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Environmental geotechnics: Brazilian experience and some hot topics

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Article

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Abstract
This paper, in commemoration of 75 years of the Brazilian Society for Soil Mechanics and Geotechnical Engineering, highlights contributions of Brazilian Geotechnicians to some selected topics of Environmental Geotechnics, i.e., design of municipal solid waste landfills, tropical soils and alternative clay liners, and remediation, and also brings forward the production of new geomaterials by means of an example of an engineered clay developed in Belgium with enhanced performance in barriers systems. The overall Brazilian expertise in geoenvironmental research, practice and design extends to many other geoenvironmental topics that were not presented and deserve another publication. Nonetheless, based on the given examples, the paper shows the essential role of Geotechnics in the well-being of the populations and in the fight against deterioration of the planet, and that challenges are continuously in progress for environmental Geotechnicians.

1. Introduction

Environmental Geotechnics has been an established branch of Geotechnical Engineering for about 40 years. Environmental issues have become a significant component of Geotechnical Engineering since circa 1980, although Geotechnical engineers have been involved with such themes for long (Shackelford, 2005). International landmarks in the scope of the ISSMGE (International Society for Soil Mechanics and Geotechnical Engineering) are the first technical session on Environmental Geotechnics that took place in the IX ICSMFE (International Conference on Soil Mechanics and Foundation Engineering) in 1977, the foundation of TC5, the Technical Committee on Environmental Geotechnics, presently TC215, in 1992, the 1st International Congress on Environmental Geotechnics in 1994, in Canada, the creation of TC307, the Technical Committee on Sustainability in Geotechnical Engineering, in 2012, and the launching of the journal Environmental Geotechnics in 2014.

The contribution from Brazilian professionals and researchers is meaningful in the many activities in this field (Boscov & Hemsi, 2020). In the scope of ABMS (Brazilian Society for Soil Mechanics and Geotechnical Engineering), a state-of-the-art report on Environmental Geotechnics was conveyed in the VIII COBRAMSEF (Brazilian Conference on Soil Mechanics and Foundations Engineering) in 1986, the Technical Committee on Environmental Geotechnics was created in 1994, and the 1st Brazilian Congress on Environmental Geotechnics was launched in 1999, as a sequence and development of the Symposia on Tailings Dams and Waste Disposal held in 1987, 1991 and 1995, and occurs since 2003 together with the Brazilian Congress on Geosynthetics.

Environmental Geotechnics was primarily concerned with safeguarding natural resources for future generations, acting mainly in waste disposal, soil contamination, and recovery of degraded areas. The pressure brought by extreme climatic events, increasing generation of waste and of more complex

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types of waste, need for decarbonization of human activities, increase of world population with consequent clamant need for food, energy, housing, transportation, among others, have all brought new challenges for Geotechnicians. The life cycle of materials, waste management, and sustainability of geotechnical projects cannot be ignored even by traditional engineers.

The main branches of activity in Environmental Geotechnics include design, operation and monitoring of waste disposal sites; use of soils and geosynthetics as construction materials for environmental protection works; use of geotechnical techniques for environmental protection; use of waste as geomaterial; prevention of, and recovery from, accidents and natural disasters; prevention of contamination of superficial soil, subsoil, and surface-and groundwater; recovery of degraded areas; remediation of contaminated land; environmental impact assessment of civil works; risk analyses; environmental licensing; energy foundations; and assessment of sustainability of earthworks, among others.

Brazilian Environmental Geotechnics has developed in many aspects, notably design, construction and monitoring of MSW (municipal solid waste) landfills, what in turn stimulated consistent research on pollutant transport through tropical soils, with emphasis to the construction of clay liners with local soils. Relevant findings about the geoenvironmental behavior of our soils, however, have not yet been effectively incorporated into the design and practice of environment-related geotechnical works. On the other hand, in the scope of waste landfills and disposal sites as well as in all other geotechnical areas, the use of geosynthetics is consolidated, thanks to the versatility of these materials and to the dynamism of the geosynthetics milieu.

Remediation projects of industrial areas are mostly developed by Brazilian branches of foreign firms, with a certain limitation of decisions taken by our engineers, despite their being undoubtedly prepared to captain the whole process and up-to-date research being carried out for three decades, as some successful examples in this paper will exemplify. Noteworthy is the resistance in our country, including the general public, to accept contamination containment by impermeable barriers, a typical geotechnical technique, as a strategy for land reclamation. In the state of São Paulo, covers, physical barriers, and geotechnical encapsulation have been applied only in 73 out of 6,270 rehabilitated or in treatment areas (CETESB, 2024), while the subsoil of a large extension of the state consists of thick layers of unsaturated soil, where these low-cost techniques are effective.

Recently much effort has been directed by research groups and enterprises all over the country to investigate the feasibility of reusing waste as geomaterials, such as construction and demolition waste, water and sewage treatment sludges and many industrial byproducts, with special emphasis on mining tailings. Nevertheless, many difficulties still hinder the application of the promising results of these investigations, e.g., acceptance from the public, environmental protection agencies, contractors and designers; logistics; lack of technical standards; among others.

Geotechnicians' contributions to the environment and human health can be even more relevant with an effort to disseminate technical and scientific knowledge, particularly that developed considering local conditions, in order to enhance acceptance of new proposals by citizens, governmental agencies and constructors.

2. Objectives

In commemoration of the 75th anniversary of the Brazilian Society for Soil Mechanics and Geotechnical engineering, this paper aims to present Brazilian contributions for Environmental Geotechnics in the scope of three selected topics, as well as to bring forward a promising theme by means of a successful example developed by a foreigner colleague. A counterpart of praxis and research was sought for each topic.

The selected topics are related to the following themes:

- Municipal solid waste landfills;
- Site remediation;
- Tropical soils and alternative clay liners;
- New geomaterials.

Reuse of waste, bioengineering, energy piles, geosynthetics and the manifold other topics were not addressed in this paper but represent a very important part of Environmental Geotechnics with substantial Brazilian contributions. However, choices had to be made.

3. Tropical soils and alternative clay liners

The international literature on waste landfill liners has for the last two decades notably focused on the behavior of bentonite, with remarkable developments in the understanding of relevant phenomena, in the mathematical modeling of these phenomena, and in obtaining design parameters (e.g., Bouazza, 2002; Katsumi & Fukagawa, 2005; Shackelford et al., 2010; among many others). This emphasis is probably due to the widespread use of GCLs (geosynthetic clay liners), a product with multiple advantages mainly for bottom liners of waste disposal sites, although often at a very high cost.

However, in developing countries, and more notably in small municipalities, the adequate use of geosynthetics in protection layers may be impaired by costs and lack of local technology for adequate construction. In Brazil, for instance, the Federal Constitution of 1988 empowered the municipalities as responsible for the organization and maintenance of local public services such as health, sanitation, and education. However, 44% of the 5,570 Brazilian municipalities have less than 10,000 inhabitants, and 23%, less than 5,000 (IBGE, 2024). These local governments very often lack specialized personnel for earthworks that demand a little more than very basic technical knowledge.

Given that compaction is a common geotechnical service and that tropical soils are ubiquitous in Brazil, research has been carried out for longer than thirty years by many research groups to disclose the geoenvironmental behavior of tropical soils. Other available materials have also been investigated, mostly various types of waste and soil-waste mixtures. Other countries have also pursued the safe and economic construction of liners with alternative materials. The literature shows sustained research about alternative liners but seldom published in the most prestigious technical and scientific journals, probably because regarded as irrelevant for the most developed countries.

3.1 Some basic concepts of tropical soils

According to the terminology proposed by the Technical Committee on Tropical Soils of the International Society for Soil Mechanics and Geotechnical Engineering (ISSMFE, 1985), a soil is classified as tropical when it was generated in the tropics or in a region of humid tropical climate and presents peculiarities in its geotechnical properties and behavior when compared to non-tropical soils (sedimentary and/or generated in temperate or cold climates) due to geological and pedological processes typical of humid tropical regions (ISSMFE, 1985). This definition, essentially technological and scientifically imprecise, otherwise takes into account geotechnical experience with these soils (Boscov, 2010).

The main groups of tropical soils are the lateritic and the saprolitic soils (or saprolites). Lateritic soils are formed by the intensive physical-chemical process called laterization, i.e., decomposition of feldspars and iron-magnesium minerals; silica and base leaching; and concentration of iron and/or aluminum. Their clay fraction is mainly composed of kaolinite (Vargas, 1982) and Fe and/or Al oxides and hydroxides, which precipitate and cement groups of clay particles, forming stable concretions that do not absorb water (Bernucci, 1995). The colloidal activity of kaolinite, the lowest among clay minerals, is further reduced in lateritic soils due to covering and cementation of clay particles, and individual grains are not distinguishable even with scanning microscopy (Boscov, 2010). In silt and sand fractions, usually quartz predominates together with lateritic concretions and heavy minerals such as magnetite, ilmenite, rutile, tourmaline and zircon; high iron concentrations may also turn concretions into solid lateritic gravel (Nogami & Villibor, 1995). There is not a typical grain-size distribution, depending on the quartz percentage and the size of the concretions, but generally the clay fraction is high, and the grading curve is not well distributed.

Lateritic soils are situated in the well-drained superficial layers in the inter-tropical latitudes of the planet, in regions of high temperatures, humid environment, abundant rainfall and water seepage. In Brazil, they cover large extents, with thickness varying from centimeters to meters. Lateritic soils are not necessarily residual soils; transported layers may also suffer laterization.

In-situ lateritic soils are porous, with low density and high hydraulic conductivity. However, when compacted, they present low hydraulic conductivity, high shear strength and low compressibility. Compacted lateritic clays have been extensively utilized in embankment dams (Cruz, 1996), pavement construction (Godoy & Bernucci, 2000), and as bottom liners and cover systems in municipal and industrial solid waste landfills, tailings dams and effluent ponds (Boscov, 2004).

A soil is classified as saprolitic, young residual soil or saprolite if it is a soil in the geotechnical sense (no need for explosives to excavate it), clearly shows relic structures of the matrix rock from which it originated; and is residual. Saprolitic soils are formed by intensive weathering of rocks; mechanical characteristics of the rock are modified during this process, but visual appearance, discontinuities, schistosity, faults, foliations and intrusions are preserved (ISSMFE, 1985). The geo-mechanical behaviour of saprolitic soils is highly influenced by matrix rock and degree of weathering. As weathering of the rock advances through fissures, with partial or total transformation of primary minerals, inherited features gradually disappear, forming a heterogeneous profile with finer material and predominance of secondary minerals near the surface, and rock fragments and coarse grains composed of primary minerals in deeper layers.

Saprolitic soils occur in Africa, America, Asia, Australia and Europe. In Brazil, large extensions are constituted by saprolitic soils, which may be exposed or covered by lateritic soils. Saprolitic soils derived from acidic rocks which occur over Brazilian and Guianas Pre-Cambrian Shields have been used with documented success as dam cores and pavement sub-bases due to adequate shear strength and low deformability obtained when compacted and confined (Stuermer et al., 2008) but may swell significantly when unconfined due to the presence of mica and kaolinite in the silt fraction (Boscov et al., 2009). Saprolites derived from basaltic rocks have also been used in earthworks in southern Brazil (Mori, 1983; Cruz, 1996).

The typical macro and microstructures and mineralogical composition of tropical soils render the traditional geotechnical systems of soil classification ineffectual to predict field behavior. Consistency limits generally fall near the A-line of Casagrande's plasticity chart, regardless of cohesion and swelling behavior. Highly-laterized fine soils when compacted may show nil volume change in contact with water and friction angles similar to those of sands, while saprolitic silts will swell drastically (but with low swelling pressure, so that some confinement may significantly hinder expansion in contact with water). Specific geotechnical classification systems have been proposed (Nogami & Villibor, 1995; Cozzolino & Nogami, 1993; Ignatius, 1988; Bernucci, 1987; Godoy & Bernucci, 2000; Arnold, 1986), whereas pedological classification may bring useful information, especially concerning solute transport through the soils.

3.2 Geoenvironmental behavior of tropical soils

Brazil's territory is mostly covered by thick layers of fine soils. These soils are generally characterized by low pH, low organic matter content (*OM*), and low cation exchange capacity (*CEC*). Fadigas et al. (2002) analyzed 162 Brazilian clayey soils and observed that $OM \leq 2.0\%$ for 91% of the soil samples; 89% had $pH \leq 6$ and 60%, $pH \leq 5$; 79% presented *CEC* lower than 10 $cmol_c.kg^{-1}$ and 96%, lower than 20 $cmol_c.kg^{-1}$. These values suggest low capacity for retaining pollutants; however, this is not the case. The low *CEC* of lateritic soils is probably related to the predominance of kaolinite among the clay minerals and the cementation of clay particles. However, the presence of variable-charged minerals, namely the iron and aluminum oxides, causes an adsorption capacity higher than expected, besides the capacity of also attracting anions.

Nationwide investigations have brought a consistent framework of knowledge about tropical soils, notably lateritic soils, which form a more homogeneous group in terms of mineralogy, texture, and chemical parameters, as well as geotechnical properties. However, there are not Brazilian technical standards for adsorption, diffusion, dispersion, and compatibility tests. Researchers and designers usually refer to international standards. Brazilian technical standards from geochemistry, agronomy and chemical areas are most helpful to fill some gaps, but a common basis would be profitable to share experiences and findings.

A survey conducted in 2012 (Boscov, 2012) on characterization results of lateritic soils from all regions of Brazil (north, south, northeast, southeast, and central-west) showed as main minerals kaolinite, gibbsite, iron oxides (magnetite, ilmenite, goethite), quartz, and sometimes also rutile and anatase, *pH* lower than 6, $OM \leq 2.8\%$, and $CEC \leq$ 5.3 $cmolc.kg^{-1}$, with one exception with CEC = 12 $cmolc.kg^{-1}$. The dataset is composed of the following references: Boscov (1997), Boscov et al. (1998), Leite et al. (1998), Farias et al. (1999), Leite (2001), Corrêa et al. (2004), Hemsi et al. (2002), Leite & Paraguassú (2002), Ribeiro & Lollo (2002), Tsugawa et al. (2002), Costa et al. (2003), Nascentes (2003), Anjos (2003), Gurjão (2005), Gabas (2005), Nascentes et al. (2008) Floss et al. (2010), Ritter et al. (2009) and Braga et al. (2010).

For saprolitic soils, in the same survey, similar values were observed: *pH* lower than 5.36, $OM \leq 0.7\%$, and $CEC \leq$ 7.0 $cmolc.kg^{-1}$, with one exception with CEC = 19.5-33.8 $cmolc.kg^{-1}$, however with a diversity of mineralogical compositions, since the soils were collected in regions of different geologies (Siedlecki, 1998; Paula et al., 1999; Carrara et al., 2002; Corrêa et al., 2004; Mendonça et al., 2002; Ferrari, 2005; Gurjão, 2005; Stuermer et al., 2008).

Most of these investigations determined hydraulic conductivity, sorption and diffusion and/or dispersion parameters, or compatibility with pollutant solutions, for compacted soils to be used in MSW landfill liners by means of laboratory tests with synthetic solutions or, less frequently, with real leachates and effluents.

The cited and further investigations have attested the high retardation coefficients attained by tests with metals solutions, organic compounds, and radionuclides; the capacity of simultaneously adsorbing anions, due to the variable-charge iron and aluminum oxides (Hemsi et al., 2002; Grubb & Hemsi, 2002; Mendonça et al., 2002; Borges, 2024); the relevant contribution of specific adsorption, which is time-dependent and irreversible, and closer to covalent than to Coulomb electrostatic bonds (Gabas et al., 2016).

However, the only truly eliminating parameter yet is the maximum allowed hydraulic conductivity (*k*) to water, generally equal to 10^{-9} m/s. This limit is not always met by compacted tropical soils (e.g., Cruz, 1996; Boscov, 2010; Seun et al., 2016). The high capacity of adsorbing cations and anions could compensate for a slightly higher k-value and ensure an adequate performance as pollutant barriers; however, performance-oriented design is seldom accepted by Brazilian environmental regulation agencies. Long-term performance of clay liners permeated by different synthetic solutions and leachates were the focus of Tsugawa et al. (2002), Corrêa et al. (2004), Zanón et al. (2014), Hamdi & Srasra (2013), Faraghati et al. (2019), among others.

Kinetics of adsorption in tropical soils and the influence of the double-porosity on the long time required to reach equilibrium in batch tests have also been investigated (Vega, 2016; Cavalcante et al., 2024; Borges, 2024).

3.3 Clay liners with alternative materials

Besides continued investigations about the use of tropical soils in clay liners, alternatives have been explored: soil-bentonite mixtures and waste-soil mixtures.

Bentonite addition aims chiefly at reducing the hydraulic conductivity of permeable soils and also enhancing their adsorption capacity. Waste-soil mixtures have the purpose of improving soil properties and also of giving a beneficial destination to waste. In this case, once achieving a composition that guarantees adequate behavior (according to technical specifications), the environmental safety under future solicitations and environmental conditions must be verified.

Some articles about soil-bentonite mixtures or alternative clays for liners should be mentioned: Silva et al. (2023) - local bentonitic soil; Zarime et al. (2023) - marine clay; Priyankara et al. (2013) - expansive clay, Amadi & Eberemu (2013) - fine grained lateritic soil with bentonite, Boscov et al. (2009) – saprolitic soil with bentonite, among others.

More recently, addition of various types of waste have been investigated: phosphogypsum and bentonite (Borges et al., 2023), water treatment sludge (Boscov et al., 2021; Roque et al., 2022; Marchiori et al., 2022), bagasse ash (Eberemu, 2013), wood ash (Osinubi et al., 2017), lime-mud and agricultural gypsum (Bressan Junior et al., 2022); biosolids (Goonewardena et al., 2011), biomass ashes and water treatment sludge (Studart et al., 2023; Marchiori et al., 2022), construction waste (Lukiantchuki et al., 2021), ettringite (Rios et al., 2024), PVDF fibers (Vieira et al., 2023), kraft black liquor and composite cement (Queiroz et al., 2021), among others.

Articles about real applications of these materials in engineering practice, regrettably, are very few. This remarkable framework must be put into practice to stimulate sustainable and eco-friendly solutions for waste management and disposal sites.

4. Municipal solid waste landfills

4.1 Engineering aspects of tropical landfills

In tropical landfills, environmental conditions characterized by daily average temperatures generally above 25ºC, yearly accumulated rainfall usually above 2,000 mm, and high air relative humidity, and the waste composition, rich in food waste, boost the bio-degradation processes, resulting in a much higher decay rate than predicted by governmental and climate agencies. Decay rates higher than 0.2 year^{-1} are usually reported in the literature, whereas the methane generation potential proved to be much more modest (Santos, 2017; São Mateus et al., 2012, Machado et al., 2009a). The rapid depletion of some waste fractions results in the concentration of non-easily degradable waste components over time, which also have high calorific power. Furthermore, experimental results of waste compression in large oedometric cells (São Mateus et al., 2012) have demonstrated that considerable amounts of interstitial and composition water are drained from MSW by the self-weight of the overlying layers, resulting in a much drier waste compared to the initial moisture content, observations which were confirmed in the field and coherent with the performed water balance.

The above considerations are intrinsically linked with landfill practice and draw attention to essential aspects of landfill design in tropical regions, such as the performance of provisory or definitive landfill covers and of leachate collection and treatment systems, both profoundly impacting the landfill water balance, and the possibility of using waste for energy strategies such as landfill mining (Machado et al., 2018).

Methane (CH_4) is one of the main constituents of biogas resulting from bio-decomposition processes, with a high calorific value (about 55 MJ/kg) that recommends its use for power generation or directly as fuel, both good economic and environmental options. The fuel production can be increased by the gasification of most waste components extracted from mining activities. Gasification involves several physical and chemical interactions at temperatures higher than 600 °C (Arena, 2012). The oxygen gas (O_2) concentrations are lower than necessary for complete combustion according to stoichiometric equations. The result is a hot fuel gas (syngas) containing hydrogen gas (H_2), carbon monoxide (CO), and methane (Arena, 2011; López et al., 2014; Niskanen et al., 2013). Furthermore, the heat generated during the gasification process can be used to dry the waste mass before burning.

The cover layer is paramount in the landfill's performance as a bioreactor. An effective cover layer should reduce water infiltration while reducing fugitive emissions and propitiate a truly anaerobic decomposition environment for waste decomposition. The gases generated during aerobic digestion have no calorific power! Several Brazilian works report measurements of fugitive emissions in landfill covers (Borba, 2015; Damasceno, 2018; Maciel & Jucá, 2011). The fugitive methane emissions ranged from 0 to 356 g/(m^2·day) in a landfill in Salvador (Damasceno et al., 2019), lower than those reported in a landfill in Recife (maximum of 984 g/(m^2·day)) (Maciel & Jucá, 2011), and close to those reported in a landfill located in Rio de Janeiro (maximum of 389.0 g/(m^2·day) in cracked regions) (Borba, 2015). Studies on cover layer's effectiveness in preventing waste oxidation (dos Santos et al., 2020; Machado et al., 2021) showed that the variables depth and distance from active drains that apply suction to extract the biogas influence the environmental conditions for waste depletion, the relative abundance of bacterial communities, and the microbial diversity. For either shallower MSW layers or samples located near such drains, oxygen entry in the landfill cell is plausible, and the abundance of aerobic and anaerobic facultative organisms is higher, resulting in a higher oxidation amount in the waste depletion and reducing methane production. Furthermore, less effective cover layers can increase the leachate production beyond 350 liters of leachate per Mg of MSW entering the landfill, with huge impacts on the feasibility of the landfill operation. The inefficiency of the covering layer associated with deficiencies in the liquid drainage systems leads to an increase in the level of leachate in the waste mass and reduction in the geomechanical stability of the landfill.

An aspect not usually considered in leachate production was introduced by São Mateus et al. (2012) when analyzing the water balance in the Metropolitan Center landfill in Salvador. For large landfills, where the waste column surpasses 50 meters, the amount of water drained from waste by compression is about 40% (dry basis) of the MSW's initial water content, becoming the most important source in leachate generation in tropical landfills and corresponding to around 200 liters of leachate produced per Mg of MSW. This aspect severely impacts the overall landfill performance and must be considered in the design. The waste biodegradation and compression are responsible for reducing the initial water content from about 100% to about 50% 4-5 years after disposal (Machado et al., 2010). Based on the work of São Mateus et al. (2012), a multiplatform Python® program (Waterfill) was compiled to aid in performing water balance in tropical landfills. The program accounts for aspects such as the water expelled by the weight of the superimposed layers of waste and the release of the exceeding water of organic compounds by biodegradation, besides a simplified and empirical approach to assess the water exchanges between the cover layer and the atmosphere by evaporation. The Waterfill is a free program that can be downloaded from https://geoamb.eng.ufba.br/n/programas-desenvolvidos.

Concerning the waste thermo-mechanical behavior, at least four aspects must be considered:

a) MSW is a reinforced material, with plastic and textile components acting as fibers, with a notable impact on the shear strength parameters, which are also dependent on the type of test employed for their determination (Karimpour-Fard et al., 2011; Machado & Karimpour-Fard, 2011; Shariatmadari et al., 2017; Zeccos, 2005). The disposal and compaction procedures impose prevalent sub-horizontal fiber direction and an anisotropic waste mechanical behavior. Consequently, in direct shear tests, the fibers are almost parallel to the shear plane, and the shear strength parameters are much lower compared to triaxial results, which present a concave upward stress-strain curve, so that the shear strength parameters must be defined in terms of acceptable strains (Carvalho, 1999). However, the average inclination of the failure plane in potential slip surfaces in the field is more appropriately reproduced by the triaxial tests;
b) the MSW waste particles are compressible, and Terzaghi's effective stress equation cannot be applied directly, requiring the use of a compressibility factor (Karimpour-Fard et al., 2022; Machado et al., 2017; Shariatmadari et al., 2009), as proposed in the pioneer works about effective stresses in compressive particles porous media (Skempton, 1984). Without this consideration, results from CIU and CID triaxial tests are highly inconsistent (Carvalho, 1999);
c) The immediate waste compression by self-weight and the long-term effects of creep and mass loss must be considered in landfill design. Waste compression has beneficial impacts such as increasing the landfill lifetime and decreasing the waste slopes' steepness, however also undesirable side effects e.g. damage of cover layers and drainage structures by differential settlements. Therefore, models are necessary to correctly estimate landfill settlements over time in different landfill locations, preferably considering differences in waste composition. In this regard, modeling attempts were developed (Machado et al., 2008, 2009b) which allow considering the impact of mass loss and creep in MSW compression;
d) the temperature inside the waste mass, different from the ambient temperature, impacts the MSW mechanical behavior. Temperature increments soften the reinforcement constituents (mainly plastics) and increase waste particles' compressibility, therefore increasing the contact area between particles and decreasing the pore water pressure actuation area. Recent efforts have been made to better understand and model the MSW thermo-mechanical behavior (Khaleghi et al., 2023a, b; Yazdanpour, 2024; Yazdanpour et al., 2023), and findings of such research can be easily extended to the use of light materials such as EPS and expanded clay in geoengineering. A comprehensive thermo-bio-mechanical model was presented by Machado et al. (2025), encompassing the features of the previous models of the research group and adding some new features to allow the thermal effects to be modeled. The model was successfully used to simulate the mechanical behavior of MSW specimens with different plastic contents and under varying temperatures, both in consolidated drained and undrained conditions.

4.2 Landfill management tropicalization

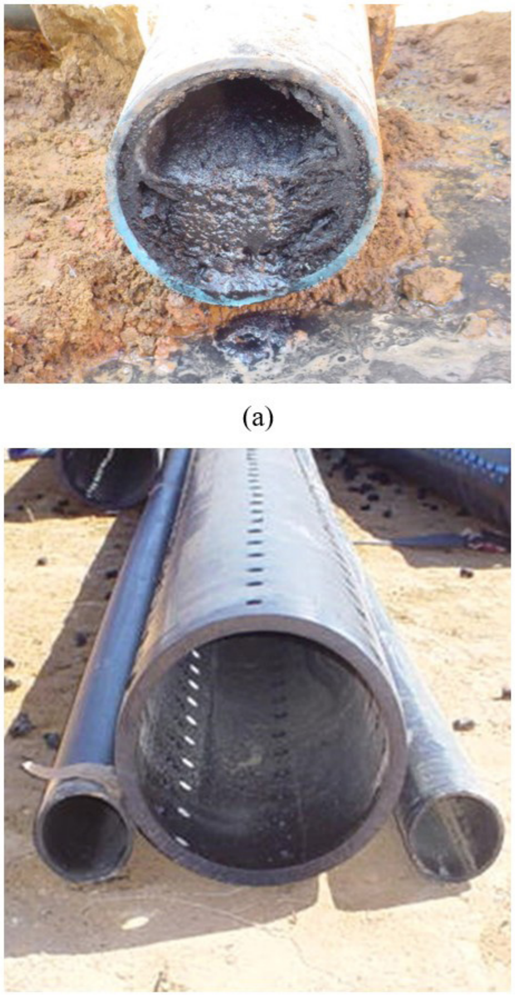
The highlighted aspects of tropical landfills - the large amount of leachate drained from waste due to compression by the weight of the overlying layers; the very quick decomposition process; and the sharp decrease in the waste moisture content over time - promote opportunities not yet properly explored in landfill management; however, some side effects must be considered in landfill design, as seen below.

Comparing similar landfills in different regions of the planet, tropical landfills will demand much more from their drainage structures. The leachate composition in the early ages carries a huge amount of organic material, which requires proper and complex treatment before final release to the environment. Furthermore, this massive organic load demands higher elevation gradients and drainage tubes of larger diameters than usual (Figure 1).

The accelerated biodegradation processes result in high waste mass settlements, which reach more than 20% of the initial height at the top cover layers. Besides, many differential settlements arise due to waste heterogeneity and in the junction between waste layers of different ages (Figure 2). Therefore, a proper final cover layer design is essential, with at least three components: a drainage layer over the waste mass, responsible for collecting the biogas that reaches the waste surface and sending it for combustion, a low permeability soil layer, and a second low permeability layer such as a geomembrane.

Furthermore, the drainage structures must be prepared for the high gas and leachate flow rates and differential settlements. Executing drainage components such as the "Ranzine" drains with usual dimensions may represent non-admissible risks of premature end of serviceable life. Figure 3 illustrates the use of a large leachate removal well composed of a central large HDPE (1.5 m) perforated tube surrounded by a vertical metallic container used for the construction of underground galleries.

The MSW characteristics and the management of tropical landfills also impact post-closure activities such as land reclamation. The accelerated decay rates of the biodegradation



(a)

(b)

Figure 1. (a) Clogging occurrence in the field; (b) Comparison between drainage tubes for tropical and non-tropical landfills.

processes would allow the landfill area to be reused in a shorter period than in temperate regions. Obviously, these post-closure uses of former landfill areas require intensive geotechnical monitoring of settlements, gas production, leachate properties, and the adoption of the appropriated risk abatement measures. Although still incipient, some examples of Brazilian land reclamation can be cited, as in the case of Salvador-BA (Figure 4), where the new metro/bus station was partially constructed over a former construction and demolition waste landfill area, and a parking area will be constructed over an ancient dump site of the city.

Finally, despite all the efforts of different segments of our society, the waste disposal scenario in Brazil, unfortunately, is still challenging and disappointing. According to ABREMA (2024), in August 2024, Brazil still had more than 3,000 dump sites, and the laws regulating waste disposal were postponed several times.

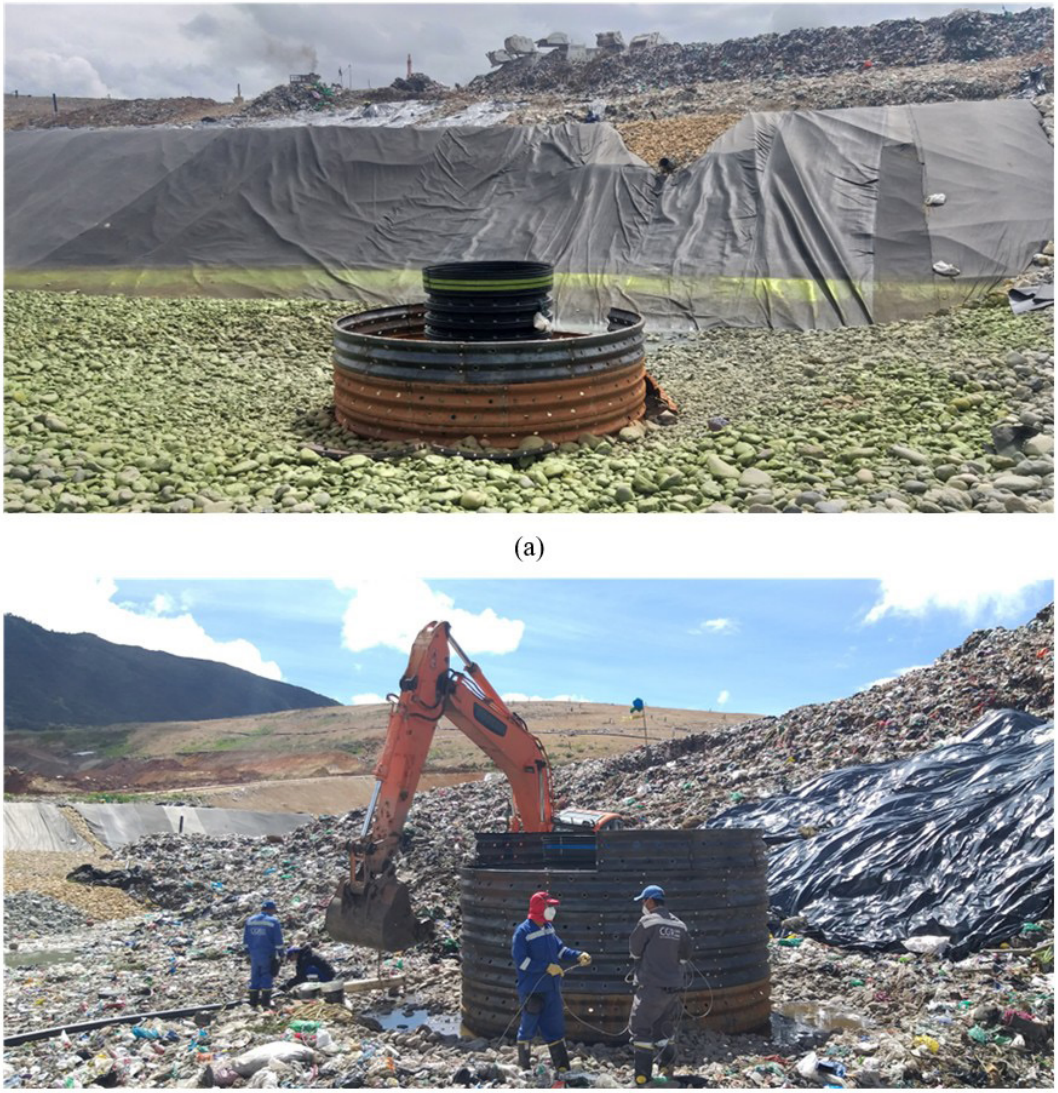
Figure 2. Cracks development in the contact between two waste uplifting with different ages.

4.3 Other important and new challenges for landfill management

There is a growing tendency to use MSW landfill facilities to also dispose of different sludge categories (Figure 5). Such materials are characterized by a physical state between liquids and solids and the possibility of flow under hydraulic gradients. Although environmentally acceptable, such co-dispositions elevate the complexity of the landfill and highlight even more the importance of a responsible geotechnical team. In such cases, much importance must be paid to the impact of sludge co-disposition on the performance of the leachate and gas collection systems and leachate treatment, as well as on the stability of the waste mass. Technical aspects and minimum criteria for the co-disposition of sewage sludge, for instance, have been discussed by Bringhenti et al. (2018). In Brazil, there are already good experiences in the management of this sludge, especially in São Paulo State, where a large part of the sludge is sent to the sanitary landfills.

5. Remediation of contaminated soil and groundwater

This chapter presents an overview of the Brazilian legal framework and management of contaminated areas



(a)

(b)

Figure 3. (a) Large leachate removal well installation in the cell base, connected to the drainage layer; (b) Large leachate removal well as a function of the disposal process.

(CAs), a brief explanation of the mostly used remediation techniques in Brazil, some examples of remediation projects totally developed by Brazilian specialists, and a numerical solution for permeable reactive barriers in heterogeneous aquifers, situation not uncommon in Brazil.

According to recent data from the National Water Agency (ANA), almost 39% of Brazilian municipalities are supplied exclusively by underground sources, and some state capitals receive most of their water supply from groundwater, such as Boa Vista, Maceio, Natal and São Luis. Even in cities where the public water supply comes mostly from surface sources, such as São Paulo, a significant portion depends on the exploitation of aquifers, which become even more vulnerable during periods of severe drought (e.g., 2014-2015).

A great number of small communities and cities depend on water supply from aquifer systems that are susceptible to contamination due to agriculture practices such as Guarani, Urucuia, Bauru-Caiuá and Barreiras. Moreover, sites with soils contaminated by mining and industrial activities, which pose relevant human health and ecological risks regardless of groundwater use, are numerous in Brazil.

5.1 Brazilian legal framework for the management of contaminated areas (CAs)

Soil contamination in Brazil has been a legal issue since the 1970s. However, effective remediation actions began in the 1990s due to Class Actions by state prosecutors (at the

(a)

(b)

Figure 4. (a) Installation of the metro/bus stations partially over a former construction waste landfill; (b) Verifying the methane levels in an investigation bore-hole.

State and Federal levels) with the support of environmental agencies and better technical and scientific base. Remediation processes have followed the state-of-the-art in recent years, with the participation of national and international research institutions and companies.

Legislation on environmental pollution control in Brazil began in the 1970s, in Rio de Janeiro and São Paulo states. The National Environmental Policy, established by Law No. 6,938/1981, created environmental licensing for effectively or potentially polluting activities, in addition to obliging the polluter to compensate or repair the damage caused to the environment and third parties. The 1988 Federal Constitution dedicated a chapter to the environment (Chapter VI, art. 225), and the Environmental Crimes Law (Federal Law No. 9,605/1998) provides for sanctions ranging from fines proportional to the damage to imprisonment for those responsible for environmental crimes.

Figure 5. Sludge being landfilled in an MSW disposal facility.

Compliance with the legislation has been supported by state prosecutors ("Ministério Publico"). This institution can assign a Public Civil Action or sign a Conduct Adjustment Agreement (TAC) in which the parties involved (responsible for the CA and public agencies) agree on the actions required for rehabilitation and the criteria for completing the remediation.

Soil and groundwater contamination in Brazil is supervised by agencies of the National Environmental System. The standards and norms to be followed are established by the National Environmental Council (CONAMA), which is a consultative and deliberative body. The management of CAs follows the guidelines of CONAMA Resolution No. 420/2009 and is carried out by state environmental agencies. In addition, the Ministry of Health and the State and Municipal Health Departments are active parties.

In Brazil, soil and groundwater are legislated under different contexts. Land use is governed at the federal level by the Urban Land Subdivision Law and the Statute of Cities and is defined in the Master Plan of each municipality. Issues related to groundwater, however, are the responsibility of the states or the federal government when two or more states or foreign territories are involved, in accordance with the Federal Constitution of 1988 and the National Water Resources Policy established in Law No. 9,433/1997.

5.2 The management of CAs in Brazil

The management of CAs in Brazil has been carried out by state environmental agencies with support, in some cases, of state prosecutors. Several States stand out, such as São Paulo (CETESB), Minas Gerais (FEAM), Rio de Janeiro (INEA), Bahia (INEMA), Alagoas (IMA-AL), Pernambuco (CPRH) and Rio Grande do Sul (FEPAM). Among these agencies, CETESB has the highest level of environmental regulation. This does not necessarily imply a higher percentage of successful remediation cases. Bureaucracy often does not allow for a more objective relationship between the parties, which prolongs remediation time and remediation decisions.

The steps for managing CAs recommended by CONAMA (based on the CETESB manual) include Identification, Diagnosis and Intervention (CETESB, 2015). After the initial investigations (Preliminary Assessment and Confirmatory Investigation), if the concentrations detected exceed the guideline values, the area is declared contaminated, and, based on risk analysis, declared contaminated with confirmed risk. The guideline values for soils and groundwater in Brazil were established in CONAMA Resolution No. 420/2009. For substances not included, the standards of the Netherlands (VROM) and the USA (USEPA) are used as reference.

The rehabilitation process should begin after the identification and confirmation of a CA, with a detailed investigation followed by a risk assessment to establish Risk-Based Remediation Targets (RBRT). These concentrations (MRBRs), less restrictive than the guideline values, guide the remediation for a specific declared use for the area.

Risk assessments in Brazil have followed the methodologies of RAGS (risk assessment guidance for superfund)/USEPA and RBCA (risk-based corrective action)/ASTM, both from the United States and focused on environmental issues. CETESB used them as a basis for its manual and the Risk Assessment

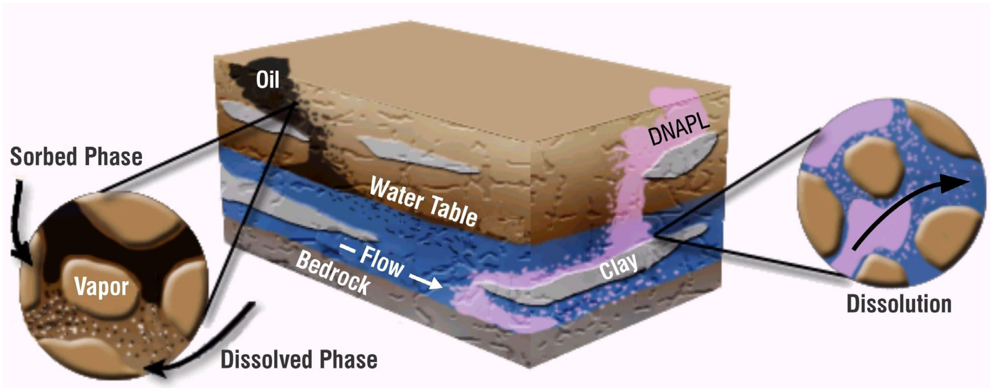


Figure 6. Block diagram with NAPL contamination (adapted from ITRC, 2011).

Spreadsheets are available on its website. In addition, the Brazilian Ministry of Health has used the public health risk assessment methodology of ATSDR (another US agency). The ATSDR protocol, in addition to following the fundamental steps of RAGS, includes specific clinical and epidemiological assessments of the potentially exposed population.

5.3 Technologies used in soil and groundwater remediation

Soil and groundwater remediation technologies have gained significant importance in recent years in Brazil. Several cases of contamination have received extensive media coverage, bringing public attention to the issue. Major remediation projects have already been conducted to meet specific risk criteria for each impacted area and in line with current Brazilian and/or international environmental standards. Additionally, preventive diagnostic work in industrial areas has enabled increasingly efficient remediation programs. Geotechnical solutions are necessary in many hydrogeological scenarios and provide effective alternatives in source control and removal as well as in linear systems such as permeable reactive barriers (Nobre & Nobre, 1997).

Soil and water contaminants include volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons, herbicides, nitroaromatics, phthalates, and heavy metals. Contamination by water-immiscible organic compounds (NAPLs – Non-Aqueous Phase Liquids) call attention due to their high toxicity and potential for migration in the gaseous, dissolved and immiscible phases (Figure 6).

The migration, mobility and persistence pathways of these compounds in the subsoil vary substantially in each case, due to the specific chemical nature of each species and geology of each site. Once infiltrated into the subsoil, the contaminant becomes susceptible to various geochemical and biological processes, which determine its mobilization or even transformation (Figure 7). These processes are intrinsically related to numerous variables, making the practice of soil and groundwater remediation complex and often expensive.

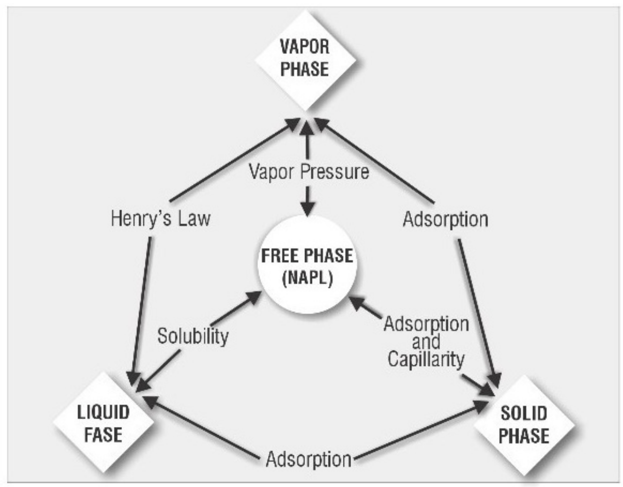


Figure 7. Phase Transformation of NAPLs.

In Brazil, the most common objectives in the rehabilitation processes of CAs have been (Nobre & Pereira, 2015):

- Containment and removal of the dissolved plume (i.e., minimizing the downgradient migration of contaminants);
- Control and removal of primary and secondary sources (i.e., minimizing additional and ongoing contamination as a result of soil leaching or secondary sources of NAPLs);
- Restoration of the aquifer to acceptable levels (based on risk), within the physical limits of the area;
- Prevention of groundwater use/consumption with concentrations above those established by the Ministry of Health and by environmental criteria according to federal (CONAMA) and state EPAs;
- Compliance with restrictions on the disposal of solid waste in the case of controlled landfills.

Table 1 summarizes the main soil and groundwater remediation techniques applied in Brazil for different contaminants.

Table 1. Soil and ground remediation technologies.

Technology	Category	Description	Application
Soil Vapor Extraction (SVE)	T	Physically removes VOCs from the saturated zone through the application of a vacuum system.	CHCs, BTEX
Bioventing	T	Accelerates the removal of VOCs through aeration on the vadose zone. Stimulates in-situ bioremediation.	CHCs, BTEX
Air Sparging (AIS)	T	Physically removes volatile and semi-volatile contaminants through aeration in the saturated zone. Stimulates in-situ bioremediation.	CHCs, BTEX, PAHs, MTBE
Bio Sparging	T	Accelerates the biodegradation of organic compounds by stimulating native microflora, through physical soil aeration processes on the saturated zone.	CHCs, BTEX, PAHs, MTBE
Permeable Reactive Barries (PRB)	T	Physical barriers that promote the passage of groundwater through “reactive gates” that enable in-situ remediation by physical, chemical and/or biological processes.	CHCs, Heavy Metals
Hydraulic Containment and Treatment (Pump-and-Treat)	C, T	Physical process of extracting contaminated water from the saturated zone and ex-situ treatment.	CHCs, BTEX
Monitored Natural Attenuation (MNA)	T	Monitoring of natural attenuation using specific parameters to validate the stabilization or shrinkage of dissolved plumes.	CHCs, BTEX, PAHs
Thermal Technologies	T	In-situ thermal processes that destroy contaminants or enable the acceleration of contaminant phase transfer in the subsurface.	CHCs, BTEX
Accelerated Bioremediation	T	Modifies the natural biochemical conditions of soil or groundwater by adding substrates to accelerate degradation by microorganisms.	CHCs, BTEX
Free Phase Recovery Systems by “Skimming”	T	Promotes the removal of the pure phase LNAPL through the application of vacuum. Stimulates bioremediation. There is no extraction of groundwater.	BTEX, Oil free phase
Incineration	T	Excavated materials are incinerated to extract volatile and semi-volatile organic compounds.	PAHs, PCBs, Pesticides
Phytoremediation	T	Specific plants are used to promote the extraction/biodegradation of organic compounds and metals from the soil.	BTEX, CHCs, PAHs, Pesticides, Heavy Metals
Soil Washing/ Reinjection/Chemical Processes	T	Washing soil with appropriate fluids to promote stripping and biodegradation. Che-mical compounds (e.g., surfactants) can be added to accelerate contaminant phase transfer.	CHCs, BTEX
Solidification/ Encapsulation/ Vitrification	C	Processes that promote the immobilization of waste through chemical and/or thermal processes.	CHCs, Heavy Metals, Radionuclides
Vertical Barriers	C	Barriers that promote containment of the secondary source (e.g., diaphragm wall).	CHCs, BTEX, Heavy Metals
Solid Waste Landfills	C	Disposal of contaminated soil in special landfills, with impermeable layers (clay liners and/or geomembranes) at the top and bottom.	CHCs, BTEX, Heavy Metals

CHCs = Chlorinated Hydrocarbon Compounds; BTEX = Petroleum Hydrocarbons (Benzene, Toluene, Ethylbenzene and Xylenes); PAHs = Polycyclic aromatic Hydrocarbons; PCBs = Polychlorinated biphenyls; MTBE = Methyl Tert-Butyl Ether; T = Treatment; C = Containment.

Pump-and-Treat (P&T), Multiphase Extraction (MPE) and Free Phase Extraction technologies have been mainly applied in the State of Sao Paulo due to the large number of active and inactive gas stations. After more than a decade of large-scale and unrestricted application of P&T systems for source control and mass removal, it became clear that this technique did not achieve, in most cases, rehabilitation in due time and at low costs. Following, soil vapor extraction (SVE) associated with in-situ soil aeration (Air Sparging/ Bio-Sparging – AIS) and oxidation/reduction of organic compounds technologies were applied to reduce operating costs; results better than expected, however, mobilized efforts to pursue innovative technologies aiming at faster, lower cost, and more sustainable solutions.

SVE is a common and efficient alternative for treating unsaturated soils impacted by volatile and semi-volatile organic chemicals. AIS uses injected air to remove VOCs from both the saturated and unsaturated zones of the subsoil and to stimulate aerobic biodegradation by increasing dissolved oxygen in the aquifer waters. Monitored natural attenuation (MNA), which relies on natural processes to remove or attenuate toxic compounds from the subsurface, may be used with other engineered systems or as a single remedial alternative; under favorable geochemical conditions, it can result in significant cost savings over more intrusive remedial options. The permeable reactive barrier (PRB) is an emplacement of reactive materials in the subsurface designed to intercept a contaminant plume, provide a flow path through the reactive media, and transform contaminants to achieve remediation goals downgradient of the barrier.

5.4 Some soil and groundwater remediation projects conducted by the authors in various Brazilian regions on industrial sites

Many soil and groundwater remediation projects were conducted by the authors in various regions of Brazil on industrial sites for the last 30 years. These include, for instance, the use of reinjection/soil washing in a 60-m deep aquifer for remediation of a DNAPL plume (Nobre et al., 1998a); implementation of a remediation strategy for the protection and rational use of groundwater resources at the Camaçari Petrochemical Complex (Nobre et al., 1998b); investigation of the use of chemical oxidants in an aquifer contaminated by DNAPLs (Nobre & Nobre, 1999); application of Air Sparging (AIS), enhanced bioremediation and risk analysis at an industrial contaminated with chlorinated organics (Nobre & Nobre, 1997, 2004a, 2008; Nobre et al., 2017); application of SVE for removal of LNAPL in the unsaturated zone (Nobre & Nobre, 2004b); implementation of a permeable reactive barrier (PRB) for a mercury plume remediation (Nobre et al., 2006); and use of drainage trenches and effluent recirculation for a barium contaminated site (Nobre et al., 2016), among other cases.

5.4.1 Application of SVE for removal of VOCs from the unsaturated zone (Nobre & Nobre, 2004b)

This treatability study comprehended the design, monitoring and remediation strategy for an SVE system installed at an industrial site contaminated with a mixture of organic compounds. The unsaturated zone at the site was very deep and presented near ideal conditions for SVE, with the contaminated portion of the porous media characterized by alluvial deposits of a well graded sand. Results of pilot-scale testing activities for an eight-month period allowed for the calibration of flow and contaminated mass transport models and the determination, in a preliminary phase, of the infiltrated mass of contaminants. The calibration of the mass transport model was implemented for different initial distributions of the contaminated mass in the subsurface, considering the existence of residual as well as pools of DNAPLs. Preliminary results suggested that the infiltrated mass at the site was in the order of 500 tons. A significant reduction in extracted vapor levels, from 400 mg/L to 80 mg/L, was verified. The simulation of various hypothetical scenarios for different operational conditions was carried out for a ten-year period. The results of the pilot-scale testing and model sensitivity analysis allowed for the definition of a unique remedial strategy for the site and design of a full-scale system.

5.4.2 Implementation of a Permeable Reactive Barrier (PRB) to contain a mercury plume (Nobre et al., 2006)

A PRB composed of a mixture of activated carbon and a granular material was proposed as a cost-effective remediation alternative to control the mercury contaminated plume due to the favorable site hydrogeological conditions and contaminant nature. Activated carbon has long been recognized for its sorption properties and is frequently used to remove dissolved organic contaminants and metals from groundwater.

The behavior, transport and fate of mercury in groundwater depend largely on its sorption reactions with sediment particles and organic matter. Therefore, laboratory batch and column tests were undertaken to establish the ideal mixture of the reactive material – with different percentages of activated carbon and sand – for treating the mercury plume, i.e., presenting a high sorption potential and, at the same time, a permeability high enough to capture the plume. The PRB would penetrate a low permeability stratum, rich in organic matter, which is a natural confining chemical layer at about 6m of depth that inhibits the vertical migration of the contaminated plume.

Groundwater flow and advective transport modeling were used during the design phase to optimize the location of the PRB as well as to assess the relative efficiency of a funnel-and-gate versus a continuous wall. A sensitivity analysis was performed to investigate the best design parameters such as wall permeability, barrier thickness, number of reactors, and PRB length that could offer the maximum capture zone to the plume at the site as well as sufficient residence time for chemical reactions to occur within the wall. The residence time was evaluated through advective transport modeling for different values of barrier thickness as well as of retardation and permeability parameters obtained by laboratory studies.

Values for the retardation parameter higher than 2,000 were obtained for mixtures with 2% of activated carbon in weight and 98% of medium sand. This composition was evaluated as the most appropriate for the PRB considering that hydraulic conductivity would remain sufficiently high, in the order of $3.0x10^{-3}$ cm/s, a value higher than that of the aquifer. Transport modeling results based on these parameters indicated that, for a 1.5m thick permeable barrier, a period of 30 years would be expected for the replacement or addition of reactive portions to the PRB based on the MCL of the mercury plume immediately downgradient the reactive wall.

5.4.3 Monitored natural attenuation and risk analysis at a site contaminated with organochlorine compounds (Nobre & Nobre, 2004a; Nobre et al., 2017)

An MNA program was implemented as part of a remediation strategy for an aquifer contaminated by organochlorines, close to an important estuarine system. A specific sampling and analysis methodology, conducted for more than five years, made it possible to verify primary and secondary lines of evidence for the intrinsic degradation of part of the dissolved mass of 1,2-dichloroethane, particularly through the reductive dechlorination process. The investigation was conducted at a site with a hydraulic containment system already in place (Nobre & Nobre, 1997). The decrease in dissolved oxygen, nitrate and sulfate levels, as well as the high methane levels in groundwater, are strong indicators of this process, including the presence of natural degradation byproducts such as vinyl chloride, ethane and ethene. A value for the average half-life of approximately 7.5 months was obtained for the natural attenuation process of 1.2-dichloroethane, based on first-order decay. RBCA analysis indicated that the transport of tracer compounds towards the nearest water body (receptor), in their dissolved phase, does not present a risk to human health and the environment due to the corrective measures implemented - and in operation - to control and remove the secondary source of DNAPL.

5.5 Remediation of residual basaltic soils of South Brazil contaminated by biodiesel

5.5.1 Basaltic residual soil

The basaltic residual soil that occurs in southern Brazil has very peculiar characteristics: clayey, structured, and with a high voids index that results in a high in-situ permeability. Table 2 presents the characterization of this soil. It can be observed that the clay content is 68%, while the permeability coefficient of 1.39×10^{-5} m/s is typical of sandy soils.

Table 2. Geotechnical and physicochemical characteristics of the basaltic residual soil from Passo Fundo and region.

Parameter	Value
Clay (%)	68
Silt (%)	5
Fine sand (%)	27
Liquid limit (%)	53
Plastic limit (%)	24
Specific gravity of grains	26.7
Natural water content (%)	34
Natural unit weight (kN/m^3)	16.3
Void ratio	1.20
Saturation degree (%)	76
pH	5.4
Organic matter content (%)	<0.8
Cation exchange capacity (cmol$_c$/dm^3)	8.6
Point of zero charge	3.1
Specific surface area $_{\text{(external+internal)}}$ (m^2/g)	33.9
Hydraulic conductivity (m/s)	1.39×10^{-5}
Soil classification	CH; A-7-5; Oxisol

5.5.2 Diesel and biodiesel remediation

Southern Brazil has the largest industrial biodiesel production plant of the country. From time to time the soil is contaminated with biodiesel, mainly due to accidents with trucks that transport this fuel. Many studies have been carried out at the University of Passo Fundo to evaluate the bioremediation of soils contaminated with biodiesel and also comparisons with diesel oil bioremediation were carried out (Meneghetti et al., 2009, 2012; Reginatto et al., 2011; Thomé et al., 2012, 2013, 2014, 2017, 2019; Decesaro et al., 2013, 2016; Cecchin et al., 2014; Schmitz et al., 2015; Kreling et al., 2018).

Bioremediation uses biological processes mediated by plants, animals or micro-organisms for soil decontamination. Many factors are important for biodegradation processes, and basic information needed includes an inventory of the residual oil concentration; population density of oil-degrading microorganisms; chemical, physical and microbiological properties of the contaminated soil; and the biodegradation potential with optimum rates of carbon (C), nitrogen (N), phosphorous (P) and potassium (K) applied as fertilizers. Efficient and low-cost methods of in-situ bioremediation include natural attenuation (natural degradation processes), biostimulation (added nutrients), bioventing (added oxygen), and bioaugmentation (introduced specific microorganisms).

Table 3 presents a comparative study of different bioremediation techniques applied to the regional soil. Figure 8 compares different treatments after 120 days and shows that natural attenuation occurs but is slower than other techniques: after 120 days the degradation of diesel was 26% and of biodiesel, 60%, while bioaugmentation achieved a degradation of 97% in the same period for biodiesel.

Cecchin et al. (2014) and Thomé et al. (2019) evaluated the influence of biostimulation with NPK and of increase in soil moisture due to rainfall on biodiesel biodegradation. Figure 9 presents one of the results; it can be observed that nutrient content directly influences the sorption capacity of biodiesel by the soil. The increase in saturation decreases the sorption capacity of clay minerals and can cause the spreading

Table 3. Tested bioremediation treatments.

Treatment	Contaminant
Natural attenuation (1)	No contaminant (Control)
Natural attenuation (2)	Diesel
Natural attenuation (3)	Biodiesel
Bioaugmentation (4)	No contaminant (control)*
Bioaugmentation (5)	Diesel*
Bioaugmentation (6)	Biodiesel*
Bioventing (7)	No contaminant (control)**
Bioventing (8)	Diesel**
Bioventing (9)	Biodiesel**

*Bacteria inoculo (~3.0×10^6 CFU mL^{-1}); **Period: four daily hours at a pressure of 280 kPa.

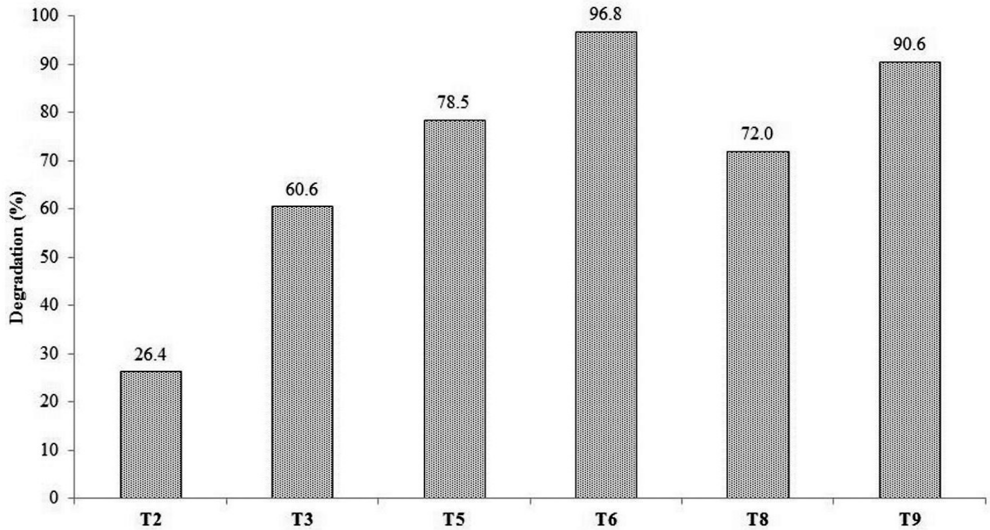


Figure 8. Percentage of total degradation of biodiesel and diesel at 120 days.

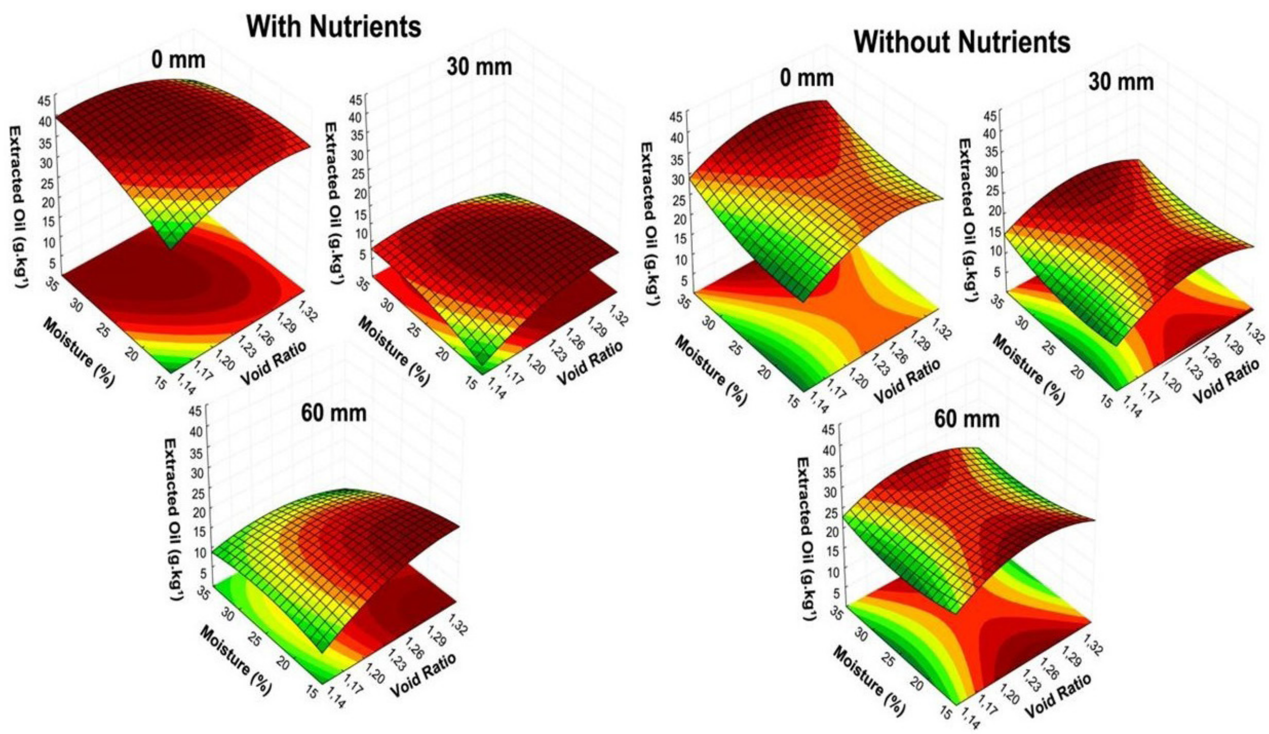


Figure 9. Extracted oil from basaltic residual soil with and without added nutrients and different amounts of rain.

of contaminants. Also, the use of biostimulation techniques with the addition of nutrients nitrogen, phosphorus and potassium (NPK) can cause significant spreading of the contamination plume if the entry of rainwater into the ground is not prevented. This has an important practical significance, because installing monitoring wells at only one fixed depth may give the false impression that biodegradation is occurring due to the reduction of contaminant amount, when in fact desorption from the soil particles and spreading of the contaminant may be occurring. Therefore, it is always recommended to use monitoring wells at different depths of the contaminated soil.

5.5.3 Nanoremediation

Zero valent metals in nanoscale (such as iron, nickel and palladium) have been widely used for the remediation of sites contaminated with various types of organic and inorganic contaminants. Due to the small particles size (generally less than 100 nm) and high surface area, this is a very stable technology for both in-situ and ex-situ implementation. Nanomaterials can stabilize or reduce inorganic contaminants such as chromium and arsenic, and dehalogenase organic compounds (Thomé at al., 2015).

Among the various types of nanomaterials, zero valent iron particles (NZVI) have been mostly used in research in southern Brazil due to their low cost of production and low toxicity (Thomé et al., 2015; Visentin et al., 2020, 2021, 2022; Reginatto et al., 2020).

Combined processes may also be used, combining physical, chemical or biological technologies to increase the efficiency of remediation, or to solve problems that may be encountered using a single technology. A novel method is the combination of the physic-chemical process of nanoremediation (fast, but expensive) with a biological process (cheap, but slow), called nanobioremediation, in which nanoparticles are used for the initial treatment of contaminated soil and micro-organisms continue the process of degradation. Nanobioremediation ends up being a more sustainable form of remediation, as it reduces the number of added nanoparticles and uses micro-organisms that are already in place (Cecchin et al., 2016, 2018).

While nanoremediation technologies have proven to be a good alternative, many uncertainties regarding long-term environmental risks have not yet been fully considered. Although some technologies have no adverse effects, such as ex-situ techniques, the toxicity, mechanism of action, and ecological risks from in-situ techniques require further investigation. Toxicity data are fragmented and incomplete, or even completely lacking. Some biological groups of organisms have been more frequently investigated, while others are unexplored. Interpretation of existing data is quite challenging due to the different forms of application of the technologies, the use of various species, the life stages or exposure conditions with different experimental designs, the consideration of different endpoints, and the lack of a database for reference (Vanzetto & Thomé, 2019, 2022; Thomé et al., 2020).

5.5.4 Sustainable remediation

Remediation, despite minimizing the effects of contamination on human health and environment, is not automatically sustainable. The awareness that application of a corrective action may have its own environmental, economic and social consequences has changed decision making on the management of contaminated areas in recent years. Initially concerns focused on the cost and ease of implementation, the time necessary for the remediation, and the efficiency to meet the decontamination targets. More recently the concept of sustainability has been incorporated; in this context, some studies have been carried out in southern Brazil (Braun et al., 2019a, 2020a; Trentin et al., 2019a, b).

Sustainable remediation is already considered an essential basis for the decision-making process. In essence, its objective is not to simply transfer the problem to another medium, geographical location or generation, or to focus only on the removal results; instead, it facilitates a long-term assessment of risks, benefits and future uses of the remediated areas (Reddy & Adams, 2015).

The methods of sustainability evaluation are systematic forms that assist in making decisions about environmental, social and economic aspects. Their concept precedes all analyses focused on sustainable remediation, since they contribute significantly and decisively to the assessment of the indicators, metrics and tools by which the sustainability of a remediation project is analyzed (Braun et al., 2019b, 2020b, 2024; Rampanelli et al., 2021).

5.6 A study of permeable reactive barriers (PRBs) in heterogeneous aquifers

The section focuses on evaluating the effect of aquifer (saturated porous medium) heterogeneity in hydraulic conductivity (*k*) on the performance of a continuous permeable reactive barrier, and presents some results from a hypothetical case study of miscible contaminant transport modeling in saturated medium under steady-state flow, with the presence of a PRB where coupled degradation reactions occur.

The heterogeneity of geologic layers is an inherent characteristic; however, it is difficult to investigate, at different scales, including the small one, and input in computer models. The unrealistic representation of *k* in the layer accounts for significant errors in model predictions when compared to field data (i.e., monitored concentrations, breakthrough times etc.).

The type of reactions chosen for this case study is the degradation of contaminants that can be represented by a sequence of first-order decay kinetic expressions, such as for two compounds A→B→:

$$\frac{d[A]}{dt} = -\lambda_A [A] \tag{1}$$

$$\frac{d[B]}{dt} = \lambda_A Y_A [A] - \lambda_B [B] \tag{2}$$

where *[A]* and *[B]* are the aqueous-phase concentrations (M/L^3) for compounds A and B, respectively, λ_A and λ_B (T^{-1}) are the corresponding 1st order decay rate coefficients, and Y_A is the stoichiometric mass ratio between compounds A and B, obtained from the chemical reaction for the degradation of A forming product B.

This type of irreversible sequential first-order decay kinetics has been used to represent the sequential reductive dechlorination of PCE and TCE to the products DCE and

VC, and, finally, to ethene, which is a non-toxic molecule. The reductive dechlorination under anaerobic aquifer conditions can occur naturally (natural attenuation), and also be catalyzed by zero valent iron (Fe^0) (Gillham & O'Hannesin, 1994; Roberts et al., 1996), which can be placed in the zero-valent iron PRBs (US EPA, 1998; Puls et al., 1999; Salles & Duarte, 2005). The reactive contaminant transport program RT3D (Clement, 1997) contains a reactive module specifically created to model the transport of four mobile species (PCE→TCE→DCE→VC) with the sequential first-order decay reactions.

Integrating in time the system of coupled initial-value problems (Equations 1 and 2), the concentrations of *A* and *B* can be ploted versus time. A numerical integration using a Runge-Kutta algorithm is commonly used. Eykholt (1999) shows an analytical solution that can also be used.

For this case study, it is considered that compound A (contaminant) is transported by advection-dispersion from a source into an aquifer, later reaching a PRB, located downgradient from the source, where it undergoes first-order degradation reaction, forming the product, compound B, which also undergoes transport and degradation.

The governing advection-dispersion-reaction equation (ADRE) for three-dimensional solute transport in saturated porous medium can be written, for compounds A and B, including the transformation kinetics described above, as (Clement, 1997):

$$R_A \frac{\partial [A]}{\partial t} = \frac{\partial}{\partial x_i}\left(D_{ij} \frac{\partial [A]}{\partial x_j} \right) - \frac{\partial \left(v_i [A] \right)}{\partial x_i} + \frac{q_s}{n}[A]_s - \lambda_A [A] \quad (3)$$

$$R_B \frac{\partial [B]}{\partial t} = \frac{\partial}{\partial x_i}\left(D_{ij} \frac{\partial [B]}{\partial x_j} \right) - \frac{\partial \left(v_i [B] \right)}{\partial x_i} + \frac{q_s}{n}[B]_s - \lambda_B [B] + Y_A \lambda_A [A] \quad (4)$$

where *t* is time, x_i and x_j can take directions x, y, and z, *[A]* and *[B]* are the aqueous concentrations for A and B, $[A]_s$ and $[B]_s$ are the source concentrations, q_s is the source discharge rate (if present), v_i is the seepage velocity vector (L/T), D_{ij} is the hydrodynamic dispersion tensor (L^2/T), R_A and R_B are the retardation factors for A and B, n is the soil's total porosity, and the reaction terms have been defined above.

The software RT3D can solve these equations, the reactions are stepwise numerically integrated using small reaction time steps contained in each transport time step, coupling transport and reactivity, in the so-called operator-splitting technique (Clement, 1997).

The one-dimensional forms (x direction) of Equations 3 and 4, assuming no sources/sinks and homogeneous medium, can be written as (e.g., Hemsi, 2024):

$$R_A \frac{\partial [A]}{\partial t} = D \frac{\partial^2 [A]}{\partial x^2} - v \frac{\partial [A]}{\partial x} - \lambda_A [A] \quad (5)$$

$$R_B \frac{\partial [B]}{\partial t} = D \frac{\partial^2 [B]}{\partial x^2} - v \frac{\partial [B]}{\partial x} - \lambda_B [B] + Y_A \lambda_A [A] \quad (6)$$

where the seepage velocity can be obtained based on Darcy's law as:

$$v = \frac{q_x}{n} = \frac{k_x i}{n} \quad (7)$$

in this equation, q_x is the specific discharge or Darcy's velocity (L/T), k_x is the hydraulic conductivity along the x direction (L/T), and *i* is the hydraulic gradient.

The hydrodynamic dispersion coefficient, *D*, includes both mechanical dispersion and molecular diffusion in the porous medium:

$$D = D_m + D^* = \alpha_L v + \tau_f D_0 \quad (8)$$

where D_m is the mechanical dispersion coefficient, given by the longitudinal dispersivity (α_L) times the seepage velocity, and D^* is the effective diffusion coefficient of the compound (in the porous medium), given by τ_f, the tortuosity factor, times D_0, the aqueous phase diffusion coefficient for the solute (electrolyte) in free solution.

Finally, the retardation factor accounting for adsorption of solutes to the surfaces of the solid phase of the soil is given by:

$$R = 1 + \frac{\rho_s}{n} K_d \quad (9)$$

where ρ_s is the dry density of the soil, and K_d is the distribution coefficient for linear, reversible, and instantaneous adsorption.

A simple-to-use analytical solution based on the one-dimensional form of the ADRE (Equation 5) can be used to pre-dimension the thickness of a PRB required for the remediation of compound A, assuming the simplifications that:

- the flow entering the PRB is uniform (aquifer is homogeneous) and can be represented by parallel flow lines, perpendicular to the PRB;
- the 1st order kinetic model can be a valid assumption to represent the degradation of compound A within the PRB;
- compound B is not a dangerous compound, such that the release of compound B is not quantified.

Under these assumptions, the analytical solution shown in Table 4 can be used for the transport and fate of compound A inside the PRB, for advection-dominated transport.

Table 4. Analytical solution for Equation 5, denoting concentration *[A]* as *C* (Bear, 1979; van Genuchten & Alves, 1982).

Initial conditions	Boundary conditions	Solution
$C(x \neq 0,0) = 0$	Upper $C(0,t) = C_0$	$\frac{C(x,t)}{C_0} = 0,5\left\{\exp\left(\frac{(v-u)x}{2D}\right)erfc\left[\frac{Rx-ut}{2(DRt)^{1/2}}\right] + \exp\left(\frac{(v+u)x}{2D}\right)erfc\left[\frac{Rx+ut}{2(DRt)^{1/2}}\right]\right\}$
$C(x = 0,0) = C_0$	Lower $\frac{\partial C}{\partial x}(\infty,t) = 0$	$u = \sqrt{v^2 + 4\lambda D}$

The analytical solution considers the PRB to be initially free of the contaminant (except at x=0), a constant-concentration (C=C_0) Dirichlet boundary condition at x=0 (inlet) and a semi-infinite boundary condition at the outlet. The analytical solution simplifies to the Ogata & Banks (1961) solution when λ=0.

Assuming steady-state transport, when concentrations within the PRB remain constant in time, the solution in Table 4 simplifies to the following analytical solution (when time is large, the first erfc → 2 and the second erfc → 0):

$$\frac{C_e(x,t)}{C_0} = \exp\left(\frac{(v-u)\,x}{2D}\right) \tag{10}$$

From where the thickness of the PRB (L_{PRB}) required to provide a certain concentration reduction ratio (C_e/C_0) can be calculated as:

$$L_{PRB} = \frac{2D}{(v-u)}\ln\left(\frac{C_e}{C_0}\right) \tag{11}$$

where C_e is the maximum allowable concentration (compound A) at the effluent side of the PRB.

A further simplified approach, applied commonly in practice (Elder et al., 2002), is to neglect dispersion and consider the passage of the contaminant through the PRB by advection only (plug flow). The solution and required PRB thickness in this case are given by:

$$\frac{C_e(L,t)}{C_0} = \exp\left(\frac{-L\,\lambda}{v}\right) \tag{12}$$

$$L_{PRB} = -\frac{v}{\lambda}\ln\left(\frac{C_e}{C_0}\right) \tag{13}$$

In this case study, it is considered that compound A (contaminant) undergoes transport and degradation in the PRB (Figure 10a). The aquifer under consideration is heterogeneous in terms of hydraulic conductivity (k) values, which have been investigated by means of the 46 sampled points depicted in Figure 10b, distributed across the area following a squared-pattern grid (k_{min} = 2.1 m/d; k_{max} = 70 m/d).

For this data set of k values, a log-normal distribution was well adjusted, with mean of lnk = 2.77 and standard deviation of lnk = 0.9, and a spherical theoretical variogram (omnidirectional) was fit to the experimental-variogram points obtained from the available values and their locations.

In the case study, the aquifer was a two-dimensional domain of 150 m in x by 100 m in y (Figure 10a), and of thickness of 1 m. The finite difference grid contained 300 columns, 100 rows and 1 layer.

For the flow model, specified hydraulic-head boundary conditions were applied at the flow inlet and outlet boundaries, x=0 line, with a constant hydraulic head of 1.5 m, and x=149.5 m line with 1.1 m. Thus, the applied regional hydraulic gradient (x direction) was 0.002676. The total porosity was adopted as uniform and equal to 0.4. The PRB was located at x = 76 to 79 m and y from 10 to 90 m. The hydraulic conductivity in the PRB was assumed to be 45 m/d.

For the transport model, a linear source was located at x = 10 m, for y from 20 to 80 m. The source remained active for the first 400 days, when the concentration of compound A remained constant at 200 mg/L. The simulation total duration was 1,000 days. The retardation factors were adopted as 1.0. The software programs MODFLOW-2000 (Harbaugh et al., 2000) and RT3D (Clement, 1997) were used for the simulations.

5.6.1 Case 1: Homogeneous aquifer with geometric-mean k value

A first simplified approach to the case study is to assume a single value of hydraulic conductivity for the

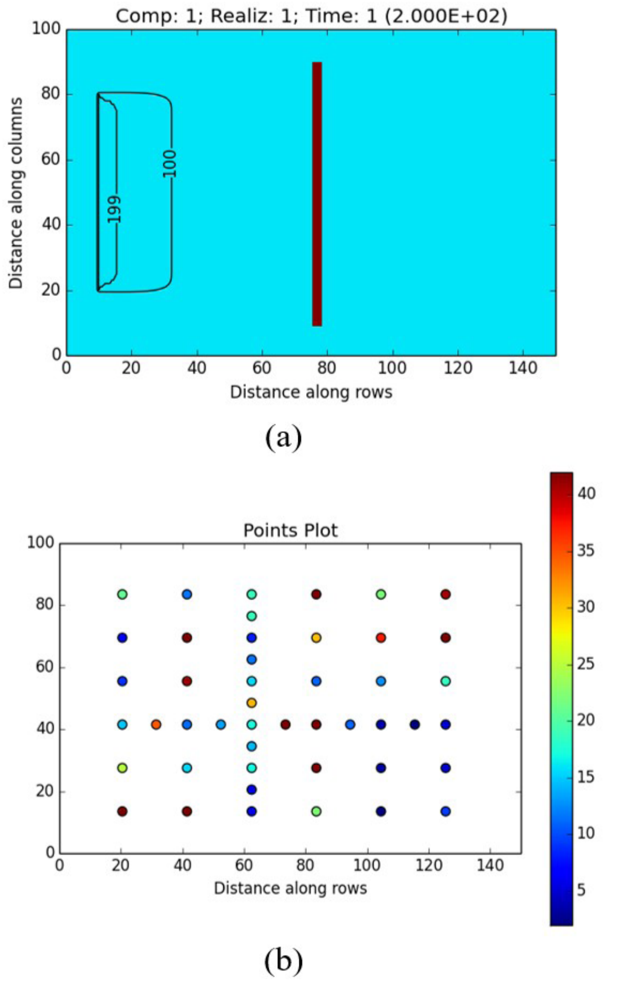


Figure 10. Plan views of (a) aquifer domain (with source and PRB); and (b) map of sampled points with k values (units: m/d).

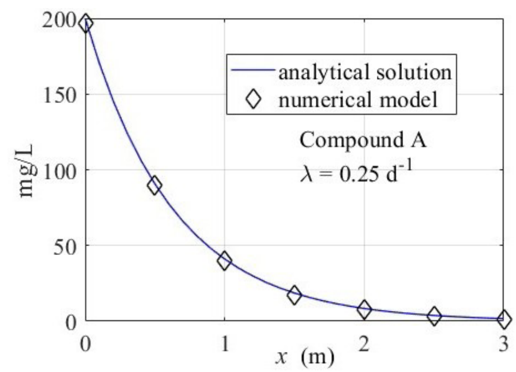


Figure 11. Compound A concentrations based on the analytical solution (Table 4) and the numerical model for case 1.

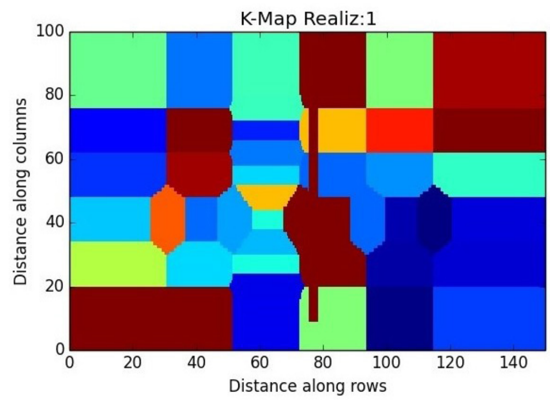


Figure 12. Plan view of the domain with k map based on the nearest-neighbor criterium (k in m/d, color bar in Figure 10).

Table 5. L_{PRB} values from the analytical solutions of Equations 11 and 13.

Equation	11	13
v (m/d)	0.107	0.107
α_L (m)	0.3	-
λ (d^{-1}) 0.25	2.9	2.0
0.30	2.5	1.6

entire aquifer, equal to the geometric mean of the k values in the sample set, 16 m/d (1.8 x 10^{-4} m/s), since they follow a log-normal distribution.

The seepage velocity results uniform and equal to 0.107 m/d. In this case, the analytical solutions of Equations 11 and 13 can be applied to pre-dimension the required PRB.

Assuming the longitudinal dispersivity, α_L, inside the PRB equal to 0.3 m and the desired C_e/C_0 ratio of 0.01, Table 5 shows the calculated required PRB thicknesses considering the values of λ = 0.25 and 0.30 d^{-1}.

Thus, based on this simple pre-dimensioning, the PRB thickness was adopted as 2.9 m, in fact, approximated to 3.0 m. The reactivity parameter for compound A inside the PRB was adopted as λ= 0.25 d^{-1}.

Exploring further the analytical solutions, Figure 11 shows compound A concentrations inside the PRB given by the analytical solution in Table 4, for the same parameter values considered for Equation 11 in Table 5. A numerical model for case 1 was also created, with v = 0.107 m/d, plug flow in the aquifer, L_{PRB} = 3 m, λ = 0.25 d^{-1} (compound A in the PRB), and longitudinal dispersivity of 0.3 m inside the PRB.

As seen in Figure 11, the numerical data points showing the decline of concentrations inside the PRB are very close to the analytical solution results. The numerical results for case 1 will be used later for comparisons.

5.6.2 Case 2: Heterogeneous aquifer using nearest-neighbor k values.

An additional numerical model was created considering another deterministic approach, which is to define the values of k in the unsampled elements of the finite difference grid using a nearest-neighbor criterium based on the 46 sampled values and their locations. Figure 12 presents the k map considered for case 2.

In this case, the porous medium is heterogeneous, and, thus, the seepage velocity is non-uniform. The same parameter values used for case 1 were adopted: L_{PRB} = 3 m, λ = 0.25 d^{-1}, and longitudinal dispersivity of 0.3 m in the PRB. The seepage velocities varied in space. Null dispersivities were adopted in the aquifer, under the assumption that mechanical dispersion was included due to flow velocity variations.

The results for cases 1 and 2 were compared in terms of the accumulated masses of compound A until 1,000 days that entered the domain, degraded in the PRB, and left, and the mass stored in the domain at 1,000 d. The homogeneous case 1 overestimated the masses that entered the domain and degraded, and underestimated the masses that passed the PRB and left the domain with respect to case 2.

5.6.3 Case 3: Set of heterogeneous aquifers generated with Sequential Gaussian Simulation (SGS)

Given the available information (set of 46 values of *k* distributed across the study area and their locations), an interesting approach is to analyze the data using geostatistics (regionalized random variable), generating a set of multiple realizations of equiprobable *k* maps for the domain, which, in turn, can be used for flow and transport simulations. This is a Monte Carlo approach, where the stochastic aspect is introduced by repeating the solution of the same problem many times, each time with a different realization of the *k* field (e.g., Elder et al., 2002; Hemsi & Shackelford, 2006; Gomes & Caicedo, 2011, 2014; Hemsi, 2021). Gomes & Caicedo (2011, 2014) used this approach not with PRBs, but to study the arrival of a contaminant, in 30 years, at a point of compliance and perform risk analyses.

A Python program named INTPRB (Machado & Hemsi, 2016) combines GSLIB packages (Deutsch and Journel, 1998) with MODFLOW-2000 and RT3D to perform data analysis, variogram modeling, kriging and sequential gaussian simulation (SGS) and generate multiple realizations of *k* fields that are run in MODFLOW-2000 and RT3D. The program works in loop, repeating the procedure for several realizations.

For case 3, 100 aquifer realizations were performed. Except for the *k* map in each run, the parameters and conditions were the same used for case 2. A null dispersivity also was considered for the aquifer, assuming that mechanical dispersion was included via the flow velocity variations. Figure 13 shows the ordinary-kriging *k* map obtained from the data interpretations and examples of simulated *k* maps generated by SGS.

The case 3 results are shown by the box plots in Figure 14. The results in Figure 14 include the accumulated masses untill 1,000 days that entered the domain, degraded in the PRB and that left, and the stored masses (at 1,000 d) upgradient, inside the PRB, and downgradient. Each box plot shows a median value (red line), the lower (25%) and upper (75%) quartiles (blue lines), a broader statistical data range (dashed lines) and,

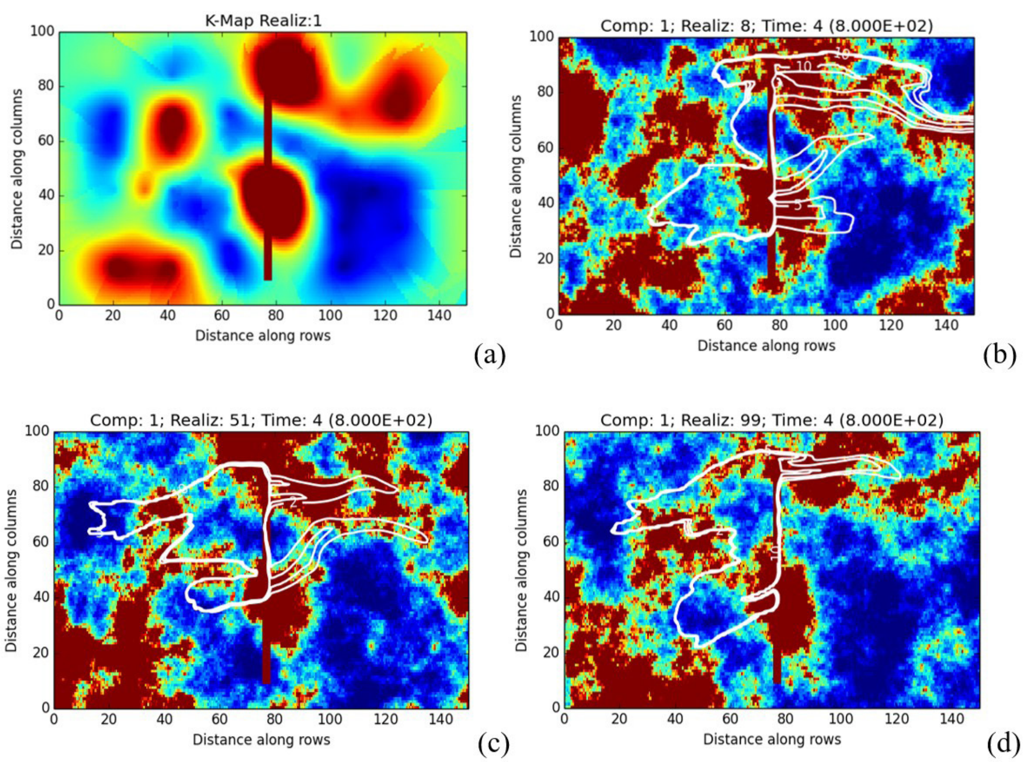


Figure 13. Plan views of (a): the ordinary-kriging *k* map, (b), (c) and (d): three examples of *k* maps generated by SGS, also showing some concentration contours (*k* in m/d, color bar in Figure 10).

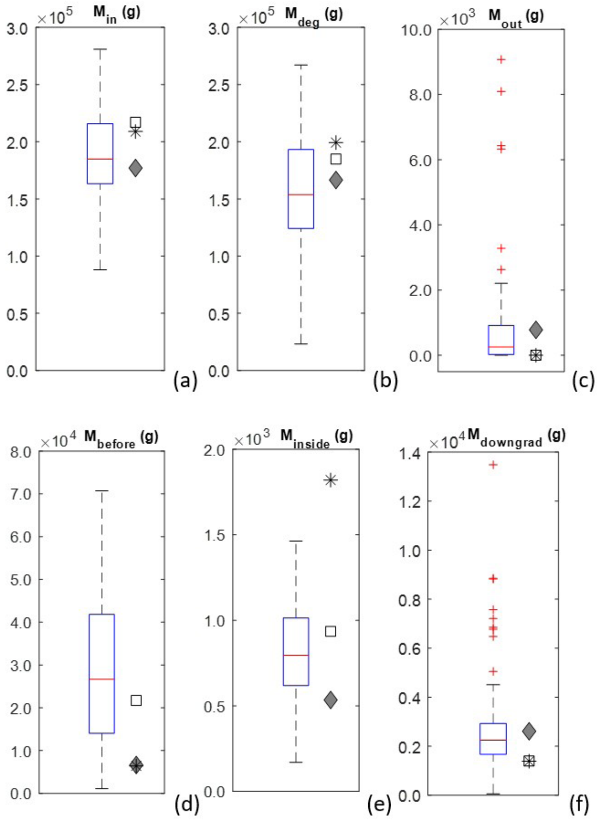


Figure 14. Simulation results (a), (b), and (c) accumulated masses until 1,000 days that entered, degraded in the PRB, and that left the domain; (d), (e) and (f): stored masses at 1,000 d upgradient, inside the PRB, and downgradient. Symbols: Case 1 (*); Case 1 with 1,0 m of aquifer longitudinal dispersivity (□), Case 2 (♦), and Case 3 (Box Plots).

if present, outliers (red crosses). For each plot, the results for case 1 (*), case 1 with 1,0 m of aquifer longitudinal dispersivity (*) and case 2 (♦) are also included for comparison.

The box plots allow us to understand the variability in the simulated results (transport and remediation) possible to have in the scenario under consideration (Figure 10), in which there are 46 points of known *k* in the layer of heterogeneous aquifer. In contrast to the deterministic results of cases 1 and 2, the SGS results (case 3) allow us to quantify variation ranges, probabilities, and outliers, associated to different variables, such as certain values of contaminant masses passing the PRB (and reaching compliance points).

6. New materials: overview of modified clays for low permeable barriers

6.1 Literature review

Hydraulic barriers containing clayey soils such as compacted clay liners (CCLs), geosynthetic clay liners (GCLs) and vertical barriers (cement-bentonite cut-off walls and soil-bentonite backfills) are widely used in geoenvironmental applications such as: impermeable barriers for the confinement of waste disposals and of polluted sites, sealing and protective layers under roads and railways, containment for above-ground tank farms, sealing under dams, canals, ponds, for waterproofing of foundations and other similar geotechnical applications. Clayey barriers are largely used for the isolation of pollutants because of their low hydraulic conductivity (*k*) to water, which, however, can be drastically increased by prolonged exposure to polluted liquids (Shackelford & Lee, 2003; Mitchell & Soga, 2005), with consequent damage to the environment and to human health.

To overcome this problem, modified clays have been introduced in the last decade in barrier applications to improve their chemical resistance to aggressive permeants (Di Emidio, 2010; Bohnhoff et al., 2013; Rowe et al., 2022; Bate & Burns, 2010; Lo et al., 1997; Lo & Yang, 2001; Malusis et al., 2010). Some examples are: multiswellable bentonites (MSB), Trisoplast, HYPER clay, dense prehydrated (DPH) GCL, and bentonite polymer nanocomposite (BPN). The potential benefits of these treated clays must be evaluated by analyzing their transport parameters and investigating the mechanisms through which the polymers and organic molecules improve the sealing capacity of a clay, to highlight the more suitable treatment techniques and to further investigate the weaknesses of the available products with the aim of future advancements and improvements.

MSB, developed by Kondo (1996), is a bentonite clay modified with propylene carbonate (PC), an organic compound able to activate the osmotic swelling capacity of the clay. PC is placed in the interlayer of the smectite and attracts numerous water molecules. MSB showed high swelling capacity and low k values for monovalent and bivalent solutions (Di Emidio, 2010; Katsumi, 2010; Mazzieri et al., 2010a, b; Onikata et al., 1999).

Trisoplast consists of a mixture of sand, bentonite and a specific mixture of polymers. The synthetic additive is a high molecular weight, hydrophilic and gel-forming polymer. Additionally, it gives rise to some internal cohesion in the sand–bentonite mixture. This mixture showed a k one order of magnitude lower than conventional compacted clay liners (Simon & Muller, 2005).

The DPH GCL is a manufactured patented GCL densified by calendaring under vacuum after the clay being prehydrated with a polymeric solution containing Na-CMC, sodium PA and methanol. DPH GCL showed excellent performance in various aggressive solutions (Di Emidio et al., 2011; Katsumi, 2010; Mazzieri & Di Emidio, 2012; Mazzieri et al., 2013). Mazzieri & Pasqualini (2008) studied the k of the DPH GCL subjected to dry/wet cycles using a 12.5 mM $CaCl_2$ solution and seawater as hydrating liquid.

Adsorption of anionic polymers onto clay surface is promoted by the presence of polyvalent cations which act as bridges between the anionic groups on the polymer and the negatively charged sites on the clay (Mortensen, 1960; Theng, 2012; Tombacz & Szekeres, 2006). Treating the clay with CMC increases its water absorption and water retention ability. Di Emidio (2010) evaluated the pollutant containment ability of clays treated with such anionic polymer, Na-CMC. This clay, named HYPER clay (HC), is mixed with a polymeric solution and then dehydrated in an oven at 105 °C to irreversibly adsorb the polymer to the clay surface. Dehydration at temperature higher than 60 °C has been proven to enhance the irreversible adsorption of the polymer onto clays' surface (Stutzmann & Siffert, 1977). After HYPER clay treatment, the anionic polymer intercalates in the interlayer region between clay platelets. The anionic polymer, negatively charged, maintains the interlayer between particles open probably due to the increased repulsion forces. This treatment method increased the water adsorption capacity of the clays, reduced k to electrolyte solutions with increasing polymer dosage, increased the heavy metals adsorption capacity, and was able to maintain performance in the long-term.

Ruehrwein & Ward (1952) stated that the adsorption of anionic polymers onto clays occurs through ionic exchange. On the other hand, Michaels & Morelos (1955) suggested that adsorption is mainly conditioned by hydrogen bonding. A third possibility would originate from the interaction between polyvalent cations acting as crosslinking agents between the clay negative surface and the anionic polymer. In the presence of salts and cations naturally present at the clay surface, the negative charges of the clay and the anionic polymer are shielded from one another allowing the polymer to coil and collapse on the clay surface. Stutzmann & Siffert (1977) compared the quality of the adsorption of anionic polymers onto montmorillonite surface for two scenarios: (a) drying the treated clay either at 60 °C and (b) drying under vacuum at 20 °C. They found that the adsorption of the polymer on the montmorillonite after drying at 60 °C can be considered as intense, irreversible fixation, corresponding to chemisorption. On the other hand, the adsorption observed with vacuum drying at lower temperatures corresponds rather to an unstable adsorption equilibrium, such as a reversible physisorption.

Bohnhoff & Shackelford (2010) studied a bentonite–polymer nanocomposite (BPN), which is a bentonite modified at the nanoscale level to maintain a low k under aggressive conditions. GCLs were assembled and directly permeated with a range of calcium chloride ($CaCl_2$) solutions known to cause large increases in the hydraulic conductivity (*k*) of natural Na-bentonite. In contrast to natural Na-bentonite tested under the same conditions, BPN maintained low hydraulic conductivities for all solutions tested.

6.2 Long-term hydraulic performance of polymerized clays for barriers

Clays show a gradual decrease in their efficiency as hydraulic barriers due to the collapse of the thickness of the diffuse double layer (DDL) of the clay particle structure. This collapse causes an increase of k and a decrease of the osmotic swelling. Both effects increase the flux of contaminants (Jo et al., 2001).

Research interest over the last years has focused on the study of clays treated with amendments to improve their chemical resistance (Bartelt-Hunt et al., 2005; Bohnhoff et al., 2014; Flynn & Carter, 1998; Di Emidio, 2010, 2016; Kolstad et al., 2004; Mazzieri & Di Emidio, 2011).

Figure 15 shows that the k of the HC to seawater was about one order of magnitude lower than that of the untreated clay, and this result was maintained after about 10 years of permeation. This test is still ongoing to further corroborate the long-term adsorption of the polymer and its long-term stability against biodegradation when HC technology is used.

6.3 Chemico-osmotic and diffusion efficiency of modified clays (Di Emidio, 2010)

Exposure of clays to high concentrations of inorganic solutions can change the clay fabric, increasing its k and destroying its membrane behavior measured by the chemico-osmotic efficiency (Shackelford, 2005). To demonstrate the potential benefits of polymer treatment concerning resistance to chemical attack, chemico-osmotic tests were performed with $CaCl_2$ solutions on some amended clays and untreated clay (UC). Unlike the UC, HC treatment maintained low k to $CaCl_2$ even in the long term, suggesting that the carboxymethyl cellulose was not flushed out during the long period of permeation with deionized water (Figure 16). The other amended clays, MSB and DPH GCL, behaved similarly to UC.

For the HC the increase of diffusion coefficient with the ionic strength was lower compared to the UC due to the presence of the polymer (Figure 17). The polymer treatment, maintaining DDL thick, restricted the availability of pores for solute transport with a consequent decrease of D*.

An approach based on the Fixed Charge theory and Donnan equations (Dominijanni & Manassero, 2011)

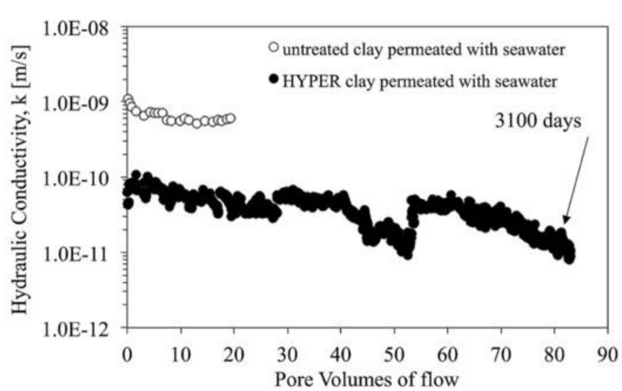


Figure 15. Hydraulic conductivity to seawater of the untreated clay and the HYPER clay.

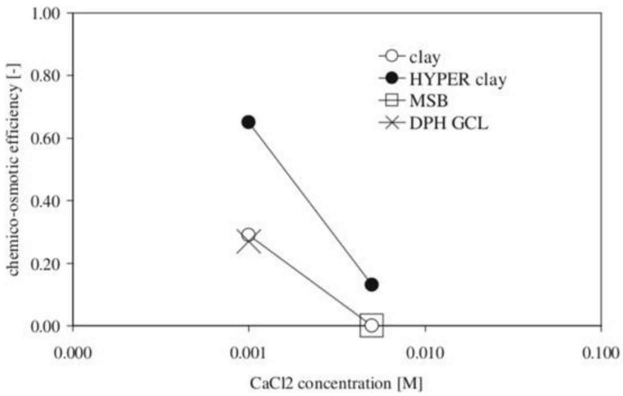


Figure 16. Chemico-osmotic efficiency overview on modified clays: HYPER clay, MSB, and DPH GCL.

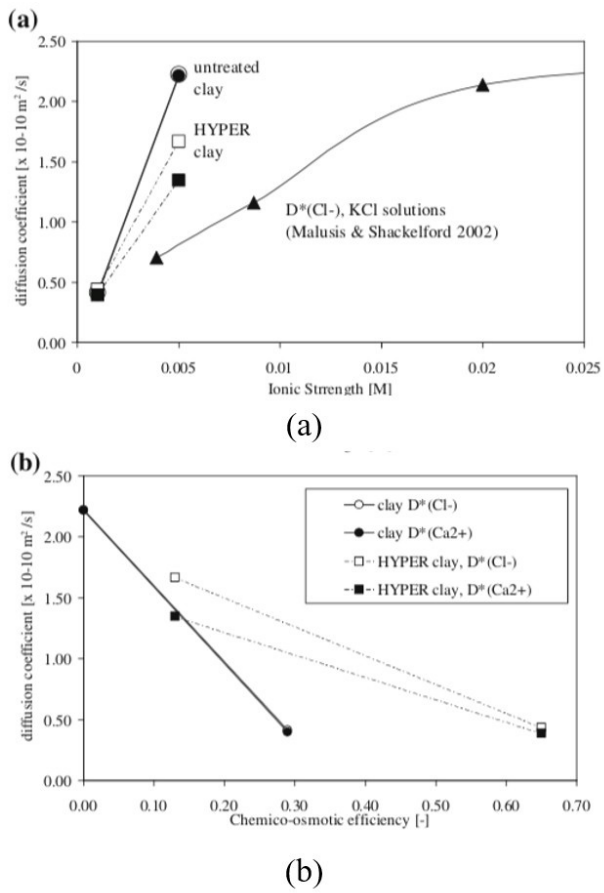


Figure 17. Effective solute diffusion coefficients plotted against: (a) ionic strength; (b) chemico-osmotic efficiency.

accounted for the effect of adsorbing anionic polymers in the nearfield of clay mineral surfaces. For the analysis of the chemico-osmotic tests performed on HC and UC, transport equations for multi-ion systems were numerically solved, and specific boundary conditions were defined to model the chemico-osmotic properties of clays in a multi-ion system. Figure 18 shows that the chemico-osmotic test results were well represented by the theoretical model. The important and new output of this simulation was that the fixed charge concentration, *CX0*, of the HC was higher than that of the UC, whereas the number of platelets per aggregate (*N*) was lower. This theoretical interpretation suggested that not only the anionic polymer maintains the interlayer open (low N), but also it increases the negative charge concentration of the clay (high CX0).

6.4 The use of novel bentonites for vertical barrier applications

Joint research (Malusis & Di Emidio, 2014; Norris, 2016; Replogle, 2016) showed that the presence of multivalent cations in the site water can result in a reduction in the quality of the slurry used for sand-bentonite backfill walls, directly affecting their viscosity and filtrate loss. This can cause an increase in the backfills' k of 2–3 orders of magnitude. The compatibility of HC was tested in the use of both bentonite slurries prepared with electrolyte solutions and sand-bentonite backfills permeated with electrolyte solutions. The k of soil-bentonite backfills containing HC is very low compared to natural clays (NC) and even to MSB (Figure 19). The red line in the figure represents the limiting maximum *k* acceptable value. The legend shows the polymer dosage of HC, the bentonite content *(%BC)* of the slurries, and the concentration in mM of NaCl and $CaCl_2$ of the mixing contaminated water.

The effect of HC impact on hydraulic and mechanical properties of cement-bentonite (CB) mixtures in contact with sulfates was also evaluated (Di Emidio et al., 2014, 2015a, b). Figure 20 shows that the k of CB containing UC increased considerably after permeation with an aggressive solution containing sulfates (25 g/L Na_2SO_4). Conversely, CB containing HC clay maintained a very low k showing its clear chemical resistance to sulfate attack.

6.5 Wet and dry ageing of polymerized clays

Several studies have evaluated the effects of wet-dry cycling on the hydraulic performance of traditional GCLs indicating significant detrimental impacts resulting from wet-dry cycling accompanied by cation exchange of divalent cations (Bouazza et al., 2006; Benson et al., 2010; Lieske et al., 2020; Mazzieri et al., 2017; Yu et al., 2020).

De Camillis (2017) and De Camillis et al. (2016, 2017) studied the performance of polymer modified GCLs when cation exchange was combined with wet-dry cycling. The performance of Na-bentonite (Na-B) after wet-dry cycling in seawater (SW) was compared to that of HC (2% and 8% by dry mass of Na-CMC anionic polymer).

As shown in Figure 21a, the swelling of HC with 8% polymer after the sixth wet-dry cycle was still considerably

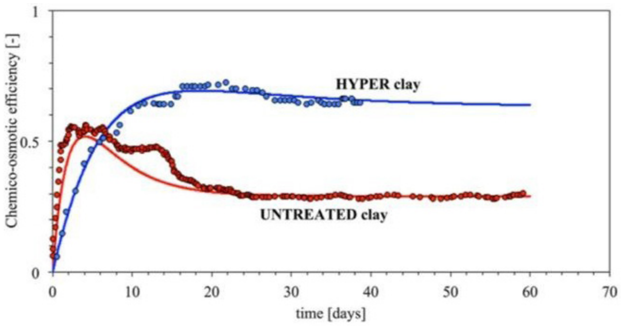


Figure 18. Comparison of the theoretical modeling of the chemico-osmotic experiments on the HYPER clay and on the untreated clay: validation of the model with experiments.

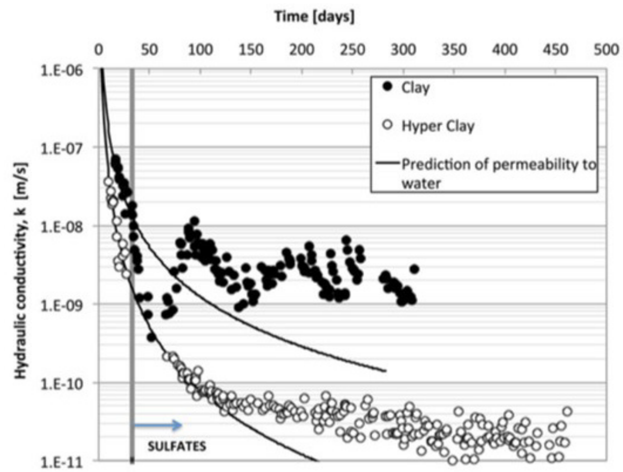


Figure 20. Hydraulic conductivity of CB containing untreated clay (Clay in the legend) and HYPER clay with a sulfate solution (25 g/L Na_2SO_4).

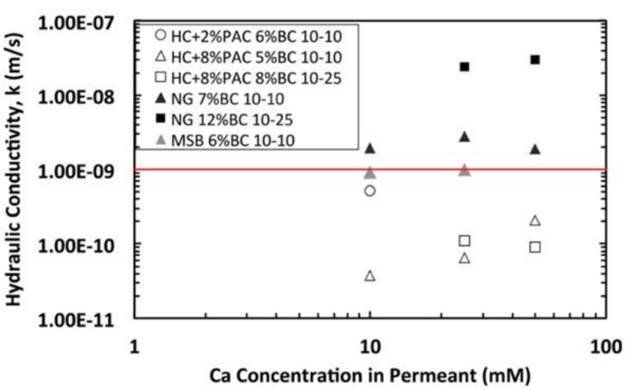


Figure 19. Hydraulic conductivity of soil-bentonite backfills containing HYPER clays (HC), untreated clays (NG) and MSB plotted here vs. Ca concentration of the permeant solutions containing also 10 mM Na.

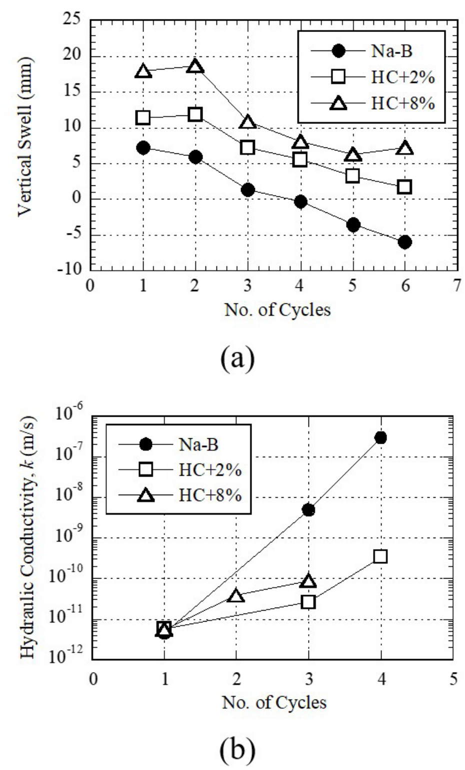


Figure 21. (a) Swell on natural Na-bentonite (Na-B) and HYPER clay with 2% or 8% Na-CMC anionic polymer (HC+2% and HC+8%) at the end of each wetting cycle; (b) Hydraulic conductivities of Na-B and Hyper clay with 2% or 8% Na-CMC anionic polymer (HC+2% and HC+8%) permeated with seawater after each wetting cycle.

greater relative to that for Na-B under the same conditions, and surprisingly similar to the swell of Na-B in contact with deionized water (DW). Also, as shown in Figure 21b, the k of Na-B increased sharply within three wet-dry cycles. Cracks formed in Na-B by drying remained intact, resulting in preferential flow paths and high k. In contrast, the HC specimens maintained a k to seawater lower than 1×10^{-9} m/s. During the fourth wet-dry cycle, the k of Na-B was three orders of magnitude higher than that of HC with 8% polymer.

A crack analysis was performed by means of micro computed tomography (μCT) scanning on Na-B, HC+2% and HC+8% after six wet-dry cycles with seawater. A better self-healing capacity (smaller volume of cracks) on the HC specimens relative to Na-B was noticed, as shown in Figure 22a, more evidently for the HC+8% specimen, wherein almost all cracks are closed. During the wetting cycle, the volume of the cracks decreased for all clays. After wetting, the volume of cracks in the Na-B, HC+2% and HC+8% specimens was 1.2%, 0.45% and 0.05% of the total volume, respectively (Figure 22b).

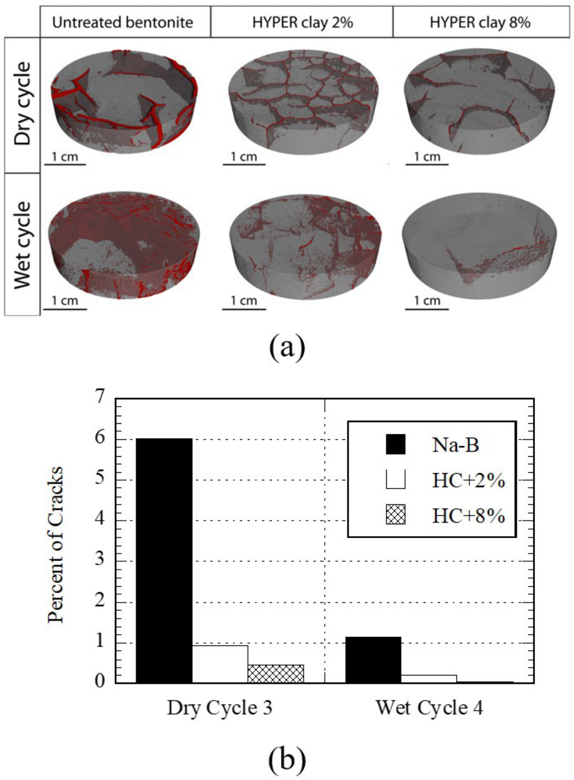


Figure 22. (a) 3D rendered results of the μCT scan of Na-B, HC with 2% or 8% Na-CMC anionic polymer after the 3rd dry cycle and during the 4th wet cycle (cracks are displayed in red); (b) Percent volume of cracking in Na-B and HC+2% and HC+8% at the end of the 3rd dry cycle and the 4th wet cycle.

7. Conclusions

This paper presented a brief state-of-the-art overview of four topics of Environmental Geotechnics written by experienced professionals and academicians, three focusing on Brazilian experience and one pointing out to cut-edge developments in the scope of new geomaterials. It demonstrates that Geotechnics can play a key role in sustainable infrastructure development and preservation and recovery of natural resources. It also highlights that such a consistent and broad research framework, which envisions practical applications, is not being fully implemented as it should be.

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Declaration of interest

The authors have no conflicts of interest to declare. All co-authors have observed and affirmed the contents of the paper and there is no financial interest to report.

Authors' contributions

Maria Eugenia Gimenez Boscov: conceptualization, writing – original draft, reviewing and editing. Gemmina Di Emidio, Alexandre de Almeida Prado Ferrari, Paulo Scarano Hemsi, Miriam de Fatima Carvalho Machado, Sandro Lemos Machado, Manoel de Melo Maia Nobre, Rosane Cunha Maia Nobre, Antonio Thomé: conceptualization, writing - original draft and review.

Data availability

No dataset was generated or evaluated in the course of the current study; therefore, data sharing is not applicable. The data used in the current study are available in the references.

Declaration of use of generative artificial intelligence

This work was not prepared with the assistance of generative artificial intelligence (GenAI).

List of symbols and abbreviations

[A]	Aqueous-phase concentration for compound A
$[A]_s$	Source concentration for compound A
ABREMA	Associação Brasileira de Resíduos e Meio Ambiente
ABMS	Associação Brasileira de Mecânica dos Solos e Engenharia Geotécnica
ADRE	Advection-Dispersion-Reaction Equation
AIS	Air Sparging
ANA	Agência Nacional de Águas e Saneamento Básico
ATSDR	Agency for Toxic Substances and Disease Registry
[B]	Aqueous-phase concentration for compound B
$[B]_s$	Source concentration for compound B
BA	Estado da Bahia
BC	Bentonite Content
BOF	Bijzonder Onderzoeksfonds (Special Research Fund Ghent University)
BPN	Bentonite Polymer Nanocomposite
BTEX	Petroleum Hydrocarbons
C	Aqueous-phase concentration
C	Carbon
C	Containment
CA	Contaminated Area
$CaCl_2$	Calcium Chloride
CAPES	Coordenação de Aperfeiçoamento de Pessoal de Nível Superior
CB	Cement Bentonite
C_e	Maximum allowable concentration
CEC	Cation exchange capacity
CETESB	Companhia Ambiental do Estado de São Paulo
CFU	Colony Forming Unit

CH_4	Methane
CHC	Chlorinated Hydrocarbon Compound
CID	Isotropically Consolidated Drained Triaxial Compression Test
CIU	Isotropically Consolidated Undrained Triaxial Compression Test
CMC	Carboxymethylcellulose
CNPq	Conselho Nacional de Desenvolvimento Científico e Tecnológico
CPRH	Agência Estadual de Meio Ambiente de Pernambuco
C_0	Constant concentration at source
CO	Carbon Monoxide
COBRAMSEF	Congresso Brasileiro de Mecânica dos Solos e Engenharia de Fundações
CONAMA	Conselho Nacional do Meio Ambiente
CWO	Commissie Wetenschappelijk Onderzoek (Commission Scientific Research Ghent University)
CX0	Fixed charge concentration
D^*	Effective diffusion coefficient
D	Hydrodynamic dispersion coefficient
DCE	Dichloroethylene
DDL	Diffuse Double Layer
D_{ij}	Hydrodynamic dispersion tensor
DNAPL	Dense Non-Aqueous Phase Liquid
D_m	Mechanical dispersion coefficient
D_0	Aqueous-phase diffusion coefficient
DPH GCL	Dense PreHydrated Geosynthetic Clay Liner
DW	Deionized Water
EPA	Environmental Protection Agency
FAPERGS	Fundação de Amparo à Pesquisa do Estado do Rio Grande do Sul
FAPESP	Fundação de Amparo à Pesquisa do Estado de São Paulo
Fe^0	Zero-Valent Iron
FEAM	Fundação Estadual do Meio Ambiente do Estado de Minas Gerais
FEPAM	Fundação Estadual de Proteção Ambiental do Estado do Rio Grande do Sul
GCL	Geosynthetic Clay Liner
GOA	Geconcerteerde OnderzoeksActie (Concerted Research Actions Ghent University)
H_2	Hydrogen Gas
HC	HYPER Clay
IBGE	Instituto Brasileiro de Geografia e Estatística
ICSMFE	International Conference on Soil Mechanics and Foundation Engineering
IMA-AL	Instituto do Meio Ambiente do Estado de Alagoas
INEA	Instituto Estadual do Meio Ambiente do Estado do Rio de Janeiro
INEMA	Instituto do Meio Ambiente e Recursos Hídricos do Estado da Bahia
IOF	Industrial Research Fund Ghent University
ISSMGE	International Society for Soil Mechanics and Geotechnical Engineering
K	Potassium
K_d	Distribution coefficient
L	Length
LNAPL	Light Non-Aqueous Phase Liquid
L_{PRB}	Thickness of the PRB
Mg	Megagram
MCL	Maximum Contaminant Level
MNA	Monitored natural attenuation
MPE	Multiphase Extraction
MSB	Multi Swellable Bentonite
MSW	Municipal Solid Waste
MTBE	Methyl Tert-Butyl Ether
N	Nitrogen
N	number of platelets per aggregate
Na-B	Na-Bentonite
NA-CMC	Sodium Carboxymethylcellulose
Na_2SO_4	Sodium Sulfate
NAPL	Non-Aqueous Phase Liquid
NG	Natural Clays
NPK	Nitrogen, Phosphorus, Potassium
NZVI	Nano Zero Valent Iron
OM	Organic matter content
O_2	Oxygen Gas
P	Phosphorus
P&T	Pump-and-Treat
PA	Sodium Polyacrylate
PAH	Polycyclic Aromatic Hydrocarbon
PC	Propylene Carbonate
PCB	Polychlorinated Biphenyl
PCE	Perchloroethylene
PRB	Permeable Reactive Barrier
R	Retardation factor
R_A	Retardation factor for A
RAGS	Risk Assessment Guidance for Superfund
R_B	Retardation factor for B
RBCA	Risk-Based Corrective Action
RBRT	Risk-Based Remediation Target
SABESP	Companhia de Saneamento Básico do Estado de São Paulo
SGS	Sequential Gaussian Simulation
SVE	Soil Vapor Extraction
SW	Seawater
T	Treatment
TAC	Termo de Ajustamento de Conduta
TCE	Trichloroethylene
UC	Untreated Clay
USA	United States of America
USEPA	United States Environmental Protection Agency
VC	Vinyl chloride
VOC	Volatile Organic Compound
VROM	Ministério da Habitação, Planejamento e Meio Ambiente da Holanda
Y_A	Stoichiometric mass ratio between compounds A and B
i	Hydraulic gradient
k	Soil hydraulic conductivity

k_{min}	Minimum soil hydraulic conductivity
k_{max}	Maximum soil hydraulic conductivity
k_x	Hydraulic conductivity in the x direction
n	Soil's total porosity
pH	Potential of hydrogen
q_s	Source discharge rate
q_x	Specific discharge or Darcy's velocity
t	Time
v	Seepage velocity
v_i	Seepage velocity vector
x_i, x_j	Directions of the coordinated axes
α_L	Longitudinal dispersivity
λ	First-order decay rate coefficient
λ_A	First-order decay rate coefficient for A
λ_B	First-order decay rate coefficient for B
μCT	Micro Computed Tomography
ρ_s	Soil's dry density
τ_f	Tortuosity factor

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