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Effects of carbon-based conductive materials on semi-continuous anaerobic co-digestion of organic fraction of municipal solid waste and waste activated sludge

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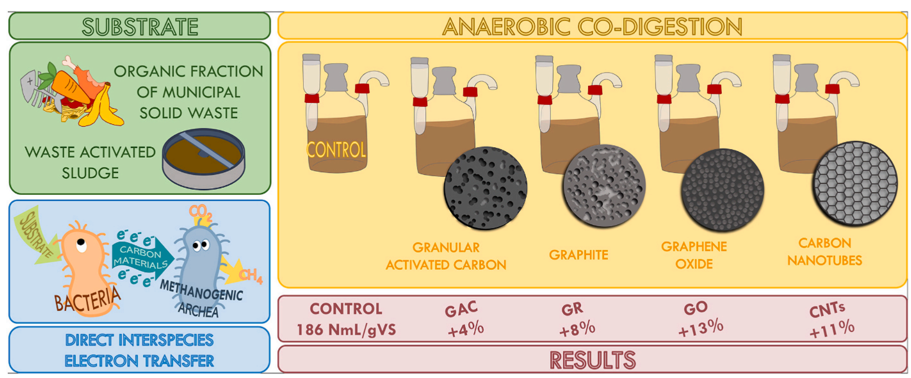
HIGHLIGHTS

- Conductive materials (CMs) were tested for co-digestion of organics and sludge.
- CH_4 yields of all reactors supplemented with C-based CMs were higher than control.
- Carbon nanotubes use led to the highest CH_4 yield increase in the short term (27.9%).
- Graphene oxide addition led to the highest CH_4 yield increase overall (13.0%).

GRAPHICAL ABSTRACT



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ABSTRACT

Organic fraction of municipal solid waste (OFMSW) and waste activated sludge (WAS) are the most produced organic waste streams in urban centres. Their anaerobic co-digestion (AcoD) allows to generate methane (CH_4) and digestate employable as renewable energy source and soil amendment, respectively, fully in accordance with circular bioeconomy principles. However, the widespread adoption of such technology is limited by relatively low CH_4 yields that fail to bridge the gap between benefits and costs. Among strategies to boost AcoD of OFMSW and WAS, use of conductive materials (CMs) to promote interspecies electron transfer has gained increasing attention. This paper presents one of the few experimental attempts of investigating the effects of four different carbon(C)-based CMs (i.e., granular activated carbon – GAC, graphite – GR, graphene oxide – GO, and carbon nanotubes – CNTs) separately added in semi-continuous AcoD of OFMSW and thickened WAS. The presence of C-based CMs has been observed to improve CH_4 yield of the control process. Specifically, after 63 days of operation (concentrations of GAC and GR of 10.0 g/L and of GO and CNTs of 0.2 g/L), 0.186 NL/g_{VS}, 0.191 NL/g_{VS}, 0.203 NL/g_{VS}, and 0.195 NL/g_{VS} of CH_4 were produced in reactors supplemented with GAC, GR, GO, and CNTs, respectively, compared to 0.177 NL/g_{VS} produced in the control process. Likewise, at the end of the test (i.e.,

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after 105 days at concentrations of C-based CMs half of the initial ones), CH_4 yields were 0.193 NL/g_{VS}, 0.201 NL/g_{VS}, 0.211 NL/g_{VS}, and 0.206 NL/g_{VS} in reactors supplemented with GAC, GR, GO, and CNTs, respectively, compared to 0.186 NL/g_{VS} of the control process. Especially with regard to GR, GO, and CNTs, results obtained in the present study represent a significant advance of the knowledge on the effects of such C-based CMs to realistic and scalable AD process conditions respect to previous literature.

1. Introduction

Anaerobic digestion (AD) plays a major role in the sustainable management of organic waste. Through AD, biodegradable matter is ultimately converted into biogas and digestate. The former is a mixture of gases (mostly methane - CH_4, 55–70% v/v, and carbon dioxide - CO_2, 30–45% v/v) that acts as a renewable energy source. The latter is the semi-liquid by-product containing stabilised organic matter and nutrients so that being potentially employable for soil amendment (Deublein, 2010). The key function of AD for circular bioeconomy is to be the last step of biorefinery process. In fact, it allows to close the circle finally transforming biodegradable waste, wastewater, and residues in biogas and digestate that can be beneficially returned to soils (Naddeo and Korshin, 2021).

Organic fraction of municipal solid waste (OFMSW) and waste activated sludge (WAS) are two organic waste streams extensively produced in urban areas. OFMSW is the separately collected organic waste from households including food and green waste, WAS is the excess sludge extracted from the secondary clarifiers of wastewater treatment plants (WWTPs) (Directive 2008/98/EC, 2008; Tchobanoglous et al., 2014). OFMSW averagely accounts for 44% of generated waste at global scale with a production estimated to rise from the current 0.9 Pg to 1.5 Pg by 2050 (Kaza et al., 2018). Its separate collection is becoming mandatory according to the EU regulation (art. 22 Directive 2008/98/EC, 2008). The global production of sewage sludge (including both primary sludge from settling tank and WAS) is estimated to be 45 Tg of dry sludge per year (Q. Zhang et al., 2017). In a perspective of circular bioeconomy, AD seems to be the most suitable solution for the direct management of such amount of organic waste. Particularly, wastewater treatment sector urgently needs solutions to seriously reduce its footprint in terms of energy demand and greenhouse gas (e.g., CO_2, CH_4, N_2O) emissions (Pahunang et al., 2021). Therefore, in this context, recovery of resource from excess sludge (e.g., residual biodegradable matter, nitrogen, phosphorous) is increasingly required.

However, AD of either OFMSW or WAS is hindered by materials' peculiar features. Specifically, OFMSW is rich in highly degradable sugars that are easily converted to volatile fatty acids (VFAs) at an early stage of the digestion process resulting in possible acidification of the system. Moreover, its high protein and lipids content can lead to inhibitory levels of ammonia, hydrogen sulphide, and long chain fatty acids, or cause digester foaming. From a technical point of view, AD of OFMSW is therefore performed at low organic loading rates (OLRs) of 2–3 g_{COD}/L•d in order to prevent process failure (Xu et al., 2018). Instead, WAS is a partially stabilised material as it originates from wastewater treatment processes often including nitrification or complete biological nutrients removal at high solids retention times (SRTs) that lead to considerable biodegradable matter depletion. As a consequence, AD of WAS usually results in low specific CH_4 production (150–300 NmL/g_{VS}) (Pangallo et al., 2023).

The use of OFMSW and WAS as co-substrates in an anaerobic co-digestion (AcoD) is increasingly applied to overcome these issues (Nghiem et al., 2017; Tyagi et al., 2018). Indeed, WAS acts as dilution medium in case of high solid content OFMSW, supplies micro-nutrients and alkalinity to the system, and balances carbon to nitrogen ratio (C/N) in presence of carbonaceous OFMSW (Xu et al., 2018). Furthermore, AcoD would represent a convenient solution for the concurrent management of OFMSW and WAS in basically every urban centre where these waste streams are normally produced. Indeed, considering the amount of OFMSW and geographic distribution of WWTP facilities, such approach has a largely untapped potential for simultaneous renewable energy production and sustainable organic waste management (Nghiem et al., 2017). Its widespread adoption is limited especially by low CH_4 yields that fail to bridge the gap between environmental benefits and true economic values of AcoD (Liang et al., 2021). In order to enhance stability and performance of AcoD, OFMSW and WAS are oftentimes preliminary subjected to pretreatments (i.e., physical, chemical, and/or biological) to facilitate their biodegradation and/or mitigate their adverse effects on the process (Ariunbaatar et al., 2014). However, recent research trend focuses more on directly boosting the biological process rather than on altering substrates' features by including pretreatment steps prior to AD. This approach would allow to ultimately increase CH_4 production without adding technical complexity to the overall system.

Among solutions such as bioaugmentation or trace elements supplementation (Choong et al., 2016; Jiang et al., 2020; Romero-Güiza et al., 2016), triggering the direct interspecies electron transfer (DIET) mechanism has gained increasing attention over the last decade. Briefly, AD process is generally divided into four stages (hydrolysis, acidogenesis, acetogenesis, and methanogenesis) respectively driven by distinct groups of microorganisms in syntrophic association. Particularly, biogas generation can occur if either acetate (produced during acetogenesis) is converted to CH_4 and CO_2 by acetoclastic bacteria or H_2 and CO_2 are used by hydrogenotrophic methanogens to form CH_4. These interactions (redox reactions) involve electrons' exchange among microbial species. The electron transfer is usually mediated (i.e., mediated interspecies electron transfer, MIET) by intermediates such as H_2 and formate acting as 'shuttle' molecules (Stams and Plugge, 2009). However, recent studies discovered that electrons can flow from one microbial cell to another through direct contacts between cells. This mechanism is known as DIET and, compared to MIET, it makes CH_4 production faster and thermodynamically more efficient (Barua and Dhar, 2017).

DIET has been evidenced to be triggered in different ways. Earliest studies experimented spontaneous direct electron transfer from electron-donating microbial species to electron-accepting one through either protein filaments (known as electrically conductive pili or nanowires) observed in defined co-cultures of anaerobic bacteria or chains of c-type cytochromes (namely, iron proteins located in microorganism membranes) (Rotaru et al., 2013, 2014; Summers et al., 2010). A longside, several studies investigated how to engineer DIET in AD systems through the addition of abiotic conductive materials (CMs) (Feng et al., 2023). Specifically, iron- and carbon(C)-based CMs have been extensively studied as electron transfer agents. The former has been observed to enrich the functional microbial species involved in DIET mechanism and further enhance CH_4 generation (Kato et al., 2012). Moreover, magnetite nanoparticles (produced via iron(II) hydroxide transformation in anaerobic environment) are smaller than microbial cell sizes so that being able to stick onto pili for enhanced electron transfer rate or aggregate to link the syntrophic cells for electrons conveyance (Wang and Lee, 2021). On the other hand, some issues related to this practice need to be addressed. For instance, metal oxides presence in biosolids is suspected to cause heavy metal contamination during digestate reuse process but even recovery techniques (such as magnetic separation) of iron particles would increase the overall costs (Wu et al., 2023).

C-based CMs, thanks to their highly porous structure and large specific surface area, provide sites where syntrophic microorganisms (smaller in size than C particles) can attach on, aggregate, and effectively exchange electrons through either cell-to-cell contact or exploiting C surface electrical conductivity. Besides triggering DIET, C-based CMs in AD systems have been observed to promote hydrolysis step (their functional groups acting as electron donors would reduce the oxygen-reduction potential) and adsorb toxic compounds inhibiting bacteria (Calabrò et al., 2019; Feng et al., 2023). Finally, they have been proven to enrich methanogens capable of DIET (Yang et al., 2017). With regard to downstream of AD process, C-based CMs are known to improve digestate dewaterability, stability, and fertiliser utilization (Wu et al., 2023; Yun et al., 2018).

Over the last decade, many relevant experiments have been conducted to investigate the effects of the addition of various C-based CMs on different AD processes. To date, activated carbon and biochar have been the most studied but especially recent studies explored also the use of graphite, graphene, carbon cloth, carbon nanotubes. Over time, several review papers exhaustively reported all these attempts in a perspective of engineering DIET in full-scale AD systems (Barua and Dhar, 2017; Lu et al., 2020; Wang and Lee, 2021). More specifically, some authors tested the addition of C-based CMs to AD of either OFMSW or WAS in order to overcome the respective aforementioned issues (Jin et al., 2022; Li et al., 2022). However, to the best of authors' knowledge, only few studies focused on improving the AcoD of OFMSW and WAS via C-based CMs amendment (results reported in (Liang et al., 2022)) and, also in those cases, activated carbon and biochar have been the most tested materials. In this context, this paper aims at separately investigating the effects on the lab-scale semi-continuous AcoD of OFMSW and WAS of four C-based CMs: granular activated carbon (GAC), graphite (GR), graphene oxide (GO), and carbon nanotubes (CNTs). Processes performances (in terms of CH_4 yield and stability) and comparisons with previous studies will be presented and discussed. Results obtained from this study would provide new insights into both i) the search for solutions to make AcoD of OFMSW and WAS more profitable and ii) the evaluation of the effects of different C-based CMs on a realistic and scalable AD process.

2. Materials and methods

2.1. Semi-continuous AcoD test equipment

The semi-continuous AcoD test was performed through a lab-scale simulation system (Bioprocess Control Bioreactor Simulator, BPC Instruments, Sweden). It includes five different reactors (designed as A, B, C, D, and E, working volumes of 1.9 L) equipped with internal stirrers and immersed in a thermostatic water bath (35 °C, mesophilic conditions). Feeding and discharge operations were simultaneously carried out five days per week. Produced CH_4 flow rate measurements and data logging of the system are fully automated. The high precision gas flow monitoring is based on a patented flow meter array using the principle of liquid displacement and buoyancy. A digital pulse is generated when a defined volume of gas flows through the device and an integrated embedded data acquisition system is used to record, display and analyse the results. The measuring range is to 10–4000 mL of gas per hour while the measuring resolution is 10 ± 0.1 mL.

Hydraulic retention time (HRT) and organic loading rate (OLR) were set at 21 days and 1.5 g_{VS}/L•d, respectively.

2.2. Inoculum and substrates

Inoculum was a residual digestate coming from a previous AcoD experiment running under the same experimental conditions set in the current test but without involving any supplement. It was maintained at 35 °C until the beginning of the AcoD test.

The OFMSW was prepared in laboratory according to the typical (average) composition of Italian organic domestic waste (vegetables, fruit, potato, meat and fish, dairy, bread, cooked rice and pasta) based on the findings of a recent literature survey (Pangallo et al., 2021). Afterwards, in order to assure its homogeneity during the whole experiment, the OFMSW underwent a drying process at 35 °C (to avoid the loss of the most volatile components) for seven days and it was shredded by using a Cutting Mill PULVERISETTE 15. To avoid an excessively long storage WAS used in the experiment was sampled every about three weeks from the sludge line of a municipal WWTP located in Reggio Calabria (Italy) serving 30′000 people equivalent (P.E.) and operating biological nitrogen removal through a pre-denitrification/nitrification scheme. More specifically, the WAS was collected after the gravity thickening of the excess sludge extracted from secondary clarifiers.

The WAS samples were quite uniform in terms of total solids (TS), volatile solids (VS) and pH except one that for reasons unknown had lower TS. However, since the loading was set in terms of VS and HRT was kept constant, this fact does not have any impact on the results. Both OFMSW and WAS samples were stored at 4 °C before their use in the AcoD test.

All the materials were characterised in terms of pH, TS, VS, and (only for WAS) volatile fatty acids (VFAs) according to the standard methods (American Public Health Association, 2012; Liebetrau et al., 2016). Results are reported in Table 1.

Appropriate quantities of OFMSW and WAS were jointly fed to all reactors according to a ratio of 50:50 (on VS basis) in order to keep the set values of the operational parameters (i.e., HRT and OLR).

2.3. Digestate analyses

In digestate samples discharged from reactors during feeding operations, pH and conductivity values were directly measured through, respectively, a digital pH meter (XS instruments) and a digital conductivity meter (HD 8706, Delta Ohm). Moreover, TS, VS, VFAs, volatile organic acids/buffering capacity ratio (FOS/TAC), chemical oxygen demand (COD), and total nitrogen (N_{tot}) contents were evaluated on weekly composite digestates. Specifically, COD and N_{tot} were determined by using pre-dosed cuvettes (Merck Millipore COD Cell Test 114555 and Nitrogen (total) Cell Test 114537, respectively, using a photometer WTW Photolab S12) whereas FOS/TAC through a two-points titration (Liebetrau et al., 2016). COD, N_{tot}, VFAs, and FOS/TAC analyses were carried out on the liquid fractions resulting from digestates centrifugation (10,000 rpm for 10 min).

2.4. C-based CMs in the AcoD test

As aforementioned, granular activated carbon (GAC), graphite (GR), graphene oxide (GO), and carbon nanotubes (CNTs) were respectively added to reactors B, C, D, and E in order to i) improve the AcoD of OFMSW and WAS performance and ii) separately evaluate their different effects on that process. An additional blank reactor (i.e., reactor A), not containing any additive, acted as benchmark (Table 2). The concentrations of the C-based CMs in the reactors were determined based on relevant literature. The overall test was divided into two distinct phases: in the first one, C-based CMs concentrations were set at 10 g/L for reactors B and C supplemented with GAC (Liang et al., 2022;

Table 1
Materials' characterisation.

Material	TS [%]	VS [%TS]	pH	VFA [mg/L]
Inoculum	0.9 ± 0.07	68.8 ± 0.84	7.9	–
OFMSW[a]	21.7 ± 0.56	96.2 ± 0.13	6.1	–
WAS	2.2 ± 0.07	78.9 ± 0.30	6.6	375.9
	2.9 ± 0.11	80.2 ± 0.24	6.4	877.7
	2.0 ± 0.14	79.8 ± 0.72	6.5	615.0
	0.7 ± 0.10	79.5 ± 0.41	7.1	134.0
	3.2 ± 0.20	79.2 ± 1.23	6.4	547.8

[a] Referred to wet sample before drying.

Table 2
Experimental setup.

Reactors	C-based material added	Concentration [g/L]	
		I phase	II phase
A	–	–	–
B	Granular Activated Carbon (GAC)	10.0	5.0
C	Graphite (GR)	10.0	5.0
D	Graphene Oxide (GO)	0.2	0.1
E	Carbon Nanotubes (CNTs)	0.2	0.1

Lu et al., 2020; Wang and Lee, 2021) and GR (Wang and Lee, 2021), respectively, and 0.2 g/L for reactors D and E supplemented with GO (Lu et al., 2020) and CNTs (Lu et al., 2020; Wang and Lee, 2021), respectively. In order to reach and maintain those set dosages, specific quantities of C-based CMs were added at the beginning of the test (i.e., 19.0 g and 0.4 g, respectively of GAC and GR, and GO and CNTs) and then loaded along with substrates during feeding operations (i.e., 0.90 g per day and 0.018 g per day, respectively of GAC and GR, and GO and CNTs). After 3 HRT (i.e., about 63 days), the loading of the C-based CMs was suspended for 14 days in order to halve their concentrations inside the reactors. Afterwards, the second phase of the test started and C-based CMs were added to respective reactors again to maintain the halved concentrations. The purpose of this phase was to assess how the reduction in concentration would have affected the CH_4 yield. The overall experimental setup is summarised in Table 2.

2.5. C-based CMs characteristics

Basically, C-based CMs present an assortment of the atomic structure of carbon atoms giving them unique properties (Madima et al., 2020; Mashkoor et al., 2020). Due to their dimensionality, size and shape, the carbon materials can be arranged as zero-, one-, two-, and three-dimensional nanomaterials.

CNTs are the most important one-dimensional carbon nanostructures, characterised by a tube-like configuration resulted from the rounded graphene sheets and, depending on the number of graphene sheets, they can be single, double or multi walled. Among the possible synthesis method, balancing yield and costs, chemical vapour deposition is the most widely used for their production. GO is instead a two-dimensional carbon network with oxygen-containing functional groups; it can be produced starting from graphite using different chemical methods, yielding a high production with low costs. GR and GAC can be tagged as three-dimensional structures. GR is composed of weakly coupled graphene layers, whereas GAC can be defined as a disorganised graphite form. GR and GAC are available at very low costs on the market. Therefore, in this work, GR and GAC were purchased by Sigma Aldrich, whereas CNTs and GO were synthesized following well-established procedures. In particular, commercial GR was of high purity and sized ≤100 μm; GAC had instead particles size ≤150 μm and very low percentage of impurities. GO was prepared by exfoliation of graphite powder following a modified Hummers' method, allowing to obtain flakes of few microns in dimension (Hummers and Offeman, 1958). Multi-walled CNTs were synthesized in large scale using a similar procedure as reported elsewhere (Milone et al., 2014) with an average length of 2 μm and nanometric diameters. Main morphological features of carbon materials are evidenced in SEM micrographs (Fig. 1). GAC appeared with a rough surface, across which macropores were clearly identifiable. CNTs displayed the typical tubular structures whereas GR was characterized by particles having thin plate shapes. Finally, GO was

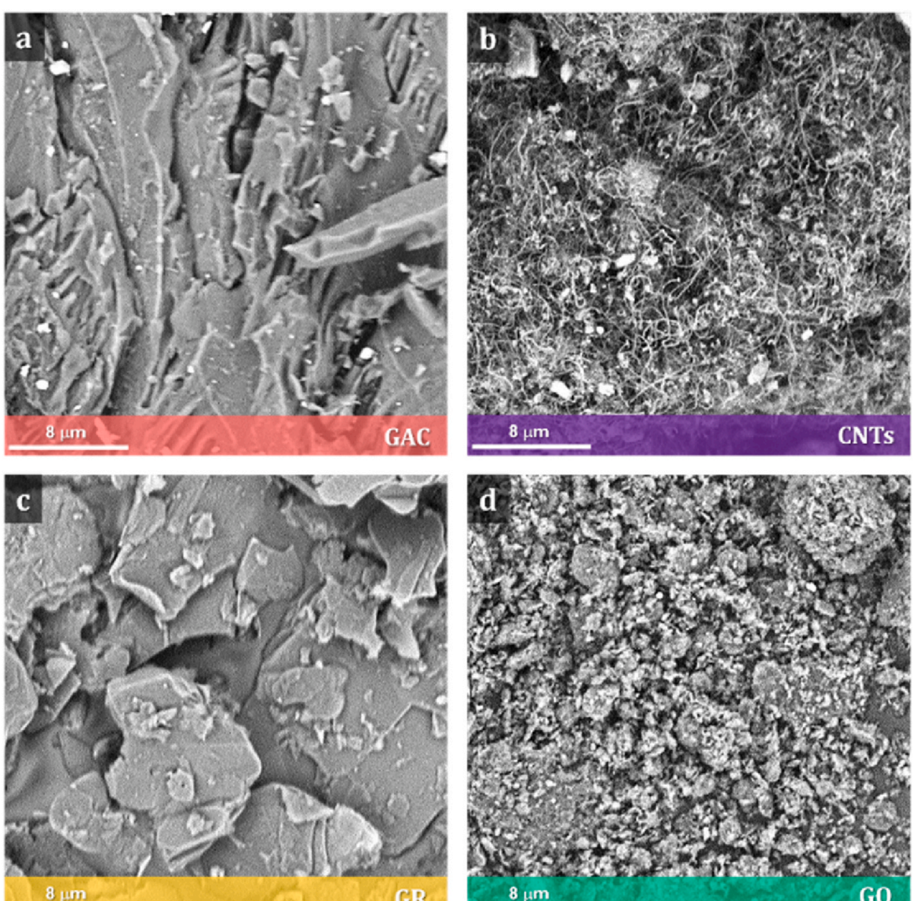

Fig. 1. SEM micrographs of C-based CMs – Legend: a) GAC = Granular Activated Carbon; b) CNTs = Carbon Nanotubes; c) GR = Graphite; d) GO = Graphene Oxide.

organized in very small and aggregated flakes.

The formation of on-purpose synthesized structures, as well as the features of commercial materials were studied by means of X-ray diffractometry, using a Panalytical Empyrean S-2 diffractometer, with a Cu Kα radiation (1.54056 Å) at 40 kV and 40 mA. The patterns were recorded in step scan mode from 5° to 60° 2θ angles in steps of 0.02° and a count time of 5 s per step and peaks were attributed in accordance with available database. C-based CMs show similarity in diffraction patterns as reported in Fig. 2. The XRD spectrum of GO showed the sharp and characteristic diffraction peak at 2θ equal to 9.5°, shifted to lower angles with respect to the strong and sharp diffraction signal of the GR at 2θ equal to 26.7°, suggesting that, due to the presence of oxygen-containing functional groups between the layer of graphite, GO sheets are loosely stacked (Fathy et al., 2016). CNTs evidenced indeed a wide peak at 2θ around 26° due to the graphitic carbon, while lower is the position of the very broad diffraction peak in GAC material, revealing a predominantly amorphous structure.

3. Results and discussion

3.1. Performances of AcoD of OFMSW and WAS processes

The performances of AcoD of OFMSW and WAS processes were initially evaluated on the basis of their respective specific CH_4 yields (Fig. 3a).

What immediately emerges is that the presence of C-based CMs improved the conventional AcoD of OFMSW and WAS (i.e., reactor A). Indeed, in the I phase of the test, reactors A, B, C, D, and E stabilised on CH_4 yields of about 0.177 NL/g_{VS}, 0.186 NL/g_{VS}, 0.191 NL/g_{VS}, 0.203 NL/g_{VS}, and 0.195 NL/g_{VS}, respectively. Likewise, at the end of the test (i.e., after 105 days), CH_4 yields were 0.186 NL/g_{VS}, 0.193 NL/g_{VS}, 0.201 NL/g_{VS}, 0.211 NL/g_{VS}, and 0.206 NL/g_{VS}, respectively for reactors A, B, C, D, and E (Fig. 3a). Thus, the average performance sequence in terms of CH_4 yield was D (GO supplemented reactor) > E (CNTs supplemented reactor) > C (GR supplemented reactor) > B (GAC supplemented reactor) > A (control). Conversely, in terms of lag phase, the supplementation of C-based CMs seemed not to alter the initial stage of microbial adaptation likely because, as mentioned in materials and methods section, the inoculum derived from a previous test carried out on the same substrates was used.

Improvements due to the supplementations of C-based CMs are depicted in Fig. 3b in terms of percentage increases respect to CH_4 yield of blank reactor A. The picture shows that the beneficial effects in terms of CH_4 production of the addition of the C-based CMs to the respective processes are evident but unsteady over test time.

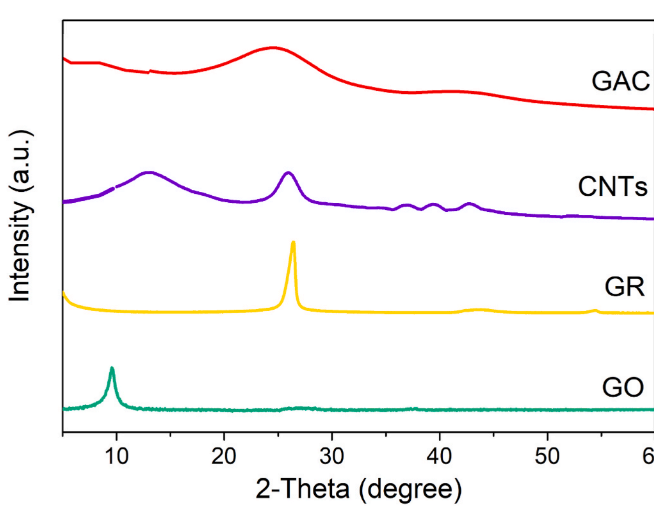

Fig. 2. XRD patterns of C-based CMs – Legend: GAC = Granular Activated Carbon; CNTs = Carbon Nanotubes; GR = Graphite; GO = Graphene Oxide.

With regard to the I phase of the test (i.e., days 1–63), reactors D and E supplemented with GO and CNTs, respectively, showed a good response with significant CH_4 yield increases in the first 10 days of the test (peaks of +25.7% in reactor D, on day 9, and +27.9% in reactor E, on day 8). Afterwards, downward trends have been recorded until day 44. Similarly, the trend of CH_4 yield increase observed in reactor C, where GR was added, showed a peak of +20.3% on day 15 followed by a steep decrease until day 48. Lastly, reactor B supplemented with GAC needed more time to exhibit CH_4 yield improvement (peak of +10.6% on day 32). The trends in CH_4 yield increases basically stabilised after two HRT (i.e., 42 days). As already mentioned, after 3 HRT, the addition of C-based CMs was suspended for two weeks (from day 64–77) with the purpose of reducing by half their concentrations in respective reactors. As a result, reactor C (supplemented with GR) was the only process that showed a sort of recovery in the two weeks of feeding suspension (Fig. 3b). Nevertheless, in the II phase of the test, the average CH_4 yield increase respect to control process were +3.8%, +7.2%, +13.3%, and +10.0% for reactors B, C, D, and E, respectively, much lower than respective peaks recorded in the I phase. The fluctuation evident in Fig. 3b on days 92–95 was recorded during a weekend and was probably due to a power supply failure to the thermostatic bath. As mentioned in materials and methods section, C-based CMs were loaded to respective reactors at the beginning of the test in quantities (i.e., 19.0 g and 0.4 g, respectively of GAC and GR, and GO and CNTs) set to reach the desired regime concentration. Then, they were periodically loaded along with substrates during feeding operations (i.e., 0.90 g per day and 0.018 g per day, respectively of GAC and GR, and GO and CNTs) considering the loss with digestate in completely stirred conditions. Especially for GO and CNTs, it can be hypothesised that good responses of the respective systems were due to the larger quantity of virgin material present in the reactors at the beginning of the test rather than during regime supplementation. Possibly, microorganisms took time to adapt and colonise virgin materials (see the first eight days of acclimation in Fig. 3a and b) and then DIET mechanism was established (see peaks in CH_4 yield increases, Fig. 3b). Once materials were 'exhausted', their regime replenishment was not sufficiently efficient to guarantee the boosting effect of the first weeks likely because of the low set dosage (i.e., 0.2 g/L). To overcome this situation it could be possible to increase the set dosage but at the risk of reaching toxic concentrations thresholds and with higher costs. Indeed, several studies reported an inhibiting effect at high GO concentrations (i.e., 0.5–1.4 g/L), likely due to the consumption of the electrons by GO reduction instead of methane formation (Ponzelli et al., 2022). Similarly, due to the relative high cost of CNTs, low quantities were used in all the studies present in scientific literature while no toxic effect was observed. Another possible solution, would be to supplement virgin GO and CNTs not continuously but semi continuously evaluating the optimal feeding pattern. Thus, it would be likely possible to maintain as much as possible the boosting effect observed in the first weeks.

It can be noted that the three-dimensional carbon structures, namely GAC and GR, showed a different behaviour with respect to the two-dimensional and one-dimensional carbon materials, GO and CNTs, respectively. The characteristic response can be attributed to the different reactivity of those materials conferred by their featuring morphologies. Indeed, GO and CNTs can be considered nanomaterials, GAC and GR can be considered bulk materials. In the first case the very low dimension of GO flakes and the tubular structure of CNTs determined in both materials a higher density of defects inducing high reactivity in the process, with respect to GAC and GR, whose activity was instead lower. The different response of C-based structures is even more evident when their concentrations [g/L] are considered over the cumulative CH_4 production. Very low GO and CNTs quantities (i.e., 0.2 g/L) were used, in accordance with literature reports, compared to GAC and GR (10 g/L, namely two orders of magnitude higher).

In order to evaluate process stability, pH was periodically measured

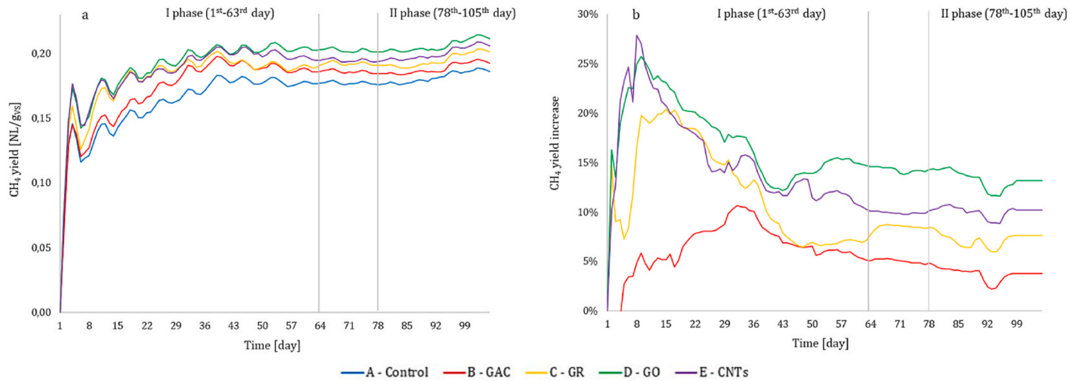

Fig. 3. a) CH_4 yields, b) CH_4 yield (percentage) increases compared to control process (i.e., reactor A) – Legend: GAC = Granular Activated Carbon; GR = Graphite; GO = Graphene Oxide; CNTs = Carbon Nanotubes.

through a portable pH meter and VFAs contents were determined in weekly composite digestate samples (Fig. 4a and b, respectively).

What emerges from these analyses is that promoting DIET resulted in more stable processes as control process (i.e., reactor A) averagely showed the lowest pH values and the highest VFAs contents over test time (even if these values never exceeded respective tolerance thresholds). On account of that, it can be stated that the larger CH_4 yields of reactors B, C, D, and E are due to a faster and more efficient microbial conversion of acids to CH_4 as expected as consequence of DIET establishment. Peaks in VFAs content recorded in reactor E on weeks 10 and 11 (of 838 mg/L and 952 mg/L, respectively) would confirm the sensitivity of the system to CNTs. Indeed, those VFAs content increases were recorded precisely when CNTs addition was suspended.

Weekly composite digestates were also analysed in terms of TS (Fig. 4d) and VS (data not shown). Particularly for reactor C supplemented with GR, probably due to its relatively high unit weight, TS tend to increase probably due to imperfect mixing. Indeed, digestate samples coming from reactor C were visibly more dense and darker than the others. As a consequence of the period of suspension of the CMs addition (i.e., weeks 10 and 11), TS concentration in reactor C sharply decreased from 3.1% to 2.1% in accordance with the others. This occurrence could explain the recovery in CH_4 yield of reactor C observed during the suspension of GR addition previously described as if the likely GR accumulation was detrimental for the process. Further research, based on continuous or semi-continuous experiments and not on batch ones, to evaluate possible toxicity problems related to high CMs addition is strongly needed.

Finally, conductivity of the systems was also measured in digestates through a portable device (Fig. 4c). Also in this case, it emerges the influence of CMs on digestate characteristics since those from reactors B, C, D, and E had slightly higher conductivity than control process (i.e., reactor A). In particular, GO and CNTs (supplemented in reactors D and E, respectively) were responsible for the highest conductivity values over test time in spite of their far lower set concentration compared to GAC and GR (0.2 g/L versus 10 g/L). This is in accordance with the higher increases in CH_4 yields measured in reactors D and E (Fig. 3b). Higher conductivity signalizes a more intense electrical activity in those systems where a more intense DIET mechanism occurs.

With regard to FOS/TAC, COD, and N_{tot} parameters, data has not been shown as negligible variations among processes have been recorded. Just in FOS/TAC case, it is noteworthy that ratio values were lower than 0.2 over test time in all processes (except for reactor E on weeks 10 and 11). Such occurrence would point out that the biomass input was low according to (Mézes et al., 2011). For this reason, in future research, larger OLRs could be tested.

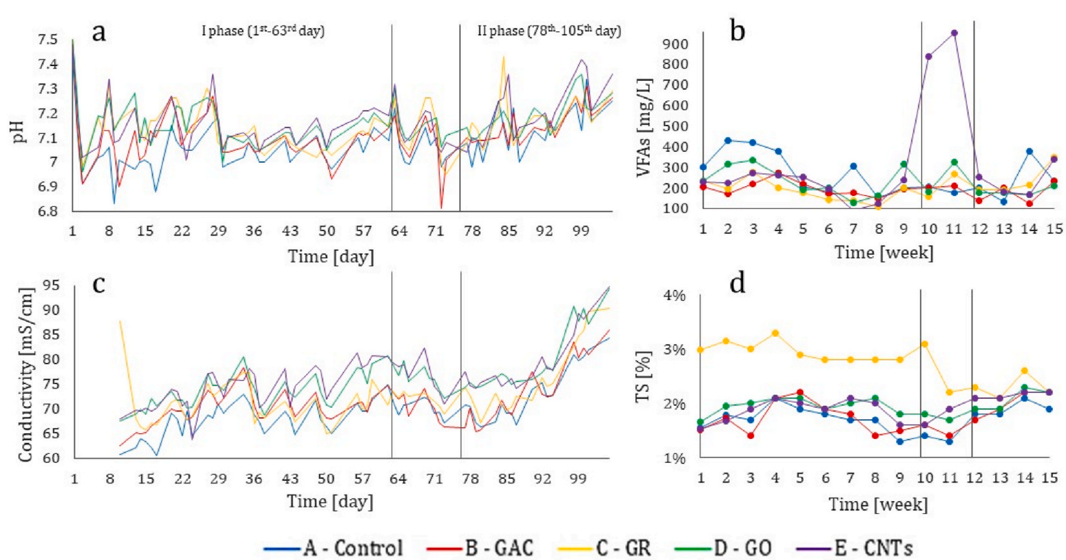

Fig. 4. Digestate characteristics on: a) pH and c) conductivity (daily samples) and b) VFAs content and d) TS content (weekly composite samples) – Legend: GAC = Granular Activated Carbon; GR = Graphite; GO = Graphene Oxide; CNTs = Carbon Nanotubes.

3.2. Comparisons with results from literature

The CH_4 yield of the benchmark process (i.e., AcoD of OFMSW and WAS in reactor A) was modest (0.177 NL/g_{VS}) but anyway consistent with literature in which CH_4 yields range from 0.16 NL/g_{VS} to 0.6 NL/g_{VS} (Mehariya et al., 2018; Xu et al., 2018). The large difference from the highest values of the range is likely due to differences in substrate and to different technical and operational conditions. First of all, thickened WAS used in this test had predictably poor content of easily biodegradable matter since it was collected in a municipal WWTP where biological nitrogen removal (pre-denitrification/nitrification) is performed, moreover it was also subjected to a long thickening (HRT = 3–6 days). In (Dai et al., 2013) (maximum CH_4 yield of 0.32 NL/g_{VS}), for instance, food waste was anaerobically co-digested with a mixture of WAS and primary sludge so that it was richer in easily biodegradable matter. Moreover, results from this study were obtained through a semi-continuous digestion mode unlike previous similar studies (e.g., (Prabhu and Mutnuri, 2016), biochemical CH_4 potential of 0.49 NL/g_{VS}). Indeed, CH_4 yields in (semi-)continuous processes are expectedly lower than biochemical CH_4 potentials determined with batch tests, even at the same feeding conditions (Ruffino et al., 2015).

The delayed response of the AD process to the GAC addition (i.e., reactor B) was consistent with a previous study of the same research group but using a different substrate (i.e., orange peel waste) (Calabrò et al., 2019). In that case, although the beneficial effects of GAC addition compared to control process were observed after 30 days (as in this test), the increase in terms of CH_4 yield was considerably higher (+68%) because GAC (added in the larger concentration of 20 g/L) likely adsorbed D-limonene (a monoterpene naturally present in orange peels known to inhibit microbial activity) so that preventing its accumulation and subsequent process failure. In this test, the presence of toxic compounds at such concentrations to cause process failure can be reasonably excluded, in fact control process (i.e., reactor A) run stably over all the test time (i.e., 105 days, 5 times the set HRT). In this context, GAC would have not exerted its function of adsorbent but it 'just' provided sites to be gradually colonised by syntrophic microorganisms. In a previous attempt of enhancing the AcoD of food waste and WAS, (Zhang et al., 2018) reached a CH_4 yield of 0.34 NL/g_{VS} with an increase of +88.9% due to the addition of 15 g/L of GAC at OLR and HRT far larger than in this test (gradually increased up to 5.2 g_{VS}/L•d and 30 days, respectively) in a semi-continuous mode. (Wang et al., 2020) recorded instead an increase in CH_4 production of +12.3% (consistent with this study) but by adding 1 g/L of GAC in the AcoD of food waste and WAS after 20 days of operation in batch mode; at a higher GAC concentration (i.e., 5 g/L), improvement was negligible.

To date, GR has not been thoroughly tested as DIET promoter (especially in semi-continuous mode) and its beneficial role is consequently still uncertain. Preliminary, GR was observed to enhance the CH_4 production from semi-continuous AD of ethanol, propionate, and butyrate by promoting DIET mechanism (Zhang et al., 2019; Zhao et al., 2015). Conversely, no significant optimization on methanogenesis was recorded when GR was added to AD of dog food and sewage sludge (batch tests) (Dang et al., 2016; Pan et al., 2020). Conversely, CH_4 yield increased by +38.3% in batch mode AD of WAS amended with GR flakes at a dosage of 10 g/L. However, the lag phase of the process resulted prolonged from 5.2 days to 8.4 days compared to control likely because of microbial adaptation. Moreover, authors discovered that GR promoted acetoclastic methanogenesis (Lü et al., 2020). This latter evidence was also observed by (He et al., 2021) as a result of the addition of 0.2 g/L of nano-GR to the batch mode AcoD of fat, oil and grease and WAS (increase of +14% respect to the cumulative CH_4 production of control process). Other instances of batch AD/AcoD tests supplemented with GR were made by (Muratçobanoğlu et al., 2020; Wu et al., 2022; Xie et al., 2020; Tsui et al., 2020) observed that GR enhanced the AD of food waste leachate in upflow anaerobic sludge bed at high OLR (i.e., biogas production increase of +38%). In this context, results obtained in the present study represent a significant advance of the knowledge on the effects of GR to semi-continuous AD processes.

Graphene-based nanomaterials are known to have dose-dependent cytotoxicity. Previous studies revealed that GO presence in WAS at high concentrations ranging from 50 to 300 mg/L inhibits microbial activity in aerobic batch tests (Ahmed and Rodrigues, 2013; Zhang et al., 2017) observed CH_4 production reductions by 13.1%, 10.6%, 2.7%, and 17.1% at GO concentrations of 5 mg/L, 50 mg/L, 100 mg/L, and 500 mg/L, respectively, in the batch AD of swine manure. Similarly, both (Dong et al., 2019; Casabella-Font et al., 2023) reported that the CH_4 production from the AD of WAS (batch test) dropped with increasing GO additions (0.025 g/g_{VS}, 0.054 g/g_{VS}, 0.075 g/g_{VS}, and 0.108 g/g_{VS}, dosage in this paper is of 0.006 g/g_{VS}). Adverse impacts of GO presence on the biological treatment of WAS are observed to be i) production of reactive oxygen species from the interaction of GO with WAS, ii) adsorption of the soluble organic matter of WAS (so that decreasing its availability in the AD system), iii) inhibition of co-enzymes involved in the methanogenesis, and iv) consumption of electrons for GO reduction instead of CH_4 formation (Ahmed and Rodrigues, 2013; Dong et al., 2019; Ponzelli et al., 2022). Nevertheless, other batch experiments were able to enhance AD performance through GO addition in terms of CH_4 production of 74% and 65%, respectively (Kaushal and Baitha, 2021; Muratçobanoğlu et al., 2021). To the best of authors' knowledge, (Muratçobanoğlu et al., 2022) represents the only attempt of semi-continuous AD improvement due to GO addition. Indeed, authors evidenced that GO concentration of 20 mg/L in the AD of OFMSW increased the CH_4 rate [NmL/L•d] compared to control process from +24% to +110% at increasing OLRs (from 0.5 g_{VS}/L•d to 4.0 g_{VS}/L•d). When digester was stressed with higher OLRs (i.e., 6.0 g_{VS}/L•d and 10.0 g_{VS}/L•d) system failed for acidification in spite of the GO presence.

(Wang et al., 2021) separately tested the effects of the addition of GR and GO on the AcoD of food waste and sewage sludge. Unlike the present test, experiment described was conducted in batch mode (18 days of operation), under thermophilic conditions (i.e., 55 °C), and C-based CMs were added after being added to deionised water (at a dosage of 50 mg/L) and then sonicated (at 600 W, 25 °C, 40 kHz, for 30 min). As a result, the cumulative CH_4 potential respect to the control process was increased by +27.3% and +17.6% due to the addition of GR and GO, respectively, in contrast to the present test in which GO presence guaranteed a higher increase in CH_4 production (Fig. 3a and b).

CNTs have been observed to stimulate DIET mechanism in defined co-cultures of anaerobic bacteria within certain concentrations (i.e., 5.0 g/L) (Salvador et al., 2017). Nevertheless, CNTs amendment of AD of complex substrates (organic waste) was not extensively tested due to high cost of purchasing and evidences of cytotoxicity of the material (Chen et al., 2019; Wang and Lee, 2021)'s review reports studies in which CNTs enhanced butyrate and propionate oxidation and benzoate degradation in AD systems. (Li et al., 2015) studied the reaction of anaerobic microorganisms to CNTs in complex environmental matrix. In particular, single-walled CNTs were added to the AD of methanogenic sludge at a dosage of 1 g/L. Although no significant improvement in terms of CH_4 yield was recorded, however, authors observed that extracellular polymeric substances excreted from the anaerobic sludge closely interacted with CNTs particles so that preventing nanoparticles from piercing into cells and thus reducing their cytotoxicity. CH_4 production improvement due to CNTs supplementation was recorded by (Ziganshina et al., 2022) in the batch AD of chicken manure. Specifically, CNTs at a dosage of 5.0 g/L increased the CH_4 yield of +15%. In this context, the present study represents one of the few instances of engineering DIET through CNTs addition at realistic and scalable AD process conditions.

One of the reasons of the limited utilization of CNTs in biotechnology applications so far is represented by the high cost of this C-based nanomaterial. Indeed, according to the current average market costs, multi-walled CNTs and GO prices are about 50–150 \$/kg_{CNTs} and 75–200 \$/kg_{GO}, respectively, versus, for instance, the far lower price of

GAC (i.e., 0.3–3.0 $/$kg_{GAC}$) (Wang and Lee, 2021) and GR (50–70 $/$kg_{GR}$). On account of that, and regarding the aforementioned cytotoxic properties of CNTs, in the present study, the dosage of CNTs in the anaerobic reactor was lower than similar previous studies, specifically 0.2 $g_{CNTs}/L_{reactor}$. In order to constantly maintain this dosage at a set HRT of 21 days, 0.018 g of CNTs have been daily added to the 1.9 L reactor working volume (i.e., about 0.009 $g_{CNTs}/L_{reactor}$•d). Thus, the cost of CNTs supplementation to the AcoD process (according to the presented solution) would be approximately of 0.95 $/$m^3_{reactor}$•d versus 0.79 $/$m^3_{reactor}$•d in case of GAC addition (dosage of 10 $g_{GAC}/L_{reactor}$). At a biomethane selling price of 0.33–0.55 $/$m^3_{CH4}$ (D'Adamo et al., 2023), the revenues guaranteed by the yield increases registered in this research could not compensate the cost of CMs additions. However, the cost of CNTs is continuously reducing while low-cost alternatives represented by biochar and GR recovered from the recycling of Li-ion batteries (Liu et al., 2022; Premathilake et al., 2023) could be tested for GAG and GR, respectively.

4. Future prospects

As a consequence of both the results previously presented and the comparison with similar previous studies, future research is needed to optimise the dosage (in terms of quantity and mode of operation) of the C-based CMs on the AcoD of OFMSW and WAS. Especially for nanomaterials (i.e., GO and CNTs), it could be advisable to investigate the effects of higher dosages (up to one order of magnitude higher). Moreover, in order to persistently maintain the maximum boosting effect on CH_4 yield recorded in this test different procedures of addition of CMs might be tested (e.g. semi-continuous addition). Finally, taking into account that no signs of process stress were recorded for control process (i.e., reactor A) and that engineering DIET seems to be more effective at higher substrate inputs as observed in previous studies, in further experiments, larger OLRs could be tested with the aim of increasing the overall CH_4 production but preventing acidification of the systems thanks to CMs presence.

5. Conclusions

To sum up, through the presented experiment, the beneficial effects of the addition of CMs (i.e., GAC, GR, GO, and CNTs) to the AcoD) of OFMSW andWAS) have been demonstrated under the process conditions of OLR of 1.5 g_{VS}/Ld, HRT of 21 days, concentrations of GAC and GR of 10.0 g/L (I phase) and 5.0 g/L (II phase) and of GO and CNTs of 0.2 g/L (I phase) and 0.1 g/L (II phase). The average performance sequence in terms of CH_4 yield was GO supplemented reactor > CNTs supplemented reactor > GR supplemented reactor > GAC supplemented reactor > control process. CH_4 yields in supplemented reactors at regime conditions were regularly higher than control process (i.e., 0.186 NL/g_{VS}, 0.193 NL/g_{VS}, 0.201 NL/g_{VS}, 0.211 NL/g_{VS}, and 0.206 NL/g_{VS} for control, GAC, GR, GO, and CNTs reactors, respectively, at the end of the test). Compared to current literature in which especially for GO and CNTs additions to AD inconsistent results have been observed likely because of dose-dependent cytotoxicity of such nano C-based CMs, the present study represents a significant advance of the knowledge. Moreover, respect to previous studies mostly performed under batch conditions, the present test is one of the few attempts of engineering DIET through GR, GO, and CNTs addition at realistic and scalable AD process conditions. Nevertheless, as a result of a rough economic evaluation, it would seem that the revenues guaranteed by the yield increases could not compensate the cost of CMs additions. However, further research could overcome this issue by increasing the overall CH_4 production of the AcoD of OFMSW and thickened WAS through testing i) higher dosages of the C-based CMs (especially GO and CNTs), ii) different procedures of additions to the reactors, iii) larger OLRs.

CRediT authorship contribution statement

Filippo Fazzino: Data curation, Formal analysis, Investigation, Methodology, Writing – review & editing. **Patrizia Frontera:** Writing – review & editing, Data curation, Methodology. **Angela Malara:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Altea Pedullà:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Paolo S. Calabrò:** Writing – review & editing, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

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.

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