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Integrated Earth Resistivity Tomography (ERT) and Multilevel Sampling Gas: A Tool to Map Geogenic and Anthropogenic Methane Accumulation on Brownfield Sites

by

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Abstract

Soil gas emissions of methane and carbon dioxide on brownfield sites are usually attributed to anthropogenic activities, however geogenic sources of soil gas are often not considered during site investigation and risk management strategies. This paper presents a field study at a redeveloped brownfield site on a flood plain to identify accumulations of methane biogas trapped in underlying sediments. The investigation is based on a multidisciplinary approach using direct multi-level sampling measurements and Earth Resistivity Tomography (ERT). Resistivity imaging was applied to evaluate the feasibility of identifying the size and spatial continuity of soil gas accumulations in anthropogenic and naturally occurring deposits. As a result, biogas accumulations are described within both anthropogenic deposits and pristine organic sediments. This result is important to identify the correct approaches to identify and manage risks associated with soil gas emissions on brownfield and pristine sites. The organic rich sediments in Quaternary fluvial environments of São Paulo Basin in particular the Tietê River, biogas reservoirs can be generated and trapped beneath geogenic and anthropogenic layers, potentially requiring the management of brownfield developments across this region.

Keywords
Methane, Quaternary sediments, São Paulo Basin, earth resistivity tomography

1.Introduction

The management of risks associated with the development of brownfield sites usually focuses on anthropogenic sources of contaminants. In some cases however, there can be both anthropogenic and geogenic sources all of which need to be correctly identified in order to effectively manage risks (Lundegard et al. 2000; O'Sullivan et al. 2010). Occurrences of soil biogas (predominantly methane and carbon dioxide) at old waste disposal sites on the plains of the Tietê River in the city of São Paulo, Brazil, have required risk management and remediation measures as development has encroached. The source of methane formation had been regarded as former waste disposal sites, with gas traveling through soils to buildings and other environmental receptors. Despite this pollutant linkage being valid in many cases, it does not explain all occurrences of soil biogas. A key problem in site characterization of such highly disturbed brownfield terrains is to identify where the sources of methane are, and in what sub surface units reservoirs of biogas are accumulating.

Methane is also an important greenhouse gas and the correct attribution of its source as anthropogenic or geogenic is important in modelling studies (Bousquet et al. 2006; Miller et al. 2013), with anthropogenic sources accounting for 50 - 65% of emissions in the 2000s (France et al. 2013). In some cases however, there can be both anthropogenic and geogenic sources all of which need to be correctly identified in order to effectively manage risks.

A major difficulty in studying landfill methane production is determining the vertical distribution of gas accumulation (Nastev et al. 2001; Scheutz et al. 2014). Multi-level monitoring wells usually are applied to groundwater studies and can be designed to track gas composition and distribution with depth and in some cases may act as preferential pathways for gas transport (Dumble et al. 2006).

This paper presents a field study to outline the distribution of soil gas at a brownfield site with a complex history on the Tietê River plain, using direct sampling from multi-level monitoring wells and geophysical data from Earth Resistivity Tomography (ERT). ERT has been applied in many landfill studies to estimate the rate of gas emission (Georgaki et al. 2008), identify fire-prone areas in municipal waste landfills (Frid et al. 2009), and track methane migration from subsurface chambers until emission sites at surface (Johansson et al. 2011). This study combines direct sampling with ERT imaging to get information about continuity of subsurface accumulations, which are difficult to determine by direct sampling methods only.

2.Materials and Methods

2.1 Study site

The study area (Figure 1) has a complex history, the site has been developed on a flood plain that contains a series of anthropogenic deposits and activity (~4 m), that overlies Quaternary fluvial sediments (~6 m) which in turn overlies Neogene sandstones. The Quaternary sediments are organic rich clays and sands, in some cases storing up to 40% weight of organic carbon. These sediments were mined and reworked for building aggregates, this created void spaces in the flood plain. Engineering works controlled the route of the river within the flood plain. The recurrent flooding in wet seasons allowed sediments to settle out in old excavation pits forming localized anoxic pools. The excavation pits were also reused as municipal waste dumps prior to the enactment of environmental legislation. Spoil from river dredging and engineering was also routinely deposited onto the Tietê flood plain. The area was then affected as part of major infrastructure works in São Paulo, with spoil from tunneling works and river dredging added to fill in the swampy flood plain for the construction of major highways.

At the site place, sediments were pumped to a large settlement pond (25 hectares) where they were allowed to dry out and the process repeated until a 4 m high terrace was constructed that capped the original flood plain. By 1980 the flood plain had been leveled, capped and partially covered with impermeable soils and paving. Until the early 2000s the area was derelict brownfield land, when a series of public initiatives began to redevelop the strip of land still available along the Tietê River. This region was developed to provide leisure options, green parks and public schools. In 2005 a new campus of the University of São Paulo was established. Methane occurrences were recognized in preliminary studies for environmental planning applications and then attributed to anthropogenic sources. This required specialized design for buildings to prevent methane gas accumulation and invasion, and a comprehensive program for gas monitoring and venting wells.

2.2 Sediment sampling and analysis

A direct push rig (Model 6620DT, Geoprobe) was used to drill two monitoring wells (well-1 and well-2). The rig used 1.5 m long rods, outer diameter of 57 mm, housing polycarbonate liners (DT22, Geoprobe) for continuous sampling of sediments. Maximum sampling depth is limited by ground resistance. For the test site it was limited to depths between 10 m and 11 m when higher resistance Neogene sandstones are met. To capture variations in the sedimentary section denser sampling (0.5 m) for total organic carbon (TOC) analysis was taken in finer and darker sediments from well-1 (instead of 1.0 m for well-2). For grain size analysis, otherwise, sampling was denser (0.5 m) in well-2 and coarser (1.0 m) in well-1. Grain size analyses were done with diffraction laser technique (Mastersize 2000, Malvern) with results presented in terms of volumetric sand fraction, e.g. that fraction in which mean grain diameter between 76.1 µm and 2.5 mm. TOC analyses were based on dry sample weight before and after oxidation with hydrogen peroxide (15% H_2O_2) the results expressed as a mass fraction (%) subjected to oxidation.

2.3 Multi-level monitoring wells

Despite differences in construction details most multi-level devices are composed by a bundle of sampling tubes, each one reaching a specific depth down the well. The system illustrated in Figure 2 combines modular supporting elements with continuous sampling tubes. The modular elements are composed by connectable 3 m long segments of 50 mm PVC (polyvinyl chloride) onto which 15 polyethylene sampling tubes (outer diameter of 3 mm) are attached. The sampling tubes extend continuously from the ground surface until equidistant terminations downhole, 0.6 m apart in the test site. The base of each sampling tube is inserted into a porous cap to prevent obstruction with sediments and debris. The porous cap is sandwiched by neoprene rings that hold the sampling port at the desired position and protect it against abrasive damage as it is inserted in the borehole.

A novel approach was to attach ring electrodes to the multilevel well at depths of 4.2, 8.2 and 10.2 m, each one wired to borne terminations at the well head. This allowed geological, groundwater and soil gas sampling to be used to benchmark resistivity imaging. Figure 3 shows field operations related to the installation of the sampling device, procedures for gas and water sampling and aspects of ERT data acquisition.

2.4 Earth Resistivity Tomography (ERT)

ERT is a direct current electrical geophysics method with theoretical and investigative applications covered by many sources, e.g. (Revil et al. 2012; Ustra et al. 2012; Adhikary et al. 2014). This approach is often combined with other intrusive monitoring applications (Binley et al. 2002; Johansson et al. 2011; Schütze et al. 2012). ERT employs a computer controlled data acquisition system in which a set of electrodes at the ground surface and in boreholes take resistivity measurements. Up to a hundred electrodes can be installed and selectively activated in groups of four electrodes, two of them to input electrical current into ground (current electrodes) and the other two to measure the medium response as electric potential difference (potential electrodes). As applied here, the acquisition of ERT data employed a multi-electrode resistivimeter (StingR1-IP, Agiusa) with 33 electrodes, 28 of them staked at the ground surface each 2 m along a profile crossing wells 1 and 2. Additional electrodes were installed in the wells, two electrodes in well-1, and three electrodes in well-2. Electrode activation was done according to wenner and dipole-dipole arrays (Revil et al. 2012) providing a database with 481 readings. Each reading was repeated four times to compute statistics by adjusting current intensity to achieve better signal to noise ratio in measured transfer resistances. Transfer resistance is obtained by the ratio of the response potential to the input current for each electrode configuration, thus providing data set for resistivity imaging.

The entire data set was then subjected to geophysical data inversion with software EarthImager2D (AGIUSA, 2009) under constraints of smoothness and resistivity range (5 to 200 ohm m) for models. The resistivity model simultaneously allowing data fitting and satisfying model constraints can be accepted as solutions of the inverse problem and, as such, as possible representations for the subsurface materials. Sensitivity analysis, based on derivatives of inverted models, allows recognition of what portions of the medium better are recovered by available data. This defines low sensitivities at bottom right and left corners of the profile section where electrode coverage is poorer. In the other portions of the section electrode coverage and data sensitivity provide a unique resistivity model that can be subjected to interpretation.

A starting point in interpreting a resistivity section is to calibrate or benchmark between the resistivity domains in the ERT section and the geological units identified by coring. This procedure outlines size and continuity of geological features identified by coring or multi-level sampling of groundwater or soil gas. Close to coring or sampling locations, geological units identified by the resistivity section can be regarded as 'proven' or benchmarked, in the sense it is validated with information from direct sampling. ERT features that are distant from the sampling points but can be linked to validated units are regarded as 'inferred' (marked as dashed lines), thus outlining targets in a further rounds of intrusive investigation. As shown here, distinguishing proven and inferred targets employs no rigorous statistical analysis but instead discriminates features supported by well data from those inferred on the resistivity section alone.

2.5 Water and gas sampling

Water samples were retrieved with a 60 mL disposable syringe connected to a three-way stopcock to apply increasing suction pressure at the sampling port. In most cases three syringe volumes were enough to retrieve water samples. To remove stagnant water the first syringe volume was discarded and the subsequent volumes sampled in the absence of air, by diverting with the three-way stopcock the sampling tube directly to the bottle. Low purging volume is required due to small inner diameter (3 mm) of the sampling tube. Solid materials were left to decant and the supernatant liquid sampled and divided in two portions. In the first portion, specific conductivity (Micronal conductivimeter) and pH (pH-meter Metrohm 654 with combined glass electrode) were measured. The second sub-sample was micro-filtered (Millex 0.22 μm pores) and subjected to chromatographic analysis (Metrohm 850 System) for the ions Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^- and SO_4^{2-}.

Sampling ports with gas were identified by suction testing, done with the same apparatus used to obtain water samples. At gas dominated sampling levels no pressure gradient (as observed in saturated levels) is sustained and suction can proceed for multiple syringe volumes, with no water yield or discernable pressure drop. We termed these ports as “gas only” ports, in which gas concentration could be measured still in the field by using a portable gas analyzer (Landtec, GEM-2000) with sensors for O_2, CO_2, and CH_4. Gas pressure in such ports could be taken (LabQuest2, Vernier with pressure sensor GPSBTS) using a three-way stopcock to alternate between port and open air to evaluated pressure gradient between trapped gas and the atmosphere.

3.Results and Discussion

3.1 Geology and hydrogeology

The anthropogenic deposits are composed by dredged sediments produced when the course of Tiete River was altered. Sediments were pumped to a pit surrounded by earth dykes where it was left to settle and dry. The place was kept as a selective recipient of the dredging and as such not used to dump garbage or other spoil (construction or industrial wastes). As the work was concluded a terrace of about 4 m high was raised mostly composed by alternating layers of sand and clay sediments.The piezometric surface follows the base of the anthropogenic deposits with a hydraulic gradient of 0.0082 towards the Tietê River. The anthropogenic deposits are highly permeable (12-62 cm/day) meaning that it is not an efficient sealing unit for soil biogas.

Geogenic deposits are composed of Quaternary fluvial deposits consisting of clays, silts, sands and gravels with high lateral and horizontal variation. The Quaternary unit overlies Neogene bedrock sandstones. These fluvial sediments were deposited at wide alluvial plains with low energy meandering channels that preserved buried organic matter at abandoned meanders. Plastic, dark sediments with high organic carbon content (30% or higher) are found in many places along the plains. The Quaternary sequence is about 5 m thick, mainly composed of dark, organic clays with embedded lenses of sand. The top of Neogene sandstone is rather uniform, being intercepted at 11 m depth below the terrace surface by a set of 20 wells. The organic carbon fraction found in the Neogene bedrock is considerably less than the unconsolidated sediments, appearing as trace in TOC measurements.

3.2 Soil Gas within Piezometers

As shown in Figures 4 and 5 there is an alternation of horizons with free phase gas along the wells. The shallower horizon corresponds to the vadose zone which in the well-1 shows a smooth transition in gas composition, starting from atmospheric air in near surface until 3:2 methane and carbon dioxide mixtures down hole. Two gas horizons are found below the water table, both of then embedded in an organic-rich layer of the Quaternary São Paulo Basin. The shallow horizon in this unit is detected by wells 1 and 2 and its pressure shows no gradient with the atmosphere. The lower horizon, however, is associated with a sandstone lens and shows confining pressures of about 4.2% above the atmosphere (0.96 atm at the moment of the measurement). These results show that methane pockets in the clay layer can be kept either under confinement or in relative equilibrium with the atmosphere. The fact that a common gas composition was measured in all gas ports in well-2 suggests that the over pressured gas trapped in the sandstone could be migrating along the annulus of the sampling probe in the borehole providing an uniform gas composition in sampling ports (Dumble et al. 2006).

3.3 Groundwater and soil geochemistry

Well 1 sampled groundwater at two saturated horizons, 3 samples from 3.5, 4.0 and 4.5m, then 3 samples at 7.25, 7.75 and 8.25 m (Figure 4). The shallower saturated horizon had a chemistry that is indicative of a landfill leachate with ammonia increasing with depth (up to nearly 50 ppm) and sulphate decreasing with depth (from 45 ppm to nearly 0 ppm). This coupled with decreasing oxygen in gas filled ports above suggests that this particular set of sample locations is crossing aerobic/anaerobic redox boundaries (Prommer et al. 2006). The pH across the this horizon is constant at around 6 but conductivity and ionic strength of anions and cations increases with depth, suggesting a concentration of groundwater contamination above the clay layer. This shallower region of groundwater indicates an anthropogenic source of groundwater contaminants and related soil gas most probably from wastes carried within the dredged sediments.

Within the gas-rich clay layer (5-8 m depth) that separates the upper and lower groundwater the amounts of total organic carbon (TOC) can be as high as 35% but also shows expressive vertical and lateral variations suggesting an irregular distribution.

Ionic composition of groundwater is more uniform at lower levels of the wells and with the overall ionic trend reversed. Conductivity and ammonia decrease with depth and sulphate begins to increase with depth in well-1 (Figure 4).This suggests that although the Quaternary sediments have a considerable amount of organic matter that is also degrading anaerobically and producing methane gas, the base of the Quaternary sediments are becoming more aerobic. This could be attributed to oxygenated groundwater coming from the permeable Neogene sandstones into the overlying Quaternary deposits.

Well 2 sampled groundwater at the same depths below ground level as well 1. As shown in Figure 5, the shallower horizon had elevated levels of Ca, Mg and K, which decreased with depth with levels of Na and Cl increasing with depth. This may be indicative of leaching from dredged sediments by rainwater ingression. The base of the shallow groundwater horizon has a similar chemistry to the deep groundwater samples with the exception of sulphate, which is depleted in the deeper groundwater horizon suggesting a more anaerobic environment at depth. This suggests that the top of the shallow groundwater horizon is aerobic and is affected by an anthropogenic materials leaching downwards. Well-2 is similar to well-1 in that soil TOC is elevated below the gas pocket below shallow groundwater horizon suggesting that this could be a possible source of methane. The base of this well is in a Quaternary sand lens, with methane kept under confining pressure relative to atmosphere. This would agree with the proposition that the soil gas measured in well 2 is predominantly geogenic and originates at depth and is using the borehole annulus as a preferential pathway. A source for the methane in the Neogene sandstone is unlikely due to its lower TOC content (below detection limits).

3.4 ERT section and quantified conceptual model

ERT results are presented in Figure 6. Sediment analysis and core description identifying main layers crossing by the wells were used to interpret the resistivity section (Figure 6a) provided by the ERT survey. Figure 6b shows the sensitivity map (AGIUSA, 2009) for the resistivity imaging showing places of the sections better resolved by the measured data set. These places define a neighborhood with respect to the electrode distribution, either near the ground surface as at a range from the boreholes electrode. Portions at bottom left and right of the section have lower sensitivities, meaning that these portions are not properly resolved. As shown in Figure 6c, most structures under interest are within portions with high or intermediate sensitivities, meaning they can be regarded as properly imaged. Figure 7 shows the cross plot of measured and calculated data, showing that resistivity model allows data fitting to measured data.

The benchmark for the resistivity contrast was defined by comparing sampling data with resistivity values from the section. The contact of the landfill layer with Quaternary sediments is marked by contrasting resistivity observable all along the section. Organic sediments show low resistivity values between 5 and 25 ohm m, in contrast with gas bearing lenses with resistivity between 80 and 110 ohm m. Higher resistivity for gas pockets can be explained as a result of gas accumulation and subsequent expulsion of lower resistivity pore fluids (Christophersen and Kjeldsen. 2001; Johansson et al., 2011; Schütze et al., 2012).

Some features identified by the wells are not clearly recognized in the resistivity section. The water table interface is not associated with resistivity changes and is outlined mainly based on well data. The resistivity contrast caused by the water table was not uniform along the section. This can be attributed to lateral inhomogeneity in the landfilled matrix, which varies from a highly porous matrix (sand and gravel materials) to one dominated by clays. A sharper resistivity contrast is expected in a more porous matrix where bulk resistivity is dominated by pore fluid conductivity. This is not the case for clayey matrix where the resistivity contrast is blurred or even absent. This lack of resistivity contrast suggest that resistivity of the vadose zone, mostly encompassing the landfilled layer, depends more on the composition of landfilled materials than pore fluid resistivity. No resistivity contrast is also observed between areas of contrasting Total Organic Carbon. Lenses of coarse and unsorted sandstones are embedded in the lower section of the Quaternary bed, suggesting a depositional sequence that fines upwards. Despite distinctive lithological aspects the resistivity contrast between Quaternary and underlying sandstones is not high enough to allow their distinction in the ERT section.

The integration of well data and resistivity imaging allows a new and extended schematic model (Figure 6c) compatible with all available data.
According to this model gas pockets are positioned within thick organic layers, suggesting it generates and stores biogas. Storage in sandstone facies suggests soil gas transport in which biogas migrates to more permeable medium pushing out and replacing pore fluids. Gas pockets in the clay horizons are occupying fissures after pore fluid expulsion by incoming soil gas. These processes can be difficult to identify from core descriptions and sediment analysis alone, but geophysical modelling allows interpretation of biogas transport beyond the boreholes.

4.Conclusions

Despite the small scale of the geophysical and borehole transects, the results from the combined application of multi-level sampling and resistivity imaging provide useful information to characterize methane emission and identification of biogas reservoirs in the subsurface. Such accumulations are outlined as more resistive targets (80 to 110 ohm m) in ERT imaging, in contrast with lower resistive background (5 to 25 ohm m) provided by organic sediments. In the case of this site, the naturally occurring organic clays serve either as sources and as traps for soil biogas. Biogas tends to accumulate in more permeable horizons such as sandstone lenses and fissured clays. Isolated permeable units, lacking connectivity towards the ground surface, can develop into over-pressured accumulations, as within sandstone lenses.

A conceptual model for gas generation and accumulation in such anthropogenic and geogenic environments is required. A thick (5 m in our case) impermeable organic rich layer is necessary in order to confine and prevent aerobic processes dominating. In this particular case the natural anaerobic environment is given additional protection by the anaerobic degradation of anthropogenic wastes above it. This anaerobic environment can be preserved even considering locally high permeabilities and soil variation. This anaerobic condition induces methanogenesis which produces biogas that can migrate to more permeable horizons. Persistent production of biogas creates an advancing front of gas that expels pore water out of permeable formations. This explains the over-pressured gas levels observed. Where migration pathways to surface are found, the gas pressure levels to that of the atmosphere, allowing the deposits to continuously act a source of greenhouse gases. Confined accumulations pose high risks to future land use since they can abruptly leak and in principle quickly fill confined spaces in housing stock at surface. This poses both an explosion and asphyxiation risk to inhabitants of developments in wider Tietê River region where anthropogenic deposits cap the organic sediments of the flood plain. Risk management practices should be oriented to consider confined geogenic as well as anthropogenic accumulations where biogas is found to occur.

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Figure captions

Figure 1 - Test site with a resistivity section (yellow line) where two monitoring wells 20 m apart were installed (not displayed); landfill extension (white) as in 1958 and 2008 aerial photos. Present straightened course of the Tietê River and other reference constructions area marked in the 1958 photo (dashed lines) for access strong landscape changing meanwhile. Mining pits for sand and gravel are recognized in the 1958 photo, with minor remains in the 2008 photo. Sediments dredged when the channel was opened were partly used to level lowland depressions with exceeding load pumped to the landfill site (white polygon) of about 22 hectares. Images freely available from www.geoportal.com.br Accessed 10 January 2012.

Figure 2 - Schematics of the multi-level sampling device installed in monitoring wells and photo of a sized down prototype. Stainless ring electrodes and connections are used to install down hole electrodes for resistivity imaging. 1) well head tube organizing plate according sampling depth; 2) tube cap; 3) borne for electrical connection; 4) continuous sampling tube; 5) copper wire; 6) modular PVC tube supporting the installed devices; 7) filter ring to prevent tube obstruction with sediments; 8) protective rings for the sampling termination; 9) additional modular tube; 10) stainless electrode.

Figure 3 - Field work: a) head of multi-level sampling device; b) multi-level 10 m long device, with 15 sampling ports each 0.6 m; c) direct push rig; d) liners and sampled sediments; e) insertion of the sampling device into the well; f) multi-electrode resistivimeter; g) connection to a surface electrode; h) connection to down hole electrodes; i) gas analysis at a sampling port; j) water sampling from ports at water saturated levels.

Figure 4- Analytical results for Well-2. Gas composition (for ports yielding only gas); total organic carbon, sand grain fraction in cored sediments, physico-chemical parameters (pH, specific conductivity) and ion composition for water

samples. Sand fraction corresponds to grain size between 76.1 µm and 2.5 mm measured with laser diffraction

Figure 5 - Analytical results for Well-2. Gas composition (for ports yielding only gas); total organic carbon, sand grain fraction in cored sediments, physico-chemical parameters (pH, specific conductivity) and ion composition for water samples. Sand fraction corresponds to grain size between 76.1 µm and 2.5 mm. Extended (1.5 m long) filtering interval installed between 8.5 and 10.0 m to sample the basal sandstone layer.

Figure 6- a) Resistivity section form ERT with surface and downhole electrode; b) sensitivity map for the resistivity image; c) conceptual model constructed by integration ERT imaging, sediment coring and from multi-level sampling. Position of electrodes downhole are slightly shifted for display purposes, to not overprint symbols denoting sampling ports

Figure 7 Cross plot of measured and calculated data from ERT imaging. Total of 434 data points, data fitting with RMS (root mean square) of 4.0%