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Impact Assessment of 2020 COVID-19 Lockdown on Landfill Leachate Treatment in Rio de Janeiro

Avaliação do impacto do Lockdown de COVID-19 em 2020 no Tratamento de Lixiviado de Aterro Sanitário do Rio de Janeiro

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In 2020, Brazil adopted different Non-Pharmacological Intervention measures (NPIs) to reduce the levels of transmission and contagion of the novel Coronavirus SARS-CoV-2, the pathogen causing the COVID-19 pandemic. The objective of this work was to evaluate the impact of Non-Pharmacological Intervention measures on the composition and treatment efficacy of Landfill Leachate treated by Reverse Osmosis in the city of Campos dos Goytacazes, Rio de Janeiro. It was observed that the Lockdown and the massive use of personal hygiene/cleaning materials, as well as the increase in food consumption, provided an unprecedented 140% overload in the daily generation of Municipal Solid Waste (MSW) in the Campos dos Goytacazes Landfill. The Leachate from this Landfill presented a slightly alkaline pH (pH ≈ 8.0), high concentrations of dissolved organic matter (BOD_5 and COD), and ammoniacal nitrogen. In addition, it was observed that 23% of the parameters in the raw leachate exceeded the respective maximum allowable values recommended by the federal legislation. However, in the period evaluated by this study, the Reverse Osmosis was technically and environmentally effective in treating the raw leachate, not being observed any result in the treated leachate that exceeded the Resolution CONAMA 430/2011.

Keywords: COVID-19 pandemic; lockdown; non-pharmacological intervention; landfill leachate; reverse osmosis.

1. Introduction

December 2019 saw the first reports of a significant increase of a new respiratory illness that was subsequently identified as COVID-19 (Coronavirus Disease 2019), caused by the new SARS-CoV-2 Coronavirus (Severe Acute Respiratory Syndrome Coronavirus 2), with its epicenter in the megacity Wuhan, capital of the highly cosmopolitan and globalized Hubei province in China.[1] Consequently, the World Health Organization (WHO) declared, on March 11, 2020, that COVID-19 has spun out of control and become a severe global zoonotic pandemic,[2] with 281,815,055 million infections and 5,422,435 million deaths from COVID-19 recorded worldwide as of December 27, 2021.[3] Brazil being the country with the disgraceful second-highest number of fatalities from the disease to date.[3] Since then, the vast majority of governments in different countries around the world have complied with WHO recommendations to adopt Non-Pharmacological Intervention measures (NPIs) to reduce the levels of transmission and contagion of SARS-CoV-2: Lockdown, quarantine, social distancing, massive use of face masks, constant hand sanitization, decontamination of packaging, restriction of public transportation, closure of public education facilities, the shutdown of non-essential industrial activities, etc.[1,4-9]

The adoption of NPIs by international governments has had significant impacts, both positive and negative, as well as direct and indirect, on society in general and the environment in particular, making COVID-19 an emblematic disease of the Anthropocene,[1,2,10] the geological Epoch in which humankind has become an unprecedented geomorphological force.[11] The social and economic impacts caused by NPIs have resulted in the closure of educational facilities (schools and universities),[4,6-9] increased unemployment, precarious jobs,[7] decreased tax collection, and others. However, it has been observed that NPIs have provided improvements in some environmental parameters (increase in atmospheric quality, decrease in greenhouse gas concentrations, decrease in noise pressure on marine life, decrease in water pollution, etc.) in some cities and regions, in Brazil and around the world.[1,12,13] Moreover, since the official announcement of the pandemic by the WHO, extremely relevant negative environmental impacts have been identified in large international urban centers (increase in the generation of Urban Solid Waste - USW, increase in the generation of Healthcare Waste - HCW, paralysis of waste recycling programs, etc.).[1,5,10,14]

During the height of the COVID-19 outbreak in Wuhan, it was observed that the average daily HCW jumped from 40 to 240 tons per day,[1] making it necessary to urgently build a new HCW treatment plant in the city.[1,14] Additionally, it has been observed in Brazil that due to the Lockdown and other NPIs, the population has changed their eating habits and started consuming more industrialized ultra-processed foods and/or foods produced by restaurants and purchased through online shopping, thus providing an average increase of over 10% in the generation of USW in the country,[1,15] due to the unprecedented addition of plastic packaging and organic waste in the mass composition of USW.[15,16] Also, the inadequate disposal by the Brazilian population of used and unserviceable Personal Protective Equipment (PPE) (cloth/plastic face masks, latex gloves, sanitizing product packaging, etc.) has contributed to the increased generation of USW that was improperly sent to Municipal Solid Waste (MSW) Landfills.[5,16] These facts have constituted a huge challenge in the public and private management of USW, which had to adapt to the sudden and unexpected extra reception of waste, as well as to the eventual significant modification of the mass composition of the waste they were used to receiving at the landfills.[16,17]

Among the greatest challenges of adequate final disposal of USW is the treatment of Landfill Leachate (LL),[18,19] which is the effluent that results from the decomposition of organic waste (biodegradable and/or recalcitrant)[20-22] augmented by rainfall, moisture content of the waste mass and/or natural water sources (surface or underground).[20-22] Each ton of USW disposed of in MSWs has the potential to generate approximately 0.2 m^3 of LL.[22,23] Therefore, the increase in USW generation indirectly provided by NPIs, especially by the Lockdown, has the potential to overload the USW treatment systems of the MSWs, which, from night to day, are forced to receive a greater input of USW and, consequently, to treat a greater volume of USW, which may even present composition of chemical pollutants different from those that were considered in the project originally proposed for the respective Landfill Leachate Treatment Plants (LLTP). Since the enactment of the federal law that established the National Policy for Solid Waste (NPSW, 12. 305/2010), several different technical alternatives for the treatment of LLs have been emerging in Brazil,[20,21] and in the State of Rio de Janeiro, the treatment technique by Reverse Osmosis (RO) is being adopted in preference to other internationally recognized technologies (nanofiltration, ultrafiltration, photo-Fenton, geobags, among others).[20,21,24,25]

To date, no research conducted by the international scientific community has been identified to evaluate the impact of NPIs on the ability to treat LL by any type of technique during the COVID-19 pandemic. Therefore, this study aims to evaluate the technical and environmental efficiency of landfill leachate treatment in a period before/during the Lockdown and other NPIs (February to September 2020), by a Reverse Osmosis system installed at the Conselheiro Josino Sanitary Landfill, in the city of Campos dos Goytacazes, the northern region of the State of Rio de Janeiro.

2. Material and Methods

2.1. Study area

The city of Campos dos Goytacazes (7621098 N and 878897 W) is the largest and seventh most populous in the state of Rio de Janeiro, with 511,168 inhabitants distributed in a total area of 4,032 km^2.[25,26] Moreover, it has the tenth largest Gross Domestic Product (GDP) of Rio de Janeiro, mainly due to the performance of the services, commerce, farming and cattle raising, civil construction, and transformation industry sectors, and to the royalties obtained from hydrocarbon exploration in Campos Basin (southeastern region of Brazil). Moreover, Campos dos Goytacazes has a humid tropical climate (Aw, according to the Köppen-Geiger classification), with an average annual temperature of 24 °C.[25-27] The average annual rainfall is 1,073 mm, with higher precipitation during the wet season (summer: October to January) and lower during the dry season (winter: February to September),[27] as can be seen in Figure 1.

All USW generated daily in Campos dos Goytacazes, about 357 tons day^{-1}, as well as that of large commercial generators, has been sent to the Conselheiro Josino Sanitary Landfill (CJSL) since 2011.[19,26] In addition, the CJSL acts as a properly licensed regional landfill also receiving USW (mostly Class II) generated in different municipalities of the north and northwest regions of Rio de Janeiro (Cardoso Moreira, Italva, Lage do Muriaé, Miracema, São Francisco do Itabapoana and São João da Barra), totaling estimated disposal around 550 ton day^{-1} at CJSL,[18,25] (useful life ≈ 30 years),[19] possessing good operational evaluation by the State Environmental Institute of Rio de Janeiro (Portuguese acronym: INEA) in the 2013-2015 triennium.[19] Finally, due to local climatic conditions the estimated LL generation, in the period before the COVID-19 pandemic, was 15 m^3 day^{-1} in the dry season and 45 m^3 day^{-1} in the rainy season.[26]

2.2. Landfill Leachate Treatment Station (LLTS)

For the current study, the performance of the Landfill Leachate Treatment Station (LLTS) Model "AST OI-03-60 System" manufactured and supplied by the company AST "*Soluções e Serviços Ambientais*" was evaluated, which has a maximum capacity to treat LL at a flow rate of 60 m^3 day^{-1} and is composed of the following integrated systems: physical pre-treatment (filtration), RO in three stages and degasser (gas scrubber), as can be seen in the flowchart of Figure 2.[24,28] The separation by membranes occurs by size

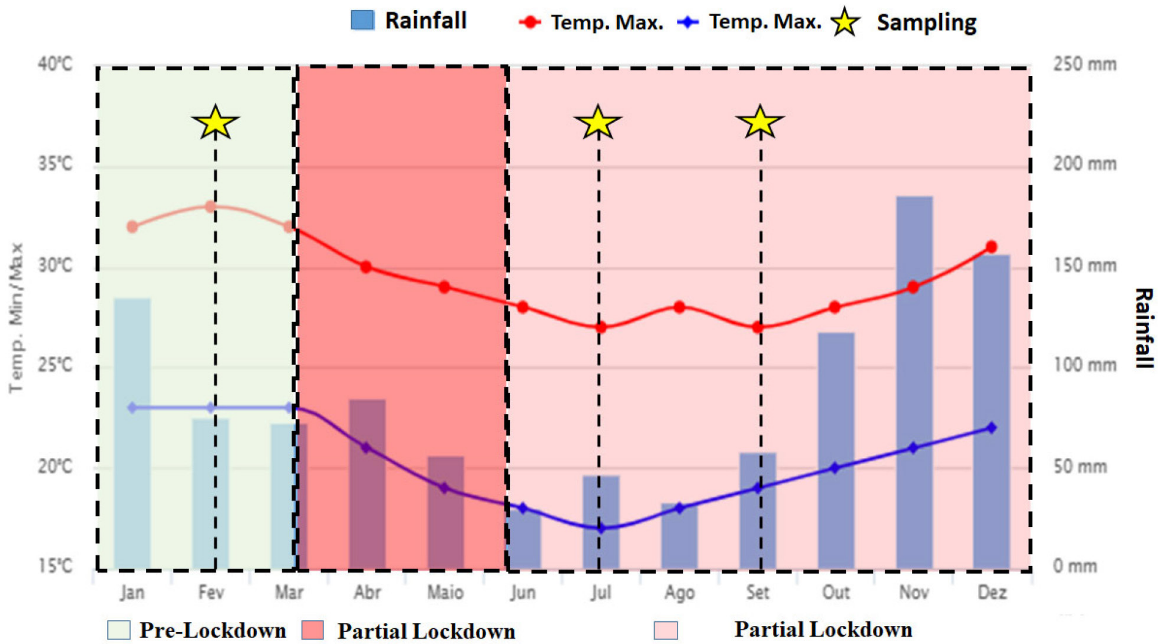

Figure 1. Rainfall and average monthly temperatures in the Pre-Lockdown period and during partial Lockdown in Campos dos Goytacazes during 2020[27]

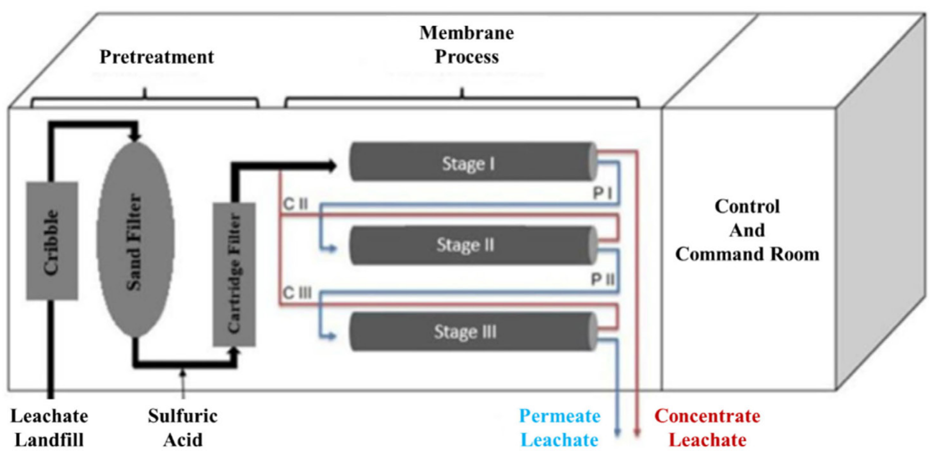

Figure 2. Flowchart of the Landfill Leachate Treatment Station (LLTS) installed in Campos dos Goytacazes

exclusion mechanism between the particles and the transport is done through porous membranes (model GE Osmonics Desal membranes SC8040F1012).[24,28,29]

The LLTS at CJSL started operating in 2015 and has a filter pretreatment to ensure the minimum necessary standards for feeding the effluent to the membrane system (Figure 2).[28,29] Initially, the raw LL constantly stored in an open-air storage lagoon in the CJSL area is pumped to a pretreatment tank. Next, the effluent is led to a sand filter and pumped to the membrane system where RO occurs.[30] The treated effluent (final permeate) is reused for humidification of the CJSL's internal paths, without any type of release to local water bodies. The concentrate generated after treatment by RO (approximately 30% of the volume of the input from the RO)[23] is the fraction where the substances present in the LL are retained, and is usually treated using: reinjection/recirculation to the CJSL since 2015; and may have as alternatives: evaporation; incineration and/or stabilization with sludge from Sewage Treatment Plants (STP).[23,31-33] In the LLTS evaluated at CJSL, the alternative adopted is the continuous recirculation of the concentrate to the waste mass,[27] as it is the most economically feasible solution.[31,34]

2.3. Reverse osmosis technology fundamentals

RO is a physical separation method with membranes in which they act as a selective barrier to transport for the separation of two fluid bases. As is well known, natural osmosis water moves from a hypotonic to a hypertonic medium through a semipermeable membrane until the mediums become isotonic. On the other hand, in reverse osmosis, using a high external pressure higher than the osmotic pressure, the reverse flow of water is forced, that is, a highly concentrated solution is forced through a membrane into a region of low solute concentration.[30,34] Thus, two feed streams are obtained in the system: concentrate and permeate (treated).[23] Furthermore, retention of even monovalent ions, such as the Cl^- anion, is guaranteed due to the membrane's high separation limit (< 0.1 nm).[23,35] This also guarantees that the RO can remove the SARS-CoV-2 Coronavirus, if present in the LL, since the mean diameter of this pathogen is between 100-120 nm.[35]

2.4. LL sampling and characterization

For the evaluation of the indirect environmental impacts provided by the adoption of NPIs for the containment of the pandemic COVID-19, on the composition and technical efficiency of the treatment of LL from the CJSL, three sampling campaigns of raw and treated (permeate) LL were conducted, in months with rainfall rates and temperatures close to the dry season during the year 2020: February = 75 mm and 23°C (Pre-Lockdown); July = 47 mm and 19 °C (Lockdown) and September = 59 mm and 21 °C (Lockdown).[27] The analysis of the physical, physical-chemical, and chemical parameters was performed against the Maximum Permissible Concentrations (MPC) established by CONAMA Resolution 430/2011 for the release of effluents from USW final disposal systems of any origin.[36] All samples were stored in sterilized plastic bottles and kept in thermal boxes at a temperature of 4.0±2.0 °C, using ice conservation and with constant monitoring. It should be noted that all samples were collected, preserved, and analyzed according to the Standard Methods for Examination of Water and Wastewater of the American Public Health Association.[37]

The collected samples were sent to private physical, physical-chemical, and chemical analysis laboratories accredited by INEA and duly accredited by the National Institute of Metrology, Quality, and Technology (Portuguese acronym: INMetro), obeying strict preservation conditions, and analyzed according to the established expiration dates. The entire chain of custody was evaluated and validated.

The following parameters were evaluated for the characterization of the LL (raw and treated) from the CJSL: pH, Electrical Conductivity (k), Temperature (T), True Color (TC), Total Alkalinity (TA), Total Hardness (TH), Sedimentable Solids (SS), Total Dissolved Solids (TDS), Total Solids (TS), Biochemical Oxygen Demand (BOD_5) and Chemical Oxygen Demand (COD), Total Phosphorus, Ammoniacal Nitrogen ($N_A = NH_3 + NH_4^+$), Total Nitrogen (TN), Vegetable Oils and Animal Fats (VOAF), Mineral Oils (MO), Total Oils and Greases (TOG), Surfactants, Aluminum, Arsenic, Antimony, Barium, Boron, Cadmium, Calcium, Lead, Cyanide, Free Cyanide, Total Chloride, Free Residual Chlorine, Cobalt, Copper, Dissolved Copper, Total Chromium, Chromium(III), Chromium(VI), Tin, Total Phenols, Dissolved Iron, Fluoride, Magnesium, Manganese, Mercury, Nickel, Nitrate, Nitrite, Potassium, Silver, Selenium, Sulfide, Sodium, Zinc, as well as Volatile Organic Compounds (VOCs = 1,1-dichloroethene cis-1,2-dichloroethene; trans-1,2-dichloroethene; benzene; chloroform; styrene; ethylbenzene; (o, m, p)-xylenes; carbon tetrachloride; toluene; 1,1,2-Trichloroethene).

2.5. Contamination potential of the LL and pollutant removal capacity

The Leachate Pollution Index (LPI) was determined for both raw and treated LL, as recommended by Kumar and Alappat, according to the equation 1:[38]

$$\mathrm{LPI} = \sum_{i=1}^{n} wipi \tag{1}$$

where, wi = is the weight of the i^{th} polluting variable; pi = is the value ofthe sub-index value of the I^{th} leachate pollutant variable; n = number of variables used in the calculation of the LPI.

In addition, the Pollutant Removal Efficiency (E%) of the treated LL was evaluated using the methodology proposed by Almeida *et al.* expressed by equation 2:[39]

$$\mathrm{E}(\%) = \left(\frac{\mathrm{C_0} - \mathrm{C}}{\mathrm{C_0}}\right) \times 100 \tag{2}$$

where, C_0= is the concentration of the pollutant in the raw LL and, C = is the concentration of the pollutant in treated LL.

2.6. Quality and analytical control

For all the parameters evaluated, the following quality controls were used: analysis blanks and samples fortified with a known concentration of the analyte of interest (spike). For volatile organic compounds (VOCs), we also used the method of tracer analysis (surrogate), which consists in adding a substance of known concentration whose chromatographic behavior is similar to the compounds under analysis but is not present in the sample.

2.6.1. Limits of detection and quantification

The limits of detection (LOD) and quantification (LOQ) were calculated by equations 3 and 4:

$$\mathrm{LOD} = 3.3\frac{sd}{b} \tag{3}$$

$$\mathrm{LOQ} = 10\frac{sd}{b} \tag{4}$$

where, *s* = the standard deviation of the response; *b* = the slope of the calibration curve.

2.6.2. Sample variability

The extent of the variability of the results obtained for the different analytes in the different sampling campaigns was determined by estimating precision at the reproducibility level.[40,41] Reproducibility was obtained by using the estimated Relative Standard Deviation - RSD (%):[40]

$$\mathrm{RSD}(\%) = 100\,\frac{sd}{x} \quad (5)$$

where, sd = the sample standard deviation; x = sample mean.

For the analysis of environmental samples, reproducibility is considered acceptable when the RSD(%) ≤ 25.00%, as recommended by Weber and Juanico when evaluating the treatment efficiency of different parameters of the Kishon Complex, a large (15 Mm^3 $year^{-1}$) agricultural irrigation STP in Israel.[40]

2.7. Statistical analysis

The results obtained were organized and analyzed using the software Statistica® version 12.0 and presented as the arithmetic mean, standard deviation (SD), and RSD(%). The student's t-test was performed at a 95% confidence level to verify the existence of statistically significant differences between the mean results obtained for some parameters of the samples of the raw LL and the treated LL.

3. Results and Discussion

3.1. Figures of merit

The respective values of the LODs and LOQs of all analytes evaluated by the different analytical techniques used are shown in Table 1S in the Supplementary Material (SM). It can be observed that all the LODs and LOQs readily met the recommendations in current federal legislation.[36] Furthermore, all the parameters were identified in the samples of raw or treated LL, except mercury; 1,1-dichloroethene; cis-1,2-dichloroethene; trans-1,2-dichloroethene and 1,1,2-Trichloroethene.

3.2. Characterization and Biodegradability of LL during COVID-19 pandemic

As can be observed in Table 1, the pH values of the LL (raw and treated) of the CJSL were slightly alkaline (pH ≈ 8.0) and statistically similar to each other, according to Student's t-test ($p < 0.05$). Furthermore, the predominance of the methanogenic phase of microbiological degradation was evidenced, which is fully consistent with the operating time of the CJSL (≈ 9 years) and the long recirculation period of the concentrated effluent obtained after LL treatment by RO (≥ 5 years). [21,24,29-31] The CJSL started receiving more than twice as much USW during the COVID-19 pandemic, as an indirect impact of the implementation of NPIs for the containment of the SARS-CoV-2 virus,[42] but this was not sufficient for a significant change in the predominant phase of biological activity. It appears that the LL from the newly disposed of USWs in the operations plaza installed on top of the CJSL, due to the Lockdown and other NPIs, is subjected to aerobic biogeochemical processes only superficially, that once being leached in depth will be subjected to anoxic conditions prevailing along with the waste mass, in which the methanogenic archaea will consume the carboxylic acids generated by acidogenic bacteria and, associated with the constant recirculation of the concentrate, will favor the elevation of the pH of the raw LL.

Slightly alkaline pH values were also obtained in young landfills of two cities in the Metropolitan Region of Rio de Janeiro (MRRJ) that have the same climatic classification as the CJSL (Aw) and treat their respective LL with RO technology supplied by the same manufacturer as in this study.[24,39] Soares *et al.*,[24] when evaluating the efficiency of RO to treat raw LL from the landfill in São Gonçalo (2,500 ton day^{-1} of USW; 120 m^3 day^{-1} of LL and < 5 years of operation) and Almeida *et al.*,[39] when evaluating the technical efficiency of RO for the treatment of raw LL from the largest landfill in Rio de Janeiro State (Seropédica: 11,000 ton day^{-1} of USW; 1,000 m^3 day-1 of LL and > 5 years of operation) identified that recirculation of concentrated LL back to the landfill waste mass, as observed in this study, provides a shortening of the acidogenic degradation phase and promotes, prematurely, the emergence of a more accelerated biological stabilization in young or intermediate stage landfills. On the other hand, the *El-Hammam* Landfill located in Alexandria, Egypt (2,700 ton day^{-1} USW; > 18 years of operation and Köppen-Geiger: Cfb);[43] the Curitiba Landfill, Brazil (1,500 ton day^{-1} USW; 600 m^3 day^{-1} LL; > 11 years of operation and Köppen-Geiger: Cfb)[44] and the *Dhapa* Landfill in Kolkata, India (3,000 ton day^{-1} USW; 400 m^3 day^{-1} LL; > 11 years of operation and Köppen-Geiger: Aw)[45] corroborate the tendency for old landfills (> 10 years) to have pH > 7.00; regardless of USW input, climate classification, or international geographic location.

There was a wide variation in the concentrations of the parameters evaluated in Table 1; ranging from < LOD for the SS of the treated LL to 17,744.21 mL L^{-1} for TDS of the raw LL, respectively. Only the concentration of surfactants in the raw LL (2.07±2.54 mg L^{-1}) exceeded, by 7%, the MPC recommended by the federal environmental legislation.[36] This may be a significant indication that NPIs indirectly provided an increase in detergent consumption in the homes of families in Campos dos Goytacazes, as it became an extremely adopted habit by the Brazilian population during the COVID-19 pandemic to massively sanitize supermarket shopping packages with alcohol gel and/or household detergents, with subsequent regular disposal of these packages to landfills. Moreover, although the concentrations of oils and grease (VOAF, MO, and TOG) did not exceed the respective MPCs, they were 11 times higher than those found in a landfill in Curitiba,[44] and 140% higher, on average, than the results found in a landfill in MRRJ.[24] This corroborates, even if indirectly, that there was

Table 1. Physical, physicochemical, and chemical characterization of the treated and raw LL

Parameters	Unit	Landfill Leachate				E(%)	MPC
		Raw		Treated			
		Mean Values ± SD	RSD(%)	Mean Values± SD	RSD(%)		
pH		8.11±0.08	0.96	8.14±0.30	4.11	------	5 ≤pH≤ 9
K	(mS cm^{-1})	24.39±18.38	*75.37*	0.193±0.871	*44.90*	99.27	------
T	(ºC)	36.10±5.28	14.60	28.90±2.40	8.41	------	40.0
TC	(mg PtCo L^{-1})	3,086.67±2,046.86	*66.31*	1.33±2.30	*173.21*	99.96	------
TS	(mL L^{-1})	17,744.21±2,102.80	11.85	45.33±39.30	*86.71*	99.74	R ≥ 20%
TDS	(mg L^{-1})	17,544.33±2,305.94	13.14	19.67±34.10	*173.21*	99.89	------
SS	(mg L^{-1})	0.27±0.15	*57.28*	< LOD		≈100	1.0
TA	(mg L^{-1})	1,2261.33±1,591.62	12.98	80.67±23.41	*28.90*	99.98	------
TH	(mg L^{-1})	1,094.67±167.44	15.30	5.33±9.21	*173.21*	99.57	
VOAF	(mg L^{-1})	42.86±59.89	*139.73*				50.0
MO	(mg L^{-1})	17.13±14.85	*86.70*	< LOD		≈100	20.0
TOG	(mg L^{-1})	60.99±66.67	*109.31*				------
Surfactants	(mg L^{-1})	**2.07±2.54**	*122.94*				2.0

Electrical Conductivity (k), Temperature (T), True Color (TC), Total Solids (TS), Dissolved Solids (TDS), Sedimentable Solids (SS), Total Alkalinity (TA), Total Hardness (TH), Vegetable Oils and Animal Fats (VOAF), Mineral Oils (MO), Total Oils and Greases (TOG). In **Bold** are the results that exceeded the respective MPCs of CONAMA Resolution 430/2011. In *italic* are the RSD(%) > 25.00%.

a change in the food consumption pattern of the Brazilian population during the pandemic of COVID-19 because during the Lockdown and other NPIs it was observed that a significant fraction of the population started to eat lunch exclusively at home, thus consuming even more vegetable oil and animal fat for food production.

The extremely dark coloration of the LL ([True Color] > 3,000 mg PtCo L^{-1}), as well as the high values presented by k, TH, TA, and solids (TS, TDS, and SS) indicate a strong presence of chromophore substances (humic substances and/or clay-mineral colloids),[44] hydroxides and highly soluble inorganic salts, accelerated dissolution of organic and inorganic solids present in the USWs,[23] as well as desorption of cations and anions present in the mineral matrix of the CJSL USWs topsoil (Table 2). [24,39] Generally, high concentrations of solids are preferentially seen in young landfills, but it cannot be ruled out that during the COVID-19 pandemic the extra input of USW rich in the organic matter provided an increase in these parameters, which were about 21% above the typical range observed for Brazilian MSWs in a pre-Pandemic COVID-19 period.[44]

The nitrogenous nutrients, P and organic matter (BOD_5 and COD) contained in the LL showed a wide range; ranging from 0.01 for NO_2^- of the treated LL to 3,050.00 mg L^{-1} for N_T of the raw LL, respectively (Table 2). The highly toxic character of the raw LL is further reinforced when it is observed that P and N_A exceeded their respective MPCs by 18 and 104 times, respectively. The total phosphorus contents of this study were 10% higher than those presented by a sanitary landfill in Paraná[44] and complement what was previously stated: the extra input of USWs provided the biological degradation of newly deposited organic matter

Table 2. Characterization of the nutrient contents present in treated and raw LL

Parameters	Unit	Landfill Leachate				E(%)	MPC
		Raw		Treated			
		Mean Values ± SD	RSD(%)	Mean Values ± SD	RSD(%)		
P	(mg L^{-1})	**18.97±4.93**	*25.99*	0.015±0.01	*37.55*	99.92	1.0
NO_2^-	(mg L^{-1})	0.49±0.09	18.37	0.01±0.01	*173.21*	97.96	------
NO_3^-	(mg L^{-1})	0.98±0.15	15.31	0.40±0.3	*173.21*	55.10	
N_A	(mg L^{-1})	**2,106.60±1,692.52**	*80.34*	10.92±8.2	*74.67*	99.49	20.0
N_T	(mg L^{-1})	3,050.00±841.13	*27.58*	21.53±4.2	19.62	99.29	------
BOD_5	(mg L^{-1})	1,404.54±437.06	*31.12*	3.77±3.5	*92.62*	99.73	R ≥ 60%
COD	(mg L^{-1})	8,753.33±2,576.54	*29.43*	13.00±8.7	*67.11*	99.85	------
BOD_5/COD		0.18±0.11	*59.00*	0.32±0.41	*124.83*	------	

Total Phosphorus (P), Ammoniacal Nitrogen ($N_A = NH_3 + NH_4^+$), Total Nitrogen (N_T), Biochemical Oxygen Demand (BOD), and Chemical Oxygen Demand (COD). In **Bold** are the results that exceeded the respective MPCs of CONAMA Resolution 430/2011. In *italics* are the RSD(%) > 25.00%.

containing phospholipids and phosphoproteins (food scraps),[45] as well as the increased use of powder detergents due to the adoption of NPIs significantly impacted the composition of the raw LL by changing some parameters (P and Surfactants). As expected, N_A (the most toxic species of nitrogen)[23,44] was the predominant in the raw LL, due to the aerobic and anaerobic decomposition of USWs[45] and having typical and comparable results to those presented by several MSWs predominant in the methanogenic phase, in intermediate and mature stages in Brazil[21,24,32,39,44] and the world.[29-31,33-35,38,43]

Sanitary Landfills that recirculate concentrated LL from ROs show a great acceleration in the degradation of biodegradable organic constituents by hydrolysis and fermentation of nitrogenous substrates (amino acids, polypeptides, etc.) from USWs,[32,39] leading to a strong increase in N_A concentrations when compared to MSWs that do not recirculate concentrated LL.[23,30] In addition, it should be considered that the significant extra input of USWs resulting from the adoption of NPIs provides even more fresh organic matter, which once exposed to the anaerobic conditions of the sub-surface layers of USWs, buried under the topsoil, will prevent an eventual conversion by oxidation of NH_3 to NO_3^- or NO_2^-,[30,32] as can be evidenced in Table 2. N_A is hardly degraded by the anaerobic microbial organisms typically present in landfills.[23,32] Therefore, the tendency is for a considerable increase in the concentration of $N_A$at the raw LL, while the CJSL is receiving the extra input of USWs rich in animal and vegetable proteins (food scraps), as an indirect consequence of the adoption of NPIs and is recirculating the concentrated LL from the RO. Finally, the higher the concentration of N_A in the LL, the greater the buffer effect on pH and the greater the ionic contribution to the *K* of the fresh LL, keeping it alkaline and electrically charged due to the presence of the NH_4^+ cation.[23]

As can be seen in Table 2, the parameters that characterize organic matter in the raw LL were extremely high: 1,404.54 mg L^{-1} for BOD and 8,753.33 mg L^{-1} for COD, respectively. These results indicate a USW composition highly enriched by organic matter and are very similar to those presented by two young MSWs in MRRJ that treat LL by RO and recirculate the treated LL,[24,39] as well the LL from an old MSW in Paraná.[44] The high concentration of COD and the dark coloration of the raw LL (Table 1) indicate that the organic matter is preferentialrecalcitrant and with high molecular weight (humic acids and fulvic acids).[23,32] Although the BOD presents a lower concentration, it should not be considered negligible, on the contrary, because it presents a relatively high value, indicating that there is still biodegradable organic matter (precursor of carboxylic acids by acetogenic bacteria) in a considerable amount.[32,44] Generally, intermediate and old MSWs that are in the methanogenic phase and recirculate concentrated RO LL tend to present lower BOD than COD.[23,31,38,43] In addition to the parameters characterizing the Organic Matter, a low BOD/COD ratio (0.18±0.11) was observed; indicating moderate biodegradability.[23,28-34,38,43] It appears that the CJSL is rapidly evolving towards high biological stability (BOD/COD < 0.1), in which the organic matter in the LL will be practical of humified and recalcitrant nature due mainly to the recirculation of concentrated LL from the RO.[23,31-33,39]

3.3. LL toxicity during the COVID-19 pandemic

As can be seen in Table 3, the Volatile Organic Compounds (VOCs) ranged from < LOD for chloroform from the treated LL to 105.00 mg L^{-1} for Toluene from the fresh LL. In addition, it was identified that all the VOC present in the fresh LL exhibited high toxicity as they exceeded their respective MPCs,[23,36] with chloroform exceeding them by 11% and toluene by 87.5 times, respectively. VOCs are also known as low molecular weight xenobiotic organic substances, are highly toxic and carcinogenic, and have industrial and domestic origins (solvents, paint removers, cleaning products, degreasers, varnishes, detergents, etc.).[23,46] The city of Campos dos Goytacazes is strongly inserted in Brazil's petroleum production chain, with numerous refining industries. Therefore, although the CJSL is only licensed to receive USW (Class II - Non-hazardous),[19,26] it may have received or be receiving significant quantities of VOC-rich waste (Class I - Hazardous) by individual household contributions or by irregular disposal from commercial or industrial

Table 3. Characterization of xenobiotic organic compounds present in treated and raw LL

Parameters	Unit	Landfill Leachate				E(%)	MPC
		Raw		Tratado			
		Mean Values ±SD	RSD(%)	Mean Values ±SD	RSD(%)		
Chloroform	(mg L^{-1})	**1.11±2.00**	*200.00*				1.0
Styrene	(mg L^{-1})	**15.00±2.51**	16.80				0.07
Ethylbenzene	(mg L^{-1})	**68.00±6,21**	9.20	< LOD		≈100	0.84
Toluene	(mg L^{-1})	**105.00±11.30**	10.80				1.2
(o,m,p)-Xylenes	(mg L^{-1})	**83.00±8.00**	9.60				1.6
$Phenols_{(Total)}$	(mg L^{-1})	**6.32±7.84**	*124.07*	0.08±0.078	*93.23*	98.73	0.5

In **Bold** are the results that exceeded the respective MPCs of CONAMA Resolution 430/2011. In *italics* are the RSD(%) > 25.00%

establishments, as observed in the United States of America.[46]

As can be seen in Table 3, the RO installed at the CJS has proven the effectiveness of this treatment technique in completely or partially removing these xenobiotic substances from the raw LL.[23,46] However, these substances are not degraded and will still be present in the concentrated OR LL, which once recirculated in the CJSL will be hyper-concentrated in the raw LL and will increase the concentration of COD, because although, naturally, MSWs increase the humification of dissolved organic matter over time; still, a significant fraction of this COD is of synthetic aromatic compounds, phenolics among others.[23] Soares *et al.*,[24] identified that the total phenols parameter was the only one that did not fit in the federal legislation[36] when evaluating the treatment of LL in an RO installed in the MRRJ.

The inorganic macro-components ranged from < LD for Ba in the treated LL to 4,700.00 mg L^{-1} for Cl^- in the raw LL (Table 4). Generally, raw LL from MSWs situated in the methanogenic phase presents low concentrations of potentially toxic metals as a result of sorption reactions, precipitation at alkaline pH,[23] and association with different geochemical fractions (hydroxides, carbonates, and sulfides) of the cover soil.[45] The high Cl^- concentrations would justify the high TDS and COD results found in the raw LL, but it seems that SO_4^{2-} would be due only to the composition of the USWs or the organic matter present in the topsoil,[23,45] and that were indirectly increased due to the implementation of the NPIs. Furthermore, the high concentrations of Na^+, K^+, Ca^{2+}, and Mg^{2+} justify the high values obtained for TH in the raw LL, because alkaline and alkaline earth cations constitute the exchangeable bases of soils and are easily leachable by rainwater.[23] If the degradation of the anions [Cl^-] and [SO_4^{2-}] or the cations [Na^+], [K^+], [Ca^{2+}] and [Mg^{2+}] by RO is not possible, they will present a conservative character in which the concentrated LL will accumulate high concentrations of these ions providing a K > 0.100 (mS cm^{-1}) when recirculated in the CJSL and thus further increasing the concentrations of these cations and anions in the raw LL.[23]

Table 4. Characterization of anions and potentially toxic elements present in treated and raw LL

Parameters	Unit	Landfill Leachate				E(%)	MPC
		Raw		Treated			
		Mean Values ± SD	RSD(%)	Mean Values ±SD	RSD(%)		
Cl^-	(mg L^{-1})	4,700.00±676.4	14.40	1.83±0.3	15.75	99.96	-------
$CN^-_{(Total)}$	(mg L^{-1})	**4.29±5.83**	*136.07*	0.02±0.01	*94.37*	99.53	0.2
$CN^-_{(Free)}$	(mg L^{-1})	**2.98±4.43**	*148.48*				1.0
F^-	(mg L^{-1})	0.57±0.9	*87.05*	< LOD		≈100	10.0
SO_4^{2-}	(mg L^{-1})	63.33±35.12	*55.45*				-------
S^{2-}	(mg L^{-1})	**1.27±0.87**	*68.98*	0.01±0.01	*173.21*	99.21	1.0
Al	(mg L^{-1})	1.47±0.95	*64.67*	0.047±0.001	*30.91*	96.80	-------
As	(mg L^{-1})	0.05±0.04	*86.61*	0.002±0.001	*86.94*	96.00	0.5
B	(mg L^{-1})	2.33±2.18	*93.57*	0.80±0.1	11.14	65.66	5.0
Ba	(mg L^{-1})	0.13±0.05	*40.46*	< LOD		≈100	5.0
Ca	(mg L^{-1})	70.17±29.69	*42.31*	0.4±0.1	*36.83*	99.45	-------
Co	(mg L^{-1})	0.04±0.03	*87.08*				-------
Cr^{3+}	(mg L^{-1})	0.11±0.19	*173.21*	< LOD		≈100	1.0
Cr^{6+}	(mg L^{-1})	0.08±0.14	*173.21*				1.0
$Cr_{(Total)}$	(mg L^{-1})	0.24±0.20	*84.41*				1.0
$Cu_{(Dissolved)}$	(mg L^{-1})	0.007±0.01	*105.19*	0.001±0.001	*173.21*	89.71	1.0
$Cu_{(Total)}$	(mg L^{-1})	0.02±0.01	*36.98*	0.005±0.001	*173.21*	97.51	-------
Fe	(mg L^{-1})	1.787±1.56	*87.21*	0.065±0.1	*122.701*	96.6	15.0
K	(mg L^{-1})	1,351.03±1,108.6	*82.06*	8.18±4.6	*56.59*	99.39	-------
Mg	(mg L^{-1})	99.29±73.84	*74.37*	0.14±0.1	*93.67*	99.86	
Mn	(mg L^{-1})	0.16±0.03	18.28	0.005±0.01	*173.21*	96.88	1.0
Na	(mg L^{-1})	1,595.6±1,122.35	*70.34*	6.56±3.7	*56.24*	99.59	-------
Ni	(mg L^{-1})	0.18±0,16	*86.76*				2.0
Pb	(mg L^{-1})	0.02±0,03	*173.21*	< LOD		≈100	0.5
Sn	(mg L^{-1})	0.06±0.05	*86.62*				4.0
Zn	(mg L^{-1})	0.55±0.35	*63.59*	0.053±0.01	*31.33*	90.36	5.0

In **Bold** are the results that exceeded the respective MPCs of CONAMA Resolution 430/2011. In *italics* are the RSD(%) > 25.00%.

Although the F^- anion presented low concentration (0.57±0.49 mg L^{-1}) in the fresh USW, the same cannot be said of the S^{2-} and CN^- anions, which varied between 1.27 and 4.29 mg L^{-1}, thus exceeding the respective MPCs by 27% and 21.45 times. The presence of SO_4^{2-} in the USWs or the organic matter of the covering soil provides a substrate for sulfate-reducing archaea present in the deeper layers of the USWs, in an anaerobic environment, to reduce sulfur to S^{2-}.[30]

The phenomenon called the "Sulfide Barrier" affects the concentration of the other metals present in the fresh LL, due to the formation of metal sulfides with the low constants of the solubility product (K_{sp}). In addition, the alkaline pH (pH ≈ 8.0) will provide the precipitation of these metals as hydroxides, as well as when recirculated in the concentrated LL they may be adsorbed by the highly humified organic matter, as corroborated by the high [COD] contents. The high concentration of Cl^- present in fresh LL, is to make metal complexation the predominant mechanism.[30] Soares *et al*.,[24] also found low concentrations of metals in raw LL and found that all parameters of treated LL met the MPCs of the federal legislation, as in this study. Calabró *et al.*,[30] also found low concentrations of cations and anions in the LL of MSW*Il Fossetto* located in Italy, where the raw LL is treated by RO and the concentrated LL is recirculated to the landfill waste mass.

3.4. Impact of the COVID-19 pandemic on the LL composition variability

Tables 1, 2, 3, and 4 present the average RSD(%) of each parameter evaluated by type of LL (raw and treated) in the CJSL. It was observed that the raw LL presented almost ¾ of the evaluated parameters with high variability (RSD(%) > 25.00%), and the remaining results with low variability (RSD(%) ≤ 25.00%). On the other hand, the treated LL showed the following distribution regarding the variability of the parameters evaluated: 51% of the parameters with RSD(%) > 25.00%; 20% of the parameters with RSD(%) ≤ 25.00% and 29% of the parameters with results below the LOD, respectively. As can be seen in Figure 1, the periods of collection of samples of the treated in raw LL were purposely chosen in the dry season, considering the months with close rainfall (average rainfall of sampling campaigns = 60.33 ± 14.04 mm and RSD(%) = 23.28%), This would eliminate the well-known effect of seasonality that strongly influences the variability of the composition of the LL and would evaluate the weather station in which the pollutants are more concentrated in solution, constituting a "worst-case scenario" to be researched. Therefore, it would be expected that the variability of the parameters in the composition of the evaluated LL would be low, due to the similar meteorological conditions of the different sampling campaigns in the dry season (Figure 1). However, according to the results obtained in this study one can justify some hypotheses for the high variability trends identified:

a) The concentrations of environmental parameters obtained in the raw LAS presented high variability as an indirect consequence of the NPIs adopted by the State of Rio de Janeiro and the municipality to contain the spread of the SARS-CoV-2 Coronavirus in Campos dos Goytacazes during the Lockdown period.
b) During the first year of the COVID-19 Pandemic, the city of Campos dos Goytacazes saw its USWs generation double, from 20,000 to 40,000 tons per month^{-1}, which is equivalent to 1,333 ton day^{-1} of USW collected in the municipality and sent to the CJSL.[42] Obviously, the variability in the concentrations of pollutants in the fresh USWs reflects the unexpected increase in the input of USWs destined for the CJSL, added to the fact that it is a USW with an increased volume of plastic packaging, food scraps, and inadequately disposed of USWs.[1,5,14,15] Thus, with an atypical gravimetric composition of the garbage, when compared to the pre-pandemic period of COVID-19.[16,17]
c) The first sampling campaign was carried out in the pre-pandemic period of COVID-19, the second campaign was carried out after the first decree issued by the State of Rio de Janeiro that stipulated Lockdown in the state as NPI, but already under the first decree that relaxed some restrictions of the state Lockdown; in turn, the third sampling campaign was carried out after the second decree that further relaxed the state Lockdown, but did not terminate it under any circumstances.[13]
d) NPIs had a heterogeneous socio-economic impact throughout the different Lockdown times in the 92 municipalities of Rio de Janeiro State, entailing severe and varied changes in the volumetry and gravimetric composition of the USW treated throughout 2020.[1,16,24]
e) The concentrations of treated LL were very low, with 16 parameters below the LOD. Thus, unlike the fresh LL, which naturally presents extremely high concentrations for most parameters, any small variation in the concentration of analytes in the treated LAS can be extremely significant for the composition of theRSD(%), given the low concentrations obtained after treatment by RO.

High variability in several parameters of LL raw was also identified by Soares *et al.*,[24] when evaluating a landfill in São Gonçalo (RJ) that also treats the LL raw with RO technology, Model "AST System OR-120" manufactured and supplied by the company AST "*Soluções e Serviços Ambientais*" (same manufacturer of this study). On the other hand, the same was not observed by Almeida *et al.*,[39] when evaluating the raw LL to be treated, also by RO technology, Model "AST System OR-1000" manufactured and supplied by the company AST "*Soluções e Serviços Ambientais*", in the largest landfill in the state of Rio de Janeiro, located in the municipality of Seropédica. Both landfills cited above also have the same Köppen-Geiger climate classification of the CJSL (Aw) but had their collection campaigns performed in different climatic periods (dry and rainy seasons) before the

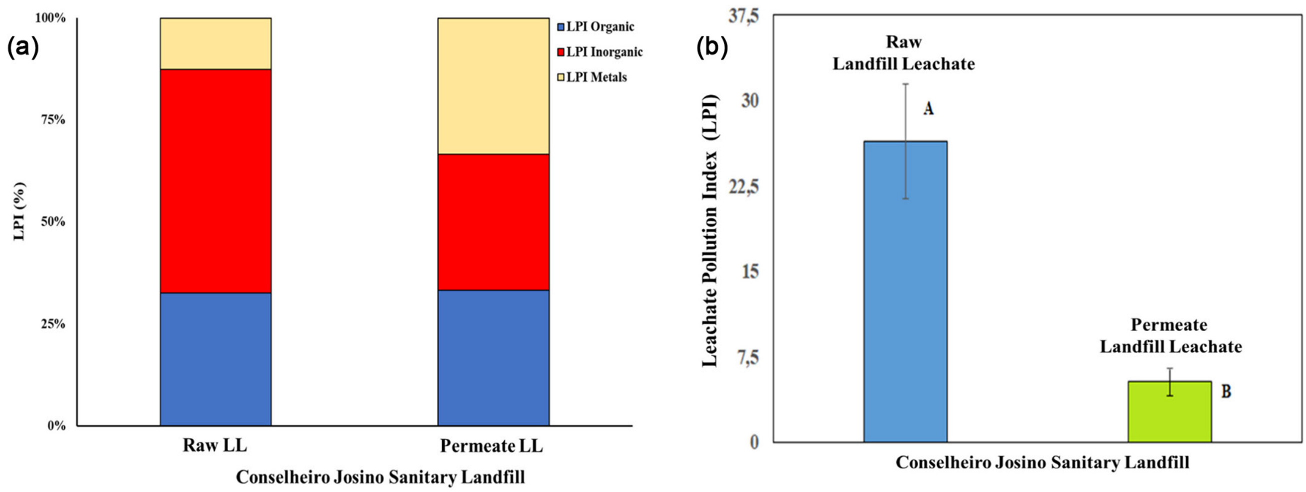

Figure 3. The potential comparison of landfill leachate contamination in Campos dos Goytacazes (RJ) before and after reverse osmosis treatment during the first year of the COVID-19 pandemic. a) Composition (%) of the CJSL Leachate Pollution Index; b) CJSL Global Leachate Pollution Index. Different capital letters indicate a significant difference (P < 0.05) by Student's t-test.

COVID-19 pandemic. [24,27,38] Furthermore, high variability of raw LL was also found a landfill located in Alexandria (Egypt)[43] and in a landfill in Curitiba (PR),[44] which unlike the CJSL had a different Köppen-Geige climate classification (Cfb) and had collections over different rainfall periods (dry and rainy seasons). Thus, regardless of the climatic classification of the landfill, it is to be expected that the variability in the parameters evaluated is mainly due to seasonal campaigns of sampling performed in antagonistic seasons of the year (dry and rainy seasons), which is not the case of the study in question, which presents high variability in the parameters evaluated in the rawLL due to the indirect impacts of the NPIs adopted by the government of the State of Rio de Janeiro and by the municipality of Campos dos Goytacazes.

3.5. Technical and environmental efficiency of reverse osmosis during the COVID-19 pandemic

When employing RO technology for the treatment of raw LL at CJSL, high rates of Pollutant Removal Efficiencies (E(%) > 90%) were observed in approximately 94% of the cases studied, and extremely high Pollutant Removal Efficiencies (E(%) > 99%) in 80% of the parameters that could be evaluated (Tables 1, 2, 3 and 4). Therefore, the high variability in the composition of raw LL, resulting from indirect socio-environmental impacts caused by the Lockdown and other NPIs required by the government of the State of Rio de Janeiro and the municipality of Campos dos Goytacazes, indirectly resulted in a significant increase of 140% of USWs sent to the CJSL[40] and changes in the composition of the raw LL, was not sufficient to reduce the efficiency of the treatment capacity of raw LL by RO technology at CJSL. Soares *et al.*,[24] also identified E(%) > 90% in many parameters when evaluating a landfill in São Gonçalo operating since 2012. Almeida and colleagues,[39] showed that RO can achieve E(%) > 90% for some parameters in the landfill of Seropédica (RJ) that operates since 2011.

Kumar and Alappat[38] developed the Leachate Pollution Index (LPI) with the initial intention of classifying the hazardousness of raw LL from landfills.[46] To do so, they used three subgroups of pollutants: Organic LPI, Inorganic LPI, and Metals LPI. However, LPI has also come to be adopted as a quality control criterion for different treatments of raw LL.[38,46] As can be observed in Figure 3.a, Inorganic LPI accounted for 54.7% of the Overall LPI due to the expressive contribution of N_A, while Organic LPI accounted for 32.6% of the Overall LPI mainly due to COD and Metals LPI accounted for only 12.6% in the composition of the Overall LPI, respectively. Furthermore, it was observed that LPI of the RO treated LL showed equal contributions (33%) of the three sub-indices (LPI Organic: 5.0; LPI Inorganic: 5.0 and LPI Metals: 5.0), thereby fully meeting the arbitrated LPI values for discharge of treated LL into surface waters (LPI Organic: 7.03; LPI Inorganic: 6.57 and LPI Metals: 7.89).[46,38] When evaluating the Overall LPI (Figure 3.b), it was observed that the raw LL from CJSL presents a 420% higher pollution potential (Student's t-test, p<0.05) than that of RO-treated LL. However, it still exhibits a Global LPI 56% lower than that of the raw LLfrom MSW from Kolkata, India.[46]

4. Conclusions

In an unprecedented disruptive moment, as an indirect consequence of the implementation of the Non-Pharmacological Intervention Measures for the reduction of SARS-CoV-2 transmission levels, the Campos dos Goytacazes Sanitary Landfill started receiving approximately 1,333.00 ton day^{-1} of Urban Solid Waste, a significant increase of almost 140% in the daily average registered in all its nine years of operation. Consequently, if this new daily average of waste receipt is maintained in the post-Lockdown period (during the pandemic of COVID-19), only the adoption of the Non-Pharmacological Intervention measures would have decreased the forecast of the remaining useful life of the Conselheiro Josino Sanitary Landfill from 21 to 19 years.

The Conselheiro Josino Sanitary Landfill is in the Methanogenic Phase of microbiological degradation, with the pH of the raw leachate slightly alkaline (pH ≈ 8.0) being buffered by high concentrations of ammoniacal nitrogen (> 2,000.00 mg L^{-1}), as well as presenting dark coloration (> 3,000.00 mg PtCo L^{-1}), high concentrations of dissolved organic matter (BOD > 1,000.00 mg L^{-1} and COD > 8,000.00 mg L^{-1}) and several parameters above the respective maximum permissible values of the federal legislation (Surfactants, Total Phosphorus, Ammoniacal Nitrogen, Volatile Organic Compounds, CN^- and S_2^-). In addition, the raw leachate presents high electrical conductivity due to the high concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+},Cl^- e SO_4^{2-}. It appears that the toxicological characteristics of this leachate are affected by the recirculation of the concentrated leachate from the Reverse Osmosis, which is recirculated to the waste mass, as well as the significant extra input of Municipal Solid Waste indirectly coming from the adoption of Non-Pharmacological Intervention measures during the pandemic of COVID-19.

The change in consumption and hygiene habits adopted by the Brazilian population as a response to Non-Pharmacological Intervention measures, with special emphasis on Lockdown, has boosted the consumption of detergents and other cleaning materials, which directly affects the concentrations of surfactants and total phosphorus present in the raw leachate. In addition, a greater consumption of food produces a larger amount of waste rich in organic matter that once disposed of in landfills will increase the concentrations of phosphorus and dissolved organic matter (BOD and COD). However, in the period evaluated, the Reverse Osmosis presented itself as efficient in treating the rawleachate, causing all parameters evaluated in the treated leachate to be below the respective maximum permissible values of the Brazilian federal legislation, according to Resolution CONAMA 430/2011.

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