

Research Article

MAPPING GROUNDWATER CONTAMINATION FROM GAS STATIONS USING GROUND PENETRATING RADAR (GPR) IN AMAZON, BRAZIL

Mapeamento da contaminação de águas subterrâneas de postos de gasolina usando radar de penetração no solo (GPR) na Amazônia, Brasil

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RESUMO: O Radar de Penetração no Solo (GPR/ GSSI-SIR 3000-Common-offset) foi utilizado para detectar e dimensionar estruturas subterrâneas e plumas de contaminação por hidrocarbonetos em torno de postos de gasolina no município de Santarém (Pará/Amazônia/Brasil). Dois postos de combustíveis foram escolhidos dentre os mais antigos, localizados em áreas populosas, o que pode representar grande risco de contaminação do subsolo e das águas subterrâneas por vazamentos de combustíveis fósseis. A presente pesquisa utilizou janelas de tempo de 100 ns e 175 ns, com antena de 270 MHz, e os dados recebidos foram processados via Software Reflex Win/ V4.5.5. Os resultados mostraram que os tanques de combustível são mais bem refletidos na configuração de 100 ns, enquanto o lençol freático e a pluma de hidrocarbonetos foram mais claramente identificados na configuração de 175 ns devido à maior faixa de profundidade. Portanto, o mapeamento raso do subsolo com o método GPR, utilizando as duas configurações complementares, foi eficaz na localização e delimitação de plumas de hidrocarbonetos até a profundidade de 4 metros. Assim, o método mostrouse eficiente na obtenção de dados importantes para orientar os serviços de saneamento e saúde pública e a elaboração de políticas públicas ambientais e licenciamento de empresas que lidam com atividades potencialmente poluidoras.

Palavras-chave: Método Geofísico; Água Subterrânea; Hidrocarbonetos; Poluição; Centro Urbano.

ABSTRACT: The Ground Penetration Radar (GPR/ GSSI-SIR 3000-Common-offset) was used to detect and scale underground structures and hydrocarbon pollution plumes around gas stations in the municipality of Santarém (Pará/ Amazon/ Brazil). Two of them are among the oldest in the city and they are located in a very populated area, which can represent a great risk of subsoil and groundwater contamination because of fossil fuel leaks. The present research used 100 ns and 175 ns time windows, with a 270 MHz antenna, and the received data were processed via Software Reflex Win/ V4.5.5. The results showed that the fuel tanks are best reflected in the 100 ns configuration, while the water table and the hydrocarbon plume were more clearly identified in the 175 ns configuration due to the greater depth range. Therefore, the shallow mapping of the subsoil with the GPR method, using the two



complementary configurations, was effective in locating and delineating hydrocarbon plumes up to a depth of 4 meters. Thus, the method proved to be efficient in obtaining important data to guide sanitation and public health services and the elaboration of environmental public policies and licensing of companies dealing with potentially polluting activities.

Keywords: Geophysical Method; Groundwater; Hydrocarbons; Pollution; Urban Center.

INTRODUCTION

Environmental contamination is a common problem in urban areas and the leakage of Underground Fossil Fuel Storage Systems (UFFSS) is a matter of great concern due to their content of dangerous and highly toxic oil products (CETESB, 2017). The longterm exposure can affect both the population and the ecosystem in addition to the potential risk of explosions and fires. The most frequent causes of fossil fuel leakage are oxidation and corrosion of the underground tanks and pipelines, oil product spills during drilling, defective installations, and leak or disruption of the fuel distribution pipelines (FERREIRA et al., 2004; GALANTE, 2008).

Although direct observation of contamination in the subsoil is difficult, noninvasive geophysical investigations are recognized as efficient tools in the detection of different underground features and are widely accepted due to their low costs and positive results (LAMBOT et al., 2008). Thus, leakage and contamination plumes can be successfully detected by using Ground-Penetrating Radar (GPR), an electromagnetic method, used since the 1990s, to detect buried targets and mapping of subsurface contamination of hydrocarbon plumes (ATEKWANA et al., 2000; PORSANI et al., 2006; MORAES & OLIVA, 2019). The GPR technique uses highfrequency electromagnetic waves (usually between 10 MHz and 2.5 GHz) and is based on the different physical properties and conductivities of hydrocarbons and the sediments of the subsoil (CARCIONE et al., 2000; PEDROSA et al., 2006; GALVÃO et al., 2018).

Santarém city, located in the Brazilian Amazon, has been undergoing an intense process of urbanization, increasing the demand for groundwater. However, the public administration is not able to meet the social demand, especially in the peri-urban zones which are not connected to the urban water supply network, resulting in unregulated chaotic private use of groundwater (OLIVEIRA et al., 2000; CPRM, 2012). About 50% of the Santarém population has no sewage service collection and treatment, and in the last twenty years the increase in demand, without enhancement of the public distribution system, has raised illegal and technically improper water extraction of the aquifer (ANA, 2017). Only the urban area of the Santarém region presents a high risk of groundwater pollution, related to septic tanks, landfills, cemeteries, improper construction of wells, and leaking fuel stations (TANCREDI, 1996). Thus, this scenario causes great concern regarding the environmental conditions of the Alter do Chão Aquifer System.

The Alter do Chão aquifer, with unconfined and confined aquifer properties, is the main source of the public water supply and provides a crucial source of private supplies for domestic and agricultural use in suburbs and the rural zone. In general, groundwater assumes physical-chemical and biological qualities along its course through aquifers, and such characteristics are due to the type of rocks, permeability,



porosity, and water filtration capacity (LEMOS et al., 2015). The protection of groundwater is based, mainly, on the preservation of its quality, education, and awareness of the population. An effective way to protect the quality of groundwater is using maps of the vulnerability of aquifers, which should support development plans and guide human activities to minimize impacts (CPRM, 2012). So, groundwater flow maps are important in environmental studies, such as sites with landfills, refineries, and fuel tanks, indicating concentration zones and direction of possible contamination plumes (BRAGA, 2006). These maps make it possible to verify the groundwater flow pattern, express the general behavior of the flow, and determine its sense and direction. It should be noted that the potentiometric maps are functional when aquifers are sedimentary–free or confined, and not when they are fractured.

In this context, the use of GPR in the identification of underground contamination and structures is strategic because there are no records of such investigation concerning the gas stations in the urban region of Santarém and the unconfined Aquifer System has a lack of monitoring wells and knowledge about its geology and vulnerability (GALVÃO et al., 2012; ANP, 2019). Thus, this study aimed to identify subsurface leaks of hydrocarbon from underground tanks using GPR profiles. It can be used as scientific data for public policies related to regulating companies that deal with toxic products that can pollute soils and water resources. In addition, the present work provides information about the trend of the direction of the potentiometric surface around the stations, correlating with the contamination of hydrocarbon plumes that could compromise the natural quality of the water. So, studies of an interdisciplinary, innovative, and educational approach within the area of Geosciences and Technologies are essential in a region where the installation of gas stations and the exploration of groundwater proliferate without meeting the necessary legal criteria.

MATERIALS AND METHODS

Study site

Santarém is the third largest city of the state of Pará with a population of 308.339 according to the 2021 IBGE census, with a total area of 17.898,389 km² and located at 2°26'22" latitude and 54°41'55" longitude in western Pará (Figure 1). The present work focused on two of the seven studied gas stations, and both gas stations are among the oldest in the city and located in an urban area, which can represent a great risk of contamination of the subsoil and groundwater by fuel fossils. Data collection was carried out during the month of September, during the dry season of the year, for better logistics in the use of GPR considering that the speed of the electromagnetic wave is severely affected by the presence of water (BRADFORD & SAWYER, 2002). This may present a limitation of the method in differentiating the transition zones between unsaturated and saturated sediments.

The region is situated above the Amazonian Paleozoic Alter do Chão Formation area, that covers an area of about 1,000 km² and is 600 m thick (CAPUTO, 2014). The Alter do Chão aquifer, one of the largest subterranean freshwater reserves in Brazil, consists of sandstone, claystone, conglomerates, and levels of laterite (CPRM, 2012). In the Santarém region, the aquifer is made up of fine to medium sands from the Quaternary, as well as fine to medium sandstones from the Alter do Chão Formation dated in the Cretaceous. Moreover, the geomorphological features greatly influence



the hydrogeological conditions of the Alter do Chão aquifer. The plateaus with elevations of 200 m, located further south of the Amazon Basin, are made up of clay on the surface (50 m) and present the hydrostatic level of groundwater from 35 m in depth, constituting themselves as recharge zones from precipitations. On the other hand, the lower altitudes areas, as in the northernmost portion where Santarém is located, are configured as discharge zones, with groundwater flow towards the Tapajós River, Amazonas and the streams (TANCREDI, 1996).

According to Köppen the climate is classified as Am-type, predominantly hot and humid characteristic of Tropical Forests (SUDAM, 1984). The temperature varies little from an annual average of 25°C and the average relative annual humidity is about 69%. The groundwater temperature in this region reflects the climatic conditions, with values between 25 a 29,5 °C (CPRM, 2012). There are two main seasonal periods of the year, (i) the so-called raining period with an average precipitation between 170 mm to 300 mm, from December to April, and (ii) the dry period from May to November with a mean rainfall below 60 mm, being August to October particularly hot and dry. The average annual rainfall is about 1800 mm (INMET, 2019).



Figure 1. Location, hydrology and lithology of the study area.

According to the National Petroleum Agency (ANP), there are about 40,000 gas stations in Brazil, 7.2% of them located in North Region, and around 1.058 in the State of Pará, many of them built during the 1970s (CORSEUIL & MARINS, 1997; ANP, 2022). Currently, the city of Santarém has sixty-one authorized gas stations (Figure 2),



selling special gasoline, regular gasoline, S10 diesel (special), S500 diesel (common) and, less frequently, ethanol. According to the Brazilian standard regulation NBR 13785/2003, recently replaced by NBR 16161/2019, there are ordinary, bipartite, or tripartite tanks, which contain one, two or three different types of fuels, respectively, and they are classified as ecological steel or carbon tanks with single or double walls and as cylindrical tanks that are covered by a second layer of fiberglass.

Around 30% of gas stations in Santarém are in operation for more than fifteen years, according to the National Agency of Petroleum, Natural Gas and Biofuels (ANP, 2022). This period of time exceeds the limits of the governmental CONAMA Resolution n^o 237 and are submitted to environmental protection regulations (CASTRO & BRANCO, 2003). Moreover, most establishments still do not meet the standard national regulation, such as the absence of basic safety instruments and tools such as monitoring wells and containment ditches.



Figure 2. Posts authorized for operation from 2001 to 2021 in the Municipality of Santarém (PA), Brazilian Amazon.

GPR data acquisition and processing

The data acquisition was performed by using a GSSI Utility Scan DF SIR 3000 System Multi-Frequency GPR of the Federal University of Western Pará (UFOPA). To get a better resolution in the shallow portion of the subsoil, the GPR has been integrated with a dual-frequency digital antenna with a 270 MHz mono-static principle. The acquisition parameters of the GPR sections were defined after a number of tests in loco aiming at a better relationship between the depth of effective investigation and the vertical and horizontal resolutions of the GPR imaging. Therefore, two temporal windows of 100 and 175 ns were used to get the subsoil imaging at a range of depths up to 4.0 and 11.0 m and the GPR imaging signature of the underground tanks is composed of a sequence of different reflectors and zones with electromagnetic attenuation at different levels.

To calibrate the GPR equipment, the GPR sections were made along the surface where the storage tanks are located. Thus, two gas stations in the urban area of Santarém were selected according to their operating time, geology of the area,



operational and accessibility facilities for obtaining GPR profiles. The data acquisition was made around fuel tanks at gas stations with a total of 51 2D-sections. To carry out a successful 2D or 3D GPR survey it is necessary to acquire a dense grid of GPR traces. So, a regular spacing of 1.0 m in the grid of the 100 ns survey and of 0.5 m for the 175 ns has been performed.

Image processing and treatment consisted of (a) importing raw data, (b) static correction (set time zero), (b) removing header gain, (c) vertical gain, (d) removing background, (e) running average and (f) conversion of time into depth. These corrections were performed with the aid of Reflex Win software Version 4.5.5, to improve (i) the signal/noise ratio of the images acquired by the antennas, (ii) the visualization of anomalies of interest, (iii) enhancing features and (iv) reducing attenuations and noise generated by artificial or natural sources occurring during the propagation of the radar pulse in the subsoil (ORTEGA, 2006).

Potentiometric map

In the present work, the potentiometric surface has been delimited based on data from wells close to the two presented fuel stations. To access the information, we used the database of the Groundwater Information System of the Geological Survey of Brazil (SIAGAS/CPRM), whose access is free for all researchers. The altimetry data of the wells were acquired by using the Digital Elevation Model (Topodata/ INPE) and ArcGIS 10 Software. The values of the selected static levels (NE) were subtracted from the altimetric elevation of the tubular well, establishing thus the hydraulic load (h). The interpolation of all h's generated the potentiometric surface by means of the Surfer 12 Software, interpolated by the kriging method to establish a map with hydraulic load isolines (LÖBLER et al., 2014). The potentiometric map was built by using the Surfer 12 Software which estimates the direction of groundwater flows from higher to lower potentiometric areas (HEATH, 1983).

RESULTS AND DISCUSSIONS

In the GPR sections, the storage tanks were observed from 1.5 m deep, whose reflectors showed high amplitude and hyperbolic shape in the radargrams (Figure 3 to 5). These structures were well observed using the 270 MHz antenna with a 100 ns time window and, comparatively, less prominent in the 175 ns configuration, in which they showed low reflection. Additionally, the areas in which the tanks are located represented by the attenuated zones with low reflection at 0.5 to 4.5 depth (radargrams 8 and 9) are confirmed by manufacturing standards (Figure 4 and 6).



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Figure 3. Gas station 01. Radargram 1 (line 8) using 100 ns time window, processed and interpreted, Santarém, Pará.

The GPR signature of the geological substrate and / or other related materials was identified at a depth of 4.5 m in the radargrams using a 175 ns time window configuration, characterized by high amplitude reflectors (Figure 4 and 6). The gas stations of Santarém are located on sandstones, conglomerates and pelites of the Alter do Chão Formation covered by lateritic levels (CAPUTO, 2014). However, the 270 MHz antenna did not provide further details regarding the existence of any sedimentary structures of the material below the storage tanks.

The water table was detected at gas station 01 (radargram 8) at a depth of 7.0 m, using a 175 ns time window (Figure 2). The reflected pattern exhibited continuous lines with good reflection and high amplitude, in which the water table was characterized by an oscillation of two or three bands in the wave pattern.

The high amplitude is caused mainly by the increase in the dielectric constant due to the greater volume of interstitial water in the subsoil material and this result is characteristic of waves emitted by the GPR that meet water (CRUZ, 2015). Furthermore, the transition from dry to saturated sand below the reflection of the water table waves occurs gradually and not abruptly due to capillary effects (AL-SHUHAIL, 2006).

Gas station 01 is located near the Tapajós river (about 20 m away from the river) in an area periodically subject to temporary flooding of the river during the rainfall season (during the months of March and April). Therefore, the GPR signature of the substrate below the tanks, with heterogeneous attenuation of the electromagnetic signal may indicate a soaked soil due to the rise of the water table in the area. At this gas station, it was not possible to identify any type of leak, but only a more saturated region probably related to the flow of water from the Tapajós River.



Figure 4. Gas station 01. Radargram 8 (Line 11) using 175 ns time window, processed, and interpreted, Santarém, Pará.

At the gas station 02, an attenuated region was observed in the area below and on the side of the storage tank, delimited by the red dotted line, which indicated a fuel leak from 2.5 m deep. This type of reflection was first observed using the 270 MHz antennae, with a 100 ns time window (Figure 5). With the 175 ns configuration, with a



range of 11.0 m in depth, the attenuation pattern of the electromagnetic signal was best observed in radargram 9 from 5.0 m in depth (Figure 6). Underground, the material composed of hydrocarbons derived from fossil fuels responds to the GPR signal with such type of attenuation, characterized by whitish spaces and noise, due to its high electrical conductivity (CRUZ, 2015).

Therefore, the hydrocarbons can accumulate around the underground tank due to the absence of a waterproofing system in the porous substrate, and then easily move from the unsaturated zone to the water table below it. Thus, such hydrocarbon plumes also can contaminate the water wells of the neighborhood.



Figure 5. Gas station 02. Radargram 2 (Line 5) using 100 ns time window, processed and interpreted, Santarém, Pará.



Figure 6. Gas station 02. Radargram 9 (Line 20) using 175 ns time window, processed and interpreted, Santarém, Pará.

Both gas stations 01 and 02 are among the oldest ones of Santarém according to the ANP (2022). The first one is in operation since 2003 and the second one since 2005, respectively. In their corresponding radargrams, the signals emitted by the underground tanks (these in the form of hyperbole; radargrams 1, 8 and 2) are like the interfering signals of the iron covers existing in the surface area of most tanks (Figure 3 to 5). The signs of the iron covers are of good amplitude in column form from the land surface of the area over the tank to 11.0 m in depth and were observed in practically all radargrams, except for radargram 9 of which the tank area had no iron cover (Figure 6).



The potentiometric surface of the area indicates that some drawdown cones are concentrated in the map, because of the topographic variation found in the study area, since altimetry is considered in the calculation of potentiometry, as well as pumping wells in the center of Santarém (Figure 7). Therefore, the convergences and divergences in the directions of flows can be visualized by the change from the region with a higher potentiometric surface to those with a lower potentiometry. Some vectors indicate lower regions (north and northeast portion) close to the Tapajós river and nearby the stream (west and southwest portion).

The map presented that gas station 01 is located in a high susceptibility region with a low potentiometric value since the natural direction is to meet the Amazon and Tapajós rivers (TANCREDI, 1996). Considering that this area temporarily floods, underground tanks may be exposed to groundwater, which could be a cause for concern as corrosion of the material can occur and cause leakage, and consequently, release toxic material directly into the environment. On the other hand, gas station 02 is in a region with an intermediate potentiometer that is influenced by regions with higher potentiometers, which requires greater attention, not only in areas of potential points of contamination but also in those where the flows are directed, in case of contamination plumes. Therefore, considering that the gas stations were close to different domestic pumping wells and as the water flows to regions with lower potentiometries, it suggests a vulnerability of the aquifer to fossil fuel contamination.



Figure 7. Potentiometric surface indicating groundwater flow directions in the municipality of Santarém, Pará.

Previous work (CORSEUIL & MARINS, 1997; NASCIMENTO DA SILVA, 2014) reported that many fuel storage tanks are still made of iron, which are most vulnerable to corrosion. This may be the case for gas station 02, which had not undergone any renovation since construction and, in addition, did not have a monitoring well or containment ditches. It should be noted that the legislation at the time of registration of

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both gas stations (NBR 13785: 1997/2003), currently replaced by NBR 16161/2019, defined, as one of the security measures, the non-metallic wall as a type of material for making the tanks. Therefore, factors such as the location in the flood area and the time of operation of the project reinforce the attention to the present study to portray the vulnerability of the aquifer to the contamination of the water table by chemical elements harmful to humans.

FINAL CONSIDERATIONS

The GPR proved to be an excellent geophysical tool in the study of the vulnerability of Alter do Chão groundwater to contamination by fossil fuels, mapping the underground gas stations in the municipality of Santarém (Pará). The use of the GPR allowed both the identification of the water table at station 01 (in operation since 2001) which is located in an area of periodic temporary flooding by the Tapajós river, as well as the hydrocarbon plumes at station 02 (from 2005), whose reflective signs were attenuated, due to the high electrical conductivity of the organic component in the subsoil. The combined use of two different GPR antenna configurations was crucial to identify the underground features that are sharper and better defined using the 100 ns configuration limited to depths of 3.5 m maximum. While the 175 ns setting was ideal for detecting, delineating, and sizing the oil-derived hydrocarbon contamination plumes, the local water table and fuel storage tanks. Therefore, the results obtained confirmed the importance and reliability of this geophysical tool in the identification of environmental problems involving underground water resources in urban areas, such as Santarém, whose sedimentary subsoil comprises one of the largest aquifer systems in the Amazon, the Alter do Chão, which in the region is the main source of public and private water supply with increasing abstraction without complying with the national well-drilling legislation.

Despite the numerous advantages such as the indirect, non-invasive, quick and low-cost identification of structures, materials and underground features with the GPR technique, it is necessary to carry out other investigation methods to better characterize the contaminants quantitatively and qualitatively and the different groundwater materials and thus provide a decision-making tool based on the best available scientific data, to direct groundwater protection efforts, so that most environmental and public health benefits are achieved at the lowest cost.

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