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EVALUATION OF POTENTIAL METHANE GENERATION IN THE INVESTIGATION OF AN ABANDONED CONTAMINATED LANDFILL IN SANTIAGO, CHILE

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Abstract - This study presents the environmental evaluation of an abandoned and potentially contaminated landfill using analyses for the presence of heavy metals and for methane generation potential. The site is located in the city of Santiago, Chile, and was used as a rural landfill for domestic, industrial and construction waste until 1978, but is now in a heavily urbanized area and surrounded by houses. Analyses performed on 24 samples taken in and around the site show Potential Methane Generation (PMG) values between 1.6% and 11.3% of maximum projected levels. These low values, compared to those of an active landfill, indicate that waste material stored in the site has a low capacity to generate methane. Concentrations of heavy metals in the surface and deep soil are similar to typical levels for these metals in normal soil, according to international USEPA standards, and do not present imminent risk to human health. The use of the PMG test technique for the study of the health risk of an abandoned landfill is a new contribution to the Chilean evaluation methodology and management program for Abandoned Sites with Potential Presence of Contaminants (SAPPC). As part of the environmental management strategy for the site, two of the five operable units studied were transformed into a park after this study.
Keywords: Abandoned landfill; Methane; Solid waste; Contaminated sites.

INTRODUCTION

Studies related to the potential presence of contaminants in soils at a site are varied and usually develop as a prior and fundamental step in the evaluation of remedial alternatives, recovery of the site for different uses, or both, depending on the characteristics of the location. However, there is no definitive, common methodology applied to all situations. To deal with this limitation, several comparative models have been developed to establish the presence of abnormal levels of contaminants at a specific site under study (Aslibekian and Moles, 2003; Muhlbachova *et al.*, 2015; Rodríguez *et al.*, 2015; Wen *et al.*, 2015; Khan *et al.*, 2008). For example, since 1995 in the United Kingdom it has been known that soils within 1 to 3 km of metal smelters may contain up to 15 times the natural values of Pb in the soil and also may present high concentrations of Cd at distances as far as 40 km from the originating industrial activity (Aslibekian and Moles, 2003).

In polluted soils, interactions between heavy metals, organic matter content and microorganisms have been correlated (Muhlbachova *et al.*, 2015). Increasing concentrations of metals in the urban environment have been studied, wherein concentrations of

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Cd, Ni and Cr measured in plant leaves in 2012 exceeded those reported in 1941 for the same species by factors of 10, 13 and 16, respectively (Rodríguez *et al*., 2015). The increase of these pollutants in the urban atmosphere was related to human activity changes during a period of more than 70 years. The anaerobic biodegradation of domestic and industrial waste in landfill sites goes through a complex process and therefore it is not easy to estimate the biological conversions involved. Measurements at these sites must be performed carefully taking into account different waste sources such as pharmaceutical residues, plastic products, antibiotics, and complex organic compounds (Wen *et al*., 2015; Khan *et al*., 2008; Kumar *et al*., 2004; Aguilar-Virgen *et al*., 2011; Aguilar-Virgen *et al*., 2012; Angelidaki and Sanders, 2004; Stergar and Zagorc, 2002; ISO 11734, 2012; Kolstad *et al*., 2012; Gartiser *et al*., 2007; El-Mashad *et al*., 2012; Angelidaki *et al*., 2006).

Human activities in Chile have generated locations known as Abandoned Sites with Potential Presence of Contaminants (SAPPC), such as old landfills, uncontrolled dumpsites, or industrial waste sites. When abandoned, these sites may be converted to new land uses without additional regulation. Studies of contaminants in soils at these and other sites have been performed considering the type and extent of pollutants in the involved area (Romero *et al*., 1999; Ginocchio *et al*., 2004; Molina *et al*., 2009; Escudey *et al*., 2007; Badilla-Ohlbaum *et al*., 2001; Palma-Fleming *et al*., 2000). The systematic evaluation of SAPPC in Chile began only 5 years ago, in 2010, and has targeted defined areas that have been environmentally impacted by one or more potentially polluting activities, which ended at some point without a proper site closure process.

In 2012, the Chilean Government began to apply a national methodology (Chilean Government, 2012) to identify and confirm the presence of contaminants at these sites. This methodology contains an ordered sequence of activities whose first step is the application of criteria to identify and prioritize SAPPC sites within each region. Subsequently, in step two, the Preliminary Investigation collects and analyses site historical information. In step three, the Confirmatory Investigation collects and analyses site samples. See Figure 1.

The Confirmatory Investigation of the SAPPC methodology, as shown in Figure 2, is designed to determine representative concentrations of pollutants present at the potentially contaminated site, which are then compared with reference criteria to confirm whether or not the suspected contaminant levels pose a preliminary risk to potential receptors.

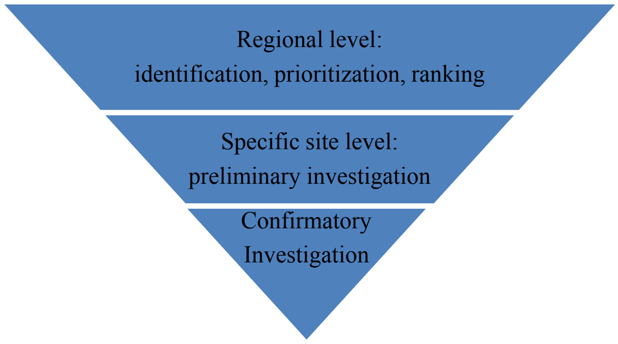


Figure 1: Illustration of the Chilean SAPPC.

Evaluation Methodology developed and conducted by the Ministry of Environment.

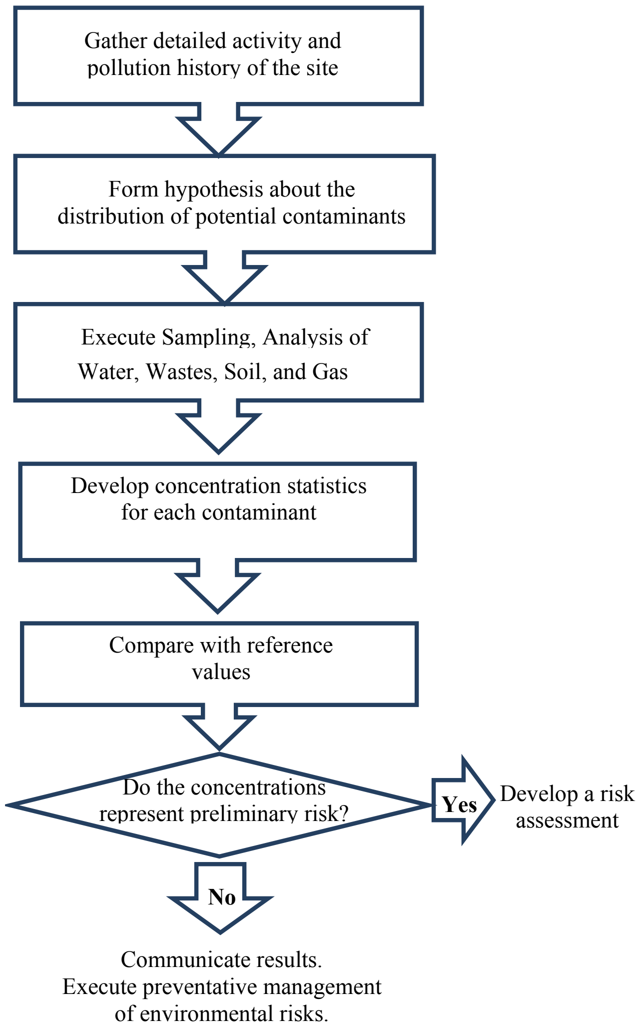


Figure 2: Flow Diagram for the SAPPC Confirmatory Investigation.

Suitable methods of analysis for this study were selected for quantifying the presence of heavy metal contaminants. To evaluate an abandoned landfill under the SAPPC methodology, it is crucial to establish the levels of landfill gas (biogas, consisting of CH_4,

CO_2, H_2S, N_2). However, the current Chilean SAPPC methodology does not contain details about how to establish these levels.

This paper presents a case study including the evaluation of Potential Methane Generation (PMG) in the Confirmatory Investigation of an abandoned landfill in the city of Santiago, Chile. This study of PMG as a methodology for determining the landfill gas levels is an important contribution to the advancement of the Chilean SAPPC Evaluation Methodology.

METHODS

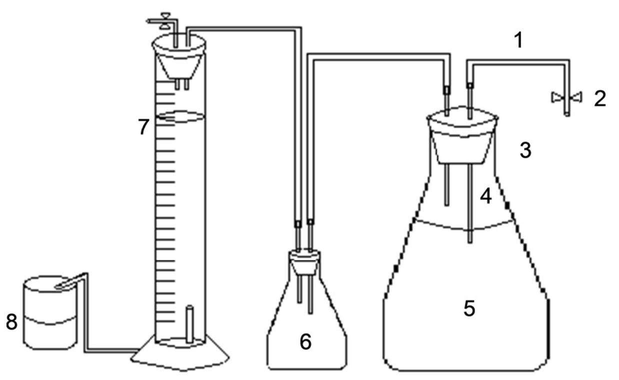
The SAPPC known as La Cañamera in Santiago, Chile, had an area of 25 hectares and received residential, industrial and construction waste between 1962 and 1978. Wastes deposited there were from five municipalities located in the southern part of the city and were placed using a rudimentary landfill technique that did not include the use of impermeable barriers or other special protective measures. After the closure of the landfill, the land was parceled out and used for different objectives, including the construction of residential housing. It was expected that soils in a landfill such as this one might contain heavy metals, gas and other pollutants.

The test area was divided into six operable units, taking into account the different uses and owners of each sector. 30 representative sampling points were identified throughout the area, with each sample point consisting of a surface zone, a trash zone and a deep soil zone. 80 soil samples and 24 trash samples were collected for later evaluation of PMG, metal concentrations, pH levels and extrinsic toxicity. The extrinsic toxicity was measured using the Toxicity Characteristic Leaching Procedure (TCLP) according to USEPA Test Method 1311, established in the Resource Conservation and Recovery Act (RCRA) of the United States and in the Chilean regulations for industrial wastes.

Abandoned landfills are characterized by a heterogeneous distribution of domestic and industrial waste. Similarly, the generation of methane or landfill gas is a problem in these sites and can manifest weakly at the surface while the sub-surface trash can retain a significant capacity for generating gas via anaerobic biological conversion. The presence of heavy metals is related to their environmental persistence following deposition of metal-containing wastes from industrial and domestic sources.

Potential Methane Generation was evaluated according to the principles described in the 2009 Columbian Technical Standard NTC4233 “Environmental Management. Water quality. Evaluation of the ultimate anaerobic biodegradability of organic compounds in digested sludge. Method by measurement of the biogas production”, with some minor modifications for solid residue evaluation. The method consists of carrying out the anaerobic digestion of solid residues in 500 ml batch reactors at 35° C, which have been previously inoculated with microorganisms adapted to these residues. A solution of trace micronutrients was used in this study to ensure the best conditions for anaerobic digestion. The main compounds of this solution were $MnCl_2.4H_2O$, H_3BO, $ZnCl_2$, $CuCl_2$, Na_2MoO_4, $CoCl_2.6H_2O$, $NiCl_2.6H_2O$ and Na_2SeO_3, according to the requirements for methanogenic archaea microorganisms (Milán *et al.*, 2010; Ortner *et al.*, 2014; Pereda *et al.*, 2006).

Figure 3 shows the experimental procedure setup schematic for one reactor. To achieve the anoxic conditions inside the reactor, the system was flushed with nitrogen gas for 20 minutes before the beginning of digestion, thereby eliminating any oxygen that might have been present in the reactors. The pH of the reactor medium was adjusted with NaOH to 7 ± 0.2. Methane production during digestion was



1. Silicone tubing;
2. Controlled leak silicone tubing;
3. Rubber plug,
4. Glass capillaries;
5. 500 ml anaerobic reactor (Schott glass bottle);
6. Beaker (500 ml) for receiving any NaOH leaving the cylinder;
7. 500 ml graduated cylinder containing NaOH to scrub CO_2 from the generated gas;
8. Displaced NaOH solution (equal to the volume of methane generated).

Figure 3: Experimental setup for the anaerobic reactor.

measured by displacement of a saturated solution of NaOH during 35 days of anaerobic digestion. The measured methane volume was compared with the expected theoretical volume. In this case, it was determined that the amount of landfill-waste methane generated would be 170 L per kg of waste. Since the reactor would work with 100 g of waste, it would therefore be expected to generate a maximum biogas volume of 17 L. The Potential Methane Generation (PMG) result was expressed as a percentage of the theoretical value.

The CH_4 and H_2S levels in the biogas were determined using a Gas Chromatograph, model Perkin Elmer GC Clarus 500/580. The determination of metal concentrations in both the soil samples and garbage samples was performed using a Perkin Elmer Optima 3000 ICP according to standard protocols based on official methods of the US-EPA (US-EPA, 1992; US-EPA-a, 2007; US-EPA-b, 2007; US-EPA-c, 2007). Figure 4 shows the physical installation for the experimental anaerobic digesters

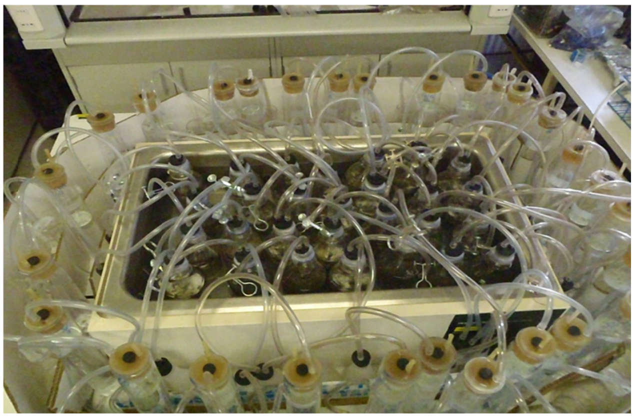


Figure 4: View of anaerobic reactors in a thermostatically controlled bath (35 ºC).

Potential Methane Generation was calculated as PMG(%) = (Vg(L)·100)/17 L.

RESULTS AND DISCUSSION

Figures 5 to 9 contain the location maps and results tables for the operable units in the study. Each table includes a short description of the current land-use for the unit and the thickness of each excavated layer according to the following description: Layer 1-Unconsolidated solid surface material, Layer 2-Trash, Layer 3-Deep soil. Other tabulated results include the pH range in each layer, range of PMG % and range of H_2S in Layer 2, and the metal concentration range in each layer. The results observed for Operable Unit Parcels 3 and 4 (Figure 7), and Operable Unit Residential Within Abandoned Landfill (Figure 9) are not shown, but are similar to the results shown for the other units.

Garbage was found in 24 of the 30 perforated pits, with differing levels of degradation, in thicknesses ranging between 40 and 370 cm across the sector. No garbage was found at the sample points located outside of the landfill boundary.

In 8 of the 24 samples analysed, the biogas levels recorded were zero, indicating that in these areas the garbage had completely stabilized. The non-zero measured PMG values were between 1.6% and 11.3%. The maximum PMG value found (11.3%) belonged to a sample with the characteristic odor and oily black appearance of industrial garbage and was explained by the extended stabilization time expected for this type of garbage.

In general, all observed PMG values are low compared to an active landfill. This indicates that the trash has achieved an advanced state of degradation, but still maintains a low capacity to generate methane under appropriate conditions. These results are consistent with previous studies conducted at the site in 2004, and are relatively similar to those reported in the study by Kristman (2009) in the area called Parcel 5, showing that the garbage had a low capacity to decompose. It is important to take into account that there is ample evidence that La Cañamera received wastes more than 35 years ago and, as such, there is not expected to be any further increase in the anaerobic degradation rate of the garbage stored in the site.

At the end of each sample digestion process, the methane and hydrogen sulfide content of the resulting biogas was determined. The methane volume fraction varied between 50% and 55% while the hydrogen sulfide volume fraction was generally near the detection limit of 0.02%. The low H_2S concentration in sanitary landfill biogas has been reported in other studies. Desideri *et al*. (2003) found that the concentration of hydrogen sulfide (0–200 ppm) and carbon monoxide (0–500 ppm) in sanitary landfill biogas was very small and therefore negligible for their study. Jaffrin *et al*. (2003) also found that the hydrogen sulfide concentration in sanitary landfill biogas was very low at only 100 ppm (0.01%). Themelis and Ulloa (2007) stated that sanitary landfill biogas has an H_2S volume fraction less than 1%, while Nikiema *et al*. (2007) found a level of 0–0.2%. The low proportion of sulfur compounds present in municipal solid waste explains these low H_2S concentrations (Zhou *et al*., 2014; Hla and Roberts, 2015).

For all the metals tested (Cd, Zn, Cr, Cu, Pb, Al, Ni, V, Se, As, Ba, B, Co, Mo, Mn, Fe, Hg), the highest concentrations were found in the garbage layer, when compared to the concentrations found in the unconsolidated solid surface and deep soil layers. This is because domestic and industrial wastes often contain metallic components.

The garbage layer in the site was covered with a layer of filler soil approximately one meter thick. The concentrations of heavy metals found did not exceed either the natural metal concentrations in Chilean soils or the human health risk standards defined by the United States Environmental Agency (USEPA) in its Superfund Site Remediation program.

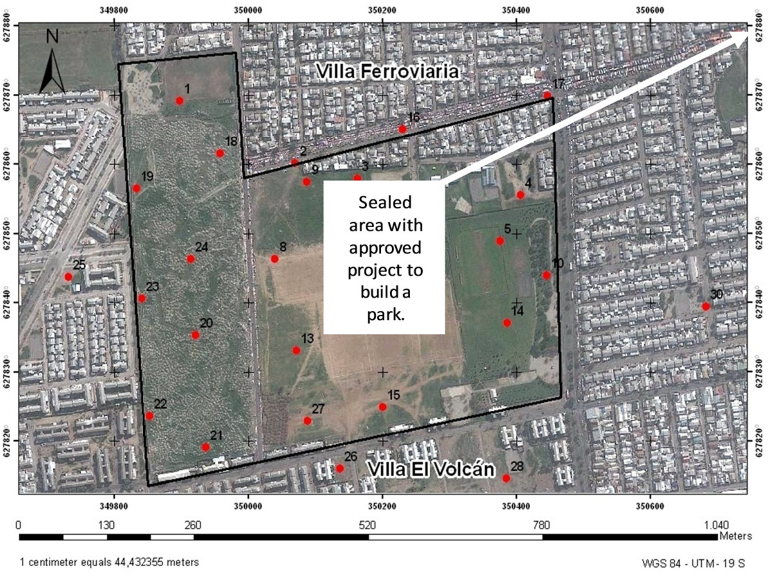


Table 1: Range of Values for Operable Unit Parcel 5			
	Layer 1	Layer 2	Layer 3
Thickness (cm)	40–70	40–280	40–210
pH	6.57–7.53	6.65–7.98	6.85–7.23
PMG (%)	-	0–6.0	-
H_2S (%)	-	<0.02	-
Cd mg/kg	<0.06–0.85	<0.06–2.63	<0.06–0.64
Zn mg/kg	18.1–148.2	341–1156	29.6–515
Cr mg/kg	13.4–28.3	10.9–420.6	11.2–253
As mg/kg	<2.39–22.8	<2.39–23.3	<2.39–12.6
Cu mg/kg	44.4–128,1	242–1085	49.5–392
Pb mg/kg	10.9–28.8	102–1478	15.2–243.4
Al mg/kg	3159–16076	4924–18562	4729–11764
Se mg/kg	<1.8	<1.8	<1.8
Ni mg/kg	<0.38–18.0	<0.38–29.0	<0.38–21.3
V mg/kg	198.9–182	74.4–138.8	95.1–198.2
Ba mg/kg	10–50.1	24.9–249	26.6–59.2
Co mg/kg	11.8–29.9	11.8–24.5	12.9–30.7
Mo mg/kg	<0.31–1.32	1.29–9.94	<0.31–3.35
B mg/kg	<0.46–99.4	<0.46–144.8	1.98–180.4
Fe mg/kg	9288–57523	8025–76629	1655–41467
Mn mg/kg	232–871	339–990	307–844
Hg mg/kg	0.04–59.7	0.41–12.7	0.05–1.07

Figure 5: Location and Results for Operable Unit Parcel 5.

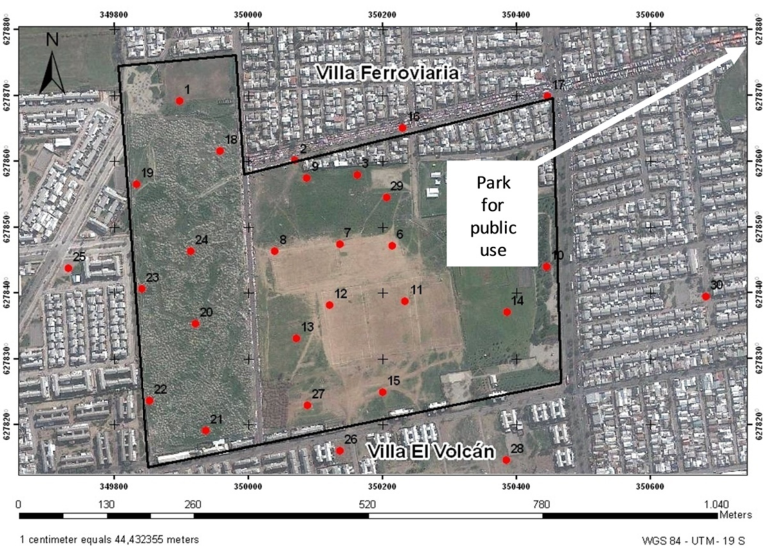


Table 2: Range of values for Operable Unit Current Park			
	Layer 1	Layer 2	Layer 3
Thickness (cm)	9–10	120–190	80– 20
pH	7.65–7.99	7.48–7.73	7.69–7.94
PMG (%)	-	0–5.8	-
H_2S (%)	-	<0.02	-
Cd mg/kg	1.38–2.51	0.78–1.93	<0.06–1.24
Zn mg/kg	551–713	587–838	87.7–280
Cr mg/kg	122–236	43–468	25.5–79.4
As mg/kg	5.31–19.9	5.06–9.83	<2.39–5.29
Cu mg/kg	359–2411	99–2312	66.4–221
Pb mg/kg	301–807	30.4–1082	<0.27–104
Al mg/kg	4610–9501	5536–7367	3610–79663
Se mg/kg	7.81–18.2	6.87–16.6	4.56–14.9
Ni mg/kg	25.9–43.0	13.6–63.2	10.0–17.1
V mg/kg	83.9–116.2	62.6–102.3	92.6–120.4
Ba mg/kg	20.3–57.5	43.6–48.7	15.4–292.5
Co mg/kg	15.7–25.3	11.6–22.4	14.8–19.2
Mo mg/kg	9.71–16.5	2.64–4.11	<0.31–1.63
B mg/kg	100–297	114–241	75.5–150.4
Fe mg/kg	25693–46569	30547–40477	20766–40602
Mn mg/kg	487–640	567–6227	413–865,8
Hg mg/kg	0.60–2.88	0.65–65.4	0.12–32.0

Figure 6: Location and results for Operable Unit Current Park.

At present, the operable units identified as Parcel 5 and Actual Park have been transformed into a park, while maintaining the contained garbage, and have chimneys installed for monitoring the emissions of methane gas that may be released from the trash layer. The land-use change management, for the other sectors of the site, is planned for execution at later dates due to differing legal and environmental action requirements.

The results obtained in this study are consistent with the general principles discussed by the WHO study of contaminated sites (WHO Regional Office for Europe, 2000). Therefore, this testing shows that the Potential Methane Generation (PMG) methodology should be added to the standard methodology for the assessment of contaminated soils in Chile.

Figure 7: Location of Operable Unit Parcels 3 and 4.

Table 3: Range of values for Operable Unit Property Reserve

	Layer 1	Layer 2	Layer 3
Thickness (cm)	70–170	120–230	80–130
pH	7.03–7.54	6.36–6.96	6.85–8.16
PMG (%)	-	2.3–11.3	-
H_2S (%)	-	<0.02	-
Cd mg/kg	<0.06–0.45	<0.06–9.02	<0.06
Zn mg/kg	72.9–130.4	264–1313	49.5–109
Cr mg/kg	21.9–32.6	101,7–853.3	19,1–34.8
As mg/kg	<2.39–8.0	<2.39–15.4	<2.39–7.0
Cu mg/kg	52.3–95.7	199.6–1319.5	56.9–60.8
Pb mg/kg	15.4–44.4	178.5–360.7	5.69–35.8
Al mg/kg	7211–7930	4961–27085	6376–11788
Se mg/kg	<1.8	<1.8	<1,8
Ni mg/kg	<0.38–17.0	<0.38–21.5	<0.38
V mg/kg	124–131	68.7–108.3	121–146.2
Ba mg/kg	27.9–52.4	46.1–166.2	35–45
Co mg/kg	16.4–20.1	9.5–16.1	15.1–18.4
Mo mg/kg	0.51–2.92	3.9– 5.4	<0.31–1.63
B mg/kg	87.6–176	20–141	25.6–146.8
Fe mg/kg	21199–29789	32322–41350	19998–39490
Mn mg/kg	265–428	520–685	303–401
Hg mg/kg	0.06–0.18	0.65–1.16	0.12–0.68

Figure 8: Location and results for Operable Unit Property Reserve.

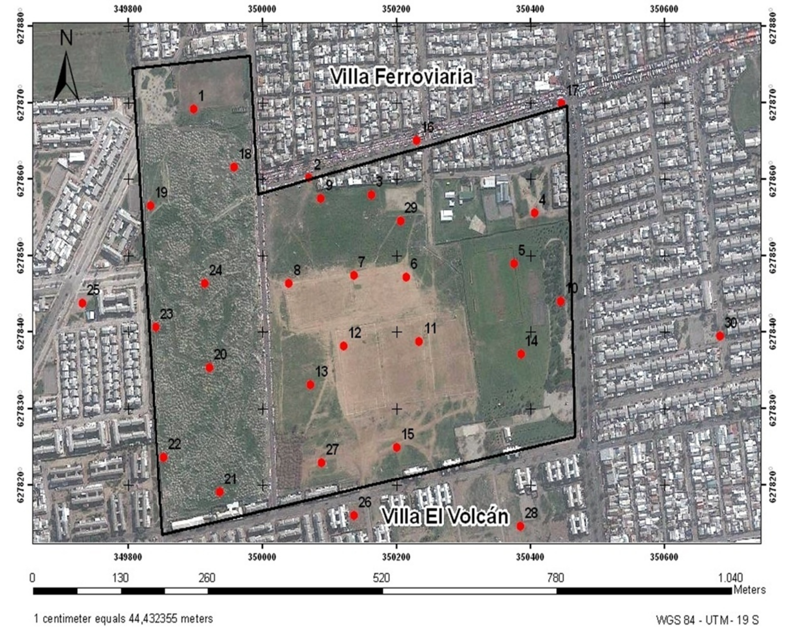


Figure 9: Location of Operable Unit Residential Within Abandoned Landfill.

CONCLUSIONS

Study development and analyses have identified PMG values between 1.6% and 11.3% in 16 of the 24 garbage samples taken at the landfill site. These values are low compared with an active landfill, which indicates that the trash stored in the site has achieved an advanced state of degradation, but still maintains a low capacity to generate methane under appropriate conditions.

Concentrations of heavy metals in the surface material layer that covers the trash and in the deep soil layer do not represent imminent risk to human health and do not exceed the normal levels of metals in soils.

The usefulness of the Potential Methane Generation (PMG) test for risk assessment of abandoned landfill sites has been demonstrated, which is an important contribution to the Chilean methodology for evaluation and management of SAPPC. After the completion of this study, the environmental management plan for the site transformed two of the operable units studied into a park.

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