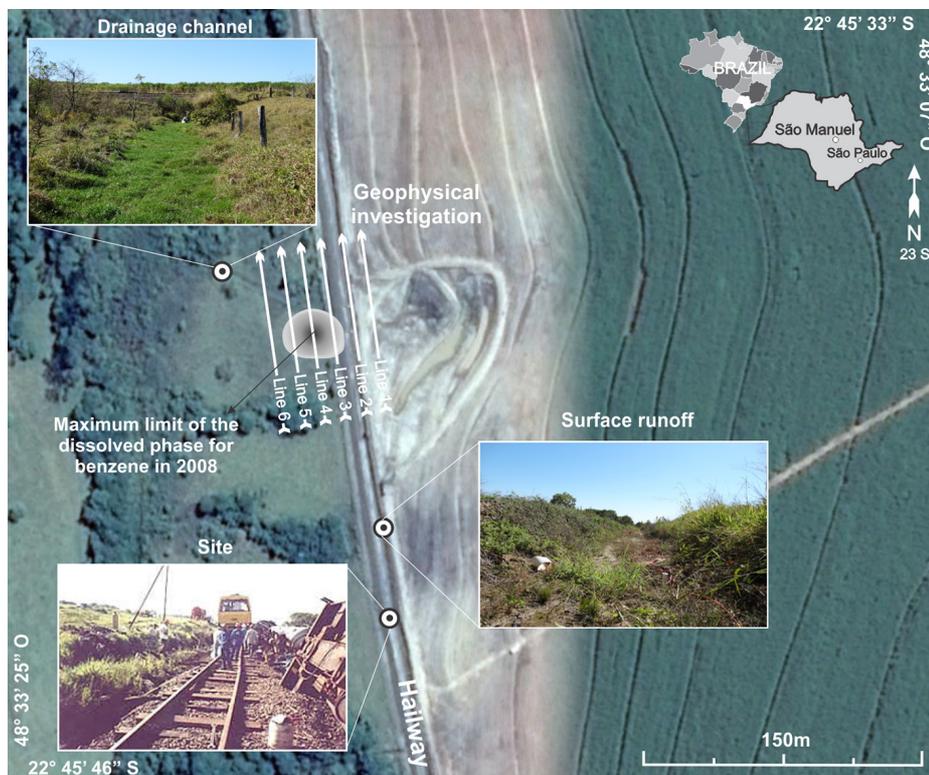


Figure 2 – Study area, with site, surface runoff, drainage channel under railway, lines of geophysical investigation and maximum limit of the dissolved phase for benzene in 2008 (based in CETESB, 2008).

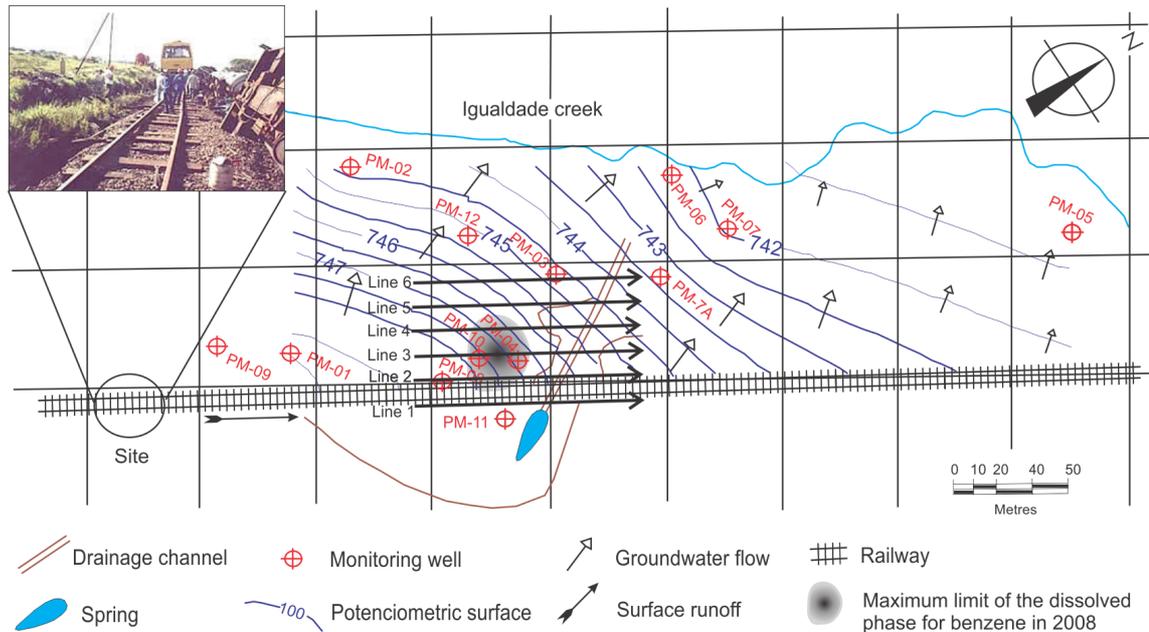


Figure 3 – Groundwater flow based on monitoring wells and location of the geophysical survey lines (based on CETESB, 2009).

Table 1 – Evolution of the benzene in aquifer (in $\mu\text{g/l}$) (CETESB, 2006 and 2011).

Well	Date								
	11/2000	12/2003	08/2006	03/2008	10/2008	06/2009	11/2009	10/2010	03/2011
PM-01	<L.D.	*	<5.0						
PM-02	<L.D.	<5.0	*						
PM-03	<L.D.	<5.0	<5.0						
PM-04	420	76	55.1	15	2.81	5.7	4.1	<5.0	5.8
PM-05	<L.D.	<L.D.	<L.D.	*	*	*	*	*	*
PM-06	—	<L.D.	<L.D.	*	<L.D.	<L.D.	<L.D.	<5.0	<5.0
PM-07	—	<L.D.	<L.D.	<L.D.	<L.D.	<L.D.	<L.D.	<5.0	<5.0
PM-08	—	350	20.1	3	28.2	<1.0	<L.D.	"	<5.0
PM-09	—	—	<L.D.	<L.D.	<L.D.	<L.D.	<L.D.	<5.0	<5.0
PM-10	—	—	24.3	<L.D.	<L.D.	<L.D.	<L.D.	<5.0	<5.0
PM-11	—	—	<L.D.	<L.D.	<L.D.	*	*	*	*
PM-12	—	—	<L.D.	<L.D.	<L.D.	<L.D.	<L.D.	<5.0	<5.0

L.D.: Limit of detection. *Destroyed well.

describes the presence of benzene in maximum values of $5,8\mu\text{g/l}$ for dissolved phase in the well PM-04 (Table 1).

The resistivity data was processed in the software Res2dinv and presented in sections of electrical resistivity in terms of distance and depth, with logarithmic graphical scale and intervals of interpolation of values in colors.

This program automatically determines a two-dimensional model of the subsurface. The 2D section divides the pseudo-section into rectangular blocks, which will represent the pseudo-

section by the adjustment of the field measurements. This optimization aims to reduce the difference between the apparent resistivity values, calculated and measured in the field, by adjusting the resistivity of the block model, with the difference expressed by RMS (Root Mean Square) (Griffiths & Barker, 1993; Loke & Barker, 1996).

The inversion models were standardized on a single scale of values and colors, so that comparative analyzes of lines are permitted. The processed data reveal a range of values between

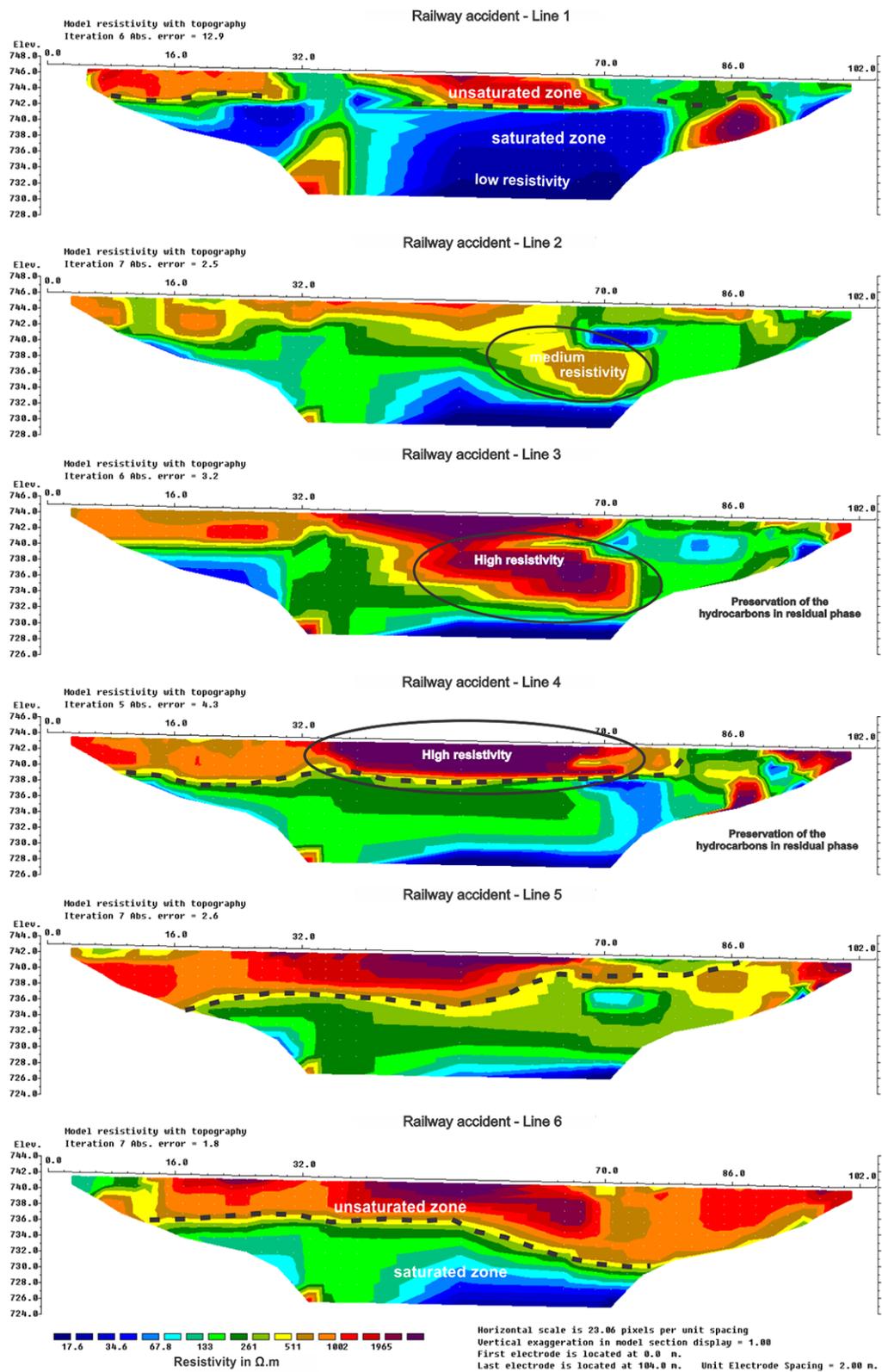


Figure 4 – Inversion resistivity models.

17Ω.m and 1965Ω.m, with a predominance of high values in the near-surface portions and gradual reduction on values with increasing depth (Fig. 4).

The line 1 presented high and intermediate values close to the surface and a wide zone of low resistivity in depth. High resistivity values close to the surface are attributed to dry sandy soil whereas intermediate values are regions of moisture accumulation. The values below 60Ω.m predominates in depth, and are indicative of the saturated zone or highly degraded dissolved phase hydrocarbons, since in this region there was accumulation of diesel.

The line 2 was carried along the runoff channel where the flow of diesel leaked in the accident. Between 60m and 80m positions the topsoil is extremely humid, characterized in the section by resistivity values between 260Ω.m and 511Ω.m. In depth, there are relatively higher values, between 511Ω.m and 750Ω.m and values below 60Ω.m only in the deeper portions. This increase in resistivity values and the proximity to the area with higher concentrations of hydrocarbons in the aquifer may be indicative of the presence of residual phase in intermediate conditions of degradation.

The lines 3 and 4 cross the area of greatest concentration of hydrocarbons in the residual phase, characterized by intervals with resistivity higher than 1000Ω.m at the central band in depth, which may be related to the presence of hydrocarbons in high concentrations in the soil and in the matrix of the saturated zone. Due to the polymeric architecture of the carbon and hydrogen atoms of the hydrocarbons, these compounds typically have high resistivity values. The existence of residual phase in the aquifer after 14 years from the leakage of diesel in the area is attributed to releases of hydrocarbons accumulated in the pores of the soil and saprolite, whose degradation allows their migration to the dissolved phase in the aquifer level.

The patterns of resistivity that characterize lines 5 and 6 are indicative of increasingly intensive action of the degradation processes, in face of the drop of resistivity values in the aquifer interval towards the line 6, positioned downstream of the place of accumulation of hydrocarbons. This gradual decrease in resistivity may yet indicate the more effective conversion of residual phase to the dissolved phase, besides the gradual return to the natural conditions of the area through the complete consumption of hydrocarbons by the action of natural attenuation processes.

The data generated after the 2D inversion were gathered into a single file, later used as a database for generating 3D models and depth maps. This process was developed in the Oasis Montaj platform where 2D inversion results obtained from the Res2Dinv

program were interpolated and modeled using the kriging method, for enhancement of extreme values in a model of three-dimensional blocks, where ERT lines were positioned (Fig. 5).

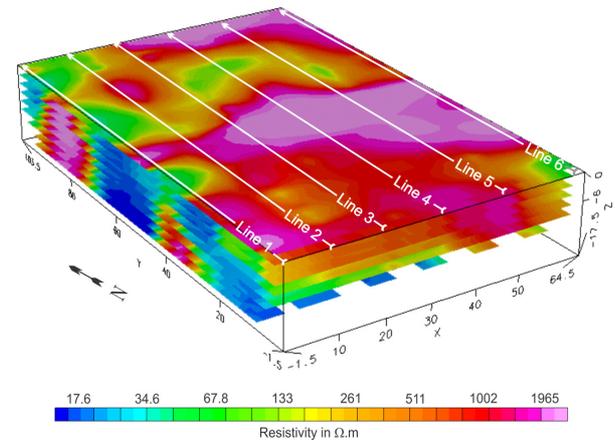


Figure 5 – Model of the electrical resistivity, with location of the acquired lines.

In sequence, the initial data were modeled by the minimum curvature method that smoothed the core values in relation to the limits of the investigated area, and then presented in maps or slices for various depths. The range of 1800Ω.m values was modeled as a 3D surface in an attempt to assess the shape of the high resistivity zone, where the unsaturated and saturated zone contact were also shown (Fig. 6).

The product of the 3D integration of 2D sections allows a clear view of the contaminant in the subsurface. The high resistivity values found on the surface, above the water table, demonstrate the behavior of the residual hydrocarbon in the unsaturated zone. The residual phase persists in the unsaturated zone in regions where there is probably slow renewal of oxygen and other electron acceptors that trigger degradation.

In the saturated zone, however, the hydrocarbon is trapped in the soil pores, and the different permeabilities of the underlying layers created conditions for the hydrocarbon to take a conical and concentrated shape.

CONCLUSIONS

Fourteen years after the railway accident the results show the persistence of hydrocarbons in residual and dissolved phase in soil and groundwater. The history of geochemical data gathered from analyzes of groundwater reveals a decreasing trend in the levels of benzene for almost all the wells. However the well PM-04 shows a concentration reduction until 2008, followed by increasing values in the following years (2009 and 2011). Geophysical lines 3 and 4, near well PM-04, presented the higher

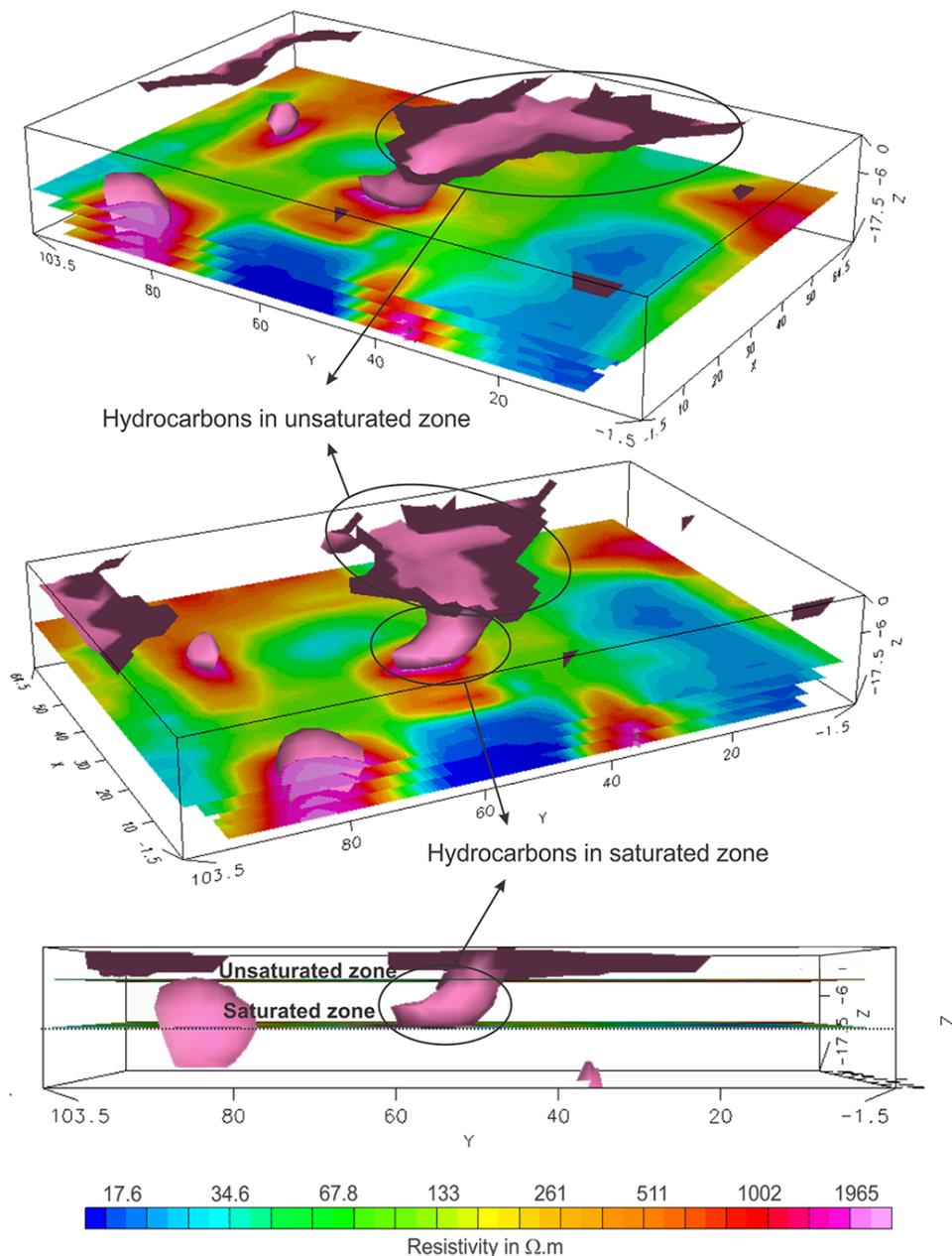


Figure 6 – 3D isosurface of $1800\Omega.m$, with saturated and unsaturated zone.

resistivity associated to hydrocarbons in residual phase in the aquifer (Fig. 2).

This indicates the action of natural attenuation processes, which result in various degrees of degradation of hydrocarbon in residual phase and sporadic release into the aquifer, in the form of dissolved phase compounds. In this sense, the input of the dissolved phase will definitely finish only after the complete consumption of the residual phase of hydrocarbon retained in the

pores of the aquifer.

The highest levels of dissolved hydrocarbon phases were characterized by high resistivity values ($>1000\Omega.m$), indicative of the presence of high concentrations of residual phase, in face of the typical high resistivity of low degraded hydrocarbons. Similar patterns are described in the work of Cassidy et al. (2001), Menezes et al. (2011), Porciúncula & Lima (2012), Delgado-Rodríguez et al. (2014).

The integration of the 2D sections in 3D visualization provides an accurate analysis of the current behavior of contaminants in the area. The isosurface of $1800\Omega\cdot\text{m}$ resulted in a volume of high resistivity contrasting between unsaturated and saturated zone. The volume defined by the isosurface should represent accumulations of remaining hydrocarbons present in dissolved and residual phase, after natural attenuation processes.

The integration of geochemical and the geophysical data enabled a comprehensive and effective diagnosis for understanding the current situation of the contaminated area, and enabled the identification of priority sites for remedial action on the ground, something extremely difficult only from groundwater chemical analyzes. The 3D visualization models are particularly relevant as complementary procedure in monitoring contaminated areas, given its similarity to conceptual models for hydrocarbon contamination in the soil.

The application of remediation techniques in places with high levels of residual phase should certainly accelerate the decontamination and extinguish the input of dissolved phase in the aquifer.

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