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Evaluating the Operation of a Full-Scale Sequencing Batch Reactor–Reverse Osmosis–Evaporation System Used to Treat Landfill Leachates: Removal of Pollutants, Energy Consumption and Greenhouse Gas Emissions

Konstantinos Tsompanoglou [1,2], Olga P. Koutsou [2] and Athanasios S. Stasinakis [2,*]

1 Regional Association of Solid Waste Management Agencies of Central Macedonia, Fragon 6-8, 54626 Thessaloniki, Greece; envm22045@env.aegean.gr
2 Department of Environment, University of the Aegean, 81100 Mytilene, Greece; koutsou@env.aegean.gr
* Correspondence: astas@env.aegean.gr; Tel.: +30-2251036257

Abstract: Limited information is available in the literature regarding the energy consumption and the greenhouse gases emitted during landfill leachates treatment. A full-scale landfill leachates treatment system that included primary sedimentation, biological treatment in sequencing batch reactors, reverse osmosis and mechanical vapor recompression evaporation was monitored and evaluated for the removal of major pollutants, energy consumption and greenhouse gas emissions. Samples were taken during a period of two years from different points of the system, while the actual power consumption was calculated considering the available mechanical equipment and the hours of operation. The quantities of greenhouse gases emitted were estimated using appropriate equations and based on the operational characteristics of the system. According to chemical analyses, biological treatment resulted in partial removal of COD and total nitrogen, while the removal of BOD_5 and NH_4-N was significant, reaching 90 and 98%, respectively. Use of reverse osmosis increased the removal of all pollutants, satisfying the requirements of the legislation on wastewater discharge into the environment. Power consumption was calculated to be 35.3 KWhr per m^3 of treated leachate, while mechanical vapor recompression evaporation was responsible for 60.5% of the total energy required. The contribution of other processes to energy consumption was as follows, in decreasing order: sequencing batch reactors > reverse osmosis > primary treatment. The roots blower vacuum pump used for mechanical vapor recompression evaporation, and the blowers providing air to the sequencing batch reactors, were the most energy-intensive pieces of apparatus, contributing 44.2% and 11.3% of the required energy, respectively. The quantity of greenhouse gases emitted was estimated to be 27.7 Kg CO_{2eq} per m^3 of treated leachates. Among the different processes used, biological treatment and mechanical vapor recompression evaporation contributed to 45.7% and 44.1% of the total emissions, respectively. The findings of this study reveal that an integrated landfill leachate treatment system that combines biological treatment and reverse osmosis can assure the protection of the aquatic environment by producing high-quality effluent; however, further research should be conducted regarding the sustainable management of reverse osmosis concentrate. Mechanical vapor recompression evaporation contributes significantly to the environmental footprint of the landfill leachates treatment system due to both high energy consumption and elevated emissions of greenhouse gases.

Keywords: landfill; leachate treatment; energy consumption; GHGs emissions; reverse osmosis; SBR; mechanical vapor recompression evaporators

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1. Introduction

The transfer of municipal solid waste to landfills represents the most common practice for solid waste management in Greece [1]. In 2020, over 75% of the produced solid wastes were disposed in 82 landfills around the country [2]. Their landfilling produces high amounts of leachates, mainly due to the inherent water content of solid waste, rainwater percolation and the water produced during waste decomposition [3]. Landfill leachate is characterized as a saline and highly contaminated wastewater with a low ratio of biochemical oxygen demand (BOD_5) to chemical oxygen demand (COD), high levels of ammonium nitrogen (NH_4-N) and increased concentrations of various micropollutants such as pharmaceuticals, perfluorinated substances and personal care products [4,5].

Management of leachates is one of the most significant challenges to address during the design, construction and operation of a landfill site [6]. Conventional leachate treatment often includes recirculation of the produced leachate into the landfill body [7,8], co-treatment of leachates with municipal sewage in centralized Wastewater Treatment Plants [9] and on-site aerobic or anaerobic biological treatment [10,11]. In the case of on-site treatment, chemical or/and physical treatment processes such as coagulation–flocculation, chemical oxidation, hydrodynamic cavitation, carbon adsorption and use of membranes are sometimes combined with biological processes to achieve a high-quality effluent [12–18]. The treatment systems that are commonly used in the larger Greek landfills combine primary treatment, aerobic biological treatment in activated sludge bioreactors and reverse osmosis (RO) in series. Furthermore, in the landfills of the two largest Greek cities (Athens and Thessaloniki), MVR evaporators have also been installed for the treatment of RO concentrates. This process achieves important condensation of the RO concentrate which, due to its toxic nature, is typically recirculated to the landfill.

The energy consumption of the different processes should be a significant factor in selection of the methods to be applied, as it significantly affects the operating costs of a landfill site. It is widely known that biological treatment in activated sludge reactors is an energy-intensive process which requires amounts of energy ranging between 0.36 and 0.67 KWh/m^3 for municipal wastewater [19]. A major part of the required power is used for aeration of the mixed liquor in aerobic tanks [20]. As regards RO treatment, the predominant source of energy consumption is the high-pressure membrane process [21], while operation of the evaporators requires significant amounts of energy to heat the treated liquids [22]. Despite the importance of energy consumption in the operational costs of Leachate Treatment Plants (LTPs), limited relevant information based on full-scale systems is available in the literature. Di Maria et al. [23] studied the energy consumption of a full-scale LTP in Italy and estimated that for each m^3 of treated leachate, the MVR evaporator consumed 45.5 kWh of electricity. This amount corresponded to 65% of the total power consumption. Zhang et al. [24] studied the use of membrane bioreactors (MBRs) for landfill leachates treatment in China and concluded that the energy consumption ranged between 20 and 30 kWh per m^3. These levels of power consumption were much higher than those observed during municipal wastewater treatment, and this was due to the much higher concentrations of pollutants in leachates and the lower biodegradability of the organic matter.

In addition to the energy requirements, greenhouse gas (GHG) emissions are also an important matter in the treatment of heavily contaminated effluents, because they add to the environmental footprint of the LTP. Previous studies have reported that treatment of landfill leachates is a major source of N_2O and CH_4 [25,26]. However, most of the relevant published information originates from lab-scale or pilot-scale studies. Additionally, previous studies have typically focused on the GHGs emitted by biological processes, while the role of advanced treatment processes is usually ignored [27]. Specifically, Nuansawan et al. [28] studied N_2O and CH_4 emissions in a lab-scale MBR leachate treatment system and discussed the effect of sludge recirculation and hydraulic retention time (HRT) on the emitted gases. In other lab-scale studies, Boonnorat et al. [29] compared emissions of CO_2, N_2O and CH_4 in lab-scale activated sludge reactors, and Wang et al. [30] compared GHG emissions arising from the treatment of young and aged landfill leachate. Chiemchaisri et al. [27] reported information on N_2O and CH_4 emissions from a pilot-scale LTP consisting of an anoxic and an aerobic MBR. Wang et al. [25] measured the emissions of N_2O and CH_4

in three full-scale LTPs in China and estimated a total N_2O emission factor equal to 8.55 g N_2O-N per capita and year. Finally, Hua et al. [31], using IPCC guidelines, calculated GHG emissions from the treatment of heavily contaminated leachates, comparing a system that consisted of anaerobic lagoons and aerobic ponds with another where an up-flow anaerobic sludge semi-fixed filter was coupled with a membrane bioreactor.

Based on the above, the main objectives of this study were to analyze the performance of a full-scale LTP consisting of primary treatment, biological treatment, RO and MVR evaporation in series, and to calculate the power consumption of each treatment process. The on-site and off-site GHG emissions of different treatment processes were also estimated using appropriate equations and the carbon footprint of the system was discussed. The Mavrorachi Sanitary Landfill serving the Thessaloniki metropolitan area (Greece) was used as a case study, and was monitored for a period of two years.

2. Materials and Methods

2.1. Landfill Leachates Treatment Plant

Mavrorachi landfill is a sanitary landfill serving the metropolitan area of Thessaloniki. It has been operational since 2008, receiving on average 1000–1200 tons of solid waste per day. For the period of the study, the average daily production of leachates was approximately 350 m^3/d. The existing LTP (Figure 1) includes pretreatment (grit removal), primary treatment with no coagulant addition and biological treatment with nitrogen removal in four sequencing batch reactors (SBRs). Effluent from the SBRs is transferred to a discharge tank (Figure 2) where settling occurs, and the supernatant from this tank is introduced to the RO unit. The RO concentrate is condensed via MVR evaporation. The sludge from the primary treatment, the discharge tank and the excess sludge from the SBRs is thickened in a sludge tank.

(**a**) (**b**) (**c**)

Figure 1. Photographs of the studied Landfill Leachates Treatment Plant: (**a**) the four parallel SBR reactors, (**b**) the one-stage reverse osmosis unit with capacity equal to 350 m^3/d, (**c**) the two evaporators with total capacity equal to 120 m^3/d.

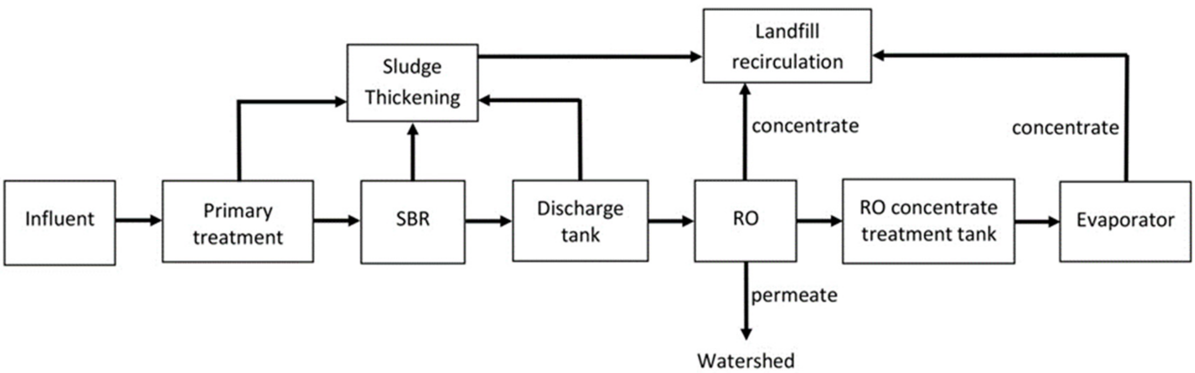

Figure 2. Flow chart of the studied Landfill Leachate Treatment Plant.

The final concentrate from the evaporators and the excess amounts of RO concentrate are reinjected into the landfill (Figure 2), while the RO permeate is discharged to a nearby stream. Further information on the operation of the SBRs, the RO and the evaporators is provided below.

For the period of the study, the four SBRs operated in parallel in an 8 h cycle and each reactor had a capacity of 2100 m^3. The hydraulic retention time (HRT) had been set to 17.5 days, while the sludge residence time (SRT) was practically infinite, as there was no discharge of excess sludge. The suspended solids concentration in the bioreactors was approximately 4000–4500 mg/L. During the 8 h operational cycle, aerobic and anoxic periods in the SBRs were alternated to achieve nitrogen removal through nitrification and denitrification. Because of the limited presence of biodegradable organic matter in the landfill leachates, glycerol, a by-product of biodiesel production, was added to the influents to enhance endogenous denitrification.

Concerning the RO, a one-stage RO unit was used. It was equipped with disc-tube modules that contained brackish water polyamide membranes. The secondary treated leachate was initially prefiltered through a sand filter and then passed through cartridge filters (nominal pore size equal to 10 μm). The capacity of the RO unit was 15.5 m^3/h, the operational pressure was between 30 and 40 bars and the recovery ratio ranged between 60 and 70%. As regards MVR evaporation, two evaporators (TC60000, Led Italia, Pordenone, Italy) were used, with a capacity of 120 m^3/day of pre-concentrated leachate. The pre-concentrated liquid could reach a concentrate of approximately 20% TS, which is a pumpable fluid. In these units, boiling temperature is maintained using thermal resistances, while pressure is maintained using a blower vacuum pump [23]. The nominal concentration ratio of the evaporators was 1:5.5, contributing to the leachate storage capacity of the landfill. The pH of the concentrate was adjusted with HCl and an antifoam agent was also used to prevent foaming.

2.2. Monitoring the System

The LTP was monitored for a period of two years. Samples were collected on a weekly basis from the following points of the system: influent, primary treatment, SBR effluent and RO permeate. The collected samples were analyzed for BOD_5, COD, total suspended solids (TSS), total nitrogen (TN), NH4-N and conductivity.

As regards energy consumption, the actual power consumption was calculated taking into account the electrical input power, the motor efficiency of all power-consuming devices in each treatment process and their hours of operation. The total power required by a three-phase motor consists of the real power and the reactive power (the non-working power caused by the magnetizing current). At the studied LTP, the power factor (the ratio between real power and apparent power) is enhanced by capacitors. Due to this correction technique, the power factor has been improved and the amount of apparent power has been reduced [32]. In the current study, the value of the power factor used in calculations of energy consumption is considered to be 0.90.

2.3. Analytical Methods

All the analyses of collected samples were performed according to standard methods [33]. For the analysis of TSS, an aliquot of the collected leachate samples was vacuum filtered through glass fiber filters (GF Type A/E, nominal pore size 1.0 μm, diameter 47 mm). Other aliquots of the samples were used for COD, BOD_5, NH_4-N, TN and conductivity measurements. Determination of the BOD_5 was undertaken using respirometric measurements (OxiTop® system, WTW, London, UK). The COD, TN and NH_4-N were determined with a Hach-Lange DR 3900 Spectrophotometer (Hach-Lange, Düsseldorf, Germany). Conductivity was measured using a Hach-Lange HQ440d conductivity meter (Hach-Lange, Düsseldorf, Germany).

2.4. Empirical Model for GHG Emissions

Off-site and on-site GHG emissions of the biological treatment were estimated based on the Bridle modelling methodology that had been applied in several previous studies [20,34–36]. The different processes that were considered for GHG emissions estimation and the equations used are presented in Table 1. Conversely, GHG emissions stemming from the tertiary treatment (RO evaporators) were estimated using an empirical equation which is also presented in Table 1.

Table 1. The processes used for the calculation of GHG emissions in the studied Landfill Leachates Treatment Plant and the relevant equations.

		Processes 1	Equations Used for GHG Emission Estimation ($kg\ CO_{2eq}/d$)
Biological treatment	On-site GHG emissions	Production of GHGs from biomass decay	$CO_{2,biomassdecay} = X_{decay} \times 1.947$ ($X_{decay} = Q \times HRT \times MLVSS \times b_H$)
		Production of GHGs from BOD oxidation and production of biomass	$CO_{2,BODoxidation} = R_{O2} \times 1.1$ ($R_{O2} = [f - \frac{K_e \times Y_H}{1 + b_H \times SRT_{total}}] \times E_H \times Q \times F_o$)
		Consumption of GHGs from nitrification	$CO_2 = 0.308 \times N_{nitro}$ ($N_{nitro} = N_{total} - N_{bio} - (NH_4\text{-}N)_{out} - N_{orgout}$)
		Production of GHGs from nitrification–denitrification	$N_2O_{emission} = N_{total} \times 0.005$ ($CO_{2,eq} = N_2O_{emission} \times GWP_{N2O} = N_2O_{emission} \times 296$)
	Off-site GHG emissions	Production of GHGs from net power consumption	$CO_{2,electricity} = E_{required} \times \Sigma(F_i \times EF_i)$
		Production of GHGs from treated leachates discharge to the aquatic environment	$N_2O_{emission} = N_{Effluent} \times EF_{Effluent} \times (44/28)$ ($CO_{2,eq} = N_2O_{emission} \times GWP_{N2O} = N_2O_{emission} \times 296$)
Tertiary treatment		Operation of the RO and evaporators	Emissions (CO_2, CH_4, N_2O) = Electrical energy consumption × emissions (CO_2, CH_4, N_2O) per electrical energy consumption × Q ($CO_{2,eq} = CO_2 + (28 \times CH_4) + (296 \times N_2O)$)

[1] Processes and equations for the biological treatment received by Koutsou et al. [36].

Where:

X_{decay}: biomass degraded per day (kgVSS/d); Q: average influent flowrate (m^3/d); HRT: hydraulic retention time (d); MLVSS: concentration of VSS in the mixed liquor (kg/m^3); b_H: rate of endogenous decay (d^{-1}); R_{O2}: oxygen consumption in kg O_2 d^{-1}; SRT_{total}: sludge residence time in the biological gradient (d); K_e: $BOD_{ultimate}$ of biomass (equal to 1.4); E_H: BOD removal efficiency; F_o: primary effluent BOD_5 concentration (mg/L); f: $BOD_{ultimate}/BOD_5$ ratio (equal to 1.6); Y_H: yield coefficient; N_{nitro}: daily nitrified mass of N (kg/d); N_{total}: total daily influent mass of N (kg/d); N_{bio}: daily mass of N taken up for biomass synthesis (kg/d) (assumed to be 15% of N_{total}); $NH_4\text{-}N_{out}$: daily effluent NH_4-N mass (kg/d); $N_{org\ out}$: daily effluent organic N mass (kg/d); $E_{required}$: consumption of electricity in the LTP (KWh/d); F_i: the contribution (%) of fuel i to the electricity that is produced in Greece; EF_i: the emission factor of GHG for fuel in electricity produced (gr CO_{2e}/KWh); $N_{Effluent}$: mass of effluent N that is released into the aquatic environment (kg N/d); $EF_{Effluent}$: N_2O emission factor for the discharged wastewater (0.005 kg N_2O-N/kg N); Emissions (CO_2, CH_4, N_2O): emitted amounts of CO_2, CH_4, N_2O (kg/d); Electrical energy consumption: kWh/m^3 of treated leachates; Emission (CO_2, CH_4, N_2O) per electrical energy consumption for Greece: for CO_2, CH_4, N_2O, respectively, in kg/kWh

3. Results and Discussion

3.1. Characteristics of Landfill Leachates and Performance of the System

The qualitative characteristics of the raw leachates as well as the effluents of the primary treatment, SBR and RO processes are described in Table 2. The raw leachates were characterized by high conductivity (23,015 ± 2973 μS/cm) and high concentrations of total TN and NH_4-N. These characteristics are typical for raw leachates originating from an active landfill site with a small accumulation history [37].

Table 2. Characteristics of the samples collected from different points of the Leachate Treatment Plant (LTP). The values are reported as mean ± sd. Standard deviation values are given in parentheses.

Type of Sample	Conductivity (μS/cm)	pH	TSS (mg/L)	COD (mg/L)	BOD_5 (mg/L)	TN (mg/L)	NH_4-N (mg/L)
Raw leachate	23,015 (2973)	7.5 (0.2)	166 (97)	3286 (1910)	909 (1242)	1360 (300)	941 (224)
Primary treatment	19,278 (1274)	8.4 (0.1)	363 (127)	2953 (950)	1014 (598)	997 (266)	785 (200)
SBR effluent	16,719 (2117)	8.0 (0.3)	217 (158)	1302 (418)	90 (50)	747 (210)	22 (46)
RO permeate	405 (163)	7.3 (0.5)	4 (5)	15 (8)	11 (7)	15 (9)	<1

The biological treatment of landfill leachates resulted in a significant decrease in organic loading, as well as ammonium nitrogen concentrations. As a result, an average COD removal of 60% was calculated, while the removal of BOD_5 and NH_4-N was equal to 90% and 98%, respectively (Figure 3).

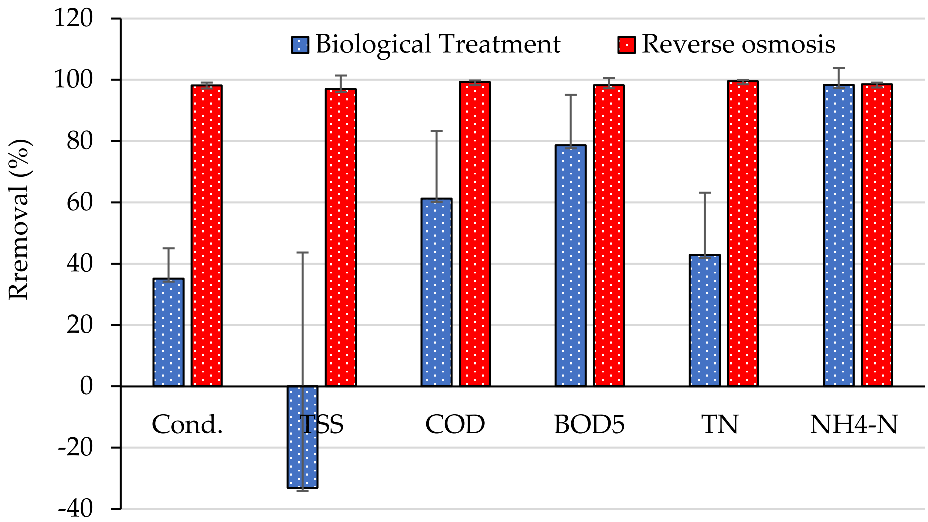

Figure 3. Removal efficiency (%) of the major pollutants after biological treatment in sequencing batch reactors (SBRs) and after reverse osmosis (RO) in the studied Landfill Leachates Treatment Plant.

The partial removal of COD relative to that of BOD_5 is to be expected, as a significant portion of the organic compounds detected in landfill leachates are not biodegradable and cannot be removed via biological treatment [38]. On the other hand, concentrations of TSS in the effluents of the bioreactors were higher than those in the influents (217 vs. 166 mg/L), indicating poor settling capacity in the mixed liquor in the SBRs (Table 2). The use of RO as an advanced treatment step resulted in a significant decrease in the conductivity of the final effluents (405 ± 163 μS/cm), while the removal of COD, BOD_5, TN and NH_4-N reached 99%, 99%, 99% and 100%, respectively (Figure 3). Consequently, the final effluent of the RO was characterized by low concentrations of all major pollutants (Table 2), satisfying the environmental requirements of EU Directive 91/271 concerning the treatment of effluents for disposal in the aquatic environment.

3.2. Energy Consumption

The total energy consumption of the LTP was calculated to be 4,507,690 KWh per year or 35.3 KWh/m^3 of treated leachate. The contribution of the different treatment processes on the energy consumption of the LTP studied is shown in Figure 4. For the energy consumption calculation, 8000 operating hours per year were assumed. The energy consumption in the primary treatment unit represented 5.5% of the total LTP real power consumption, a value which was equal to 1.7 kWh/m^3 of treated leachate (Figure 4).

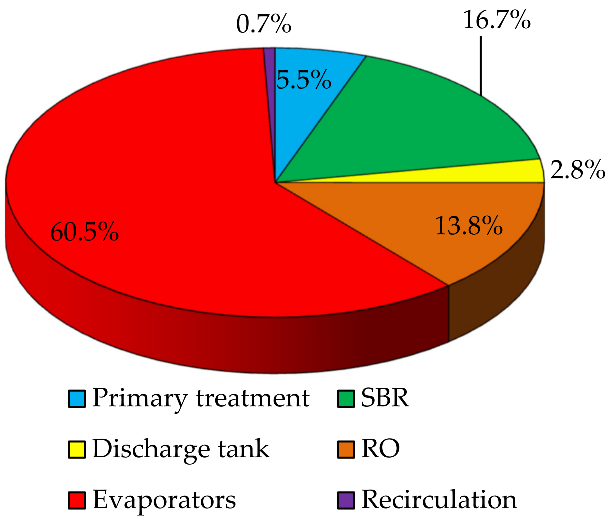

Figure 4. Contributions of the different processes to the total real power consumption in the studied Landfill Leachates Treatment Plant.

Regarding secondary treatment, operation of the SBRs accounted for 16.7% of the total LTP real power consumption (5.1 kWh/m^3 of treated leachate). The principal consumers in the biological treatment were the blowers (roots lobe) used to aerate the reactors, which consumed 443,475 KWh/year (11.3% of the total amount of energy consumed). Operation of the RO unit accounted for 13.8% of the total real power consumption, namely 4.2 kWh/m^3 of treated leachate. In RO, the main source of energy consumption was the high-pressure membrane, i.e., the plunger pumps and the booster modules, which consumed 433,620 KWh/year (11.1% of the total amount of energy consumed). The major part of the power consumed was due to MVR evaporation of RO concentrations. Specifically, the evaporators (two units) contributed 60.4% of the total LTP real power consumption. For each m^3 of RO concentrate treated, the evaporators consumed 54.1 kWh of electricity. The highest consumer during instrumental operation was the roots blower vacuum pump (1,734,480 KWh/year; 44.2% of the total amount of energy consumed). On the other hand, the recirculation of the leachates into the landfill contributed only 0.8% of total LTP real power consumption (3.6 kWh/m^3).

3.3. GHG Emissions

The GHG emitted from the processes of the studied LTP (expressed as kg CO_{2eq}/d) are presented in Table 3. Based on the results, the total amount of emitted CO_{2eq} from this LTP was equal to 9686.9 Kg CO_{2eq} per day or 27.7 Kg CO_{2eq}/m^3 of treated leachates. The biological treatment contributed to 45.7% of the total emissions (4422.7 kg CO_{2eq}/d). On-site GHG emissions were responsible for 88.7% (3921.4 kg CO_{2eq}/day) of the total GHGs generated by the biological treatment, while off-site GHG emissions made up the remaining 11.3% (501.3 kg CO_{2eq}/day). Among biological treatment processes for leachates, the highest amounts of GHGs were emitted due to biomass decay (64.7% of the GHGs in secondary treatment), while the lowest were due to the treated effluents discharged to the aquatic environment. Furthermore, the contribution of other processes such as oxidation of BOD, biomass production, net power consumption and nitrification–denitrification ranged between 10.8% and 15.9%.

Table 3. Estimated GHG emissions (kg CO_{2eq}/d) in the studied Landfill Leachates Treatment Plant.

		Processes	**GHG Emission (kg CO_{2eq}/d)**
Secondary treatment	On-site GHG emissions	Production of GHGs from the decay of biomass	2862.1
		Production of GHGs from BOD oxidation and production of biomass	477.2
		Consumption of GHG from nitrification	−122.4
		Production of GHGs from nitrification–denitrification	704.5
	Off-site GHG emissions	Production of GHGs from net power consumption	492.0
		Production of GHG from treated leachates discharge to the aquatic environment	9.3
		GHG emissions from the biological treatment	**4422.7**
Tertiary treatment		GHG production from RO	988.4
		GHG production from evaporators	4275.8
		GHG emissions from the advanced treatment	5264.2
		Total GHG emissions	**9686.9**

In a previous study, Hua et al. [31] estimated GHG emissions for two different leachate treatment systems—a system combining anaerobic lagoons and aerobic ponds and a system coupling an up-flow anaerobic sludge semi-fixed filter with a membrane bioreactor—and reported values of 209.4 kg CO_{2e}/m^3 and 61.3 kg CO_{2e}/m^3, respectively. These values are much higher than the 27.7 Kg CO_{2eq}/m^3 of treated leachates calculated in this study. The observed difference is mainly due to the high levels of contamination of the leachates in the older study (mean concentrations of BOD, COD and TN equal to 35,000 mg/L, 68,000 mg/L and 11,600 mg/L) and the different treatment processes applied. In the current study, calculation of GHG emissions per m^3 of treated liquid showed that the emissions from secondary treatment and RO were 12.64 and 2.82 kg CO_{2eq}/m^3 of treated leachate, respectively. The value calculated for RO is comparable to the RO emissions reported by Ab Hamid et al. [39] in the application of a forward osmosis aerated membrane bioreactor (FOAeMBR) connected with RO in urban wastewater treatment. On the other hand, the evaporators emitted 35.63 kg CO_{2eq}/m^3 of treated RO concentrate.

4. Conclusions

Due to the chemical characteristics of landfill leachates, biological processes and advanced treatment technologies are usually applied in series to achieve efficient leachates treatment. Despite the wide use of these technologies at full scale, there is as yet limited information available on their energy consumption and GHGs emitted. A holistic approach was applied in this study for the evaluation of a full-scale landfill leachates treatment system analyzing samples for the removal of major pollutants, measuring energy consumption and estimating GHG emissions. According to the results, the combination of biological treatment and RO achieved sufficient treatment of landfill leachates to fulfil the legislative requirements for treatment of leachates for discharge in the environment. However, questions have arisen regarding the sustainable management of RO concentrates, as the MVR evaporation applied in the studied LTP was responsible for the consumption of more than 60% of the total energy, while it also contributed significantly to the carbon footprint of the plant, emitting more than 44% of the GHGs. The operation of SBRs and RO required 16.7% and 13.8% of the total energy, while SBRs contributed significantly to the total GHG emissions (45.6%) mainly due to on-site emissions and the process of biomass decay. Future studies should focus on the application of alternative technologies and practices for the sustainable management of RO concentrates. As the volume and characteristics of leachates produced vary during the year, the impact of seasonality on energy consumption and GHG emissions should also be studied. Data on the overall cost of the different processes should also be collected.

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