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A 35-year monitoring of an Italian landfill: Effect of recirculation of reverse osmosis concentrate on leachate characteristics

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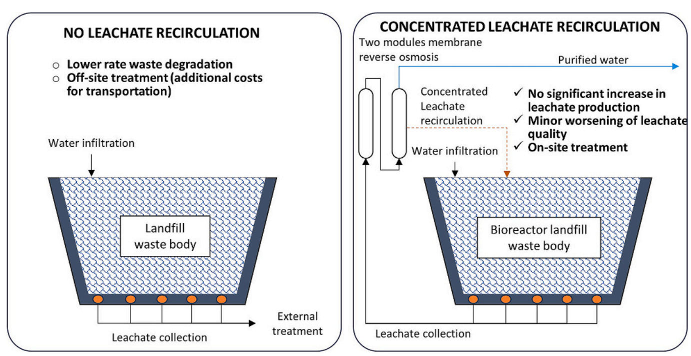
HIGHLIGHTS

- "Fossetto" landfill operates since 1988 following the state-of-the-art.
- Since 2006 concentrated leachate is recirculated into the landfill body.
- The rise in leachate production cannot be totally attributable to recirculation.
- The concentration of NH_4^+ and Cl^- increased by 60 % and 58 %.
- This increase did not influence the performance of the treatment plant.

GRAPHICAL ABSTRACT



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ABSTRACT

"Fossetto" landfill (Monsummano Terme - Tuscany, Italy) started operation in 1988 as a controlled landfill accepting mixed municipal solid waste collected without any attempt of recycling. Then, progressively, following the evolution of the state-of-the-art, it adopted biogas extraction and valorisation systems and mechanical-biological treatment for incoming waste (both since 2003). Finally, since 2006, in the plant is performed on-site reverse osmosis leachate treatment with the concentrated leachate being recirculated back into the landfill body. Recently a new landfill cell, separate from the others, was put in operation adding a capacity of 200,000 m^3 to the already available 1,095,000 m^3. This plant can provide long term leachate composition data to study the evolution and impact of changing landfill technology and waste composition on various parameters. The rise in leachate production (+84 % in 2018–2022 respect to the period before recirculation) cannot be totally attributable to recirculation but could be also linked to the increase in the amount of landfilled waste. The concentration of certain parameters (NH_4^+, Cl^- and to a less extent of COD) increased (+60 %, +58 %, +17 % respectively in the last five years with respect to the period before recirculation); however, this increase did not influence the performance of the treatment plant. Nevertheless, the overall leachate management would benefit from an optimized reinjection system.

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1. Introduction

Even though the last decades have been characterised by the increase of innovative technologies related to recycle and recovery of urban and industrial wastes, landfills still play an important role in the management of residual waste (i.e. that cannot be otherwise valorised). On the other hand, leachate and biogas represent the main issues related to the disposal of waste in landfill, as they are both generated by the degradation of the residual organic fraction of municipal solid waste (MSW). In particular, landfill leachate (LL) is generated from water infiltration (generally rain based on local climatic and pedological conditions) and subsequent percolation of the waste's moisture itself (Frikha et al., 2017; Ghaffariraad et al., 2021; Yesilnacar and Cetin, 2005) that occurs when the moisture content in waste exceeds its water holding capacity (Salati et al., 2013). LL is characterised by high concentration of organic pollutants, heavy metals and ammonia-nitrogen (Ghaffariraad et al., 2021; Ma et al., 2021). Leachate physico-chemical composition depends, apart from the type of waste, on the landfill operating conditions (Ghaffariraad et al., 2021; Moody and Townsend, 2017; Renou et al., 2008) and landfill age (Li et al., 2010; Salati et al., 2013). As a consequence of its characteristics, LL accidentally released can cause a decrease in soil and water quality and long-term environmental impacts with negative effects on human health (Budihardjo et al., 2021; Ma et al., 2021; van Turnhout et al., 2018).

Environmental pollution related to landfilling still represents a crucial issue in waste management, even though a huge work has been done in the last fifty years in the search for reducing the environmental impact from landfilling. At legislative level, the European Directive 1999/31/EC (adopted in Italy since 2003), in order to minimise landfill environmental impacts both in active and post-closure period, was intended to progressively reduce the disposal of biodegradable waste and to only allow the landfilling of mechanically-biologically treated (MBT) waste (Calabrò and Lisi, 2014). The purpose of the mechanical biological treatment, indeed, is to increase waste biological stability (Evangelou et al., 2016; Tran et al., 2014).

With regard to management operations, leachate recirculation in the landfill body is a technique widely used not only as a treatment for LL itself, but also to improve organic matter biodegradation (by establishing optimal moisture conditions for microbial growth) and, consequently, to enhance biogas recovery and to make waste biological stabilization faster (Aromolaran and Sartaj, 2023). Landfills in which leachate recirculation is used for this purpose are, indeed, referred to as (anaerobic) bioreactor landfills (Budihardjo et al., 2021). Leachate resulting from LL recirculation is reported to have lower COD concentration compared to landfill without leachate recirculation (Budihardjo et al., 2021). Moreover, leachate recirculation can significantly influence metal and chloride behaviour in the landfilled waste since these compounds cannot be biochemically transformed (Lee et al., 2023). Chloride affects the release of metals through the of binding the metals on humic acids and through the adsorption of metals (Begeal, 2008; Damikouka and Katsiri, 2021; Lee et al., 2023). Other treatments, such as biological (aerobic and anaerobic) and/or physico-chemical ones, can be additionally adopted in combination with LL recirculation (Ma et al., 2021). The first generally show good performances in the treatment of the young leachate due to the presence of high amounts of easily biodegradable organic matter (Fazzino et al., 2021; Luo et al., 2020), while mature LL could be better treated by physico-chemical processes (such as flotation, coagulation, adsorption, advanced oxidation processes and membrane filtration) (Calabrò et al., 2021; Fazzino et al., 2021; Luo et al., 2020; Renou et al., 2008; Torretta et al., 2017). Concentrated leachate, that is the polluted effluent derived from reverse osmosis treatment, is a dark brown solution characterised by the presence of recalcitrant organic matter (such as humic and fulvic acids), ammonia, heavy metals and high chemical oxygen demand (COD) (Calabrò et al., 2018, 2010; He et al., 2015; Zhang et al., 2013). Recirculation into the landfill is the most simple and economically viable treatment technology for concentrated leachate (Calabrò et al., 2010; Liu et al., 2008; Qu et al., 2008; Renou et al., 2008; Sluiter et al., 2012; Wiszniowski et al., 2006) that should be otherwise treated by other expensive processes (such as evaporation, distillation, coagulation and advanced oxidation) (He et al., 2015).

The specific studies present in scientific literature on this practice are not numerous and opinions are often conflicting (de Almeida et al., 2023; He et al., 2015; Heinigin, 1995; Zhang et al., 2013).

This paper aims to advance the knowledge published previously on the same topic (Calabrò et al., 2018, 2011, 2010; Calabrò and Mancini, 2012). Specifically, we analysed the long-term effect of concentrated leachate recirculation in an Italian landfill (Fossetto landill in Tuscany) where the reverse osmosis technology to treat leachate is used since September 2006. In particular, we aimed to analyse the effect of the recirculation of the reverse osmosis plant on the qualitative and quantitative leachate characteristics analysing data available over a period of 16 years. For this reason, the statistical analysis was based on grouping leachate quality measurements prior to 2006 (no recirculation) and after 2006 (recirculation period) using several sampling locations per period.

2. Materials and methods

2.1. The landfill "Il Fossetto" (Pistoia Province, Tuscany, Italy)

"Il Fossetto" landfill has been active since 1988. It is located in Monsummano Terme (Pistoia Province, Tuscany, Italy); it reached over the years a total authorized volume of about 1,295,000 m^3 thanks to the recent (end of the year 2020) addition of a 200,000 m^3 new cell (Cell 8).

At the beginning of its operation, this landfill was used to dispose of mixed municipal waste and then also non-hazardous bottom ash and slag coming from a municipal incinerator. In accordance to EU directives, the direct landfilling of mixed municipal waste was banned in 2003 when a mechanical and biological treatment (MBT) plant was put in operation. In 2011 also the disposal of incineration bottom ash and slag was stopped. In addition to the MBT plant, a biogas recovery and energy production and a leachate treatment plant are operating in the landfill.

The Landfill is divided into sub-landfills (Fig. 1), Landfill 1 is (the smallest and the first entered into operation), Landfill 2 and Landfill 4 (Landfill 4 was partially built over Landfill 3; for analysis purposes, Landfill 3 is considered part of Landfill 4) that includes the recently authorized and already mentioned Cell 8 (adjacent to Landfill 4 but completely separated from it). In "Il Fossetto" landfill, leachate collected by the drainage system is extracted by several wells, their number increased over the years when new disposal areas were added to the landfill or when some well loses efficiency.

Landfill 1 was closed on 1989 and does not generate any leachate. Landfill 2 was closed on the beginning of 2015. Its final cover was recently built and leachate is still collected.

Until 2006, all the leachate produced by the landfill was transported to external plants mainly for co-treatment with municipal sewage in wastewater treatment plants. Since September 2006, most of the extracted leachate is treated on site in a reverse osmosis plant. Purified water obtained by leachate treatment is discharged into a small nearby channel, while the generated concentrated leachate is recirculated back into the landfill by four vertical wells constructed in Landfill 4. At the moment 4 reinjection wells are used. The generated concentrated leachate that is reinjected into the landfill, at the beginning of operation of the leachate treatment plant it represented about 30 % of the total incoming leachate, for management reasons linked to the worsening of its quality especially during summer, in order to avoiding excessive membrane fouling and allowing easier recirculation, this amount for limited periods of time increased up to 50 % in the last years.

This study uses landfilled waste data from a database of 35 years (1998–2022) and leachate data from a database of 24 years (1999–2022).

For more information on “Il Fossetto” landfill see available literature (Calabrò et al., 2018, 2010).

2.2. Monitoring activities

According to the requirements of the Control Authority (Pistoia Province), an extensive monitoring program is being regularly carried out in “Il Fossetto” landfill.

All the monitored physical and chemical parameters were measured using internationally recognized methods that were adopted by the control authority and are regularly updated following best available techniques.

The following data are recorded:

- meteorological parameters (e.g. temperature and rainfall; Meteorological station Siap+Micros s.r.l. Castello Roganzuolo di San Fior – Treviso - Italy);
- amount of waste landfilled (detailed for each single type – certified truck scale);
- leachate and concentrated leachate produced (certified flowmeters – several models and types);
- MBT waste basic characteristics (Nappi et al., 2000);
- Chemical characteristics (pH, COD, ammonia nitrogen, chloride, As, total Cr, Cu, Hg, Ni, Zn) of leachate from each recovery well, one sampling per year (APAT and IRSA-CNR, 2003; APHA et al., 2012);
- Chemical characteristics (pH, conductivity, suspended solids, COD, BOD_5, ammonia nitrogen, chloride, sulphides, total Cr, Ni, Zn, As, Hg, Cu) of leachate in the equalization tank - four samplings per year (APAT and IRSA-CNR, 2003; APHA et al., 2012);
- Chemical characteristics (pH, conductivity, suspended solids, COD, BOD_5, ammonia nitrogen, chloride, sulphides, total Cr, Ni, Zn, As, Hg, Cu) of recirculated concentrated leachate - four samplings per year (APAT and IRSA-CNR, 2003; APHA et al., 2012);
- Extracted biogas amount and basic composition (average CH_4 percentage, O_2 and other gases only for more recent years) (certified flowmeters and portable gas analysers e.g. Geotechnical Instruments GA2000 plus);
- Estimation of fugitive biogas emissions from the landfill surface, one measurement per year since 2008, (Marshall et al., 2010).

A limitation is that for several parameters (e.g. leachate characteristics from each well) only one measurement per year is available and therefore the uncertainty of the estimation is quite high, however this is counterbalanced by the availability of long series of data that allow to evaluate the trends with a sufficient reliability.

2.3. Statistical analysis

The means of each quality parameter prior to and after 2006 were calculated using measurements that were performed once per year. Typically, around 4 to 8 measurements (treated as replications) were available prior to 2006 depending on when monitoring of the leachate started in each leachate well or other sampling spot (usually on 2000 or 2001). The annual measurements after 2006 ranged from 10 to 18. The comparison of the means prior to 2006 (period of no recirculation) and after 2006 (recirculation period) was made with an independent *t*-test (with equal variances) at $\alpha = 0.05$. The normality of the data had been checked with the Kolmogorov – Smirnov test of normality. Most data in both periods ($<$ 2006 and $>$2006) did not differ significantly from that which is normally distributed (at $\alpha = 0.05$), so normality was assumed to exist throughout, and, thus, the t-test was applied for the comparisons in all cases. All statistical tests performed with Minitab® v21.

3. Results and discussion

3.1. Leachate generation

Fig. 2.a puts in relation the cumulated landfilled waste and the cumulated leachate collected for the periods before (1988–2006) and after (2007–2021) the beginning of the recirculation of concentrated leachate, an increase in leachate production after the implementation of the recirculation is evident. The comparison with the same analysis limited to the data available until the year 2016 (Calabrò et al., 2018) showed that the slope of the regression line for the data after the recirculation beginning was lower (0.788 vs 0.973) and therefore the tendency to the increase of leachate production respect to the landfilled waste is more evident.

Additional analysis (Fig. 2.b) demonstrates that the increase in leachate production is not due to rainfall since while the rainfall tended

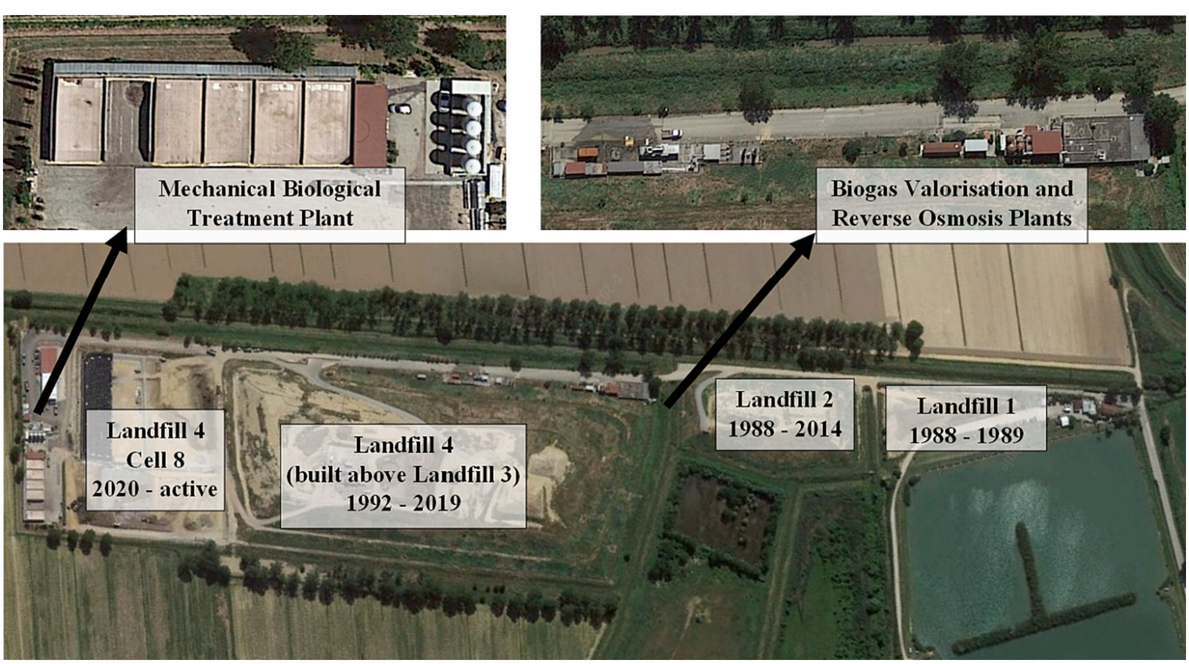

Fig. 1. Il Fossetto Landfill.

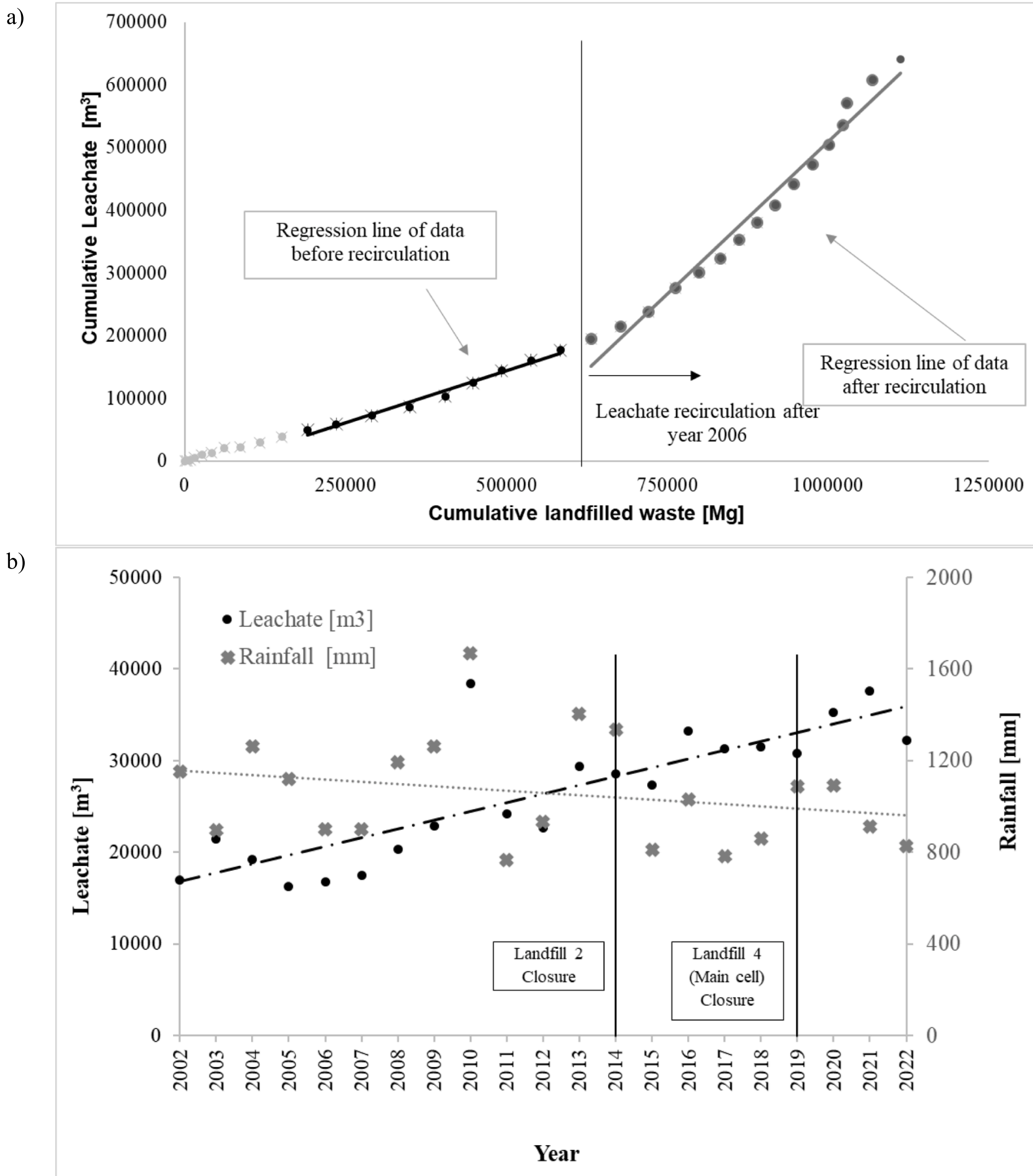

Fig. 2. Cumulative Leachate recovery vs. Cumulative landfilled waste in the period 1988–2022 (a) and comparison between Leachate collected and Rainfall (b).

to slightly reduce in the period 2002–2022 the leachate production tends to increase. In fact, in the first five years after recirculation beginning, increase in leachate production was about 36 % and it rose progressively to about 84 % in the period 2018–2022 respect to the five years prior recirculation beginning. However, the increase in leachate production is slightly lower than the cumulated landfilled waste: total landfilled waste increased by 90 % in 2022 respect to 2006, therefore the increase in leachate production cannot be attributable to recirculation but could be also linked to the increase in the amount of landfilled waste. Another issue to be considered is the impact of the ban of landfilling of bottom ash and slag on leachate production. According to (Yu and Rowe, 2021), the ban of this type of waste could increase waste permeability and, as a consequence, increase leachate flux in the upper landfill layers.

The additional landfilling area added in 2020 (Cell 8) increased landfill surface exposed to rainfall by about 20 % but this did not change proportionally leachate production.

3.2. Leachate composition

3.2.1. Raw leachate quality before recirculation (equalization tank)

Table 1 reports leachate composition from “Il Fossetto” landfill according to the sampling carried out in the equalization tank where the flow from all the wells is mixed and that therefore is representative of the whole production.

The equalization tank was built prior to the reverse-osmosis leachate treatment plant; therefore, the sampling period of the leachate data from that tank were obtained over a two-year period, a period considered still representative of the leachate composition there. The composition is typical of a mature landfill (Renou et al., 2008; Wiszniowski et al., 2006) with alkaline pH, high concentration of COD, ammonium and chlorides and moderate presence of heavy metals and Arsenic (Table 1).

3.2.2. Raw leachate quality after recirculation (equalization tank) and concentrated recycled leachate

Table 1 clearly shows that the leachate from "Il Fossetto" landfill equalization tank tended to worsen its quality in the first period after recirculation beginning especially for COD and some metals and metalloids (Cu, Ni, Zn, As) for Ni, Zn and As the increase was especially high.

Since 2011 this tendency changed completely, only ammonium and chloride concentrations increased significantly respect to the period before the recirculation beginning while all the metals and Arsenic showed a clear tendency to a decrease (Calabrò et al., 2018).

This decrease in metals concentration could be linked to the ban of landfilling of incinerator bottom ash and slag (since 2011), to the highly efficient separate collection that prevent the landfilling of materials containing heavy metals and metalloids (e.g. electric and electronic equipment, batteries).

Sulphate has been analysed in the leachate from equalization tank only since 2019 (4 years) but it is an extremely important parameter because, in this stage of the landfill metabolism (mature landfill in stable methanogenesis) (Kjeldsen et al., 2002), it should not be present in the leachate in significant quantities and therefore it is only due to the recirculation process (average concentration in the equalization tank in the last three years equal to 8090 mg/L) being one of the main components of the concentrated leachate (sulphuric acid is added during the treatment – average concentration in the recirculated leachate in the last three years 19,600 mg/L). Moreover, its concentration is highly correlated (correlation coefficient of 0.99 and 0.98 respectively) with ammonium and chlorides. Therefore, it is likely that the increase of the concentration of these latter compounds in leachate is strictly linked to the recirculation process.

An improvement of the leachate characteristics was observed over the last couple of years (2020−2022) in which leachate recirculation would be realized. In fact, the levels of certain parameters (e.g. COD, ammonium and chlorides) tended to reduce to levels of the period 2012–2016. This could be linked to the higher dilution of the concentrated leachate recirculated back to the landfill. During summer, raw leachate was always quite dense. For this reason, the ratio between concentrated and raw leachate during summer was increased to avoid clogging and damages to the membrane due to the high solids content.

Metals presence, both in raw and concentrated leachate, tends to reduce in recent years also for those metals (Cu, Ni, Zn) whose concentration significantly increased in the period immediately after recirculation beginning (Table 1). This is especially true for As, whose concentration increase was dramatic in the period 2006–2011. This change was probably due to both the ban of landfilling of incineration ash and slag since 2011 and to the metals precipitation. In fact, at this pH, most likely, sulphates introduced with concentrated leachate, were reduced to sulphide by sulphate reducing bacteria thus enhancing the formation of insoluble metal sulphides. According to literature (Lewis, 2010), this process at the pH of LL is less efficient for As and Zn whose concentration is in fact declining more slowly.

3.2.3. Raw leachate from the newly activated cell 8

On 2021, disposal of MBT wastes into the new cell 8 that belongs to landfill 4 was initiated. This cell represents a unique occasion to obtain leachate data from landfilled MBT waste, despite the small sample size (4 annual measurements). Therefore, data from this new cell were finally included in the statistical analysis since this was the only cell that did not accept neither untreated waste, nor incineration ash/slag nor any leachate recirculation was performed. Table 2 presents the very first data available for the leachate of cell 8.

Moreover, it shows the average leachate composition in wells (42.1, 43.1, 44.1) activated in the period 2000–2003, in newly opened cells, and where raw mixed municipal waste and incineration ash and slag were landfilled.

The well known benefits on leachate composition of MBT are evident form the analysis of the data, after less than one year after the beginning of waste landfilling the pH is neutral or slightly acidic; with a very few exceptions a very mild content of COD, ammonium and chloride is evident and metals concentration is limited. It is noticeable that larger COD and sulphate concentrations are associated as in typical young LL.

These benefits are clear if leachate data from cell 8 (MBT cell) are compared to the data from the cells of Landfill 4 that had been accepting untreated waste in the period 2000–2003 (i.e. before MBT initiation). The latter are considered as reference values. Specifically, in the MBT cell derived leachate, there were decreases equal to 18 %, 129 %, 825 % and 239 % for pH, COD, NH_4^+ and Cl^-, respectively. Similar concentration reductions were recorded for total Cr and Ni. On the other hand, Cu and As had slightly higher values in the MBT waste derived leachate cell compared to the raw waste leachate.

As resulting from a study carried out on the analyses of data obtained by leachate samples collected from European landfills, leachate derived from MBT waste are characterised by a lower polluting potential with respect to mechanically sorted organic residues (Robinson et al., 2005), especially in terms of organic compounds. The expected lower concentrations of organic matter in the leachate from MBT waste compared to that of untreated waste, is related to the nature of the aerobic treatment itself, which must lead to a reduction of the organic substances (Tran et al., 2014). For instance, in a study of (Salati et al., 2013), leachate derived from treated waste were characterised by a reduction of 54 %, 69 %, 77 %, 70 %, 81 % and 16 % for NTK, NH_3, TOC, COD, BOD_5 and total heavy metal contents, respectively, compared to leachate obtained from untreated waste. Even though MBT wastes are characterised by a higher metals proportion (caused by the loss of the organic matter, (Molleda et al., 2020)), many metals join the organic matter from the waste matrix after the biological stabilization process reducing the metal leaching potential (Farrell and Jones, 2009; van Praagh et al., 2009).

The reduction of ammonia nitrogen is particularly important in bioreactor landfills as high ammonia concentration leads to the inhibition of the anaerobic biodegradation (Aromolaran and Sartaj, 2023; Chamem et al., 2020). Possible accumulation of ammonium nitrogen and chloride in LL over the time could be explained by the recirculation of raw leachate or concentrate leachate resulting from reverse osmosis process (Chamem et al., 2020; Ma et al., 2021). In some studies, indeed, the concentration of the ammonium nitrogen in leachate from MBT waste remained high for long periods of time (Molleda et al., 2020; Salati et al., 2013; Tran et al., 2014). Possible increase of the COD concentration could be due to an excess of leachate recirculation which may lead to the leaching of organics in the leachate (Ma et al., 2021). In this case, the increase of the ammonia nitrogen could be related to the hydrolysis of organic nitrogen (Ma et al., 2021), thus explaining its high concentration and long-term presence in leachate (Jiang et al., 2007; Long et al., 2009).

The lower organic loading of leachate obtained from landfilled MBT waste allows for a more rapid establishment of the methanogenic phase in landfill degradation as the acidogenic phase would be considerably shorter (Molleda et al., 2020; Robinson et al., 2005; Siddiqui et al., 2012). However, a quite long lag-phase, attributed to the high concentration of volatile fatty acids (VFAs) and of ammonia, was observed also in biomethanisation tests carried out on MBT waste (Pantini et al., 2015).

Since the aim of the mechanical biological treatment is the reduction of the organic matter and not of the ammonia nitrogen (Tran et al.,

Table 1
Characteristics of Leachate in Equalization tank and of Recirculated Concentrated leachate.

	Equalization Tank							Concentrated Leachate			
	Average Before Recirculat. (2005–2006)	Average in the first 5 years of Recirculat. (2007–2011)	Average in the period 2012–2022	Average in the period 2018–2022	Difference period 2006–2011 respect to the period before Recirculation (2006–2006)	Difference period 2012–2022 respect to the period before Recirculation (2006–2006)	Difference period 2018–2022 respect to the period before Recirculation (2005–2006)	Average in the period 2006–2011	Average in the period 2012–2016	Average in the period 2017–2022	Average in the period 2020–2022
pH	7,69	7,98	7,62	7,59	3,8 %	-0,9 %	-1,3 %	6,05	6,50	5,84	5,77
COD [mg/L]	3366	4131	3663	3928	22,7 %	8,8 %	16,7 %	4670	4512	5401	4579
$NH4^{+}$ [mg/L]	1832	1786	2812	2926	-2,5 %	53,5 %	59,7 %	3920	4387	5309	4473
Cl^{-} [mg/L]	2179	2501	3708	3446	14,8 %	70,2 %	58,1 %	5763	5955	7108	6214
Pb [mg/L]	0,61	0,32	0,11	0,07	−48,2 %	−82,2 %	−88,1 %	0,25	0,10	0,03	0,04
Cr_{tot} [mg/L]	6,55	5,15	2,58	2,98	−21,4 %	−60,7 %	−54,5 %	2,70	2,88	3,68	3,35
Cu [mg/L]	0,25	0,33	0,12	0,10	34,7 %	−51,1 %	−58,9 %	1,22	1,49	0,13	0,17
Ni [mg/L]	0,62	0,96	0,56	0,56	53,1 %	−9,7 %	−10,1 %	1,58	0,88	0,99	0,84
Zn [mg/L]	0,87	2,09	0,45	0,47	139,7 %	−48,2 %	−45,9 %	1,92	1,95	0,39	0,43
As [mg/L]	0,06	0,28	0,06	0,07	392,8 %	3,9 %	17,1 %	0,12	0,09	0,14	0,15

Table 2
Composition of leachate from Cell 8 of Landfill 4 and comparison with Leachate data before MBT implementation.

	pH	COD [mg/L]	NH_4^+ [mg/L]	Cl^- [mg/L]	SO_4^{--} [mg/L]	Pb [mg/L]	Crtot [mg/L]	Cu [mg/L]	Ni [mg/L]	Zn [mg/L]	As [mg/L]	Hg [mg/L]
	Well 48.1											
apr-21	7.60	2125	740	1650	133	B.D.L.	1.20	B.D.L.	0.29	0.25	0.13	0.08
dec-21	7.00	471	162	N.A.	693	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
apr-22	6.6	347	158	283	N.A.	B.D.L.	0.23	0.071	0.062	0.23	B.D.L.	< 0.01
dec-22	6	95	90	N.A.	123	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
	Well 48.2											
dec-21	7.00	490	54	N.A.	53	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
apr-22	6.3	329	68	335	N.A.	B.D.L.	0.13	0.073	0.065	0.15	B.D.L.	< 0.01
dec-22	5.7	114	90	N.A.	113	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
	Well 48.3											
apr-22	6.6	< 50	1.4	13.4	N.A.	B.D.L.	B.D.L.	0.074	B.D.L.	0.097	B.D.L.	0.018
dec-22	5.6	95	81	N.A.	24.4	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
	Well 48.4											
dec-21	7.00	824	54	N.A.	162	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
apr-22	7	1025	73	1132	N.A.	B.D.L.	0.52	0.11	0.2	0.44	0.077	< 0.01
dec-22	7.2	2286	504	N.A.	2514	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
	Well 48.5											
dec-22	5.8	209	72	N.A.	50	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
	Well 48.6											
apr-22	7.1	1261	315	1272	N.A.	B.D.L.	0.45	0.12	0.19	0.93	0.079	< 0.01
dec-22	7.4	5714	1260	N.A.	2760	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
	All wells cell 8 (years 2021–2022)											
Average	6.7	1099	248	781	663	B.D.L.	0.51	0.09	0.16	0.35	0.095	N.A
	Wells Landfill 4 activated in the period 2000–2003 (42.1. 43.1. 44.1)											
Average	7.9	2522	2295	2649	N.A.	0.13	1.26	0.02	0.39	0.29	0.03	N.A
Difference	−18 %	−129 %	−825 %	−239 %	N.A.	N.A.	−147 %	78 %	−144 %	17 %	68 %	N.A.

B.D.L.: Below detection limits; N.A.: Not available

2014), a viable way to reduce nitrogen compounds on the leachate (and thus their inhibitory effects) could be the application of supplementary aeration (Ma et al., 2021; Šan and Onay, 2001; Shou-liang et al., 2008; Tran et al., 2014) (Ma et al., 2021).

Previous researchers (Huo et al., 2008; Šan and Onay, 2001) have found that aerobic conditions have a positive impact on nitrogen removal in bioreactor landfills.

It is worth mentioning that, considering the few available data (Table 2) referred to the short period immediately after the beginning of the landfilling of MBT waste in cell 8, the acidogenic phase may have already been almost completed.

In the MBT cell, all main leachate parameters were lower compared to the raw waste cells.

3.2.4. *Analysis of the characteristics of raw leachate from selected wells in the are subjected to recirculation of concentrated leachate*

As already mentioned, sulphate presence in leachate in mature landfill is generally scarce but in this case this compound derives from the addition of sulphuric acids during membrane treatment and therfore sulphate can be used as a sort of tracer to select the wells whose leachate composition is more influenced by the recirculation. On this basis, among the wells with a sufficient data set, well 42.1 can be considered basically unaffected by the reinjection of concentrated leachate (average sulphate concentration 3520 mg/L), while wells 41.1, 43.2 and 44.1 are those with the most noticeable presence of sulphate (average sulphate concentration 7050, 17,350 and 11,800 mg/L respectively).

For this reason a statistical analysis for COD, ammonium, chloride and selected metals has been carried out on these wells and in the equalization tank (Table 3).

With regard to COD, Ammonium, Chlorides, Cr, Ni, and especially As it was shown that leachate in the three wells that were thought to be significantly affected by recirculation, had statistically higher values after 2006. This indicates that in fact recirculation most probably affected leachate quality especially for specific wells also when the general leachate composition (i.e. equalization tank) is not affected.

For NH_4^+ it was shown that concentration increase was statistically significant even in the well 42.1 (the one considered not influenced by recirculation) but not in the equalization tank.

Metals (Pb, Cr, Zn, Ni) and As behaviour was not homogeneous; Pb and Zn concentrations decreased both in the wells and in equalization tank, even dramatically for lead in the wells, but this decrease was not statistically significant. As and Ni concentrations increased also in the well 42.1 and in the equalization tank but, again, the increase is not statistically significant. Also Cr, as already mentioned, increased in the wells but, in this case, the concentration in the equalization tank tended to significantly decrease and this is the only statistically significant difference for leachate sampled in the equalization tank.

The lack of statistical significance even in presence of large variations between the period before and after the recirculation beginning was the result of the large variance among the yearly measurements.

Table 3
Statistical analysis of landfill leachate from equalization tanks and selected wells before and after recirculation.

		Before recirculation Avg. Conc. [mg/L] (number of yearly observations)	After recirculation [mg/L] (number of yearly observations)	Relative difference	Statistical difference at α = 0.05
COD	Well 42.1	2219 ($n = 7$)	2714 ($n = 16$)	18 %	No
	Well 41.1	1635 ($n = 8$)	2456 (n = 16)	33 %	Yes
	Well 43.2	2566 ($n = 7$)	3807 ($n = 15$)	33 %	Yes
	Well 44.1	1753 ($n = 6$)	3410 (n = 16)	49 %	Yes
	Equal. Tank	3486 (n = 8)	3806 ($n = 64$)	8 %	No
NH_4^+	Well 42.1	1956 (n = 7)	2542 (n = 16)	23 %	Yes
	Well 41.1	1210 (n = 8)	2472 (n = 16)	51 %	Yes
	Well 43.2	2392 (n = 7)	3866 (n = 15)	38 %	Yes
	Well 44.1	1770 (n = 6)	3388 (n = 16)	48 %	Yes
	Equal. Tank	1899 (n = 8)	2483 (n = 64)	24 %	No
Cl^-	Well 42.1	2385 (n = 7)	2994 (n = 16)	20 %	No
	Well 41.1	1306 (n = 8)	2598 (n = 16)	50 %	Yes
	Well 43.2	2670 (n = 7)	4785 (n = 15)	44 %	Yes
	Well 44.1	1987 (n = 6)	4189 (n = 16)	53 %	Yes
	Equal. Tank	2321 (n = 8)	3318 (n = 64)	30 %	No
Pb	Well 42.1	0.321 (n = 7)	0.107 (n = 16)	−200 %	No
	Well 41.1	0.230 (n = 8)	0.097 (n = 16)	−137 %	No
	Well 43.2	0.269 (n = 7)	0.116 (n = 15)	−132 %	No
	Well 44.1	0.235 (n = 6)	0.130 (n = 16)	−81 %	No
	Equal. Tank	0.233 (n = 8)	0.172 (n = 64)	−35 %	No
Cr	Well 42.1	1.213 (n = 7)	1.691 (n = 16)	28 %	No
	Well 41.1	0.599 (n = 8)	1.563 (n = 16)	62 %	Yes
	Well 43.2	1.423 (n = 7)	2.640 (n = 15)	46 %	Yes
	Well 44.1	0.872 (n = 6)	2.410 (n = 16)	64 %	Yes
	Equal. Tank	6.28 (n = 8)	3.39 (n = 64)	−85 %	Yes
Ni	Well 42.1	0.416 (n = 7)	0.573 (n = 16)	38 %	No
	Well 41.1	0.599 (n = 8)	1.563 (n = 16)	79 %	Yes
	Well 43.2	1.423 (n = 7)	2.640 (n = 15)	103 %	Yes
	Well 44.1	0.872 (n = 6)	2.410 (n = 16)	151 %	Yes
	Equal. Tank	6.28 (n = 8)	3.39 (n = 64)	79 %	No
Zn	Well 42.1	0.461 (n = 7)	0.379 (n = 16)	−18 %	No
	Well 41.1	0.361 (n = 8)	0.359 (n = 16)	−1 %	No
	Well 43.2	0.501 (n = 7)	0.399 (n = 15)	−20 %	No
	Well 44.1	0.677 (n = 6)	0.372 (n = 16)	−45 %	No
	Equal. Tank	0.990 (n = 8)	0.950 ($n = 63$)	−4 %	No
As	Well 42.1	0.0328 (n = 7)	0.096 (n = 16)	193 %	No
	Well 41.1	0.0136 (n = 8)	0.0676 (n = 15)	397 %	Yes
	Well 43.2	0.0347 (n = 7)	0.114 (n = 15)	229 %	Yes
	Well 44.1	0.0247 (n = 6)	0.101 (n = 16)	309 %	Yes
	Equal. Tank	0.0567 (n = 8)	0.134 ($n = 59$)	136 %	No

4. Conclusions

The conclusions from this work are:

- The increased landfill surface that was exposed to rainfall (20 %), due to the construction of Cell 8 of landfill 4, did not affect leachate generation rates.
- The concentrations of key leachate parameters (e.g. NH_4^+, Cl^-, COD) over the last five years in in which recirulcation was practised, did not differ significantly compared to those before the initiation of recirculation.
- Based on the leachate composition of Cell 8 (in which no recirculation occurred nor incineration ashes/slag was disposed), mechanical and biological pretreatment is effective in reducing key leachate pollutant contents compared to the concentration in wells of Landfill 4 that did not accept MBT waste. In the MBT waste derived leachate, a decrease by 129 %, 825 % and 239 % were recorded for COD, NH_4^+, Cl^-, respectively.

According to our findings, recirculation of concentrated leachate can be still considered a suitable leachate treatment approach since it shows to not increase significantly pollutant contents in the leachate. Treated leachate always complied with discharge permits.

According to the results of this paper, leachate recirculation, even in landfills where only MBT waste is accepted, may reduce leachate management cost and close the treatment loop within the landfill without the need of an external plant. These benefits balance the limited worsening of leachate quality. Still, the overall leachate management would benefit from an optimized reinjection system (e.g. more reinjection points, sub-horizontal wells) to more efficiently spread the concentrated leachate over the entire landfill body.

CRediT authorship contribution statement

A. Folino: Data curation, Writing – review & editing. **E. Gentili:** Data curation, Methodology, Writing – review & editing. **D. Komilis:** Data curation, Formal analysis, Methodology, Writing – review & editing. **P.S. Calabro:** Conceptualization, Data curation, Formal analysis, Methodology, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

References

de Almeida, R., Porto, R.F., Quintaes, B.R., Bila, D.M., Lavagnolo, M.C., Campos, J.C., 2023. A review on membrane concentrate management from landfill leachate treatment plants: the relevance of resource recovery to close the leachate treatment loop. Waste Manag. Res. 41, 264–284. https://doi.org/10.1177/0734242X221116212.

APAT, IRSA-CNR, 2003. Metodi analitici per le acque. Manuali e linee guida 29/2003-Sezione 2000 Parametri fisici, chimici e chimico-fisici.

APHA, AWWA, WEF, 2012. Standard Methods for the Examination of Water and Wastewater, 22nd edition. American Public Health Association, American Water Works Association, Water Environment Federation.

Aromolaran, A., Sartaj, M., 2023. Enhancing biogas production from municipal solid waste through recirculation of blended leachate in simulated bioreactor landfills. Biomass Conv. Bioref. 13, 2797–2812. https://doi.org/10.1007/s13399-020-01260-5.

Begeal, C., 2008. The Effect of Chloride Ion on Heavy Metal Partitioning and Transport in an Urban Watershed: Patroon Creek, Albany, NY. Geology Theses and Dissertations.

Budihardjo, M.A., Ramadan, B.S., Yohana, E., Syafrudin, Rahmawati, F., Ardiana, R., Susilo, D.B., Ikhlas, N., Karmilia, A., 2021. A review of anaerobic landfill bioreactor using leachate recirculation to increase methane gas recovery. IOP Conf. Ser. Earth Environ. Sci. 894, 012013 https://doi.org/10.1088/1755-1315/894/1/012013.

Calabrò, P.S., Lisi, R., 2014. Generation of renewable energy on closed landfills to compensate for biogas emissions: a case study in the Valdina landfill (Messina, Italy). Glob. NEST J. Gen. Renew. Energy Closed Landfills 17, 130–139.

Calabrò, P.S., Mancini, G., 2012. Possible interactions between recirculated landfill leachate and the stabilized organic fraction of municipal solid waste. Waste Manag. Res. 30, 551–557. https://doi.org/10.1177/0734242X11433527.

Calabrò, P.S., Sbaffoni, S., Orsi, S., Gentili, E., Meoni, C., 2010. The landfill reinjection of concentrated leachate: findings from a monitoring study at an Italian site. J. Hazard. Mater. 181 https://doi.org/10.1016/j.jhazmat.2010.05.107.

Calabrò, P.S., Orsi, S., Gentili, E., Carlo, M., 2011. Modelling of biogas extraction at an Italian landfill accepting mechanically and biologically treated municipal solid waste. Waste Manag. Res. 29, 1277–1285. https://doi.org/10.1177/0734242X11417487.

Calabrò, P.S., Gentili, E., Meoni, C., Orsi, S., Komilis, D., 2018. Effect of the recirculation of a reverse osmosis concentrate on leachate generation: a case study in an Italian landfill. Waste Manag. 76, 643–651. https://doi.org/10.1016/j.wasman.2018.03.007.

Calabrò, P.S., Bilardi, S., Moraci, N., 2021. Advancements in the use of filtration materials for the removal of heavy metals from multicontaminated solutions. Curr. Opin. Environ. Sci. Health 20. https://doi.org/10.1016/J.COESH.2021.100241.

Chamem, O., Fellner, J., Zairi, M., 2020. Ammonia inhibition of waste degradation in landfills – a possible consequence of leachate recirculation in arid climates. Waste Manag. Res. 38, 1078–1086. https://doi.org/10.1177/0734242X20920945.

Damikouka, I., Katsiri, A., 2021. Natural attenuation in marine sediments: investigation of the effect of chloride concentration on the mobility of metals. Environ. Sci. Pollut. Res. 28, 59180–59189. https://doi.org/10.1007/s11356-020-09852-4.

Evangelou, A., Calabrò, P.S., Greco, R., Sánchez, A., Komilis, D., 2016. Biodegradation activity of eight organic substrates: a correlation study of different test methods. Waste Biomass Valoriz. 7, 1067–1080. https://doi.org/10.1007/s12649-016-9532-2.

Farrell, M., Jones, D.L., 2009. Critical evaluation of municipal solid waste composting and potential compost markets. Bioresour. Technol. 100, 4301–4310. https://doi.org/10.1016/j. biortech.2009.04.029.

Fazzino, F., Folino, A., Mauriello, F., Pedullà, A., Calabrò, P.S., 2021. Biofuel production from fruit and vegetable market waste and mature landfill leachate by an active filter-anaerobic digestion integrated system. Energy Convers. Manag. X 12, 100130. https://doi.org/10.1016/j.ecmx.2021.100130.

Frikha, Y., Fellner, J., Zairi, M., 2017. Leachate generation from landfill in a semi-arid climate: a qualitative and quantitative study from Sousse, Tunisia. Waste Manag. Res. 35, 940–948. https://doi.org/10.1177/0734242X17715102.

Ghaffariraad, M., Ghanbarzadeh Lak, M., Ebrahimi Sarindizaj, E., 2021. Sensitivity analysis of significant parameters affecting landfill leachate generation rate. Environ. Monit. Assess. 194, 12. https://doi.org/10.1007/s10661-021-09653-3.

He, R., Wei, X.-M., Tian, B.-H., Su, Y., Lu, Y.-L., 2015. Characterization of a joint recirculation of concentrated leachate and leachate to landfills with a microaerobic bioreactor for leachate treatment. Waste Manag. 46, 380–388. https://doi.org/10.1016/j.wasman.2015.08.006.

Heinigin, P., 1995. Recirculation of leachate concentrate from reverse osmosis. In: Proceedings Sardinia 95, Fifth International Landfill Symposium, vol. 2. CISA, Cagliari.

Jiang, J., Yang, G., Deng, Z., Huang, Y., Huang, Z., Feng, X., Zhou, S., Zhang, C., 2007. Pilot-scale experiment on anaerobic bioreactor landfills in China. Waste Manag. 27, 893–901. https://doi.org/10.1016/j.wasman.2006.07.008.

Kjeldsen, P., Barlaz, M.A., Rooker, A.P., Baun, A., Ledin, A., Christensen, T.H., 2002. Present and long-term composition of MSW landfill leachate: a review. Crit. Rev. Environ. Sci. Technol. 32, 297–336. https://doi.org/10.1080/1064 3380290813462.

Lee, H., Coulon, F., Beriro, D.J., Wagland, S.T., 2023. Increasing recovery opportunities of metal(loid)s from municipal solid waste via landfill leachate recirculation. Waste Manag. 158, 116–124. https://doi.org/10.1016/j.wasman.2023.01.011.

Lewis, A.E., 2010. Review of metal sulphide precipitation. Hydrometallurgy 104, 222–234. https://doi.org/10.1016/j.hydromet.2010.06.010.

Li, W., Zhou, Q., Hua, T., 2010. Removal of organic matter from landfill leachate by advanced oxidation processes: a review. Int. J. Chem. Eng. 2010, e270532 https://doi.org/10.1155/2010/270532.

Liu, Y., Li, X., Wang, B., Liu, S., 2008. Performance of landfill leachate treatment system with disc-tube reverse osmosis units. Front. Environ. Sci. Eng. China 2, 24–31. https://doi.org/10.1007/s11783-008-0024-x.

Long, Y., Hu, L., Shen, D.-S., 2009. Nitrogen transformation in the hybrid bioreactor landfill. Bioresour. Technol. 100, 2527–2533. https://doi.org/10.1016/j.biortech.2008.11.036.

Luo, H., Zeng, Y., Cheng, Y., He, D., Pan, X., 2020. Recent advances in municipal landfill leachate: a review focusing on its characteristics, treatment, and toxicity assessment. Sci. Total Environ. 703, 135468 https://doi.org/10.1016/j.scitotenv.2019.135468.

Ma, J., Li, Yijia, Li, Yuqian, 2021. Effects of leachate recirculation quantity and aeration on leachate quality and municipal solid waste stabilization in semi-aerobic landfills. Environ. Technol. Innov. 21, 101353 https://doi.org/10.1016/j.eti.2021.101353.

Marshall, R., Browell, D., Smith, S., 2010. Guidance on Monitoring Landfill Gas Surface emissionsThe Environment Agency. Out There, Making your Environment a Better Place. Environment Agency, Bristol.

Molleda, A., López, A., Cuartas, M., Lobo, A., 2020. Release of pollutants in MBT landfills: laboratory versus field. Chemosphere 249, 126145. https://doi.org/10.1016/j.chemosphere.2020. 126145.

Moody, C.M., Townsend, T.G., 2017. A comparison of landfill leachates based on waste composition. In: Waste Management, Special Thematic Issue: Sanitary Landfilling, 63, pp. 267–274. https://doi.org/10.1016/j.wasman.2016.09.020.

Nappi, P., Valenzano, F., Consiglio, M., 2000. Analisi merceologica dei rifiuti urbani Rassegna di metodologie e definizione di una metodica di riferimento. ARPA, Roma (Italy).

Pantini, S., Verginelli, I., Lombardi, F., Scheutz, C., Kjeldsen, P., 2015. Assessment of biogas production from MBT waste under different operating conditions. Waste Manag. 43, 37–49. https://doi.org/10.1016/j.wasman.2015.06.019.

van Praagh, M., Heerenklage, J., Smidt, E., Modin, H., Stegmann, R., Persson, K.M., 2009. Potential emissions from two mechanically–biologically pretreated (MBT) wastes. Waste Manag. 29, 859–868. https://doi.org/10.1016/j.wasman.2008.06.028.

Qu, X., He, P.-J., Shao, L.-M., Lee, D.-J., 2008. Heavy metals mobility in full-scale bioreactor landfill: initial stage. Chemosphere 70, 769–777. https://doi.org/10.1016/j. chemosphere.2007.07.013.

Renou, S., Givaudan, J.G., Poulain, S., Dirassouyan, F., Moulin, P., 2008. Landfill leachate treatment: review and opportunity. J. Hazard. Mater. 150, 468–493. https://doi.org/10.1016/j.jhazmat.2007.09.077.

Robinson, H.D., Knox, K., Bone, B.D., Picken, A., 2005. Leachate quality from landfilled MBT waste. In: Waste Management, 1st UK Conference and Exhibition on Biodegradable and Residual Waste Management, 25, pp. 383–391. https://doi.org/10.1016/j.wasman.2005.02. 003.

Salati, S., Scaglia, B., Di Gregorio, A., Carrera, A., Adani, F., 2013. The use of the dynamic respiration index to predict the potential MSW-leachate impacts after short term mechanical biological treatment. Bioresour. Technol. 128, 351–358. https://doi.org/10.1016/j.biortech.2012.10.101.

Šan, I., Onay, T.T., 2001. Impact of various leachate recirculation regimes on municipal solid waste degradation. J. Hazard. Mater. 87, 259–271. https://doi.org/10.1016/S0304-3894(01)00290-4.

Shou-liang, H., Bei-dou, X., Hai-chan, Y., Shi-lei, F., Jing, S., Hong-liang, L., 2008. In situ simultaneous organics and nitrogen removal from recycled landfill leachate using an anaerobic–aerobic process. Bioresour. Technol. 99, 6456–6463. https://doi.org/10.1016/j.biortech.2007.11.047.

Siddiqui, A.A., Richards, D.J., Powrie, W., 2012. Investigations into the landfill behaviour of pretreated wastes. Waste Manag. 32, 1420–1426. https://doi.org/10.1016/j.wasman.2012. 03.016.

Sluiter, A., Hames, B., Ruiz, R., Scarlata, C., Sluiter, J., Templeton, D., Crocker, D., 2012. Determination of Structural Carbohydrates and Lignin in Biomass, Technical Report NREL/.TP-510-42618.

Torretta, V., Ferronato, N., Katsoyiannis, I.A., Tolkou, A.K., Airoldi, M., 2017. Novel and conventional technologies for landfill leachates treatment: a review. Sustainability 9, 9. https://doi.org/10.3390/su9010009.

Tran, H.N., Münnich, K., Fricke, K., Harborth, P., 2014. Removal of nitrogen from MBT residues by leachate recirculation in combination with intermittent aeration. Waste Manag. Res. 32, 56–63. https://doi.org/10.1177/0734242X13512892.

van Turnhout, A.G., Brandstätter, C., Kleerebezem, R., Fellner, J., Heimovaara, T.J., 2018. Theoretical analysis of municipal solid waste treatment by leachate recirculation under anaerobic and aerobic conditions. Waste Manag. 71, 246–254. https://doi.org/10.1016/j.wasman.2017.09.034.

Wiszniowski, J., Robert, D., Surmacz-Gorska, J., Miksch, K., Weber, J.V., 2006. Landfill leachate treatment methods: a review. Environ. Chem. Lett. 4, 51–61. https://doi.org/10.1007/s10311-005-0016-z.

Yesilnacar, M.I., Cetin, H., 2005. Site selection for hazardous wastes: a case study from the GAP area, Turkey. Eng. Geol. 81, 371–388. https://doi.org/10.1016/j.enggeo.2005.07.012.

Yu, Y., Rowe, R.K., 2021. Numerical modeling of clogging of landfill leachate collection systems with co-disposal of municipal solid waste (MSW) and incinerator ash. Can. Geotech. J. 58, 83–96. https://doi.org/10.1139/cgj-2019-0851.

Zhang, Q.-Q., Tian, B.-H., Zhang, X., Ghulam, A., Fang, C.-R., He, R., 2013. Investigation on characteristics of leachate and concentrated leachate in three landfill leachate treatment plants. Waste Manag. 33, 2277–2286. https://doi.org/10.1016/j.wasman.2013. 07.021.