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Evaluation of the Efficiency of Reverse Osmosis in the Treatment of Sanitary Landfill Leachate in the Metropolitan Region of the Rio de Janeiro

Avaliação da Eficiência da Osmose Reversa no Tratamento de Lixiviado de Aterro Sanitário da Região Metropolitana do Rio de Janeiro

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The decomposition of the organic matter in municipal waste from landfills results in a highly polluted and toxic effluent, named sanitary landfill leachate. Due to the potential contamination for soil, groundwater and surface water bodies, the treatment of sanitary landfill leachate is essential to avoid environmental and public health damage. This study aims to evaluate the technical and environmental efficiency of reverse osmosis technology for the treatment of leachate in landfill from the Metropolitan Region of the Rio de Janeiro, Brazil. It was observed that the raw leachate from this landfill has highly polluting, and has chemical and physicochemical characteristics typical of mature landfills, with operation older than 5 years: pH = 7.85; ammonia nitrogen concentrations of 2,473.00 mg L^{-1}; BOD 2,545.84 mg L^{-1} and COD 4,881.81 mg L^{-1}, respectively. Except for only one sampling, with phenol parameter, reverse osmosis was technically and environmentally efficient in mitigating pollutants present in the raw sanitary landfill leachate with excellent removal rates: $E_N(\%) > 99\%$; $E_{BOD}(\%) > 98\%$, and $E_{COD}(\%) > 99\%$. After reverse osmosis treatment, the treated leachate was respecting Brazilian legislation and could be released into the local stream. During reverse osmosis treatment of 120 m^3 day^{-1} of leachate, prevented 107.84 kg $year^{-1}$ of N from being released into nature; 110.95 kg $year^{-1}$ of BOD, and 211.70 kg $year^{-1}$ of COD, avoiding eutrophication from nearby rivers that inevitably flow into Guanabara Bay. It becomes a promising technology to face the impacts that pressure the Planetary Boundaries in the Anthropocene.

Keywords: Municipal solid waste; landfill leachate treatment; Planetary Bounderies; Anthropocene.

1. Introduction

Since the second half of the XX century, a period known as “The Great Acceleration” of Anthropocene, an increase in the pollution from aquatic and terrestrial ecosystems due to the changes in habits and patterns of production and consumption is evident, also the exponential growth of the world’s population, adding to the fact of no urban planning and strong industrialization.[1-3] Therefore, the lack of management from both industrial and municipal effluents and solid wastes results in significant negative impacts on different environmental compartments: soil, air, sediments, surfaces waters, groundwater, human health, and flora and fauna.[4-7] It is a dangerous threat to the Planetary Boundaries (PB), defined as “operational safe zones for humanity development as species”,[8,9] even more so at the moment when the world is confined due to the effects of the zoonotic COVID-19 pandemic, a disease of the Anthropocene.[10,11]

The main concern about the proper disposal of municipal solid waste (MSW) is on behalf of the associated risks. The following ones stand out: biogas emission containing greenhouse gases (GHG), with particular attention to methane (CH_4) and carbon dioxide (CO_2), which contribute to the global climate changes and air pollution.[12-14] Also proliferation of disease agents and inevitable Sanitary landfill leachate (SLL).[15-16] The SLL is an effluent resulting from the organic, biodegradable, and recalcitrant waste decomposition,[17] increased by moisture and water sources, could be retained by capillary absorption, external (rainwater, surface, and groundwater), or even from bacteria that dissolve organic matter with enzymes to turn it into liquids.[12,13,18] If it inappropriately is disposed of, the SLL represents an expressive threat to public health and the environmental aspects of soil, surface, and groundwater contamination, being 200 times superior in damage than the conventional domestic sewage.[15,17,19] Besides, it is a highly recalcitrant material, being hard to suffer biological degradation, due to the complexity in their composition, which consists mostly in organic, ammonia nitrogen, inorganic ions, metals, and others toxic compounds.[19-22]

Each ton of MSW disposed of in a landfill produces an estimated volume of 0.2 m³ of SLL.[23] Therefore, according to ABRELPE based on results of 2018,[24] referring only to the fraction of MSW disposed of in Brazilian landfills (43.3 million tons), annual production of SLL is estimated around 8,660,000 m^3. Therefore, the SLL parameters depend on factors like; biological, physical, and chemical processes that occurred inside the landfill body, also the landfill age, the operational configuration including compaction level of the cover material, and the specific regional climate trends.[18,25-27] Considering that SLL has potential impacts, it justifies the importance of the effluent treatment. This is a considerable challenge due to the heterogeneity MSW disposed of, also the trend of more strict legal demands by the standards and requirements for discharging treated wastewater in water bodies.[13,26] In Brazil, legal demands for discharging are established by CONAMA's resolution nº 430/2011.[28] It stands out that the Brazilian technical regulation NBR 8419/1992,[29] describes the minimum standards for MSW landfill project presentation and demands that this kind of disposal project contains a SLL collection, drainage, and treatment system.

To comply with environmental legislation, it is customary to combine different methods of treatment, being mostly conventional: aerobic or anaerobic biological processes (activated sludge, leachate evaporation ponds, biological filters) and physical-chemical (filtration, coagulation, flocculation, adsorption, precipitation, sedimentation, ion exchange, chemical oxidation, evaporation, and incineration).[14,16] SLL can also be forwarded to the sewage treatment stations, being careful that the additional charge does not cause any damage to the treatment process.[12,13,17] Additionally, the co-treatment could influence the sewage treatment stations fitness and efficiency, due to the following issues; a significant increase of organic load at the final effluent, possibly exceeding the project's limits, compromising the legislation reach limits of discharging wastewater; potential input of meaningful ammonia levels, of biodegradable compounds and recalcitrant non-degradable substances that leachates contain and might affect the result of the final effluent. If the removal of those materials were not considered in the design project of the STS; the possible presence of toxic substances from SLL, especially if the landfill received industrial waste, might be found metallic and organic contaminants, which inhibit the kinetics of the enzymatic reactions of nitrifying, denitrifying and heterotrophic bacteria used in the biological treatment of sanitary sewage.[13,26]

Because of technical limitations from conventional treatments and the needs of subsequent treatment, the physical treatment of reverse-osmosis system (RO) appears as a proper solution for pollutants removal on SLL, especially due to retention and quality reached by the technic up through three purification stages.[12,14,19,30] However, a successful technic used in a sanitary landfill will not necessarily turn into a global solution and be automatically suitable to any others.[13,31] Therefore, technical efficiency and environmental evaluation it is necessary for each case.[21] The technical efficiency consists of optimizing the inputs used in a way that those technological possibilities are turned into useful products. In other words, the technical requirements must be following the designed use and related to the increase of productivity.[32] In regards to environment efficiency, it is expressed by the lowest potential to cause environmental damage and unsustainable use of natural resources.[32] In this context, the present study aimed to evaluate the technical and environmental efficiency in the reverse-osmosis system on treatment of leachate from a sanitary landfill located in the Metropolitan Region of the Rio de Janeiro State. Also, evaluate the impacts of concentrate leachate recirculation in the landfill body, verifying the central part of the landfill leachate in the Planetary Boundaries in the Anthropocene.

2. Material and Methods

2.1. Study area

The technical efficiency of RO for SLL treatment was evaluated based on periodical analysis results of SLL and permeated (treated effluent). The SLL is from a sanitary landfill opened in 2012 and located in the Metropolitan Region of the Rio de Janeiro state (coordinates UTM 7470700 N and 706400 O) that receives approximately 2,500 ton day^{-1} of MSWs,[21] attending large commercial generators of MSWs in nearby cities (Niterói, Maricá, and Itaboraí) and full attendance to the MSW produced in São Gonçalo city, second-most populous city of Rio de Janeiro state, with an estimate population of more than one million of citizens.[33] São Gonçalo has tropical weather, with higher pluviometry in summers than winters, and an Aw climate classification (Köppen-Geiger), 23.3°C average temperature and the annual pluviometry average of 1,257 mm (Figure 1). The average SLL production of the Landfill is about 250 m^3 day^{-1}.

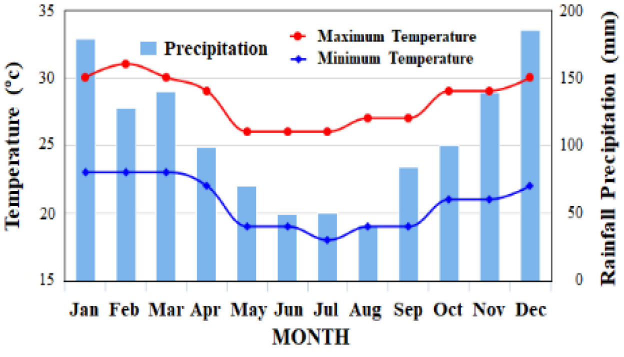

Figure 1. Climatology of the São Gonçalo city. Modified and adapted from Climatempo[34]

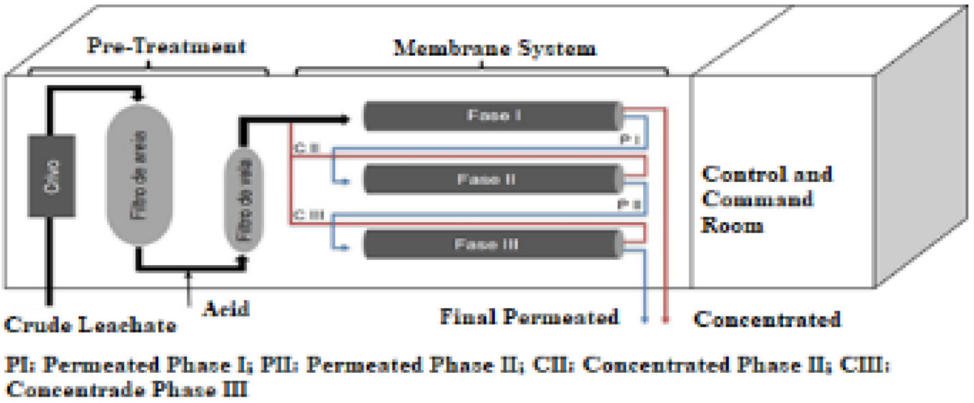

Figure 2. Flowchart of the Landfill Leachate Treatment Station (LTS). Adapted and modified from literature[21,30]

2.2. Leachate Treatment Station (LTS)

As observed in Figure 2, the leachate treatment station (LTS) for landfill has been operating since 2014, system model OR120 manufactured and provided by the company AST *Soluções e Serviços Ambientais* was evaluated, with 120 m^3 day^{-1} of treatment capacity and composed by the following integrated systems: physical pre-treatment (filtration), reverse osmosis (RO) in three stages and degassing (gas scrubber).[21,31] The membranes separation occurs by the exclusion mechanism, according to the different sizes of the particles, and the transport is done through porous membranes.[12]

The RO operation demands a pre-treatment procedure, filtering the effluent until it reaches the required feeding standards in the membrane system (Figure 2). Initially, the SLL storage in a leachate evaporation pond is pumped into a tank, where the pre-treatment process proceeds, the effluent is filtered and, then, pumped to the membrane system, where the RO occurs (Figure 3).[35] After the treatment, the treated effluent (final permeate) is reused for humidification of internal routes of the Landfill.[21] The concentrated liquid consists of portions of SLL substances that were retained at the process, being common to be treated through: recirculation/reinjection for the sanitary landfill; evaporation; incineration; solidification/stabilization with sewage treatment sludge.[17,27,36] In the LTS evaluation in this study, the adopted alternative is the traditional recirculation of the concentrated liquid to the waste mass, for being an economically viable/practical solution.[13,21,36] However, the recirculation must be applied with precautionary measures to avoid hyper concentration effects on Landfill, since high concentrations might interfere in the technic profits and act as secondary pollution sources to be treated by LTS[30,36] and compromise the sanitary landfill biogas production and/or the biological activity of microorganisms existing in the landfill body.[27]

Figure 3. Leachate Treatment Station (LTS)

2.3. Reverse Osmosis Technology

The RO consists of a process of substances segregation by membranes, it acts like selective barrier separation originating two-fluid bases. In osmosis, a physicochemical phenomenon is essential for cellular functioning, the water flows from a hypotonic medium to a hypertonic medium through a semipermeable membrane, until it balances and becomes an isotonic environment. Also, through an external pressure higher than the osmotic pressure, the opposite flow of water is forced, meaning that a highly concentrated solution is forced through a membrane into a region of low concentration of solutes.[13,35] Therefore, two supply currents are obtained from the system: concentrated and permeated (treated).[21] Therefore, even the monovalent salts retention, such as the chloride anion, is guaranteed due to the high level of separation limit of the membranes (< 0.001 μm).

2.4. Sampling and analysis of the efficiency of removal of pollutants from sanitary landfill leachate

Six campaigns to collect SLL samples (*in natura*) and treated (permeate) were realized, from July 2014 to March 2016, referring to the following campaigns: January/2014; March and November/2015; January, February, and March/2016. The analysis of the chemical and physical-chemical parameters was carried out according to the effluent discharge standards established by CONAMA 430/2011 resolution.[28] The samples were stored in sterile polyethylene bottles and kept in thermal boxes at a temperature of 4 ± 2 °C, using ice conservation and with constant monitoring. It should be noted that all samples were collected, preserved, and analyzed according to the recommendations of the Standard Methods for Examination of Water and Wastewater da American Public Health Association.[37]

The collected samples were sent to the chemical and physical-chemical laboratory accredited by the National Institute of Metrology, Quality, and Technology (IN Metro), obeying the preservation conditions, and analyzed according to the established expiration dates. To ensure that they were carried out according to the appropriate analytical method, the following quality controls were used for all parameters: analysis blanks and fortified samples with a known concentration of the analyte of interest (spike). For volatile organic compounds (VOCs), the tracer analysis method was also used, which consists of adding a substance of known concentration whose chromatographic behavior is similar to the compounds under analysis, but not present in the sample in question.

The following parameters were evaluated for the characterization of SLL: pH, electrical conductivity (k), total suspended solids (TSS), ammoniacal nitrogen ($N_A = NH_3 + NH_4^+$), Biochemical Oxygen Demand (BOD), and Chemical Oxygen Demand (COD). On the other hand, the following parameters were evaluated for the treated SLL pH, electrical conductivity (k), total suspended solids (TSS), ammoniacal nitrogen ($N_A = NH_3 + NH_4^+$), Biochemical Oxygen Demand (BOD), and Chemical Oxygen Demand (COD), phenol, total alkalinity (TA), methylene blue reactive substances (MBRS), mineral oils and greases (MOG), total oils and greases (TOG), vegetables oils and greases (VOG), arsenic, barium, boron, cadmium, calcium, lead, free cyanide, chloride, copper, total chromium, chromium(III), chromium(VI), tin, dissolved iron, fluoride, phosphorus, magnesium, manganese, mercury, nickel, silver, selenium, total dissolved solids (TDS), sulfide, sodium, zinc, as well as the volatile organic compounds (VOCs = 1,1-dichloroethene; benzene; cis-1,2-dichloroethene; chloroform; styrene; ethylbenzene; (o, m, p)-xylenes; carbon tetrachloride; toluene; trans-1, 2-dichloroethene; trichloroethene).

The efficiency (E%) of pollutant removal from SLL used the equation 1 proposed by Almeida e collaborators:[31]

$$E(\%) = \left(\frac{C_0 - C}{C_0}\right) \times 100 \qquad (1)$$

where: C_0 is the concentration of the pollutant in the raw SLL, and C is the concentration of the pollutant in the treated SLL.

2.5. Statistical analysis results

The results were organized and analyzed using the software Statistica® version 7.0. and submitted in minimum, maximum, arithmetic median, and standard deviations. To verify the existence of significant statistical differences between the average results for some parameters from SLL and the treated (permeated) SLL samples, the Student's test was performed at a 95% confidence level.

3. Results and Discussion

3.1. Sanitary landfill leachate characteristics and biodegradability

Firstly, the high variability observed for some parameters in the SLL may be related to seasonal climatic variations between the different sampling campaigns (Figure 1 e Table 1). On the contrary of what Almeida and collaborators observed,[31] on the characterization between 2016 and 2017, the SLL of Seropédica's Landfill, also located in the Metropolitan Region of the Rio de Janeiro State, it's operation started in 2011 received about 11,000 ton day^{-1} of MSW and produces about 1,000 m³ day^{-1} of SLL,[31] which is submitted by the same climatic characteristics from the Sanitary Landfill of this study, Aw (Köppen-Geiger) (Figure 1). On the other hand, it was observed a high variability on the *El-Hammam* Landfill leachate analysis results; this landfill receives 2,700 ton day^{-1}, and is located in Alexandria, Egypt.[15] Furthermore, another Sanitary Landfill

Table 1. Parameters determined in SLL from Landfill. (n = 6).

Statistics	pH	*K*	TSS	N_A	BOD	COD	BOD/COD
		(mS cm^{-1})	--------------------- (mg L^{-1})			------------------------	
Minimum	7.55	26.00	122.00	**683.50**	1,232.5	2,295.3	0.537
Maximum	8.04	30.80	451.10	**5,143.20**	4,243.37	7,526.86	0.564
Average	7.85	27.20	232.10	**2,473.00**	2,545.84	4,881.81	0.521
SD	0.26	1.00	190.00	**2,356.60**	1,968.16	2,891.92	0.061
MAV (1)	5 ≤ pH ≤ 9	--------	20%	20	60%	--------	--------

k = Electrical conductivity. TSS = total suspended solids. N_A = Ammonia nitrogen. BOD = Biochemical oxygen demand. COD = Chemical oxygen demand. SD = Standard Deviation. Maximum Allowable Values of CONAMA Resolution nº 430/2011 for effluents discharge.[28] **Bolded** values exceeded the respective MAV (Maximum Allowable Values) located in the Metropolitan Region of Curitiba-Brazil (1,500 ton day^{-1})[31] which has the same climatic classification which Egypt Sanitary Landfill on Köppen-Geiger as Cfb.[22]

Furthermore, it was observed a small variation of 6.5% on pH values, from 7.55 to 8.04 in SLL, which is proper to methanogenic bacteria development (Table 1), but theoretically not consistent with the time life of the Landfill in this study (< 5 years). Similar results were noticed by El-Salam & Abu-Zuid and Almeida and collaborators,[15,31] as Battker[22] and associated that found leachate samples with a slightly high and remaining pH from 7.00 to 8.00 during the operation of the respective Landfills older than 5 years, which indicates for this Landfill a stage of shortening the acidic phase and a near appearance of the methanogenic phase. Those pH results are also in accord with the ones reached at *Dhapa* Landfill (3,000 ton day^{-1} and > 10 years), located in the region of Kolkata (India), it reinforces the hypothesis that the organic compounds existing in the SLL may be prematurely promoted to a faster biological stabilization,[19] being the recirculation of the osmosis concentrate to the landfill body one of the most likely/possible causes.

El-Salam & Abu-Zuid evaluated the SLL parameters from the Egyptian Sanitary Landfill and found average values of electrical conductivity (*k*) and total suspended solids (TSS) in the order of 40.92 mS cm^{-1} and 27,452 mg L^{-1}, respectively. Those values are way above from the results present in this study,[15] in which the SLL's *k* is in the range of is 26.00 to 27.30 mS cm^{-1}, with 28.30 mS cm^{-1} of average, and 232.1 mg L^{-1} average result of TSS (Table 1). The *k* and TSS data are effects of the presence of inorganic compounds, especially in high concentrations of various cations and anions, as well as for highly soluble salts in water that makes difficult a SLL treatment with conventional systems and creates a biological decomposition methods resistance.[19]

In addition, anomalous analyses of ammoniacal nitrogen (N_A) were found in the samples of SLL from the following campaigns: Jul/14, Mar/15, and Mar/16 (Table 2). Therefore, N_A concentrations measured from 683.54 to 5,143.20 mg L^{-1}, typical data of mature Landfills (> 5 years; $[N_A] > 400$ mg L^{-1}), which are associated with slightly alkaline pH > 7.0, provided, according to Le Chatelier principle, the displacement of the acid-base balance (Brönsted-Lowry Theory) of ammonia ionization reaction (equation 2; $pK_b = 4.751$ or $K_b = 1.774 \times 10^{-5}$ L mol^{-1} at 25 ºC) from the products (NH_4^+ e OH^-) to reagents creation (NH_3), which is more toxic and inhibits the development of methanogenic bacteria, making the SLL treatment more complicated, as providing higher toxic effects to aquatic organisms.[15,22] Obviously, it should be noted that pH values close to or above 8.00 indicate a tendency for an even more basic medium, possibly associated with high concentrations of ammoniacal nitrogen due to the alkalization effect of the pH provided by free ammonia (equation 2).[18,22]

$$NH_{3(aq)} + H_2O_{(l)} \rightleftarrows NH_4^+{}_{(aq)} + OH^-{}_{(aq)} \qquad (2)$$

Furthermore, a higher concentration of ammonia in SLL may be a consequence of biological degradation of amino acids (polypeptides) and other nitrogenous organic compounds, which might correspond to up to 0.5% of the dry mass disposed of in a sanitary landfill.[17,18,31] A part of the nitrogen may have also been released in SLL as an ammonia compound by MSW aerobic and/or anaerobic decomposition.[19] The highly SLL toxic characteristic is reinforced in the observation of its N_A average value. It exceeded 124 times the MAV of CONAMA Resolution 430 (Table 2). According to Campos and collaborators,[18] sanitary landfills that recirculate SLL show an acceleration in the degradation of biodegradable organic constituents, tending to a considerable increase in N_A concentrations, when compared to landfills that do not use such practice.

In the present study, the range of BOD concentration varied from 1,232.51 to 4,243.37 mg L^{-1}, with a mean value of 2,545.84 mg L^{-1}, and the COD varied from 2.295.37 to 7,526.86 mg L^{-1}, with 4,881.81 mg L^{-1} of the mean value. The average result of COD is statically similar (*Student* Test t, $p < 0.05$) to the Seropédica Landfill data and has very proximity to the Cartagena City (Colombia) SLL;[31] sanitary landfill data (600 ton day^{-1}; > 50 years; COD = 4,482 mg L^{-1}),[20] which may indicate a similar composition of MSWs enriched by organic matter. The mean value of the rate BOD/COD = 0.5216 denote a remaining high biodegradability on the SLL (BOD/COD > 0.5) in the anaerobic phase, as noted by El-Salam & Abu-Zuid (2015), who identified a BOD/COD rate of 0.69, but that is early approaching to values typically presented by mature Landfills (BOD/COD < 0.5).[13,18]

3.2. Effects on biodegradability and stability promoted by concentrate recirculation on the landfill

Although obtained higher mean values of BOD and COD than those of the present study, a BOD/COD ratio = 0.63 was identified close to that of another sanitary landfill in Alexandria, Egypt (BOD: 28,833 and COD: 45,240 mg L^{-1}, respectively).[38] On the other hand, the results of the present study were different from those obtained on the SLL of the *Bordo Poniente* Sanitary Landfill (Mexico City, Mexico), which was identified a BOD/COD rate < 0.01 and mean values of BOD = 20 mg L^{-1} and COD = 5,000 mg L^{-1}, suggesting that this SLL was from a mature and well-stabilized.[39] A study was carried out in Taipei City (Taiwan) on the composition of rainwater and its effects over the lifetime of *Fuketeng* Sanitary Landfill, the results demonstrate that, after five years of operation, the BOD and COD concentrations (296 e 3,340 mg L^{-1}**,** respectively) were way below those presented in this study, which also suggests that the SLL belongs to a landfill in the mature stage.[40] Chofqi and colleagues when studying the SLL from the Landfill of Municipal *EL Jadida* (Marrocos) (150 ton day^{-1}; > 30 years),[25] identified BOD and COD concentrations of 60 and 1,000 mg L^{-1}**,** respectively, which provides a

BOD/COD rate = 0.06, indicating well-stabilized leachate and an SLL that is entirely in the methanogenic phase of anaerobic degradation.

Except for pH and N_A, from SLL results which were typical of young SLLs and also present higher BOD than mature-stage sanitary landfills. The COD concentration might reach 81,000 mg L^{-1} on young Sanitary Landfills, while the mature Landfills usually do not exceed 5,000 mg L^{-1}.[15] The BOD/COD rate in these types of Landfill, which the biological activity corresponds to the acidic phase of anaerobic degradation, can reach values of 0.85.[25] On the other hand, the mature Landfills produces highly stabilized SLL with low degradability (BOD/COD < 0.1).[15,22] According to Chen,[40] young Landfills (ages from 2 to 5 years) have a gradual tendency to decrease substrates and microbiological contents as it goes older, reflecting a change in the SLL composition. Consequently, in middle age or at the end of the Sanitary Landfill lifetime, the main organic compounds in SLL are recalcitrant long-chain carbohydrates and/or humic substances. So, a fast and significant decrease in the BOD/COD rate of that SLL is to be expected.

Generally, in young Landfills like the one in case, when the MSWs are deposited (Figure 4), the degradation of organic residues starts, which results in an SLL with a higher concentration of easily degradable components, with emphasis on VOCs. Over time, the high molecular weight of refractory compounds is found instead of degradable organic matter, with special emphasis on humic substances (fulvic and humic acids) which can form potentially toxic compounds.[31,36] For example, the BOD/COD rates on initial LL in *Futekeng* Landfill, in Taipei (Taiwan) ranged from 0.6 to 0.8, similar to this study, but after five years of operation, the values of BOD/COD rate decrease to 0.2 to 0.4,[40] the same was observed in Seropédica Landfill that after two years of operation it has a data of BOD/COD rate = 0.44 in the SLL and after seven years of operation presented a decrease on BOD/COD rate = 0.07.[31] Therefore, it is expected that, over the years of operation, the BOD/COD rate in this study area will reach lower values, consistent with the temporal evolution of the generated SLL.

Figure 4. Sanitary Landfill of São Gonçalo City (Brazil)

The recirculation of the concentrated in the landfill provided components attenuation due to the biological activity and physical-chemical reactions that occur inside the landfill body, as indicated by the high pH and N_A values.[13,35] This acceleration effect on the stability of the organic matter present in the landfill (Figure 4), has been reported by several authors and is favored by the environmental conditions of tropical regions,[13,41,42] like in Brazil, as they increase the evaporation of leachate, and as a consequence, reducing its volume. According to Abbas and collaborators,[13] the concentrate recirculation not only improves the SLL quality by increasing the moisture content, but also provides a redistribution of nutrients and enzymes between methanogenic bacteria and solid/liquid interfaces. Further, it reduces the time needed for the landfill stabilization from decades to a few years (2-3 years), as is being observed in this study.

3.3. Treated landfill leachate (permeate) by reverse osmosis characterization

The characteristics of treated SLL by RO (Figure 5) are in Tables 2 and 3. As mentioned before, during the analysis, the following parameters were also determined: arsenic, barium, cadmium, free cyanide, chromium(VI), tin, dissolved iron, fluoride, magnesium, mercury, nickel, silver, selenium, sedimentable solids, and volatile organic compounds (1,1-dichloroethene; benzene; cis-1,2-dichloroethane; chloroform; styrene; ethylbenzene; (o, m, p-xylenes); carbon tetrachloride; toluene; trans-1,2-dichloroethane; trichloroethene). These are not exposed in the respective tables because the concentrations obtained were below the limits of detection (LD) and quantification (LQ) of the respective methods of analysis in all sampling campaigns.

Figure 5. Treated Leachate Collection Pond

As observed in the SLL (Table 1), the pH values in the treated SLL samples (Table 2, Figure 5) are above neutrality, varying the pH from 6.93 to 8.07 and having an average pH = 7.71±0.42, statistically similar to the mean value of the SLL ($p < 0.05$), but knowingly complying with the current legislation.[28] The SLL from the city of *Diyarbakir* landfill (Turkey), after using RO for treatment, presented the same behavior and mean pH value of 7.50, close to this research data.[43] In *Il Fossetto* sanitary landfill located in the province of *Pistoia* (Toscana North-East, Italy) it was also observed the same behavior and mean pH value of 7.69 when RO treatment and concentrate recirculation in the landfill body was used.[35]

As seen in Table 2, the $E_k(\%)$ efficiency reduction between SLL and permeate effluent was expressed in all campaigns where both were analyzed (99.64% to 99.92%). The same behavior was observed for TSSs which $E_{TSS}(\%)$ removal rate was between 97.5% e 99.3%, almost five times above the minimum removal rate (20%) recommend by current legislation.[28] Hasar *et al.*,[43] also identified a k expressive reduction in a RO treated SLL of 7.40 to 0.02 mS cm^{-1}. The k results for treated SLL (permeate) compose a critical operational parameter because of the proportion relation with salts concentration and osmotic pressure and,[12] remaining below of 1.0 mS cm^{-1}, indirectly imply that this SLL will provide low anthropogenic impact when eventually discharged.[1,4] Almeida and collaborators[31] reached a maximum $E_k(\%)$ of 34% in Seropédica landfill, even using a treatment combining coagulation-filtration, ammonia *stripping,* and nanofiltration techniques (C-F/N-NH_3 and *Stripping* +NF), demonstrating the superiority of OR in removing salts from species with a molar mass of 500 to 2,000 Dalton.

The permeate presented a high $E_N(\%)$, between 99.2% to 99.9% (Table 2), and not a one moment exceeded the maximum permissible value (MAV) to the aquatic life safety, according to federal resolution CONAMA 430/2011 for this requirement (MAV $N_A \leq 20.00$ mg L^{-1}).[28] Otherwise, Almeida and collaborators (2019) reached in Seropédica landfill an $E_N(\%)$ of 70% maximum, even using the same combined treatment (C-F/N-NH_3 *Stripping* +NF). As in this study, Hasar *et al.,*[43] applied a RO system directly in the high ammoniacal concentration SLL (N_A = 2,620.00 mg L^{-1}) and reported a high $E_N(\%)$ (MPV $N_A \leq 5.00$ mg L^{-1}), it confirms that RO technique is applicable for SLL treatment. On the other hand, Calabró *et al.*,[35] applying a RO in a landfill that has a constantly recirculated concentrate SLL, achieved mean values of ammonia nitrogen closed to the ones reached in this study ($N_A \cong 15.00$ mg L^{-1}).

Regarding the BOD, all campaigns achieved $E_{BOD}(\%)$ between 98.37% and 99.59% (Table 3), almost 50% superior to the minimum established by the federal resolution CONAMA 430/2011 ($E_{BOD}(\%) > 60\%$).[28] The COD parameter is not mentioned in the environmental control instruments that layout requirements and standards for the discharge of liquid effluents. However, for the Jan-Mar/2016 campaigns, the COD values of the permeated SLL indicated $E_{COD}(\%)$ of 99.28% to 99.99% relating to the crude leachate values. As in this study, a high $E_{COD}(\%)$ and $E_{BOD}(\%)$ were registered in an SLL that adopted RO technique in Turkey (98% and 99% removal, respectively).[43] Otherwise, the combined process (C-F/N-NH_3 and *Stripping* +NF) used in Seropédica landfill removed at most $E_{COD}(\%) = 94\%$ from the SLL COD.[31]

The phenol parameter was the only one that did not comply with CONAMA 430/2011 resolution on all occasions, since, in the November 2015 campaign, it presented the result of 513 µg L^{-1} in the treated leachate (Table 2). However, in the following campaigns (January and March 2016), the results have already decreased significantly (55.32% and 58.40%, respectively). As previously observed, the BOD/COD rate = 0.521 denote a dominance of biodegradable

Table 2. Leachate Landfill (permeate) physical-chemical characterization (n = 6)

Parameter	Unit	Minimum	Maximum	Average	SD	E(%)	MPV[(1)]
pH		6.93	8.07	7.71	0.42	-	$5 \leq pH \leq 9$
k	(mS cm^{-1})	0.02	0.11	0.03	0.04	99.82±0.15	-
TSS	(mg L^{-1})	3,00	18.00	7.00	7.34	99.32±0.02	20%
N_A	(mg L^{-1})	3.72	14.60	10.85	4.21	99.51±0.32	20.00
BOD	(mg L^{-1})	5.00	69.00	35.67	23.43	98.85±0.65	60%
COD	(mg L^{-1})	0.081	75.22	35.08	31.17	99.42±0.51	-
Phenol	(µg L^{-1})	13.50	**513.70**	198.95	177.62	-	500
TA	(mg L^{-1})	0.046	85.66	42.78	40.45	-	-
MBRS	(µg L^{-1})	< LD	60.00	-	-	-	-
MOG	(mg L^{-1})	1.00	1.00	-	-	-	20.00
TOG	(µg L^{-1})	< LD	2.80	-	-	-	20.00
VOG	(µg L^{-1})	< LD	17.14	-	-	-	50.00

k = Electrical Conductivity. TSS = Total suspended solids. BOD = Biochemical oxygen demand. COD = Chemical oxygen demand. TA = Total alkalinity. MBRS = Substances reactive to blue methylene. MOG = Mineral oils and greases. TOG = Total oils and greases. VOG = Vegetable oils and greases. < LD = Values below the limit detection of the analytical method. E (%) = Efficiency in removing pollutants. [(1)]Maximum permissible values of CONAMA 430/2011 Resolution for effluent discharge.[28] The **bolded** values exceeded the respective MPV

Table 3. Landfill´s permeate Inorganic characterization. (n = 6)

Parameter	Unit	Minimum	Maximum	Average	SD	MPV(1)
Boron	(mg L^{-1})	0.98	1.34	1.14	0.13	5.00
Calcium	(μg L^{-1})	< LD	192.0	130.0	87.68	-
Lead	(μg L^{-1})	< LD	28.00	-	-	500
Cyanide	(μg L^{-1})	< LD	23.50	-	-	1,000
Chloride	(mg L^{-1})	0.01	4.90	1.56	1,93	-
Copper	(μg L^{-1})	< LD	26.00	-	-	1,000
Chromium	(μg L^{-1})	< LD	15.00	11.0	1.41	1,000
Chromium(III)	(μg L^{-1})	< LD	13.00	9.00	5.65	1,000
Phosphorus	(μg L^{-1})	0.02	180.00	64.00	68.77	1,000
Manganese	(μg L^{-1})	< LD	58.0	-	-	1,000
Sodium	(mg L^{-1})	3.38	26.61	11.59	9.23	-
Sulfide	(μg L^{-1})	< LD	8.00	5.40	2.07	1,000
Zinc	(μg L^{-1})	< LD	16.00	-	-	5,000

< LD = Values below of the limit detection of the analytical method. (1)Maximum permissible values of CONAMA nº 430/2011 resolution for the discharge of effluents (BRASIL, 2011)

organic matter (Table 1). Somehow, as observed in that same Table 1, the mean concentrations of COD in SLL are incredibly high (COD $\cong$ 4,881.81 mg L^{-1}), and it is known that a significant proportion of the recalcitrant material (more than 60% of total organic carbon) have humic substances,[39] mostly humic acids and the rest of the recalcitrant material proportion is composed by synthetic substances, such as: aliphatic, aromatic compounds, phenolic compounds, alicyclic, amongst others.

The other parameters in table 2 (TA, MBRS, MOG, VOG e TOG) presented the same trend of shallow results and did not conflict with the respective MPV, when applicable. The same behavior was noted on the inorganic parameters from treated leachate (Figure 5): meager results from the respective MAV (Table 3). As in this research, Calabró and colleagues also identified low results for cations and anions in an SLL frequently used in recirculation of landfills before RO treatment.[35] In addition, Hendryche e collaborators registered low concentrations of Pb, Zn, and Cr in SLL samples from Bohemia Central (Germany) Sanitary Landfills treated by OR.[27]As expected, the pH > 7.0 of LL promotes precipitation of the evaluated metals due to the recognized small solubility product constants (K_{sp}) being the primary medium.[17,22,27]

The low concentrations of inorganic parameters (Table 3) after being treated by RO can be explained both by operational and chemical criteria. The Landfill can have an inherent capacity for natural attenuation concerning to some of the pollutants present in the recirculated leachate. In a way, once reinjected, the cover layer of the sanitary landfill can provide adsorption phenomena favored by the presence of stabilized organic fraction, evidenced by the high COD values. On the other hand, the phenomenon known as the "sulfide barrier" may be occurring in the landfill, which significantly affects metals concentration. The sulfates presence in the landfill body, or even in the covering soil, can be biologically converted to the sulfide anions (S^{2-}) in the anaerobic environment of the landfill.[2,35] The formation of poorly soluble metallic sulfides can effectively reduce the presence of metals in the Landfill leachate particularly during the methanogenic phase, which has high pH values, as in this study. As can be noticed in Table 3, the SLL has an average concentration of [S^{2-}] = 5.40 ± 2.07 **μg** L^{-1}, which corroborates the existence of this precipitating reagent in the aqueous reaction environment.

3.4. Landfill leachate and the threats to planetary boundaries

The PBs are a science attempt to quantify the safe operational space for the worldwide human existence, remaining as constant as possible. The global conditions are predominantly found in the Holocene Period, currently threatened by the advent of the Anthropocene in the 20th century.[8,9,44] Each PB represents a critical process for the Earth System function, nine of which have been recognized so far: global climate changes, changes in the biosphere integrity, oceans acidification, depletion of the ozone layer, biogeochemical flows (N and P cycles), land and water use changes, atmospheric aerosols and chemical pollution.[8,9,44] Unfortunately, four of these PBs have been exceeded because of global human actions (global climate change, N and P biogeochemical flows, land-use change, and changes in the biosphere integrity).

As presented in this research, the SLL has a highly polluting and toxic character to aquatic organisms, and can even act as an element of local pressure, or regional, to several of the PBs from the Earth System (Change in the biosphere integrity, oceans acidification, biogeochemical flows of N and P, water use and chemical pollution), which becomes worrying because in times of the COVID-19 pandemic in which the partial Lockdown implemented in Rio de Janeiro provided an increase in the generation of solid urban waste, consequently increasing the generation of leachate.[11] However, the SLL treatment by RO in this Landfill, as a closed system in which all the leachate generated is recirculated to the landfill body, prevents a discharge of almost 107.84 kg year^{-1} de N_A; 110.95 kg year^{-1} of BOD e 211.70 kg year^{-1} of COD to local water bodies, which would inevitably flow into Guanabara bay, one of the most eutrophic estuaries in Brazil.[45]

4. Conclusions

The reverse osmosis technology came as a highly efficient alternative for the removal of the high concentrations of toxic pollutants present in the leachate of large landfills located in tropical regions, both young or mature stages of operation. However, one of the campaigns had the phenol concentration above the maximum permissible value required by CONAMA 430/2011 resolution, possibly due to the presence of a high concentration of the recalcitrant (humic and fulvic acids) as denoted by the variation of the results for COD. It is noteworthy that the sanitary landfill discussed disposes of both solid municipal waste and non-hazardous industrial waste authorized by the state environmental agency. As phenol constitutes several industrialized products such as surfactants, antioxidants, lubricants, oils, perfumes, paints, medicines, pesticides, plastics, etc., such change may be due to the receipt, in the period studied, of a more significant amount of waste with these chemical characteristics, possibly a more significant batch of products out of specification, treated as waste.

Once the operating parameters have been adjusted for the reverse osmosis plant, it no longer exceeds the maximum permitted values for phenol (January and March 2016 campaigns), the permeate began to have the quality that allows a more noble use than humidifying the internal routes of the landfill. It also can attend to high technological complexity industries demand (food or chemical-pharmaceutical) that need water with a high standard of purity and quality.

Reverse osmosis offers technology capable of mitigating possible negative environmental impacts from MSW's in big cities. Besides, its uses combined with concentrate recirculation for the landfill body attenuates the eutrophication of water bodies close to landfills. Therefore, it helps to ensure that different planetary boundaries are not exceeded at a local and regional level.

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