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# Boosting the circularity of waste management: pretreated mature landfill leachate enhances the anaerobic digestion of market waste

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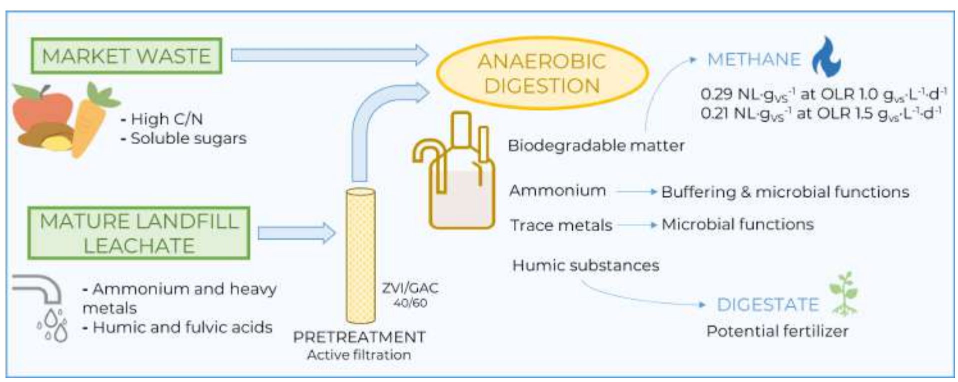
## HIGHLIGHTS

- Recovery of mature landfill leachate (MLL) from a full-scale landfill was explored.
- MLL was pretreated to prepare it for further use in anaerobic digestion (AD).
- Pretreated MLL was used as a nutrient solution for the AD of market waste.
- The process irreversibly failed in the reactor without MLL addition after 47 d.
- MLL allowed a successful AD at an increased organic loading rate of 1.5 $g_{VS} \cdot L^{-1} \cdot d^{-1}$.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Adequate waste management is essential not only to ensure healthy living conditions but also to mitigate climate change. Accordingly, the research on developing strategies to boost the circularity of waste management systems is ongoing. In this context, two waste streams are concurrently managed to recover energy and materials in the present study. Specifically, real leachate collected from a full-scale mature landfill site was preliminarily treated through active filtration to remove inhibitory substances partially and then tested, at the laboratory scale, as a nutrient solution for semi-continuous anaerobic digestion of a carbonaceous substrate represented by market waste. The results demonstrate that, at an organic loading rate of 1.0 $g_{VS} \cdot L^{-1} \cdot d^{-1}$, the process was impossible without using the nutrient solution, while the nitrogen present in the pretreated leachate could balance the carbon content of the market waste and provide the system with the necessary buffering capacity, ensuring process stability. The average methane yield (approximately 0.29 $NL \cdot g_{VS}^{-1}$) was satisfactory and consistent with the literature. Despite the increases in both the organic loading rate (up to 1.5 $g_{VS} \cdot L^{-1} \cdot d^{-1}$) and volume of added pretreated leachate (up to 100% of the dilution medium), the process remained stable with a slightly lower methane yield of 0.21 $NL \cdot g_{VS}^{-1}$, thanks to nitrogen supplementation. The potential use of produced methane as a renewable energy source and residual digestate as fertilizer would close the loop of managing these waste streams.



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**Contents**



**Abbreviations**

| | |
|---|---|
| AD | Anaerobic digestion |
| C/N | Carbon to nitrogen ratio |
| DIET | Direct interspecies electron transfer |
| EGSB | Expanded granular sludge bed |
| FOS/TAC | Volatile organic acids to buffering capacity ratio |
| FVW | Fruit and vegetable waste |
| FW | Food waste |
| GAC | Granular activated carbon |
| GHG | Greenhouse gas |
| HRT | Hydraulic retention time |
| IPCC | Intergovernmental Panel on Climate Change |
| MLL | Mature landfill leachate |
| MW | Market waste |
| OLR | Organic loading rate |
| PMMA | Polymethyl methacrylate |
| SSL | Sewage sludge |
| TS | Total solids |
| VFA | Volatile fatty acids |
| VS | Volatile solids |
| WAS | Waste activated sludge |
| WM | Waste management |
| ZVI | Zero valent iron |

## 1. Introduction

Worldwide, annually, approximately $2\times10^3$ Tg (teragram) of municipal solid waste is generated; if commercial, industrial, construction, and demolition waste sources are included, the estimated value increases to $10\times10^3$ Tg (Kaza et al., 2018). Managing this high quantity is a critical and urgent issue to tackle. Indeed, waste management (WM) is (explicitly or implicitly) included in more than half of the 17 United Nations Sustainable Development Goals (Sharma et al., 2021). In particular, WM can play a significant role in mitigating climate change. In this regard, in the latest Intergovernmental Panel on Climate Change (IPCC) report, it was stated that WM accounted for approximately 3.9% of global greenhouse gas (GHG) emissions in 2019 (namely, 2327 $Tg_{CO2\text{-}eq}$) (Dhakal et al., 2022). However, the potential contribution of better WM to mitigate climate change considerably exceeds that value. In fact, it is estimated that improving WM practices at present can lead to a 10 – 15% (up to 20%) reduction in global GHG emissions. That ambitious target can be attained primarily through waste prevention and the reuse of end-of-life products (actions that prevent waste generation) and, secondarily, through recycling and other forms of waste recovery (European Council, 2008; Calabrò et al., 2015). The latter options generate closed loops in which resources derived from waste (such as materials and energy) remain in the economy for as long as possible so that GHG emissions (related to the supply of virgin materials or energy generation) can be partially or totally avoided. The sustainable recycling of spent lithium-ion batteries (Du et al., 2022), recovery of precious metals and nutrients from wastewater (Ghomi et al., 2020; Sniatala et al., 2023); extraction of chemicals from organic residues (Espro et al., 2021), and biofuel production (Malode et al., 2021) are instances of the ongoing research conducted to enhance the circularity of WM systems.

From this perspective, in the present study, a circular-oriented strategy to concurrently manage two different waste sources: market waste (MW) and mature landfill leachate (MLL), is presented. MW consists of both edible and inedible parts of fruits and vegetables discarded in either wholesale or retail markets and is produced in large quantities worldwide (Mattsson et al., 2018). Due to its high biodegradability, MW is used as feedstock for the anaerobic digestion (AD). AD technology is a fundamental tool for circular-oriented WM because of its final products: biogas (possibly upgraded to methane) and digestate. The former is normally employed in energy generation; the latter concentrates chemical compounds that typically make it suitable for agricultural use (Horváth et al., 2016). However, the AD of MW is hindered by some MW characteristics, namely, its high concentration of soluble, simple sugars (which can lead to the irreversible acidification of the system) and lack of macro- (i.e., nitrogen) and micro- (i.e., light and heavy metals) nutrients (which are essential to microbial metabolism) (Xu et al., 2018).

Leachate results from water percolating through landfilled waste (Ehrig and Stegmann, 2018). Worldwide, approximately 40% of waste is still disposed of in sanitary landfills (Kaza et al., 2018) because landfilling often represents the cheapest solution whenever the complete recovery of some materials appears practically unfeasible (Calabrò et al., 2015). Leachate produced by so-called mature landfills (i.e., > 10 yr old) has a slightly alkaline pH and is rich in hard or non-biodegradable organic matter. Moreover, ammonium and metals are present in significant and moderate quantities, respectively (Ehrig and Stegmann, 2018). As national and EU regulations mandate that landfill aftercare should continue for at least 30 yr following their closure (European Union, 1999), the treatment of MLL is expected to be an increasing necessity in the near future, especially in developed countries with mature WM systems (Calabrò and Satira, 2020). The vast majority of MLL treatment options aim to remove pollutants to meet discharge limits (Luo et al., 2020); however, increasing research attention is being paid to recovering useful substances from leachate. Albeit technically simple, conventional processes utilized for nitrogen recovery, such as precipitation (Di Iaconi et al., 2010) and stripping (dos Santos et al., 2020), are expensive in terms of the addition of reagents (pH control agents, chemical compounds, and compressed air) and electricity consumption (intense mixing, long aeration periods, and potentially heating), thus limiting their widespread application (Cossu et al., 2019). In this context, the use of MLL in AD can represent a cost-effective alternative as MLL is observed to i) improve buffering capacity, ii) supplement trace metals, iii) dilute suspended solid content, and iv) serve as a source of nitrogen to balance the carbon to nitrogen ratio (C/N) (Lv et al., 2021).

Considering these factors, the present study investigates the possibility of completely recovering valuable compounds and energy from MW and MLL by using AD as the key process. More specifically, the soluble sugars of MW are converted to methane, and the performance of the process is enhanced by the presence of MLL as ammonia nitrogen as nutrients and by increasing the buffering capacity of the system. Additionally, nitrogen and hard or non-biodegradable compounds (i.e., humic substances) present in MLL are expected to improve the fertilizing properties of the digestate. To ensure the success of the experiment, an MLL pretreatment (through active filtration) was performed to reduce the concentration of bio-refractory and toxic compounds (e.g., the excessive amount of heavy metals), which, otherwise, would have either inhibited the process or made final digestate

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unsuitable for agriculture use. As shown in **Table 1**, only a few studies were focused on the use of MLL in AD processes over the last several years (2018-2022).

From **Table 1**, it can be concluded that the use of MLL as an additive to enhance the AD process' performance is attracting attention in the field as it appears that MLL presence positively affects the critical stages of AD: i) hydrolysis and acidification, which are known to be the rate-limiting steps of AD of sewage sludge (SSL) and waste activated sludge (WAS) (Xu et al., 2020), and ii) methanogenesis in the cases of AD of carbonaceous substrates. However, only slight increases in methane yields were recorded for the former. Conversely, the latter was observed to be considerably boosted by MLL compounds (especially nitrogen and trace metals) as they create favorable conditions for methanogenesis. On the other hand, it appears that MLL exerts beneficial effects on AD, mostly when added in modest volumes (especially with SSL and WAS, which are usually already rich in nitrogen and micronutrients).

The use of MLL in the AD of the fruit and vegetable residues produced by wholesale or retail markets (i.e., MW) is relatively unexplored in the literature. de Quadros et al. (2022), Liu et al. (2022), and Peng et al. (2022) used almost equivalent carbonaceous substrates (food waste in general with a C/N considerably higher than the optimum value); however, in these cases, the integration with MLL was limited: 3% of the total substrate, up to 20% v/v, and 40% of the dilution medium, respectively. Hence, the present study tested the effect of adding considerably higher volumes of MLL (up to 100% of the dilution medium) on the AD to maximize MLL recovery quantitatively. This study is the continuation of the authors' previous preliminary work (Fazzino et al., 2021a), where the MLL used was prepared in a laboratory considering only the essential elements (e.g., Cu, Ni, and Zn were the only metals used), thus neglecting all the possible interactions both with an active filter (during pretreatment) and microbial community (during the subsequent AD). Furthermore, the AD of the present study was conducted using a more sophisticated (and automatic) semi-continuous AD system (i.e., a standard continuous stirred tank reactor) than the previous work (Fazzino et al., 2021a), where the reactors used were very simple and the operations of feeding, discharge, and methane collection were performed manually. Finally, as aforementioned, in the present experiment, the dosage of MLL was increased up to 100% of the dilution medium (with a concurrent increase in the organic loading rate, OLR), respective to the previous studies.

## 2. Materials and Methods

### 2.1. Leachate

The leachate used in this experiment was sampled from a landfill site located in Alli (Catanzaro Province, Calabria Region, Italy), where mainly mixed waste was disposed of. Its physical properties and chemical composition were analyzed according to the standard methods (APHA et al., 2012). In **Table 2**, the main characteristics of the raw MLL are provided (the concentrations of other pollutants can also be found in *Section 3.1*). According to these analyses, the raw leachate could be considered mature based on the attributes provided by Renou et al. (2008).

**Table 2.**
Raw mature landfill leachate (MLL) characterization (main parameters).

| Parameter | Value |
|---|---|
| Unit weight [g·cm$^{-3}$] | 1.00 |
| pH | 8.31 |
| Total solids [%] | 0.6 |
| Volatile solids [%] | < 0.1 |
| $BOD_5$ [mg·kg$^{-1}$] | 762.0 |
| COD [mg·kg$^{-1}$] | 1881.0 |
| $NH_4$-N [mg·kg$^{-1}$] | 1650.8 |

### 2.2. Pretreatment test

Following our previous study (Fazzino et al., 2021b), the first step of the entire experiment consisted of the pretreatment of the MLL. In this case, using a real MLL allowed the validation of the preliminary results as all the possible interactions among the solution constituents and active materials were considered. The pretreatment was simulated at a laboratory scale through a column composed of polymethyl methacrylate (PMMA) – Plexiglas (internal diameter and height of 5 ± 0.1 and 50 cm, respectively)

**Table 1.**
Latest research studies (2018-2022) on the use of mature landfill leachate (MLL) in the anaerobic digestion (AD) processes.

| Substrate | Reactor Type | Operational Conditions | Methane Production | MLL Effect | Reference |
|---|---|---|---|---|---|
| SSL* + pretreated (through cavitation) MLL (at 5% v/v) | Semi-flow mode | - OLR: 1.20 – 1.45 $g_{VS}$·L$^{-1}$·d$^{-1}$<br>- HRT: 20 d | Highest yield: 0.28 ± 0.05 L·$g_{VS}^{-1}$ | Improving buffering capacity | Montusiewicz et al. (2018) |
| SSL + MLL (at 0%, 5%, 10%, 20% and 40% v/v) | Batch mode | - | + 36% (only for MLL at 10% v/v) than the control | Increasing methane yield (only with MLL at 10% v/v) | Berenjkar et al. (2019) |
| WAS + MLL at VS ratios of 1:0, 20:1, 10:1, 7:1, 5:1 | Batch mode | - | Highest yield: about 110 mL·$g_{VS}^{-1}$ for MLL at the VS ratio of 10:1 | Promoting hydrolysis and acidification (but hindering methanogenesis) | Gao et al. (2021) |
| MW + pretreated (through active filtration) synthetic MLL to adjust C/N at 25 | Semi-continuous mode | - OLR: 1 $g_{VS}$·L$^{-1}$·d$^{-1}$<br>- HRT: 20 d | 0.260 NL·$g_{VS}^{-1}$<br>0.302 NL·$g_{VS}^{-1}$ with GAC addition to promote DIET | Balancing C/N and providing the microbial community with macro- and micronutrients | Fazzino et al. (2021a) |
| FVW + MLL at a ratio of 9.7:0.3 to adjust C/N at 25 | Batch mode | - | 138.7 NmL·$g_{VS}^{-1}$ | Improving buffering capacity | de Quadros et al. (2022) |
| FW + MLL (at 10% or 20% v/v) | Expanded granular sludge blanket reactor | - OLR gradually increased from 0.9 to 23.6 $g_{COD}$·L$^{-1}$·d$^{-1}$<br>- HRT gradually decreased from 120 to 44 h | Highest rate: 5.87 ± 0.45 L·L$^{-1}$·d$^{-1}$ | Providing ammonium to avoid acidification and trace metals for microbial growth | Liu et al. (2022) |
| FW + MLL (from 0% to 100% v/v of the dilution medium) | Batch mode | - | The highest yield within 350 and 400 mL·$g_{VS}^{-1}$ for MLL at 40% v/v of dilution medium | Accelerating the digestion kinetics and increasing the methane yields | Peng et al. (2022) |
| FW + MLL at 40% v/v of dilution medium | Semi-continuous mode | - OLR: 3 $g_{VS}$·L$^{-1}$·d$^{-1}$<br>- HRT: 30 d | 473.95 ± 18.61 mL·$g_{VS}^{-1}$ | Enhancing ammonia tolerance | |

* *Abbreviations:* SSL: sewage sludge, MLL: mature landfill leachate, OLR: organic loading rate, HRT: hydraulic retention time, WAS: waste activated sludge, VS: volatile solids, MW: market waste, C/N: carbon to nitrogen ratio, GAC: granular activated carbon, DIET: direct interspecies electron transfer, FVW: fruit and vegetable waste, and FW: food waste.

filled with a granular mixture of zero-valent iron (ZVI) and granular activated carbon (GAC) at a weight ratio of 40:60 (300 and 450 g of ZVI and GAC, respectively). The ZVI was type FERBLAST RI 850/3.5 and distributed by Pometon S.p.A. (Mestre, Italy), whereas the GAC (type CARBOSORB 2040) was provided by Comelt S.r.l. (Milan, Italy). The sizes of ZVI and GAC particles were similar (0.85–0.425 mm). The MLL was fed to the column by a peristaltic pump (Watson Marlow 205S) under a constant upward flow of 0.68 $mL·min^{-1}$, equivalent to 0.5 $m^3·d^{-1}$ of leachate per $m^2$ of the granular filter. Through three sampling ports (i.e., at 3 cm from the inlet, in the middle of the column, corresponding to a height of 25 cm, and at the outlet), the MLL filtered through the active medium was periodically sampled during the test to monitor the changes occurring in the MLL in terms of pH (measured by a digital pH meter, XS instruments), color (determined by measuring the absorbance at a wavelength of 445 nm), COD and ammonium (measures using pre-dosed cuvettes, Merck Millipore COD Cell Test 114555 and Ammonium Cell Test 114559, respectively). The removal efficiency for these pollutants was calculated as shown in **Equation 1**:

$$R_E(\%) = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \quad \text{Eq. 1}$$

where $C_{in}$ and $C_{out}$ represent the pollutants' concentrations in the MLL at the inlet and outlet of the column, respectively.

Following 32 d of continuous operation, the column test was stopped. The pretreated MLL produced during the entire test period was stored at 4°C before its use in the subsequent AD test.

### *2.3. Semi-continuous anaerobic digestion test*

Evaluating the use of the pretreated MLL in the AD of MW represented the second step of the entire experiment, as previously mentioned (Fazzino et al., 2021a). However, this study used a more sophisticated semi-continuous AD system (Bioprocess Control Bioreactor, BPC Instruments). It consisted of two glass reactors (designed as A and B) equipped with an automatic internal stirrer and immersed in a thermostatic water bath (35 °C). The BPC system allowed for the simultaneous feeding and discharge of the reactors. Methane produced during the process was automatically measured by a patented system based on water/gas displacement.

According to an internal quality control test, the uncertainty of the measurements of cumulated biomethane production performed on a single semi-continuous AD test was in the order of 5–15%. This value can be compared, for example, using the Italian Norm (UNI/TS 11703:2018), which tolerates variations of ± 25% between the experimental and theoretical values of the biomethane potential of microcrystalline cellulose (used as the control for the procedure). This substrate is easily biodegradable, and the procedure used to perform biomethane potential tests for it is much simpler than the semi-continuous tests performed in this investigation.

The inoculum used in the AD test was a residual digestate stored at 35°C obtained from previous AD tests. As a substrate, a representative sample of synthetic MW was prepared by shredding and homogenizing common commercial fruits and vegetables, such as apples, potatoes, and carrots (45, 49, and 6%, w/w, respectively, as described previously by Fazzino et al. (2021a)). Both the inoculum and substrate were characterized in terms of pH, total solids (TS), and volatile solids (VS), according to the standard methods (APHA et al., 2012), whereas the C/N of MW was determined through an elemental analyzer TOC-LCSH (Shimadzu, Kyoto, Japan) (**Table 3**). In particular, the synthetic MW mimicked the specific characteristics reported in the literature (Bouallagui et al., 2005). The pretreated MLL obtained from the previous column test was used as the nutrient solution.

After initially loaded with inoculum and water up to the working volume of 1.8 L, both reactors were fed (usually five times per week) with 12.3 $g·d^{-1}$ of MW based on a fixed organic loading rate (OLR) of 1.0 $g_{VS}·L^{-1}·d^{-1}$. The hydraulic retention time (HRT) was set to 20 d, corresponding to a feeding volume of 90 $mL·d^{-1}$. To attain the optimal value for the C/N of 25 (Steinhauser and Deublein, 2011), in reactor A, some of the water to be added to attain the set volume was substituted with the pretreated MLL (i.e., 48.2 $mL·d^{-1}$, approximately 54% v/v). Instead, reactor B, designed as the control, was fed with MW, and only tap water was used as the dilution medium. The first 7 d of the test was considered a period of microbial adaptation. The reactor B test was stopped after 47 d (i.e., approximately 2 HRT following the acclimation period) because of the reasons explained in *Section 3*, whereas, from the same day, the OLR in reactor A was increased up to 1.5 $g_{VS}·L^{-1}·d^{-1}$ so that the pretreated MLL constituted the only dilution medium (i.e., 90 $mL·d^{-1}$, 100% v/v) maintaining the same C/N in the feeding (i.e., 25).

**Table 3.** Inoculum, market waste (MW), and pretreated mature landfill leachate (MLL) characterization.

| Parameter | Inoculum | MW | Pretreated MLL |
|---|---|---|---|
| pH | 7.50 | 4.66 | 8.50 |
| Total solids [%] | 3.0 | 15.8 | 0.7 |
| Volatile solids [%TS] | 68.5 | 92.5 | 71.4 |
| C [$mg·kg^{-1}$] | - | 413700 | - |
| N [$mg·kg^{-1}$] | - | 10920 | 1433.5 |
| C/N | - | 37.9 | - |

The performance of the AD test was evaluated based on methane production. The results were presented in terms of both methane yield (i.e., the volume of methane produced per mass of VS added to the reactor) and cumulative methane production (i.e., the total volume of methane generated in the reactor throughout the test). Furthermore, to evaluate the stability of the processes based on the digestates collected during each feeding/discharge operation, the pH level was directly measured, whereas the analyses of other parameters (i.e., TS, VS, COD, ammonium, total volatile fatty acids, VFA, and volatile organic acids/buffering capacity ratio (FOS/TAC) were conducted on weekly composite samples. TS and VS were measured for raw digestates, whereas COD, ammonium, VFA, and FOS/TAC were determined for the liquid fractions resulting from centrifugation (10000 rpm for 10 min). Moreover, VFA and FOS/TAC were determined through a four-point titration method (Liebetrau et al., 2016). To identify possible differences between the two reactors, the correlation coefficient of the daily methane production was calculated.

## 3. Results and Discussion

### *3.1. Pretreatment test*

**Table 4** presents the pH trends of filtered MLL samples obtained from the three sampling ports throughout the test. As expected, the pH values generally increased both along the column (i.e., along the granular filter) and over the test time. This behavior was caused by the dissolution of the ZVI, which occurs due to the interaction between the ZVI and MLL in anaerobic and aerobic conditions. As a result, hydroxyl ions are released according to **Equations 2** and **3** (Moraci et al., 2016):

$$Fe^0 + 2H_2O \rightarrow Fe^{2+} + 2H_2 + 2OH^- \quad \text{Eq. 2}$$

$$Fe^0 + O_2 + 2H_2O \rightarrow Fe^{2+} + 4OH^- \quad \text{Eq. 3}$$

The difference between the pH values at the bottom (i.e., 3 cm) and the outlet of the column in the final days of the test could indicate that the active granular filter was not totally exhausted yet.

**Table 4.**
pH measured in the effluents from the three sampling ports of the column used for the mature landfill leachate (MLL) pretreatment.

| Test day | Sampling port | | |
|---|---|---|---|
| | 3 cm | 25 cm | Outlet |
| 4 | 8.42 | 8.80 | 8.87 |
| 7 | 8.46 | 8.84 | 8.86 |
| 11 | 8.35 | 8.72 | 8.70 |
| 14 | 8.38 | 8.80 | 8.85 |
| 22 | 8.11 | 8.90 | 8.78 |
| 25 | 8.07 | 8.92 | 8.84 |
| 31 | 8.66 | 9.06 | 9.25 |

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In **Figure 1**, color, COD, and ammonium trends are depicted. Regardless of the sampling port, the color removal efficiency decreased over time. This was also visually evident as samples withdrawn from the outlet on day 4 were far brighter than those withdrawn from the other ports and those withdrawn on the other days. Color reduction in the first days was consistent with other studies performing the filtration of an MLL (Rohers et al., 2021). In the first days of the test, COD and ammonium were scarcely removed and only present at the bottom of the filter (i.e., 14 and 24%, respectively, at 3 cm from the bottom). COD removal efficiency exhibited a peak (i.e., 36%) at the outlet approximately halfway through the test, whereas a modest ammonium removal efficiency (i.e., 49%) was achieved at the outlet only at the end of the test.

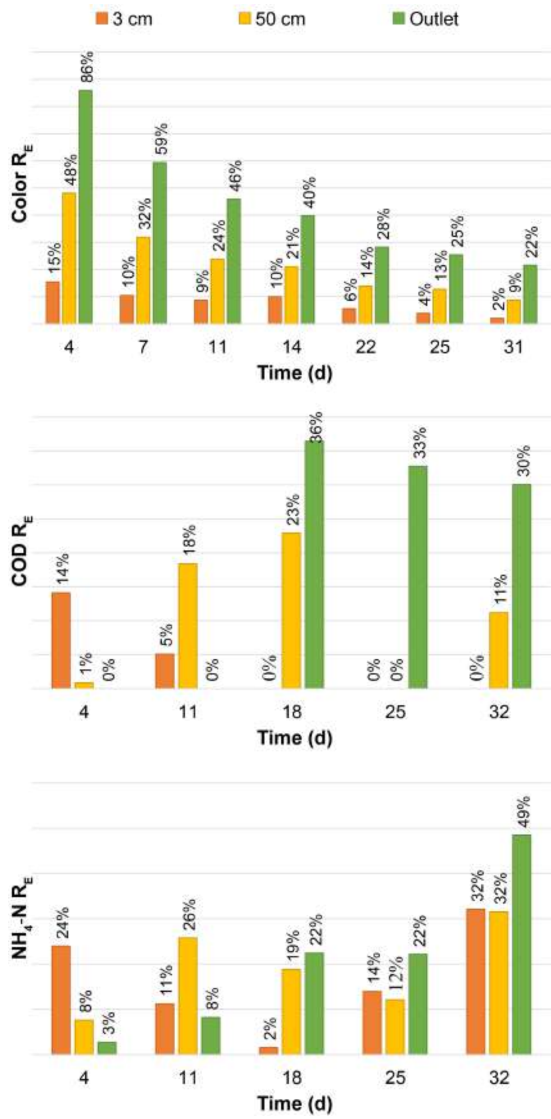


**Fig. 1.** Color, COD, and ammonium removal efficiencies determined in samples obtained from the three sampling ports of the column used for the mature landfill leachate (MLL) pretreatment process.

Contrary to the study conducted by Fazzino et al. (2021b), in which the maximum COD and ammonium removal efficiencies were achieved at the beginning of the test, in this study, the active filter took more time to react with the MLL, probably due to the presence of a plethora of constituents not considered using a synthetic MLL. Most probably, COD removal occurred through mechanisms of i) the adsorption of organic compounds in the micropores of the GAC until the exhaustion of the accumulation sites (Mohammad-pajooh et al., 2018) and ii) the entrapment in ZVI corrosion products (Zhou et al., 2014; Bilardi et al., 2020b). Instead, ammonium concentration was reduced by the ZVI action (i.e., electrostatic attraction, ion exchange, and adsorption on corrosion products (Eljamal et al., 2022), whereas GAC contribution was probably negligible as its nonpolar surface results in poor interactions with polar adsorbates (Halim et al., 2010). Thus, it can be speculated that ZVI's removal action was progressively activated in relation to the gradual formation of the corrosion products. Indeed, the pollutants' removal was firstly recorded at the bottom of the column where the contact between the MLL and granular filter began (see COD and ammonium removal efficiencies at 3 and 25 cm in days 4 and 11, respectively, **Fig. 1**), and only later at the top (greater removal efficiencies at the outlet in days 18, 25, and 32, **Fig. 1**).

**Table 5** summarizes the concentrations of pollutants (i.e., metals and bio-refractory compounds) in the MLL samples before and after the pretreatment (composite sample of the whole active filtration test). Only copper, selenium, and hydrocarbons exceeded the concentration limits for the discharge in surface water imposed by the National Regulation. However, the starting concentrations of these metals were not unusual for the MLL. Indeed, as previously mentioned, MLL management must mainly address issues related to refractory dissolved organic matter and ammonia rather than the presence of metals (Ehrig and Stegmann, 2018). Therefore, the real MLL used in the present experiment was consistent with the literature. Barium content was significantly reduced (by approximately 96%), which was in line with the results of the previous batch adsorption tests conducted using activated carbons (Kaveeshwar et al., 2018). Likewise, zinc reduction (of approximately 67%) was consistent with the average results obtained by Fazzino et al. (2021b). On the other hand, the incapability of the pretreatment to reduce the chromium content from the MLL was expected, as chromium removal from wastewater through adsorbents is promoted by acidic conditions (pH level ranging from 2 to 6) (Gupta et al., 2013; Ambika et al., 2022). Finally, the significant increase in iron concentration following pretreatment was caused by the release of iron ions, as presented in **Equations 1** and **2**. Refractory organic compounds (such as hydrocarbons and benzene) were expected to be considerably reduced by GAC due to its capacity to absorb various organic substances, even at low concentrations (Bansal and Goyal, 2005). Furthermore, fats and oils were also satisfactorily reduced (Sultana et al., 2022). To summarize, it can be stated that the performance of the active granular filter towards the main pollutants was satisfactory, thus confirming the suitability of this type of technology for the pretreatment of a real MLL.

**Table 5.**
Pollutants' concentrations in the mature landfill leachate (MLL) before and after the pretreatment (composite sample).

| Pollutant | Concentration [mg·kg$^{-1}$] | |
|---|---|---|
| | **Raw MLL** | **Pretreated MLL (Composite Sample)** |
| Aluminum | 0.8 | 0.7 |
| Arsenic | 0.1 | < 0.1 |
| Barium | 2.4 | 0.1 |
| Chromium (total) | 0.4 | 0.4 |
| Iron | 2.3 | 39.5 |
| Nickel | 0.2 | 0.1 |
| Copper | 0.1 | 0.1 |
| Selenium | 0.1 | < 0.1 |
| Tin | 0.4 | 0.2 |
| Vanadium | 0.1 | < 0.1 |
| Zinc | 0.3 | 0.1 |
| Hydrocarbons (total) | 14 | < 5 |
| Fats and oils | 14 | < 5 |
| Benzene | 0.2 | < 0.1 |
| $NH_4$-N | 1651 | 945 |
| Chloride | n.d.* | 3012 |

* n.d.: not determined.

### *3.1.2. Semi-continuous anaerobic digestion test*

**Figures 2a** and **b** present methane yields and cumulative methane yields, respectively, for reactors A and B. Methane production at the beginning of

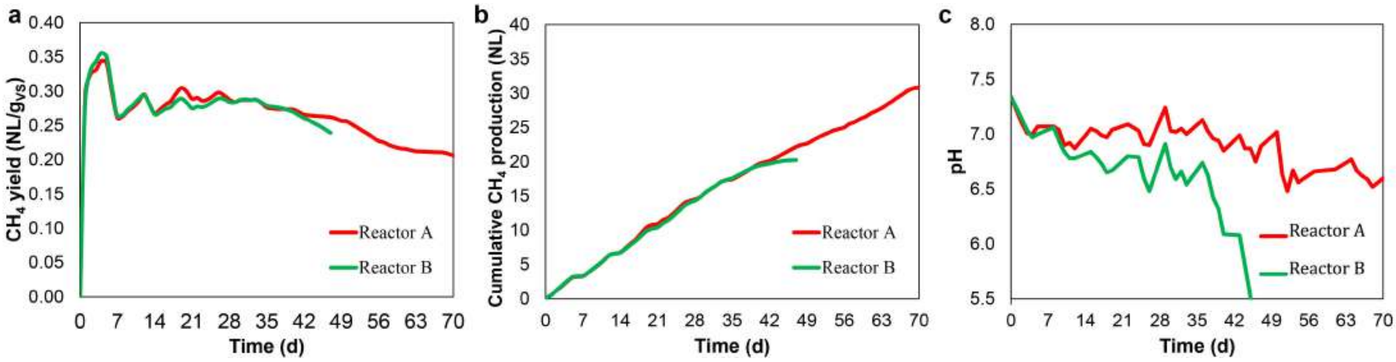


**Fig. 2.** Trends of **(a)** methane yield, **(b)** cumulative methane production, and **(c)** pH for reactors A (digestion of market waste, MW, and pretreated mature landfill leachate, MLL) and B (digestion of MW only).

the AD test (see peaks of 0.34 and 0.36 NL·$g_{VS}^{-1}$ for reactors A and B, respectively, on day 4, in **Fig. 2a**) was greater than for the rest of the test as the residual organic matter of the starting inoculum was degraded. After approximately 7 d, counted as a period of microbial adaptation, methane yields of both reactors started to fluctuate between 0.26 NL·$g_{VS}^{-1}$ (day 8) and 0.30 NL·$g_{VS}^{-1}$ (day 19), with methane production from reactor A appearing to be slightly greater than that from reactor B (see also cumulative methane productions' trends presented in **Fig. 2b**). However, from days 7 to 34, the average methane yields for both reactors were almost equal (i.e., 0.29 NL·$g_{VS}^{-1}$). From day 34 onwards, methane production of both reactors tended to decrease. Specifically, in reactor B, the methane yield showed a sharp, downward trend and stopped on day 47 for reasons explained in the following paragraphs. Similarly, the methane yield of reactor A slightly decreased compared to the previous days of the test; however, it seemed to stabilize around the value of 0.26 NL·$g_{VS}^{-1}$. In response to the OLR increase (1.5 $g_{VS}$·$L^{-1}$·$d^{-1}$) on day 47, methane production by reactor A steadily decreased until day 63, but then it remained stable around 0.21 NL·$g_{VS}^{-1}$ until the end of the test.

The correlation coefficients of the daily methane production rates of the two reactors were extremely high (>99%, using the data until the 14th day and >97% from the 15th to the 22nd days), confirming the similarity of the processes occurring in the two reactors at the beginning of the experiment. This may be explained by the presence of sufficient nitrogen and micronutrients supplied by the inoculum, which enhanced the biological processes. The differences between reactor A, receiving the pretreated leachate, and reactor B, where nutrients and especially nitrogen were lacking, began to progressively grow after approximately 1 HRT, as witnessed by the rapid decrease in the correlation coefficient that was dramatic in the final 6 days of operation for reactor B (a decrease from 0.94 to 0.85).

**Figures 2c** depicts the trends of pH level, which is essential to understand the evolution of the AD process. At the beginning of the test, the pH value was equal in both reactors (i.e., 7.34). However, after approximately a week of operation, the difference between reactors A and B became noticeable. Specifically, the pH values of reactor A were higher than those measured in reactor B over the entire test period. The pH level of reactor A remained around the optimal value for methanogens (frequently, it was higher than neutral) until day 50. Conversely, the pH trend of reactor B abruptly decreased from day 36, clearly presenting the acidification of the system. From day 47, reactor A also suffered a pH decrease because of the greater substrate input related to the increase in the OLR to 1.5 $g_{VS}$·$L^{-1}$·$d^{-1}$. Nevertheless, only once during the entire test (i.e., on day 52), it was necessary to adjust the pH level of the system with 4 g of $NaHCO_3$, as it was below the inhibition threshold of 6.5 (Speece, 1996). However, from day 52 to the end of the test, the process ran stably, with pH values ranging from 6.55 to 6.75, witnessing a progressive adaptation of the system to the new loading value.

As shown in **Figure 3a**, during the first two weeks of the test, the COD content in both reactors was similar (differences of 4 and 5%, respectively). Then, COD exhibited a decreasing trend in both reactors, with higher values determined in digestates obtained from reactor B than those measured in reactor A. However, in the seventh week, reactor B's COD content peaked at 4125 mg·$L^{-1}$, indicating the considerable presence of organic matter undigested by microbial population suffering from the acidification of the system and, as analyzed later, lack of nitrogen (see **Fig. 3b**). The accumulation of undigested material was also observed in reactor A following an increase in OLR. In fact, the COD content increased from 1710 mg·$L^{-1}$ in week 7 to 5770 mg·$L^{-1}$ during the final week of the test. However, in this case, the increase in COD did not have drastic consequences.

VFA trends were initially decreasing for both reactors (**Fig. 3c**). Then, from week 4 onwards, VFA contents in both reactors steadily increased in reactor B, regularly exhibiting higher VFA concentrations compared to reactor A. In general, the stability of the AD process is evaluated through the variation in VFA content over time rather than its absolute value. For instance, the sudden increases in VFA concentration indicate the accumulation of acids and likely acidification of the system. In light of this, the peak in the VFA content recorded in reactor B during the seventh week of the test (i.e., 2041 mg·$L^{-1}$) was a clear sign of the acidification of the system. On the contrary, in reactor A, no considerable variations in the VFA content were recorded until week 7. However, in response to the increase in the OLR, a clear accumulation of VFA occurred during the final weeks of the test, with the VFA content reaching 3276 mg·$L^{-1}$ in week 9. Similar to the COD, the VFA accumulation also seemed to have been tolerated by the system.

FOS/TAC values strictly ranging from 0.15–0.45 are a symptom of a well-balanced and stable process. Instead, the lower values indicate a lack of organic acids (essential to the AD process), possibly related to an excessively low substrate input value (i.e., low OLR), whereas higher values involve an excessive accumulation of organic acids (risk of acidosis of the system) (Weichgrebe, 2009). FOS/TAC in reactor A remained below the upper limit of the range until week 7 (**Fig. 3d**), so we decided to increase the OLR. From that point onwards, the ratio dramatically increased to the maximum value of 1.32 in week 9, but without having any severely adverse impacts on the methane production rate. Conversely, the FOS/TAC trend of reactor B exceeded the optimal range from the third week, reaching its highest value of 8.3 in the seventh week, confirming that the process had already been jeopardized.

Ammonium concentration is the key factor in understanding the two reactors' different behaviors. Both reactors were very similar from this perspective during the first week of the test (i.e., 280 mg·$L^{-1}$). Then, while the initial ammonium content was progressively consumed in reactor B, where its concentration was below 100 mg·$L^{-1}$ in the third week, in reactor A, thanks to the addition of the pretreated MLL, the ammonium remained in the range considered stimulatory for AD (Yenigün and Demirel, 2013) for the entire test period. The greatest difference between ammonium concentrations in the two reactors was observed in the fourth week of the test (i.e., 539 *vs.* 29 mg·$L^{-1}$). In week 7, the situation in reactor B was so compromised that the test was stopped. Instead, in reactor A, the ammonium concentration remained stable until the end of the test, even when the OLR was increased.

The ammonium trend was responsible for the decline in methane production and pH value of reactor B, where the microbial consortium, suffering from a lack of nitrogen, reduced the conversion of VFA to

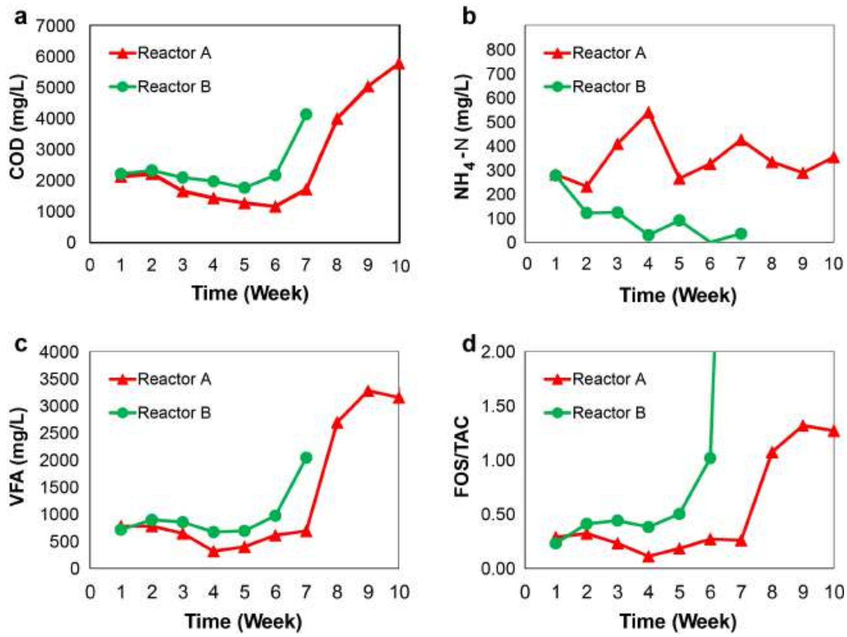


**Fig. 3.** Trends of **(a)** COD, **(b)** ammonium, **(c)** volatile fatty acids (VFA), and **(d)** volatile organic acids to buffering capacity ratio (FOS/TAC) determined during test time for weekly digestate samples obtained from the reactors A (digestion of market waste, MW, and pretreated mature landfill leachate, MLL) and B (digestion of MW only).

methane with the subsequent accumulation of these latter. Conversely, reactor A responded positively to pH fluctuations due to the presence of nitrogen added through the pretreated MLL. In fact, according to the ammonia – ammonium equilibrium, at a pH below 9.3, ammonium ions are formed with the release of hydroxyl ions ($NH_3 + H_2O \leftrightarrow NH_4^+ + OH^-$). Moreover, weak acidification is avoided due to the reaction between ammonia and VFA (represented by $C_xH_yCOOH$), as presented in **Equation 4** (Zhang et al., 2014):

$$C_xH_yCOOH + NH_3 \cdot H_2O \rightarrow C_xH_yCOO^- + NH_4^+ + H_2O \qquad \text{Eq. 4}$$

Thus, the increase in the buffering capacity of the system due to the addition of the pretreated MLL clearly implied a higher tolerance to VFA accumulation occurring in reactor A. Conversely, the lack of buffering agents in reactor B expectedly led to the acidification of the process. Previously, Liao et al. (2014) proved a similar result by performing a stable single-batch AD of food wastes (FW) with and without adding MLL. They recorded process failure in the reactor loaded with FW, only while dilutions performed with different amounts of MLL led to methane yields ranging from 369 mL·$g_{VS}^{-1}$ (MLL at 83% of the dilution medium) to 466 mL·$g_{VS}^{-1}$ (MLL at 55% of the dilution medium). In a study by Liu et al. (2022), the AD of FW performed in an expanded granular sludge bed (EGSB) reactor suffered from an inhibition of the methanogenic activity after the pH decreased to 5.48 due to VFA accumulation. By increasing the presence of MLL in the feed (from 10 to 20% v/v), the authors immediately recovered the process's performance thanks to the ammonium buffering action.

It is noteworthy, as in the present experiment, the fluctuations of pH levels followed the weekly feeding rate (**Fig. 2c**). Practically, pH trends peaked on Mondays (i.e., the first feeding day of the week), and then they decreased during the week down to the lowest values measured on Fridays (i.e., the final feeding day of the week). This behavior was related to the substrate degradation path. Indeed, substrate addition to the reactors for five consecutive days led to VFA production and unavoidable accumulation as methanogenic archaea, having a slower growth rate than other microorganisms involved in the AD process (Lim et al., 2020), were able to only partially convert all the produced VFA to methane. Accordingly, the pH levels of the systems decreased. When the reactors were not fed (i.e., on Saturdays and Sundays), methanogens completed the VFA conversion, increasing the pH levels of the systems. Concerning the AD of MW (and FW in general), this behavior is even more evident in the presence of highly degradable sugars in the substrate (Edwiges et al., 2018a). Thus, based on both the literature and the present study, it can be confirmed that the AD of FW and MW, and other similar highly degradable carbon-rich substrates, is virtually impossible without co-digestion being performed with nitrogen-rich substrates and/or the external supplementation of macro-nutrients (Xu et al., 2018). This is why the AD of food and MW is often conducted at low OLRs (below 3.0 $g_{VS}$·$L^{-1}$·$d^{-1}$) (Mata-Alvarez et al., 1992; Scano et al., 2014).

In this experiment, reactor A presented low values of both VFA concentration and FOS/TAC (**Figs. 3c** and **d**) at an OLR of 1.0 $g_{VS}$·$L^{-1}$·$d^{-1}$, indicating a possible underloading effect. The same conclusion was obtained by Fazzino et al. (2021a), where FOS/TAC values remained at approximately 0.15. For this reason, from day 47 onwards, OLR was increased to 1.5 $g_{VS}$·$L^{-1}$·$d^{-1}$ keeping the same proportions of MW and pretreated MLL to obtain the optimal C/N (i.e., 25) in the feeding input. This significantly increased the volume of added pretreated MLL until it completely replaced the dilution medium (i.e., 100%). As observed in the study, OLR increase stressed the system with consequent lower methane production (**Fig. 2**) and the accumulation of COD and VFA (**Figs. 3a** and **c**, respectively). However, methane yield (0.21 NL·$g_{VS}^{-1}$) was still satisfactory and consistent with the literature (see below), and COD and VFA contents, albeit far higher than the previous days, did not lead to process failure. This tolerance of the system was certainly due to its buffering capacity (as previously explained) but also possibly due to microbial adaptation to adverse conditions (such as high VFA contents and slightly acidic pHs levels), as also observed in the previous studies (Calabrò et al., 2018).

The average methane yield of 0.29 NL·$g_{VS}^{-1}$ calculated for reactor A without considering the acclimation phase (i.e., the first 7 days) was slightly higher than that obtained by Fazzino et al. (2021a) (i.e., 0.26 NL·$g_{VS}^{-1}$) at the same input conditions, but using simpler semi-continuous AD technology. However, in that previous study, the process performance was enhanced up to 0.30 NL·$g_{VS}^{-1}$ through the addition of GAC as a supplement triggering the direct interspecies electron transfer (DIET) mechanism. In this case, a similar result was achieved without using any supplement. As aforementioned, the literature concerning adding MLL to the AD of MW is virtually non-existent. However, the results obtained from the test conducted herein are consistent with the range of 0.16 – 0.40 L·$g_{VS}^{-1}$ (up to 0.50 L·$g_{VS}^{-1}$ determined under optimal conditions) reported by Bouallagui et al. (2005) for the AD of an almost equivalent substrate, such as fruit and vegetable waste (FVW). Edwiges et al. (2018b) tested increasing OLRs (up to 5 $g_{VS}$·$L^{-1}$·$d^{-1}$) in a semi-continuous AD of FVW. The best performance in terms of methane yield (i.e., 0.285 NL·$g_{VS}^{-1}$) was recorded at 3 $g_{VS}$·$L^{-1}$·$d^{-1}$.

The same result was obtained in this study, although at a lower OLR, thus reasonably suggesting that adding the pretreated MLL favorably affected the process. Likewise, a methane yield of 466 mL·$g_{VS}^{-1}$ determined by Liao et al. (2014) from the batch AD of food waste and MLL was only slightly higher than that obtained in the present study considering a reduction of approximately 30% due to the generally accepted scale-effects of AD tests (Ruffino et al., 2015). Additionally, Peng et al. (2022) obtained a significant result (i.e., 0.47 NL·$g_{VS}^{-1}$) from the semi-continuous AD of FW with MLL addition adopting an HRT and OLR of 30 d and 3 $g_{VS}$·$L^{-1}$·$d^{-1}$, respectively. The use of an HRT higher than that used in this study probably allowed the microbial consortium (especially methanogens) to degrade the input substrate more efficiently. However, both studies tested MLL only as a fraction of the dilution medium (55 and 40%, respectively), while in the present case, it was demonstrated that the AD process could run stably without signs of inhibition, even when the dilution medium was totally replaced by the pretreated MLL and OLR was significantly increased (although with a slightly lower methane yield).

In addition to the increase in the buffering capacity of the system, the macro- and micro-nutrients provided by the addition of the pretreated MLL played a crucial role in maintaining the stability of the process. Nitrogen is involved in the synthesis of proteins, enzymes, ribonucleic acid (RNA), and deoxyribonucleic (DNA) (Parkin and Owen, 1986), while trace metals are beneficial for the activity of some enzymes and stimulate the growth and activity of methanogens (Guo et al., 2019). Moreover, iron is a fundamental component as it also serves as both an electron acceptor and donor in oxidative-reductive reactions that occur (Lv et al., 2021). Accordingly, digesters should be provided with these elements, even if needed externally, to avoid deficiencies (Holliger et al., 2016). In this context, the partial or even total replacement of external solutions with pretreated MLL saves the considerable cost of the specific reagents and is consistent with the circular economy concept. During the feeding, nitrogen was added based on the C/N of the input, and metals' regime concentrations were lower than the inhibitory thresholds reported by Guo et al. (2019). Peng et al. (2022) determined the concentrations of metals in the digestate to be considerably lower than the upper limit recommended for organic fertilizers, and no accumulation of metals was observed during the AD process.

Furthermore, in the literature, the organic fraction of MLL is known to be dominated by humic and fulvic acids (Renou et al., 2008), which are refractory to biological transformations and are inhibitory for hydrolysis and methanogenesis (Li et al., 2019). As Liu et al. (2022) proved in their study, adding up to 20% of the feeding of MLL did not induce any inhibition from this perspective, as concentrations of humic substances remained below the inhibitory range of 0.5–0.8 g·$L^{-1}$. In the present study, although the added volume of pretreated MLL was larger, no signs of inhibition were recorded due to humic and fulvic acids' accumulation. In this case, the presence of stabilized organic fraction in the residual digestate could make it even more suitable for subsequent agricultural use.

## 4. Limitations of the present study

The present study had some limitations. Firstly, although the MW utilized in the study was representative of the typical composition of this waste stream, it was still synthetic; therefore, the study can be repeated using fruit and vegetable residues collected from wholesale or retail markets. Secondly, a statistical analysis was not conducted since the reactors were not replicated. However, replicates in studies addressing semi-continuous anaerobic digestion tests are not common in the literature (Liczbiński et al., 2022; Zhu et al., 2022) because these experiments are labor-intensive, very time-consuming, and expensive. Nevertheless, it would be useful to design at least two or three replicates per type of process in future tests. Finally, residual digestate quality was evaluated only in terms of its major characteristics (e.g., pH level, ammonium, COD, and VFA), and more specific analyses (i.e., pot tests) are required to fully confirm its suitability for agricultural use.

## 5. Practical implications of the present study

From a technical and economic perspective, a full-scale application of the overall process could significantly reduce the costs associated with MW and MLL management. First, the disposal of putrescible waste, such as MW, can be avoided, and soluble sugars can be used to generate methane. Concerning MLL, the preliminary filtration step can be directly performed on the landfill site. Exhausted filtration material can be recycled (e.g., to recover iron) or disposed of in the same landfill site. Subsequently, the pretreated MLL is dosed along with MW to the anaerobic digester according to specific volumes (which can be remarkable, as witnessed in the results of this study) to enhance process performance. The produced methane can be used for energy generation, thus partially compensating the impacts of uncontrolled landfill gas emissions and the energy used in the previous steps of the leachate treatment process. Moreover, digestate, if fully proven to be suitable, can replace the use of fertilizers due to the presence of nitrogen and humic substances derived from pretreated MLL.

## 6. Conclusions and prospects

The present study demonstrated the feasibility of using pretreated MLL as a nutrient solution in the AD of MW at a laboratory scale. From the perspective of enhancing the circularity of WM systems, the results of this study are promising. Indeed, by following a pretreatment method that reduced the concentrations of bio-refractory compounds, MLL was proven to prevent the failure of the AD of MW. As a result, methane yields of 0.29 and 0.21 NL·$g_{VS}^{-1}$ were produced at OLRs of 1.0 and 1.5 $g_{VS}$·$L^{-1}$·$d^{-1}$, respectively. In particular, the latter condition allowed the pretreated MLL to replace the dilution medium completely, thus saving the costs of water and reagents. Coupling the treatment of MLL with improved biofuel production through AD for C-rich substrates is an important step forward in the field to solve the important limitations of these forms of technology. Larger-scale experiments and life cycle analyses of the overall process would be necessary before the full-scale application of the process proposed in the present study can be realized.

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