Remediation alternatives for an abandoned gold mine contaminated with mercury in Brazil

Alternativas para descontaminação de mina de ouro abandonada contaminada com mercúrio no Brasil

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ABSTRACT

Heavy metal contamination of soils has generated two major problems, namely, loss of land value and health risk for people near contaminated sites. Studies on soil remediation are typically conducted on a pilot scale under very controlled conditions. This is a misleading approach, once the complexity of the actual biogeochemical soil conditions may causes inefficiency in full scale real-world application. In addition, environmental impacts of remediation practices themselves are often ignored, which may make them unfeasible. In 2002, an occurrence of mercury in an abandoned old gold mine was

detected in a rural area of Descoberto, Brazil. In 2014, the State Public Prosecutor required from the responsible State Environment Agency, a remediation project to the area. This study compares alternatives for remediation of the contaminated area. Also a historical approach on mercury amalgamation in gold mining in the region, in order to estimate the amount of mercury remaining in the site. The geochemical characterization of the contaminated area was performed to describe the contamination process. The results were compared with the remediation proposal advocated by the Brazilian Ministry Public, and some alternatives for decontamination of the area were studied. The electrokinetic method has displayed the lowest overall unit cost, between US$ 120.00/t and US$ 260.00/t. The residual Hg concentration after four cycles of the electrokinetic technique was estimated at 1.28 mg (Hg)/kg (soil), slightly higher than the intervention value for agricultural soil, which is 1.20 mg (Hg)/kg (soil).

Keywords: Environmental impact, contaminated area, mercury, abandoned mine, cost-benefit analysis.

RESUMO

A contaminação dos solos por metais pesados gerou dois grandes problemas, a saber, a perda do valor da terra e o risco à saúde das pessoas na circunvizinhança. Os estudos de remediação do solo são tipicamente conduzidos em escala piloto, sob condições muito controladas. Esta é uma abordagem enganosa, uma vez que a complexidade das condições biogeoquímicas reais do solo pode causar ineficiência na aplicação em escala real e no mundo real. Além disso, impactos ambientais das próprias práticas de remediação são muitas vezes ignorados, o que pode inviabilizá-las. Em 2002, uma ocorrência de mercúrio em uma antiga mina de ouro abandonada foi detectada em uma área rural de Descoberto, Brasil. Em 2014, o Ministério Público Estadual exigiu do Órgão Estadual do Meio Ambiente responsável, um projeto de remediação para a área. Este estudo compara alternativas para remediação da área contaminada. Também foi feita uma abordagem histórica sobre amalgamação de ouro com mercúrio na mineração na região, a fim de estimar a quantidade de mercúrio remanescente no local. A caracterização geoquímica da área contaminada foi realizada para descrever o processo de contaminação. Os resultados foram comparados com a proposta de remediação preconizada pelo Ministério Público brasileiro, e foram estudadas algumas alternativas para descontaminação da área. O método eletrocinético apresentou o menor custo unitário global, entre US$ 120,00/t e US$ 260,00/t. A concentração residual de Hg após quatro ciclos da técnica eletrocinética foi estimada em 1,28 mg de Hg por kg de solo, ligeiramente superior ao valor de intervenção para solo agrícola, que é de 1,20 mg de Hg por kg de solo.

Palavras-chave: impacto ambiental, área contaminada, mina abandonada, análise de custo-benefício

1 INTRODUCTION

Unlike the vast majority of organic pollutants, heavy metals cannot be degraded. Their different oxidation states determine their mobility, bioavailability and toxicity (Chen & Li, 2018). This makes remediation of soils contaminated by heavy metals

difficult and a long-term procedure, as these metals can only be transformed into more soluble, insoluble and less toxic species (Akcil et al., 2015).

Mercury is probably the most complicated of the heavy metals to deal with, due to its volatility and its strong affinity for soil particles and organic matter — ionic forms of Hg are strongly adsorbed by soil and sediment with slow desorption when under conditions favourable to leaching by weathering processes (Wang et al., 2012). Bridges (1991) points out that the residence time of Hg in the soil is around 500 to 1000 years.

Mercury takes various chemical forms: metallic or elemental mercury (Hg), inorganic mercury (Hg^{+}) and (Hg^{++}), mercuric ($HgCl_2$, HgS) and mercurous salts (Hg_2Cl_2), as well as organically bound mercury, for example methylmercury and ethylmercury (Rodriguéz et al., 2012, HE et al., 2015; CHEN et al., 2018). Generally, in soil, mercury is strongly adsorbed to organic matter, iron oxides and clay minerals. This, together with mercury low water solubility, hampers the remediation of contaminated soils. A consortium of two or more techniques is sometimes required to meet legal limits, which often constrain the process route.

Madep (1996, apud Rodríguez et al., 2012), points out that clay minerals adsorb Hg ions at pH 6.0, iron oxides adsorb Hg ions in neutral soils, whereas most Hg ions are adsorbed by organic matter in acid soils. When organic matter is not present, Hg becomes more mobile in acidic soils and can evaporate into the atmosphere or be leached, contaminating water sources and groundwater.

This study was conducted in an abandoned gold mine in the municipality of Descoberto, state of Minas Gerais, Brazil where mercury was found. Since then, the Environmental Agency hired a consultancy company to prepare an Executive Project for the Mercury Contaminated Area Intervention Plan — Serra do Grama (FEAM, 2013). The plan was approved and should be implemented by the end of 2019. The company remediation plan foresees the removal of contaminated soil and its confinement in a Class I landfill, to be located about 100 km away, as the most economically and technically viable alternative (adoption of Class I landfill is indicated as the final destination of waste classified as hazardous (those that have contaminating components, pollutants, harmful to the environment and humans).

Since this method only shifts the contamination problem to another location along with the risks associated with ground transportation. Recalling that in the scenario proposed by the consultancy company can aggravate the process of contamination of the landfill soil, as there may be reaction with other constituent elements of the soil and other

residues deposited there, which may cause greater dispersion of mercury, requiring more intensive monitoring, therefore more expensive.

Given these facts, this paper has two main objectives. The first one is to present an analysis of the possible techniques for rehabilitating contaminated soil, whereas the second one is to compare these techniques with that recommended by the Environmental Agency, in order to assess their strengths and weaknesses in relation to other options.

2 REMEDIATION OF CONTAMINATED SOILS

The US Environmental Protection Agency (US EPA) groups soil treatment technologies into in situ (biological, physicochemical, and thermal techniques), ex situ (biological, physicochemical, and thermal techniques) and other confinement-based treatments such as: impermeable barriers, sealing surfaces, stabilization and solidification (US EPA, 1997).

Gong et al. (2018) classify remediation techniques into three methods: physical, chemical and biological. Physical methods include soil replacement, vitrification, electrokinetic remediation, and heat treatment. Heat treatment and soil washing have been recommended as permanent treatment alternatives for the removal of mercury from contaminated soil (Dermont et al., 2008). Chemical methods comprise chemical immobilization, solidification/stabilization and soil washing. In turn, biological methods include phytoremediation, microbial remediation and assisted microbial phytoremediation.

In situ methods generally have a lower decontamination cost and do not involve removal of contaminated soil (Hinton and Veiga, 2001). However, they are less adopted than ex situ methods, due to the soil subsurface heterogeneity, which generates uncertainties about the efficiency of the process, even with longer cleaning times.

Ex situ remediation of heavy metal contaminated sites involves excavation to remove the contaminated portion, followed by immobilization of metals using the solidification/stabilization process and subsequent disposal of treated materials at disposal sites or in permitted landfills (Zeng et al., 2014). This modality has the disadvantage that it is not permanent and involves the transfer of contaminated, solidified or stabilized material to other locations.

In general, soil remediation based on excavation, transport and landfill is highly effective with less risk but costly (Chen et al., 2018). These procedures often result from

the remobilization of mercury compounds during the excavation process and the adoption of techniques that can remove it from the soil.

To assess the adequacy of specific corrective measures in a particular location, several criteria should be considered as benchmarks to prevent the mobility of mercury species and subsequently define the mitigation techniques to be employed. These criteria include the distribution and properties of stratigraphic units, geohydrological features of the site (together with the physicochemical properties of Hg species present in the soil).

2.1 FACTORS AFFECTING HG MOBILITY

Hg transformations such as methylation and demethylation, reduction and oxidation modify Hg speciation and, consequently, its mobilization/stabilization in soils. Geochemical parameters, such as pH and soil redox potential, strongly affect Hg mobility, changing its solubility and the biological process that affects Hg transformations (Xu et al., 2015), requiring a detailed analysis of Hg alternatives remediation to be adopted.

Similarly, natural organic matter can strongly interact with Hg, affecting its speciation, solubility, mobility and toxicity (Negrete et al., 2015). Many organic compounds have high affinity for Hg through functional groups such as hydroxyl, carboxylic, aromatic and especially containing-S ligands.

According to Reis et al. (2014), when high levels of organic matter are present in the soil, the process of formation of Hg^{2+} organic complexes predominates. Thus, organic matter is the dominant factor controlling mercury mobility in acidic soils, and for neutral to alkaline soils, it is the mineral components that most extensively influence the solubility of Hg.

The amorphous clay, oxide and hydroxide minerals of Fe, Mn and Al, as well as amorphous iron sulphide (FeS) are significant inorganic sorbents for Hg.

Reis et al. (2014) remember that ions present in soil solution, such as chlorides and hydroxides, have the ability to increase the solubility and mobility of mercury. The pH has a fluctuating influence on Hg mobilization from acid to alkaline environment. The lowest dissolution of Hg occurs at pH close to 3.0, while the highest occurs around pH 5.0 (Xu et al., 2015).

2.2 REFERENCE VALUES FOR MERCURY IN SOILS

A site is considered contaminated when the concentrations of deleterious elements or substances are above a given limit, called intervention value, which indicates the

potential risk for the human health (Table 1). In this case, immediate intervention is required, including detailed site investigation and emergency measures such as restricting access to the area and suspending the consumption of surface and groundwater, and food products from the area of influence.

Table 1 — Soil guiding values

Substance	Soil (mg. kg^{-1} dry mass)				
	Benchmark Quality Value (VRQ)	Prevention Value (VP)	Intervention Value (VI)		
			Agricultural	Residential	Industrial
Mercury	**0.05**	**0.5**	**1.2**	**0.9**	**7.0**

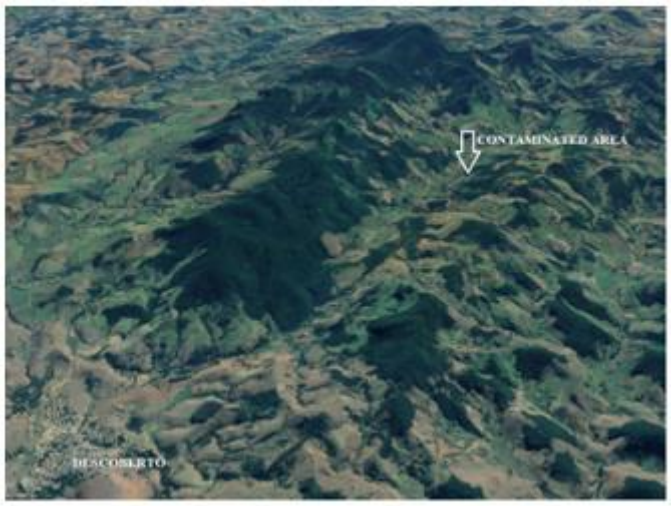
Source: (Adapted from CETESB, 2016)

The nomenclature adopted in Table 1 is explained in detail as follows: ***VRQ*** — concentration of a given substance in the soil, which defines a soil as clean; ***VP*** — Concentration of a given substance, above which changes detrimental to soil and groundwater quality may occur. This value indicates the quality of a soil capable of supporting its primary functions, protecting ecological receptors and groundwater quality; ***VI*** — concentration of a given substance in soil or groundwater above which there are potential direct or indirect risks to human health, considered as a generic exposure scenario.

3 MERCURY IN DESCOBERTO

In December 2002, residents of the rural area in the municipality of Descoberto (State of Minas Gerais, Brazil) observed the presence of metallic mercury in the soil. Mercury presence is near a tributary of the Grama creek. The waters of this stream were used for recreation, fishing and watering livestock, as well as public supply of the municipalities of Descoberto and São João Nepomuceno. Therefore, the contaminated site has the potential to jeopardize the quality of soils and waters, and may cause damage to the health and well-being of the population along the Grama creek basin, including Descoberto town (Figure 1).

Figure 1 — Contaminated area, highlighted by yellow marker. In the foreground is the small town of Descoberto.



Source: Google Earth (2019).

About 70 families live in the affected region, totalling approximately 300 people. The area of about 450 ha encompasses forests, waterfalls and several species of fauna and flora, typical of the Atlantic Forest biome (FEAM & CDTN, 2006).

As soon as mercury contamination was reported in the region, the Descoberto Town Hall and the Minas Gerais Sanitation Company — COPASA interrupted water catchment from the Grama creek for public supply. In March 2003, technicians from the State Environmental Agency (FEAM, Portuguese acronyms) and the Brazilian Aluminum Company (CBA, Portuguese acronyms) performed soil and surface water sampling in the area, whose results indicated a high concentration of mercury. Concentrations in soil of above Hg 40 mg kg^{-1} and maximum value of Hg 936 mg kg^{-1} (FEAM & CDTN, 2006) were observed at several points. As such, FEAM classified the area as contaminated and banned the water catchment.

Since then, several studies aiming to evaluate possible contamination have been conducted in the area, including Alexandre (2006); Marques et al. (2006); Durão Jr et al. (2009); Tinôco (2010) and Oliveira (2014). In the mercury emergency area, four structures known as canoes — 19th century structures used for gold extraction — were located up to 90 cm deep (FEAM & CDTN, 2006). Canoes have a thickness and width of about 30 cm and varying lengths (one 10 m, one 15 m, one 20 m and one 30 m), with contents ranging from Hg 16 mg kg^{-1} Soil to Hg 8,826 mg kg^{-1}.

The soils in the region under study have the following mineralogical composition: above 30 % quartz (SiO_2); from 10 % to 30 % goethite (FeO.OH) and kaolinite ($Al_2Si_2O_5(OH)_4$); 3% to 10% gibbsite ($Al(OH)_3$); below 3 % sillimanite (Al_2SiO_5),

muscovite ($KAl_2(AlSi_3O_{10})(OH)_2$), microcline ($KAlSi_3O_8$), albite ($NaAlSi_3O_8$), hematite ($Fe_2O_3$), ilmenite ($FeTiO_3$), magnetite Fe_3O_4), calcite ($CaCO_3$) and monazite ((Ce, La, Y, Th)PO_4) (FEAM & CDTN, 2006). These soils have a silt with clay ranging from 2.6 % to 5.3 %, i.e., they have a high sand content (Durão Jr. et al., 2009). The local hydrogenionic potential (pH) ranges from 6.53 to 5.5, presenting, therefore, low contribution to Hg mobility (César et al., 2008).

Based on the identified mineral assemblage, there is a significant amount of Fe and Al oxides and hydroxides, allied to the relative abundance of gibbsite, indicating that the local substrate suffered the action of extreme weathering processes, corroborated by the supergene deposits. bauxite in the region. Some of which are exploited by CBA.

Durão Jr. et al. (2009) state that much of the mercury that contaminated the area as Hg^o, has oxidized and is mainly linked to Fe, Mn and Al oxyhydroxides, and up to 30 % of it may be linked to organic matter present in the soil. The dispersion of mercury in soils and sediments is controlled by the distance of the mines and by the terrain slope, which favours constant erosion and transport of mercury downstream (Hinton and Veiga, 2001). In the today contaminated area, Miranda et al. (2020) reports that the dispersion of Hg is an environmental liability that began by around 1840 from artisanal mining activities.

4 MATERIALS AND METHODS

A survey of the contaminated area was performed, seeking to assess the possible correlations of Hg with the constituent elements of the soil. Following a review and evaluation of technological alternatives of remediation for the contaminated area was conducted. Then, the alternatives were compared with the proposed remediation recommended by the prosecutor. Finally, a cost/efficiency analysis was conducted to reach the best alternative to remediate the contaminated area. Following it is presented and discussed the results achieved.

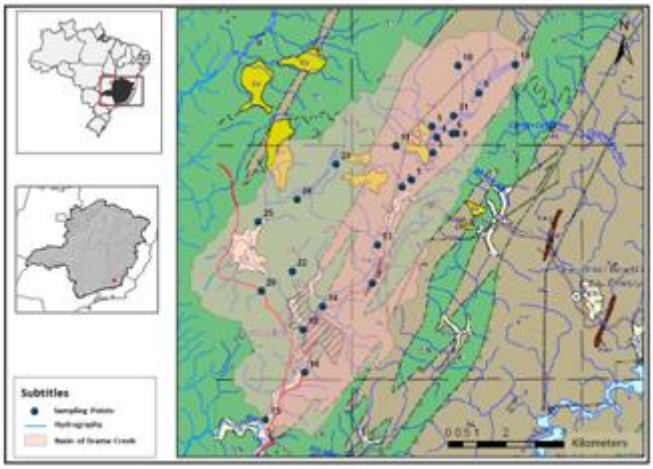
5 RESULTS AND DISCUSSION

5. 1 SURVEY OF THE CONTAMINATED AREA

To perform the correlation analysis of soil and sediment constituents throughout the Grama creek basin, soil samples were collected in three sampling campaigns - one in March 2008 (rainy season), nine samples; one in August 2008 (dry season), 26 samples; and 21 samples in March 2009 (rainy season). The first campaign was carried out with

the purpose of recognizing the basin area, checking the collection points, its difficulties and its characteristics and also collecting the first samples. The sampling mesh was established to cover the whole basin. (Figure 2).

Figure 2 — Map of Grama creek basin showing sampling points.



Chemical analysis of soil and sediment samples was performed by inductively coupled plasma - atomic emission spectrometry (ICP–AES). Statistical analysis was performed using the freeware computational package Jamovi (The Jamovi Project, 2019). Considering overall the 46 sampling points the following statistical parameters were obtained for the variable mercury content: average: 0.459 mg kg^{-1}; median value: 0.177 mg kg^{-1}; standard deviation: 1.742 mg kg^{-1}; range: from 0.000 to 11.921 mg kg^{-1}.

Regression analysis showed no significant correlation between mercury concentration and those of other commonly detected elements, as shown in Table 2. This implies that the dispersion of mercury occurred uniformly in soil and sediment components without chemical interaction (or adsorption) preferred.

Correlation analysis between mercury content and geographic coordinates of sampling points shows that this parameter should not be considered a regionalized variable, since no trend is perceived: Pearson's correlation coefficient of mercury content with geographic coordinate. X (latitude in UTM coordinates) was equal to 0.12578 (with p-value of 0.4049), whereas with Y (longitude in UTM coordinates) equal to 0.06616 (with p-value of 0.6622).

Table 2 — Pearson correlation coefficient between mercury and other typical soil and sediment elements along Grama Creek basin (using Jamovi).

Element	*Pearson's r*	*p-value*	*Element*	*Pearson's r*	*p-value*
Aluminum	-0.02521	0.86791	Nickel	-0.11387	0.45113
Barium	-0.12831	0.39544	Phosphor	-0.02746	0.85623
Calcium	0.06026	0.69076	Sulfur	0.03343	0.82544
Cobalt	-0.13285	0.37879	Scandium	-0.02712	0.85800
Chrome	-0.01018	0.94644	Strontium	-0.10335	0.49429
Copper	0.02246	0.8822	Thorium	0.14346	0.34153
Iron	-0.02624	0.86257	Titanium	-0.00382	0.97987
Potassium	-0.06414	0.67194	Vanadium	0.02266	0.88115
Lithium	-0,04451	0.76898	Yttrium	-0.14951	0.32134
Magnesium	-0.02459	0.87116	Zinc	-0.01464	0.92305
Manganese	-0.16596	0.27034	Zirconium	-0.06498	0.66789
Sodium	-0.04062	0.78868			

5.2 SOIL TREATMENT ALTERNATIVES FROM DESCOBERTO

Miranda et al. (2020), based on data compiled from various studies concerning Descoberto site (FEAM and CDTN, 2006; Alexandre, 2006; Marques et al., 2006; Durão Jr et al., 2009; Tinôco, 2010; Oliveira, 2014), estimated about 820 kg of residual mercury in the area, making remediation techniques highly recommended for this area.

The State Council of Environment of Minas Gerais, COPAM (Minas Gerais, 2017), recommends that the treatment of contaminated areas should consist of the application of techniques, aiming at the removal, containment or reduction of contaminant concentrations. Therefore, in the risk assessment stage, the remediation objectives will be defined, and the most appropriate techniques should be selected.

As the contaminated area is a farmland, chemical, physical or biological treatments, besides being expensive processes, can irreversibly affect soil properties to the point of rendering it useless as a means for plant growth (Negrete et al., 2015). Existing thermal techniques use high temperatures (for example, 600 to 800 °C) and require high energy costs (Xu et al., 2015). In addition, soil treated at this temperature is unfavourable to agricultural reuse. To address concerns about the high energy consumption associated with these temperatures, Ma et al., (2014) propose to operate the heat treatment at a lower temperature (400 °C), with $FeCl_3$ added for longer, thus ensuring soil quality. On the other hand, washing the soil to remove mercury is an alternative; however, it is a very expensive procedure and produces a hazardous waste requiring additional treatment.

There are different treatment processes for the remediation of Hg-contaminated soil, such as: stabilization/solidification techniques; in situ vitrification; electrokinetic; soil wash; bioremediation; thermal desorption; excavation, removal and disposal of the soil, among others (Sierra et al., 2016). Considering the Descoberto soil features, the five most suitable remediation alternatives to be applied in the contaminated area were evaluated, namely:

- ***Bioremediation*** — Use of vegetation to recover soil Hg. Hg recovery occurs after plant collection and incineration and depends on selected plants to clean up mercury contamination in the soil. It can also be used as a final decontamination step in conjunction with other treatment technologies (Xu et al., 2014; Hinton and Veiga; 2001; Dermont et al., 2008; Richter and Flachberger, 2010; Thakur et al. 2016; Negrete et al., 2015).
- ***Excavation***, removal and disposal of soil — Consists basically of excavation of contaminated soil and disposal of it to landfill prepared to receive contaminated waste.
- ***Heat treatment*** — this technique uses elevated temperatures to remove Hg from the soil through volatilization (Rodríguez et al., 2012; Xu et al., 2014). After its excavation, the contaminated soil is subjected to heating. The resulting gases are then condensed and the liquefied mercury is stored further (Hinton and Veiga, 2001; Chang and Yen, 2006).
- ***In situ soil scrubbing*** — Using heat and reduced pressure to volatilize mercury, followed by condensation of Hg^0 vapours (Xu et al., 2014). This technique removes contaminants from the ground without excavation, treatment or disposal waste, reducing costs and the associated risks (Chen & Li, 2018; Rodríguez et al., 2012).
- ***Electrokinetic treatment*** — This method employs an electric field gradient causing the migration of Hg compounds to electrodes placed in contaminated soil, generating accumulation halos around them, with Hg ions being subsequently removed, reducing the costs of excavations of the entire impacted area (Hinton and Veiga, 2001; He et al., 2015).

5.3. COST ANALYSIS EFFICIENCY OF PROPOSED ALTERNATIVES

Since just analysing the application of one or more techniques is not enough for an ideal choice, this paper proposes a ranking of the techniques, depending on costs and/or benefits. Theoretically, removal technologies would be most desirable because Hg can be permanently removed from the site. However, implementing these technologies can be complex and costly. Given this, the cost effectiveness of each of the methods studied was evaluated here.

Unlike the cost-benefit analysis, which compares the benefits and costs of a technique, expressed in currency units, the cost effectiveness analysis compares them, but in units of results, thus allowing a decision-making process to be chosen, and ensuring that desired result at lower costs (Miyabukuro, 2014). Thus, the intended result will be the one that presents the highest efficiency from the point of view of costs incurred to achieve the site remediation. For this purpose, it was evaluated the lowest cost technique to achieve the remediation of the contaminated area, having as a determining indicator the ratio (C/E) between the unit cost per m^3 (unit cost in US$ per m^3) and the effectiveness of soil Hg reduction (efficiency). Table 3 was prepared based on the literature to infer the base values for the calculations using the operational research technique.

Table 3 — Comparison of remediation methods for mercury contaminated areas

Remediation Technique	***Cost [US$/t]***	***Efficiency [%]***
Bioremediation	25 – 100 (Chen & Li, 2018)	24 (Chen & Li, 2018)
Excavation, removal and disposal of soil	275 (Chen & Li, 2018)	100 (Chen & Li, 2018)
Thermal technologies	460 (Chang e Yen, 2006)	98 (Chang e Yen, 2006)
Soil wash	270 – 450 (Chen & Li, 2018)	60 (Chen & Li, 2018)
Electrokinetics	30 – 65 (Rosestolato *et al*., 2015)	60 (Rosestolato *et al*., 2015)

Tables 4 and 5 show one of the comparison criteria between the methods used for soil Hg removal. By this criterion, the most advantageous method is the lowest relative cost (in US$/%), which is calculated by dividing the absolute cost for 1 t soil treatment (in US$) by the efficiency of the method (in %). The electrokinetic process presented the best result (US$ 0.50/%).

Table 4 — Comparison of the relative cost (last column of the table) for the five methods analysed using the lowest absolute cost [Cost (min) of the interval (Table 3)]

Remediation Technology	Cost (minimum) *c* [US$/t(soil)]	Efficiency *e* in %	US$/% [for 1 t of soil]
Bioremediation	25	24	1.04
Excavation, removal and disposal of soil	275	100	2.75
Thermal technologies	460	98	4.69
Soil wash	150	60	2.50
Electrokinetics	30	60	**0.50**

Table 5 — Comparison of relative cost using the highest absolute cost [Cost (max) off range (Table 3)]

Remediation Technology	Cost (maximum) *C* [US$/t(soil)]	Efficiency *e* [%]	[US$/% per t of soil]
Bioremediation	100	24	4.17
Excavation, removal and soil disposal	275	100	2.75
Thermal technologies	460	98	4.69
Soil wash	250	60	4.17
Electrokinetics	65	60	**1.08**

The electrokinetic process has presented the best result (US$ 1.08/%). One problem with the proposed treatment is that the amount of residual Hg was not taken into account. This study suggests a new procedure to indicate the lowest cost decontamination technique. For the implementation of this new comparison criterion, the number of cycles (or number of times the technique should be used) is calculated assuming a residual percentage of acceptable Hg as input, which should be used for all methods, except excavation/removal (where the percentage of residual Hg is zero for a single cycle).

The number of series cycles required (***n***) until the residual fraction of a chemical species being extracted is less than or equal to a previously specified r value, given the fractional removal efficiency at each cycle or stage (ε, adopted constant, for simplicity) can be made from the following expressions:

$$r = (1 - \varepsilon)^n \quad (1)$$

Logarithmizing equation (1) and making explicit the number of consecutive processing stages (serial operation) results in equation (2), readily usable.

$$n = \frac{\ln(r)}{\ln(1-\varepsilon)} \quad (2)$$

The total unit cost (***C***) is obtained by multiplying the number of cycles (***n***) by the absolute cost for decontaminating 1 metric ton of soil (***c***) or, in algebraic form: ***C = c x n***.

As the average soil Hg concentration in the study area is 50 g/t and the intervention value for agricultural soil is 1.2 mg Hg per kg soil or 1.2 g (Hg)/t (soil) (CETESB, 2016), you get the value of the permissible residual percentage of (1,2/50) = 2,4 %, ie r = 2,4 %. Thus, the cost per ton of soil for Hg removal, with a residual percentage of 2.4 % allowed, was calculated for each method (except excavation/removal) and the result is shown in Tables 6 and 7.

Table 6 — Comparison of the total unit cost for the five methods analysed using the lowest absolute unit cost of the interval (table 3).

Remediation Technology	**Number of cycles *n***	**Total Cost *C* [US$/t(soil)]**	**Percentage final residual**	**Residual Hg [g of Hg per t of soil]**
Bioremediation	14	350.00	2.14	1.07
Excavation, removal and disposal of soil	1	275.00	0.0	0.0
Thermal technologies	1	460.00	2.00	1.00
Soil wash	4	600.00	2.56	1.28
Electrokinetics	4	**120.00**	2.56	1.28

The electrokinetic process presented the best result (US$ 120.00/t). Also considering the highest absolute unit cost, the electrokinetic process presented the best result (US$ 260.00/t).

Table 7 — Comparison of the total unit cost for the five methods using the highest absolute unit cost of the range (Table 3).

Remediation Technology	**Number of cycles *n***	**Total cost *C* [US$/t(soil)]**	**Percentage final residual**	**Residual Hg [g of Hg per t of soil]**
Bioremediation	**14**	**1400.00**	**2.14**	**1.07**
Excavation, removal and disposal of soil	**1**	**275.00**	**0.0**	**0.0**
Thermal technologies	**1**	**460.00**	**2.00**	**1.00**
Soil wash	**4**	**1000.00**	**2.56**	**1.28**
Electrokinetics	**4**	**260.00**	**2.56**	**1.28**

6 RESULTS

Bioremediation, although presented as a promising technique for the cost effect of shallow soil remediation, is limited because there are few plant species suitable for Hg absorption, and because it requires several planting and harvesting cycles for the total removal of Hg from the contaminated area.

Heat treatment using longer temperatures may be a very viable alternative to remove Hg from Discovered soil. However, treated soil may become unfavourable for agricultural use. Although following the proposal of Ma et al. (2014), where a typical Hg removal heat desorption unit operates at temperatures of 320 °C, the method remains effective for removing Hg from the ground, even requiring high energy costs.

As a source of energy for the thermal process, it is suggested the structuring of a solar energy unit, compatible with the technique, that can later be destined for alternative generation of the school near the contaminated area, thus increasing the sustainability of the process.

At the time of the State Public Prosecution's requirement to remediate the area, the method of excavation, removal and geotechnical confinement of the soil proved to be the most viable alternative from the economic and technical point of view. However, this method only transfers the contamination problem elsewhere as it can aggravate the landfill soil contamination process via reaction with other soil constituents and even cause further Hg dispersion, requiring more careful monitoring intensive and cost effective. Remembering that at any time in the future, it will still pose the same risk as the day it was collected.

In turn, soil washing, in addition to the high cost and complexity of operating facilities, requires the use of leachate agents that cause increased Hg mobility, which may not be acceptable to regulators, due to the risk of jeopardizing soil quality for agricultural uses.

Finally, from Tables 6 and 7, one can conclude that the electrokinetic method is the lowest total cost, i.e., US$ 120.00/t and US$ 260.00/t, as the lowest and highest absolute costs are used, respectively for each method shown in Table 3. The residual Hg concentration after four cycles of the electrokinetic technique is 1.28 mg of Hg per kg of soil, slightly higher than the intervention value for agricultural soil, which is 1.2 mg of Hg per kg of soil.

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Conflicts of interest

No conflicts to declare.

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