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Research Paper

Testing sewage sludge and landfill leachate in novel biocovers: A soil column experiment to quantify landfill gas abatement

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ABSTRACT

A laboratory experiment was carried out aimed at testing novel biocovers and quantifying the mitigation of diffuse landfill gas (LFG) emissions into the atmosphere. The experiment, lasting 40 days, was designed to simulate a laboratory-scale landfill environment, using four integrated prototypes consisting of (i) a digester, representing the landfill waste body under predominant anaerobic degradation (AD) conditions, connected to (ii) a soil column, representing the landfill biocover with different mixtures of natural soil, anaerobically stabilized and dewatered sewage sludge (SS) and landfill leachate (LL). Gas samples were periodically collected from the digester headspaces and from four sampling ports located along each soil column. Experimental data showed that biodegradation processes of reduced biogas compounds increased upward along the biocover soil columns, whereas CO_2 and O-substituted compounds, being produced by degradation of former compounds, showed opposite trends. To estimate the benefits on biogas abatement provided by the soil treatments, a novel index was proposed. The index considered the biogas biodegradation referred to bacterial activity, excluding the reduction of biogas due to dilution with atmospheric air occurring within the cover soil. Noteworthily, the tested novel biocovers with soil treated only with SS showed higher biogas abatement efficiencies relative to both the untreated natural soil and that with concurrent LL addition. These results support the hypothesis that treated soils, specifically with sewage sludge as a potential resource, represent a reliable option to improve the mitigation of gaseous species and functional groups released by diffuse LFG emissions.

1. Introduction

Landfilling is the most common method for municipal solid waste (MSW) management worldwide, also in the near future (Chen et al., 2020). However, MSW landfills can strongly impact the environment and human health due to the generation of two potentially polluting and toxic streams (Zacharof and Butler, 2004): landfill gas (LFG) and landfill leachate (LL). While LL generation mostly depends on water infiltration, LFG is mainly produced by the progressive degradation of the organic fraction of deposited MSW (Alibardi and Cossu, 2015), which is primarily constituted by food waste (FW) (Zhang et al., 2007; Panigrahi and Dubey, 2019; Kumar and Samadder, 2020). Conventional landfill cover soils (LCSs), commonly allocated at the landfill surface, are expected to mitigate LL generation and LFG release into the atmosphere by limiting meteoric water infiltration into the waste body and acting as a possible biofilter for LFG, respectively (Cèbron et al., 2007; Bajar et al., 2016). When the landfill is not equipped with an impermeable bottom liner, LCSs were coupled with geomembrane liners, drainage layers and/or stratification of layers with different pore sizes to create a capillary barrier able to limit water infiltration through the cover (Chetri and Reddy, 2021).

Concerning diffuse LFG emissions, air permeation in LCSs enhances oxidizing conditions (Tassi et al., 2009; Haque et al., 2020), thus favoring the development of O_2-utilising microbial consortia, such as methanotrophs (Kightley et al., 1995; Boeckx et al., 1997; Gebert and Gröngröft, 2006; Huber-Humer et al., 2008; Sadasivam and Reddy, 2014; Kjeldsen and Scheutz, 2018) and nonmethane volatile organic compounds (VOCs) degrading bacteria (Pandey et al., 2014; Jabłońska

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and Tawfik, 2019), able to degrade LFG into CO_2, O-substituted compounds and, overall, lighter molecules (Rabus et al., 2001; Rojo, 2009; Mbadinga et al., 2011; Pratt et al., 2013; Wang and Shao, 2013; Bian et al., 2015; Randazzo et al., 2023; Randazzo et al., 2024). However, the degradation of LFG in conventional LCSs is not fully effective, and a relevant portion of non-collected LFG is regrettably emitted into the atmosphere from the soil (Barlaz et al., 2009). In this context, technical-scientific efforts to further reduce diffuse LFG emissions from the soil have been made, such as the development of biocovers as LCS systems (Huber-Humer et al., 2009) amended and optimized with sewage sludge (SS) or LL recirculation to create a favorable environment for LFG degrading bacteria to grow by providing optimal levels of moisture and nutrients (Koerner and Soong, 2000; Hilger and Humer, 2003; Thiel and Christie, 2005; Xu et al., 2012; Giri and Reddy, 2014; Feng et al., 2018; Qin et al., 2020). However, these systems have been tested individually and mainly for the abatement of CH_4 (typically consisting of 50–60 % by vol of LFG; Majdinasab et al., 2017). Accordingly, no investigation has been focused yet on quantitatively evaluating the potential capacity of a biocover integrated with both SS and LL in degrading diversified LFG components (CH_4 and nonmethane VOCs, i.e., expected small fractions of relevant alkanes, alkenes, aromatics, cyclics, terpenes, halogenated compounds, and S-substituted compounds: Randazzo et al., 2023).

The present study aimed at investigating the efficiency of novel biocovers, treated with LL spraying and different amount of SS, in degrading generated biogas components in comparison with a natural soil, focusing on both the main species (i.e., CH_4, H_2) and trace organic compounds (i.e., VOC functional groups), through a laboratory experiment simulating a landfill environment. The implemented laboratory prototypes consisted each of (i) a digester containing degrading FW as the substrate (Alibardi and Cossu, 2015; Kumar and Samadder, 2020) to generate a biogas, connected with (ii) a soil column. Three prototypes were packed with the biocovers, whereas one prototype was used as a blank. The final goal of this study was to quantitatively estimate the abatement of biogas within the tested novel biocovers relative to a conventional landfill cover; accordingly, a novel index was designed to discern the action of bacterial activity from the dilution with atmospheric air.

2. Materials and methods

2.1. Raw materials

FW components were collected from the household and food service sectors and combined in the used organic substrate according to a typical FW composition in Italy (Randazzo et al., 2023, and pertaining references therein), as follows: fruits (30.8 % by wt), vegetables (40.1 % by wt), pasta/rice (11.8 % by wt), bakery products (4.2 % by wt), meat/fish (6.3 % by wt), dairy products (1.4 % by wt), and other food products (5.4 % by wt) not included in the previous components (i.e., coffee grounds, eggs).

An anaerobically stabilized sludge collected from a municipal wastewater treatment plant located in Central Italy was used as an inoculum to provide the necessary microorganisms for promoting the anaerobic degradation (AD) process of FW (Angelidaki and Sanders, 2004).

LL and SS (the latter consisting of a dry, anaerobically stabilized sludge derived from the final dewatering step, with grain size comparable to that of soil) were collected from a landfill for non-hazardous waste and a municipal wastewater treatment plant located in Central Italy, respectively.

The natural soil was collected from a *peri*-urban woodland located in Central Italy, after removal of the surface layer (~ 20 cm thick). The soil was manually milled and sieved with a 2-mm mesh and subsequently the grain size was determined by following the American Society for Testing and Materials (ASTM) methods D 422 – 63 and D 421 – 85 (ASTM, 2007a, 2007b). The soil was classified as low gravel silty sand according to the Unified Soil Classification System (USCS), consisting of the following granulometric classes: gravel (28.9 % by wt), sand (41.3 % by wt), silt (26.1 % by wt), and clay (3.7 % by wt).

2.2. Experimental setup and run

The experimental setup used in this study (Fig. S1, Supplementary material) was updated and extended from that reported by Randazzo et al. (2023) with four laboratory prototypes, each consisting of: (i) a 20 L digester in high-density polyethylene filled with a mixture (approximately 19 L) of FW, size-reduced by using an electric blender, and inoculum (1:1 on a volatile solid basis; Carchesio et al., 2014); (ii) a dark polypropylene cylinder (105 cm height and 7 cm inner diameter) acting as a soil column, whose bottom was connected to the digester headspace with a silicon tube. The column, maintained in vertical position and filled with soil almost up to its top, was equipped with four gas sampling ports aligned along the vertical axis, and spaced 22 cm from each other, with the upper one 9 cm from the top, and the lowest one 30 cm from the bottom. Each sampling port hosted a 5.5 cm long polyamide tube, with an inner diameter of 4 mm and equipped with a three-way stopcock. A dark polypropylene plate was placed at the column bottom, while the top remained uncapped to allow free escape of biogas and interaction with above air as occurring in landfills. The digester simulated the expected AD of deposited organic waste (specifically, FW) in the landfill body, whereas the column simulated the landfill cover soil. To homogenize the generated biogas fluxes from the digester through the soil, a 15 cm gravel layer (calcareous stones) was placed at the bottom of the column. The columns were filled carefully to minimize soil compaction which could potentially interfere with the uprising of the biogas produced within the digesters.

In the experimental run, the columns were filled as follows: B column with 90 cm of untreated soil as a blank; SS1 and SS2 columns with 30 cm of untreated soil at the bottom and the upper 60 cm consisting of a mixture of untreated soil and SS in a soil:SS mass ratio of 10:1 and 20:1, respectively; and SSLL column with the SS2-type filling conditions and additionally sprayed with 100 mL (300 mL in total) of LL every 14 days, starting from the beginning of the experiment (day 0). The soil:SS mass ratios were chosen considering the potential practical application in a real landfill environment, while the volume and frequency of sprayed LL was chosen to avoid the complete saturation of the soil; furthermore, LL was sprayed slowly to minimize the soil compression due to the liquid injection. To prevent LL stagnation at the SSLL column bottom, the LL excess was collected at the column bottom through a dedicated port installed in the polypropylene plate.

The experimental run, lasting 40 days, was performed outdoors at the internal courtyard of the Department of Earth Sciences at the University of Florence during the autumn season. This duration time was reliably higher than the 30-day period for anaerobic biodegradability tests indicated by Owen et al. (1979) to ensure, in most cases, complete decomposition of biodegradable substrates (Randazzo et al., 2023). Consistently, the run duration time resulted also close to typical durations (45, 46 days) of anaerobic biodegradability tests for diversified organic substrates (Carchesio et al., 2014, 2020).

2.3. Gas sampling and analyses

Gas samples from the headspace of the digesters and the four sampling ports along the soil columns were collected every 7 days by using a 60 mL Luer Lock syringe (Beroject III®) and conveyed into 12 mL glass vials (Labco Exetainer®) prefilled with: (i) deionized water (for analysis of VOCs) and (ii) deionized water slightly acidified with ultrapure HCl (for analysis of main gaseous species) to avoid CO_2 dissolution (Randazzo et al., 2023).

The main gaseous species (CO_2, CH_4, H_2, N_2, Ar, and O_2) were analyzed using a Shimadzu 15A gas chromatograph (GC) equipped with a thermal conductivity detector (TCD). Methane concentrations < 0.05

% by vol and light hydrocarbons (C_2-C_4) were determined using a Shimadzu 14A GC equipped with a flame ionization detector (FID). Analytical errors for both GC-TCD and GC-FID were < 5 %.

The C_{4+} VOCs were analyzed using a Thermo Trace Ultra GC combined with a Thermo DSQ quadrupole mass spectrometer (MS), according to the solid-phase microextraction (SPME) method (Arthur and Pawliszyn, 1990) with a 2 cm long 3 phase fiber (Supelco; Bellefonte, PA, USA) consisting of. divinylbenzene (DVB)–Carboxen (Car)–polydimethylsiloxane (PDMS). The fiber was introduced into each glass vial via the rubber septum using a manual SPME device and exposed to the sampled gases for 30 min at 20 °C. VOCs absorbed by the SPME fiber were then desorbed at 230 °C (2 min) in the column headspace of the GC–MS instrument equipped with a TR-V1 fused silica chromatographic column (30 m x 0.25 mm i.d., 1.4 μ m film thickness), using He as carrier gas at 1.3 ml/min in constant pressure mode. The desorbed gas was injected into the gas chromatograph through a port operated in splitless mode. The column oven temperature program was: 35 °C (10 min) to 180 °C (held 3 min) at 5.5 °C/min and then up to 230 °C (held 6 min) at 20 °C/min. The quadrupole mass spectrometer operated in electron ionization mode (EI), with ionization energy 70 eV and source temperature 250 °C, whereas the GC–MS transfer line was at 230 °C. Full scan mode (m/z 35 to 400) was used. Retention times and mass spectra were both used to assign VOCs, using the mass spectra database of the NIST05 library (NIST, 2005) for comparison. Quantitative analysis was carried out via an external standard procedure using a mixture in MeOH or, alternatively, in hexane. Relative standard deviation (RSD), calculated from five replicate analyses of the standard mixtures, was < 5 % (Tassi et al., 2009).

In addition, the carbon isotope signatures ($^{13}C/^{12}C$) in CO_2 ($\delta^{13}C$-CO_2) and CH_4 ($\delta^{13}C$-CH_4), expressed as ‰ vs V-PDB, were determined by cavity ring-down spectroscopy (CRDS) using a Picarro G2201-I analyzer. Analytical error for CRDS was < 1.15 ‰ vs V-PDB.

2.4. Novel index to estimate soil treatment benefits on biogas abatement

To investigate the role of microbial activity in modifying the gaseous compositions along the tested soil columns, at first the expected concentrations of main AD-related species (CH_4, CO_2, and H_2) and VOC functional groups were recalculated, starting from those originally detected in the respective digesters, assuming that along the columns they were affected only by air dilution. To this purpose, the percentages of air within digesters and columns were calculated by dividing the sum of N_2, O_2, and Ar in respective gas samples by the typical sum of N_2, O_2, and Ar in atmospheric air (i.e., ~99.92):

$$\%Air = \frac{(N_2 + O_2 + Ar)_{ingassamples}}{(N_2 + O_2 + Ar)_{inair}} x100 \quad (1)$$

Then, the concentrations of main AD-related species and VOC functional groups expected from air dilution along the columns (S_{exp}) were computed considering the decrease in the fraction of biogas upward the columns due to the increase of the air fraction on the total gas, according to the following expression:

$$S_{exp} = S_{indigester} - \left[\frac{S_{indigester} * (\%AirC - \%AirD)}{100}\right] \quad (2)$$

where $S_{in\ digester}$ was the concentration of the S species (functional group) measured in gas samples from the headspaces of the four digesters, while $\%AirC$ and $\%AirD$, representing respectively the percentage of air within the four columns and digesters, was calculated using equation (1).

The gaps within S_{exp} and the concentrations measured in gas samples along the soil columns were inferred to be caused by biochemical processes. Thus, an initial index I_d (referred to digesters) was introduced for each gas sample and related species (functional group) to quantitatively evaluate the effects of these processes, according to the following ratio:

$$I_d = \frac{S_{meas}}{S_{exp}} \quad (3)$$

where S_{meas} was the concentration of the S species (functional group) measured within the soil, and S_{exp} was calculated using equation (2).

To estimate the benefits on biogas abatement provided by soil treatments, a complementary, novel index I_{ba} (referred to bacterial activity) has been introduced, according to the following ratio:

$$I_{ba} = \frac{I_{d,treated}}{I_{d,blank}} \quad (4)$$

where $I_{d,treated}$ and $I_{d,blank}$ represent the I_d values calculated by equation (3) for treated (SSLL, SS1, or SS2) and blank (B) columns, respectively.

3. Results

Minimum and maximum concentrations of the main gaseous species and VOCs (properly aggregated in the following functional groups: alkanes, alkenes, aromatics, cyclics, halogenated compounds, S-substituted compounds, terpenes, and O-substituted compounds) in the gas samples from digesters and B, SSLL, SS1, and SS2 columns are reported in Tables 1 and 2, respectively.

The whole dataset, including the $\delta^{13}C$-CO_2 and $\delta^{13}C$-CH_4 values and all the detected concentrations of main gaseous species and VOCs (up to 54 species) from digesters and four soil columns, is reported in Table S1 (Supplementary material).

3.1. Digesters

As expected, the compositions of biogases from headspaces of the four digesters did not show significant differences (Fig. 1), since all the digesters were filled with the same FW + inoculum mixture. At the beginning of the experiment (day 0), the main AD-related species had the lowest concentrations that increased until days 19–26 (Fig. 1), when the detected CH_4 and CO_2 concentrations resulted in reliable agreement with typical ranges expected for LFG (i.e., 30–60 % by vol for CH_4, and 15–30 % by vol for CO_2; Rey et al., 2013). After day 26, the main AD-related species showed slightly decreasing trends reflecting decreases in biogas productions (Fig. 1). The air-related species (N_2, O_2, and Ar) showed an opposite trend, exhibiting the highest concentrations on day 0; N_2 and Ar progressively decreased until days 19–26 and slightly increased during days 33–40, whereas O_2 was no longer detected from day 5 onwards (Fig. 1).

The concentrations of the different VOC functional groups, displaying total values ranging from 1.68×10^5 to 6.13×10^5 ppb during the whole experiment, depicted trends similar to those of the main AD-related species (Fig. 1). Aromatics (mostly toluene) and alkanes (mostly ethane) were the most abundant, followed by cyclics, terpenes, O-substituted compounds, and alkenes, while halogenated and S-substituted compounds occurred at subordinate amounts.

The $\delta^{13}C$-CO_2 and $\delta^{13}C$-CH_4 values slightly decreased on days 12–19, increasing during the last days close to the initial values (Table S1, Supplementary material).

3.2. Soil columns

Compositional data from B column, as well as SSLL, SS1, and SS2 columns, showed comparable temporal and vertical trends in the AD-related species, reflecting the evolutions of biogases produced inside the digesters and revealing decreasing concentrations towards the top of the soil columns.

Air-related species dominated the gas compositions all over the experiment, depicting opposite trends with respect to those of AD-related species.

Methane and CO_2 depicted decreasing trend in spatial evolution,

Table 1
Minimum and maximum concentrations of N_2, O_2, Ar, H_2, CO_2, and CH_4 in gases from digesters and B, SSLL, SS1, and SS2 columns.

Main gas species (% by vol.)		Day 0		Day 5		Day 12		Day 19		Day 26		Day 33		Day 40	
		min	max	min	max	min	max	min	max	min	max	min	max	min	max
N_2	Digester	61.30	63.30	24.80	25.10	15.90	18.50	14.50	15.50	14.90	15.20	17.50	18.90	18.40	20.10
	B	65.50	82.80	63.40	86.20	57.60	88.70	55.90	82.60	57.50	87.70	61.90	88.90	66.90	88.60
	SSLL	72.80	85.60	63.90	90.50	70.40	90.50	69.90	89.80	70.30	90.40	73.30	90.40	85.60	90.50
	SS1	76.50	88.90	63.40	89.90	65.40	90.60	60.30	90.50	62.30	89.60	70.60	90.80	77.90	90.50
	SS2	71.90	84.90	65.50	90.70	71.10	90.60	73.30	90.70	71.20	90.10	78.10	90.50	87.90	90.40
O_2	Digester	0.75	0.96	–	–	.	–	–	–	–	–	–	–	–	–
	B	4.30	9.60	2.90	8.60	0.75	5.10	2.20	8.90	0.79	6.20	1.70	6.80	1.80	7.10
	SSLL	3.50	9.60	1.70	6.10	0.45	4.10	0.85	4.90	0.75	3.80	0.85	5.10	1.10	5.60
	SS1	3.90	8.70	2.30	7.10	0.58	5.60	1.60	5.90	1.80	6.30	1.10	6.40	1.20	7.30
	SS2	3.50	11.00	2.10	7.50	0.95	7.60	1.30	7.20	1.50	8.20	1.50	8.20	1.60	8.20
Ar	Digester	0.75	0.82	0.31	0.34	0.22	0.25	0.19	0.23	0.20	0.22	0.23	0.25	0.25	0.25
	B	0.83	1.04	0.76	1.00	0.68	1.10	0.70	1.00	0.70	1.10	0.75	1.10	0.80	1.10
	SSLL	0.95	1.10	0.76	1.10	0.85	1.10	0.85	1.10	0.88	1.10	0.82	1.10	1.10	1.10
	SS1	0.96	1.10	0.75	1.10	0.78	1.10	0.76	1.10	0.79	1.10	0.85	1.10	0.95	1.10
	SS2	0.95	1.10	0.84	1.10	0.85	1.10	0.87	1.10	0.85	1.10	0.95	1.10	1.10	1.10
H_2	Digester	0.01	0.02	6.30	7.30	7.30	7.60	5.50	6.40	5.60	7.60	5.40	5.90	5.50	7.00
	B	–	–	0.05	4.10	0.05	4.20	0.15	2.60	0.25	3.90	0.25	3.10	–	2.60
	SSLL	–	0.01	0.05	3.70	0.15	2.50	0.05	2.30	0.55	3.10	0.05	1.90	0.15	1.80
	SS1	–	0.01	0.05	3.40	0.09	2.30	0.05	1.10	0.05	2.30	0.05	1.70	–	1.10
	SS2	–	0.01	–	3.10	–	1.80	–	1.40	–	1.50	–	0.85	–	0.55
CO_2	Digester	19.70	23.10	31.50	32.90	32.10	32.80	34.80	35.40	33.70	34.30	33.20	33.70	30.30	33.80
	B	2.90	17.60	2.80	9.90	3.30	15.40	5.60	15.10	3.40	13.20	2.20	11.80	2.60	13.10
	SSLL	3.30	18.80	2.20	17.30	3.60	12.70	3.10	11.00	3.60	11.10	3.10	12.10	2.50	4.30
	SS1	2.10	14.50	1.90	16.40	1.80	16.60	1.70	20.40	2.30	19.60	1.70	15.40	1.10	13.20
	SS2	2.40	19.10	0.75	16.20	0.45	13.60	0.75	11.70	0.45	15.40	0.25	10.70	0.25	4.30
CH_4	Digester	12.50	14.50	34.80	36.90	41.60	43.90	43.50	44.20	43.80	45.30	42.10	43.20	41.30	44.30
	B	1.80	10.90	1.10	19.30	2.10	21.30	2.10	23.60	1.30	24.30	1.10	20.70	0.61	15.40
	SSLL	0.45	4.10	0.05	12.70	0.55	13.10	1.10	15.10	0.55	13.90	0.25	11.10	0.15	6.10
	SS1	0.71	4.30	0.13	13.70	0.55	14.50	0.51	14.90	0.35	13.70	0.11	10.50	0.05	5.60
	SS2	0.55	4.50	0.05	12.30	0.22	11.70	0.25	11.50	0.15	9.50	0.05	7.90	0.05	4.50

remarking the occurrence of degradation processes affecting CH_4 to a larger extent with respect to CO_2. The fate of H_2 was similar to those of CH_4 and CO_2, showing decreasing concentrations upward the columns. Nitrogen and O_2 depicted opposite trends, with increasing concentrations upward the column tops.

The occurrence of O_2 within the columns influenced the composition and evolution of VOCs: $C_{2\text{-}3}$ alkanes and aromatics, including benzene, drastically decreased moving upward through the soil columns, with cyclics and terpenes showing a similar trend. Alkenes were sporadically detected in gas samples from intermediate depths of the columns, and they were no longer detected at the column tops. Halogenated compounds decreased upwards in the columns to a lesser extent relative to other VOC species. Sulphur-substituted compounds, mainly consisting of dimethylsulfoxide (DMSO), were completely degraded within the intermediate layer of soil columns. Oxygen-substituted compounds, especially aldehydes, showed increasing concentration upward along the soil columns.

The δ^{13}C-CO_2 and δ^{13}C-CH_4 values depicted temporal trends similar to those within headspaces of digesters, with a general increase in values towards the top of the soil columns.

4. Discussion

4.1. Compositional evolutions of biogases from digesters to column bottoms

The temporal evolutions of CH_4/CO_2 and H_2/CH_4 ratios observed in headspaces of all digesters (Fig. 2a,b) reflected the typical steps characterizing the degradation process of organic matter expected in a landfill body. The relatively low CH_4/CO_2 and H_2/CH_4 ratios (from 0.55 to 0.74 and from 0.6×10^{-3} to $1{,}2 \times 10^{-3}$, respectively) observed from day 0 indicated the occurrence of aerobic degradation and hydrolysis/fermentation steps, where (i) the atmospheric O_2 initially contained within the digesters was consumed to produce short-chain hydrocarbons, CO_2, and water, and (ii) carbohydrates, proteins, and lipids were hydrolyzed forming CH_4, CO_2, H_2, NH_3, and organic acids as final products (Williams, 2005; Jain et al., 2015). On day 5, the CH_4/CO_2 and H_2/CH_4 ratios increased (ranging from 1.06 to 1.17 and 0.17 to 0.21 in the four digesters, respectively), depicting the transition between hydrolysis/fermentation and acetogenesis steps, when organic acids and their derivatives started to be converted into CH_4 and CO_2 by the promoted methanogens (Williams, 2005). During the following days (12–40), the CH_4/CO_2 ratios increased (finally ranging from 1.22 to 1.46 in the four digesters), while the H_2/CH_4 ratios decreased (finally ranging from 0.12 to 0.18 in the four digesters), indicating the occurrence of methanogenesis step, when microbial consortia were expected to provide the dismutation of acetate in CH_4 and CO_2 (acetoclastic methanogenesis) but also the conversion of CO_2 and H_2 in CH_4 (hydrogenotrophic methanogenesis) (Williams, 2005). The establishment of methanogenesis step was supported by the fluctuation of δ^{13}C-CO_2 and δ^{13}C-CH_4 values (Fig. 2 c,d), likely produced by the combined influence of the mentioned acetoclastic methanogenesis (with δ^{13}C-CO_2 and δ^{13}C-CH_4 values from −30 to 0 ‰ and −30 to −15 ‰ vs V-PDB, respectively) and hydrogenotrophic methanogenesis (causing a large scattering of δ^{13}C-CO_2 and δ^{13}C-CH_4 values, from −20 to 20 ‰ and −60 to −40 ‰ vs V-PDB, respectively) (Polag et al., 2013; Polag et al., 2015;

Table 2
Minimum and maximum concentrations of VOC functional groups (alkanes, alkenes, aromatics, cyclics, halogenated compounds, S-substituted compounds, terpenes, and O-substituted compounds) in gases from digesters and B, SSLL, SS1, and SS2 columns.

VOCs (ppb)		Day 0		Day 5		Day 12		Day 19		Day 26		Day 33		Day 40	
		min	max	min	max	min	max	min	max	Min	max	min	max	min	max
Alkanes	Digesters	73,600	86,900	204,500	222,000	252,000	264,000	261,000	276,000	261,000	270,000	250,000	260,000	247,000	261,000
	B	10,500	65,800	6400	116,000	13,200	129,000	12,400	141,000	7600	144,000	6480	12,200	3450	90,000
	SSLL	2700	24,000	300	76,400	3400	79,000	6700	90,500	3900	80,200	1200	63,800	810	35,000
	SS1	4700	26,500	660	80,000	4500	84,800	4600	84,400	3500	76,600	570	56,800	91	29,600
	SS2	3100	25,400	270	71,000	1400	69,300	1300	66,400	780	53,400	260	43,800	33	24,200
Alkenes	Digesters	20	30	50	70	60	80	76	80	75	86	74	81	67	76
	B	3	20	1	30	4	38	3	45	1	41	1	37	–	21
	SSLL	–	7	–	20	–	20	1	14	–	15	–	13	–	6
	SS1	–	5	–	10	–	10	–	5	–	6	–	3	–	1
	SS2	–	6	–	10	–	10	–	5	–	6	–	3	–	1
Aromatics	Digesters	89,900	104,200	213,000	244,000	306,000	329,000	330,000	341,000	318,000	339,000	329,000	351,000	317,000	340,200
	B	11,000	79,400	6140	129,000	12,500	123,000	11,200	125,000	9000	139,000	7120	121,000	3600	87,000
	SSLL	2600	22,000	170	70,500	3300	84,000	6800	62,200	3300	114,000	1500	57,000	930	33,300
	SS1	1800	25,100	160	52,100	1300	69,900	1700	44,800	1300	54,000	220	41,200	24	23,500
	SS2	1700	20,800	120	53,700	900	66,100	1300	42,200	900	57,000	180	40,700	21	23,500
Cyclics	Digesters	520	610	1150	1300	1800	1900	2050	2340	2500	2780	2620	2700	2100	2200
	B	80	500	54	810	110	1060	110	1130	60	1160	56	1100	21	710
	SSLL	20	150	1	440	20	530	42	480	18	450	8	400	4	190
	SS1	10	130	1	280	4	290	2	300	1	220	–	200	–	100
	SS2	10	120	1	270	5	300	2	310	1	240	–	170	–	90
Halogented compounds	Digesters	4	5	10	11	13	14	13	15	13	15	12	14	12	14
	B	–	3	–	6	1	7	–	8	1	8	1	6	–	3
	SSLL	–	1	–	4	–	4	1	5	–	7	–	4	–	2
	SS1	–	–	–	4	1	5	–	6	–	5	–	3	–	2
	SS2	–	1	–	4	–	4	–	4	–	3	–	3	–	2
S-substituted compounds	Digesters	–	–	1	1	3	3	3	4	3	4	3	4	3	4
	B	–	–	–	1	–	2	–	2	–	3	–	2	–	2
	SSLL	–	–	–	1	–	2	–	2	–	3	–	3	–	2
	SS1	–	–	–	1	–	3	–	3	–	3	–	3	–	3
	SS2	–	–	–	1	–	3	–	3	–	3	–	3	–	3
Terpenes	Digesters	280	320	900	980	1100	1120	1100	1150	1100	1140	1000	1100	960	1030
	B	30	90	15	260	30	290	26	360	32	370	20	260	7	170
	SSLL	1	20	2	110	8	90	20	110	7	110	6	80	3	68
	SS1	1	20	1	90	2	50	3	47	2	50	1	34	1	22
	SS2	1	20	1	80	3	50	4	46	2	49	1	35	1	19
O-substituted compounds	Digesters	20	30	70	80	90	100	100	130	95	110	100	120	94	120
	B	90	190	110	460	200	500	160	480	190	470	160	390	200	480
	SSLL	100	1200	330	680	330	710	400	1140	300	890	300	990	270	950
	SS1	150	1100	490	1300	500	1300	640	1320	480	1200	290	1300	530	1300
	SS2	120	1300	460	1400	480	1300	590	1400	440	1140	460	1220	450	1200

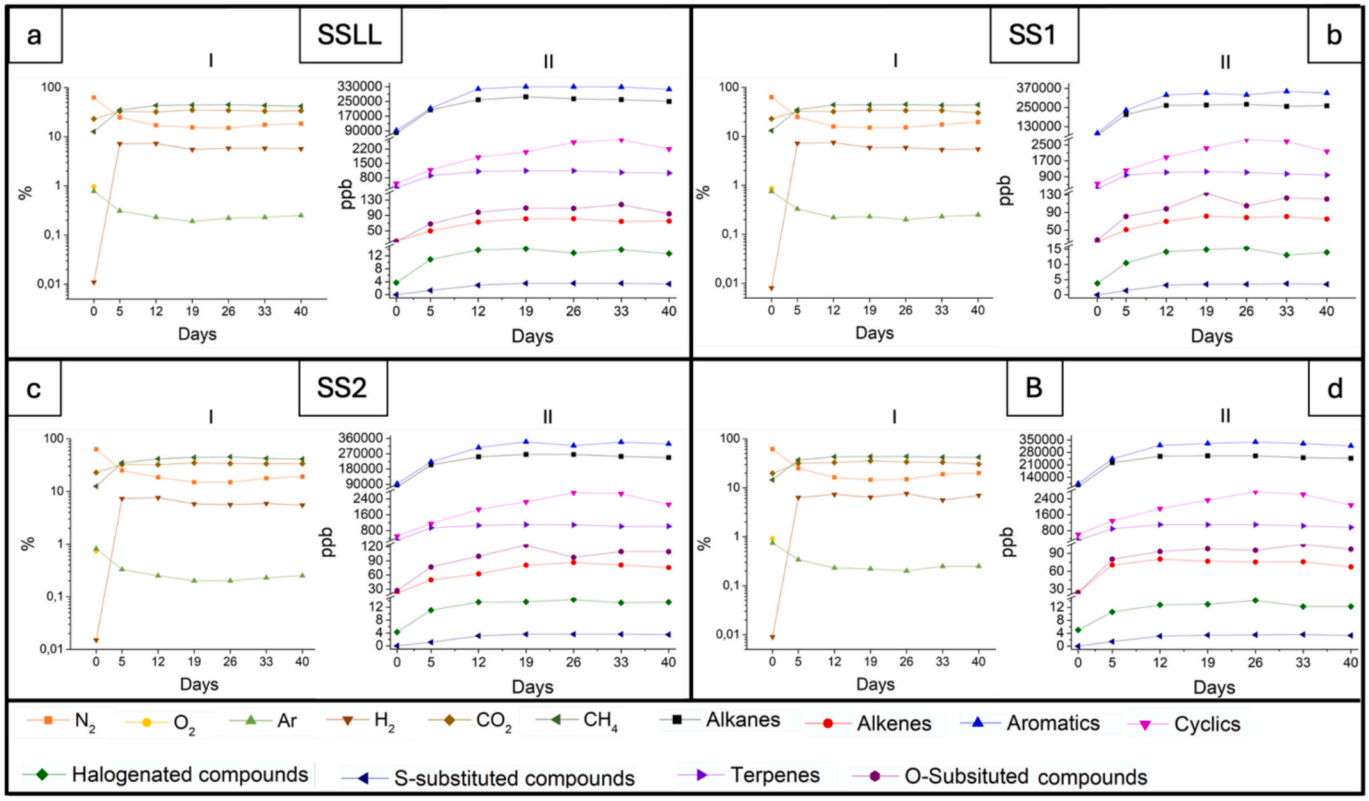

Fig. 1. Main gaseous species (I) and VOC functional groups (II) from headspaces of digesters (identified by pertaining columns: a, SSLL; b, SS1; c, SS2; and d, B).

Randazzo et al., 2020). On the whole, the δ^{13}C-CH_4 values in Fig. 2 (d) were higher than a possible range detected for LFG (from −62 to −55 ‰ vs V-PDB; Raco et al., 2014), probably due to the distinctive organic substrate (FW) tested in this study, since waste type is expected to largely influence the organic degradation pathways (Hackley et al., 1996).

The temporal evolutions of the concentrations of VOC functional groups resembled the trends observed for main AD-related species, increasing until days 19–26, when they reached a peak; in the subsequent days, alkanes, alkenes, aromatics, halogenated compounds, S-substituted compounds, and terpenes did not show significant variations, cyclics showed a decreasing trend, and O-substituted compounds depicted a first decreasing step followed by an increasing one (Fig. 1).

The temporal evolutions of the concentrations of main AD-related species and VOC functional groups, in all referable to the generated biogases, at the bottoms of the soil columns generally reflected the trends revealed from the headspaces of digesters (Table S1, Supplementary material). However, the resulting predominance of air-related species all over the experiment in the bottoms was likely referable to both (i) the air initially present within the columns prior to the beginning of the experiment and (ii) the air permeating from uncapped tops of the columns. Indeed, air dilution was not the only process governing the composition of main AD-related species at the column bottoms. The CH_4/CO_2 (Fig. 3a) and O_2/N_2 (Fig. 3b) ratios at the column bottoms were generally lower relative to the biogases from the digesters (Fig. 2a) and the atmospheric air ratio (~0.27), respectively. This suggests that (i) CH_4 was strongly affected by O_2-consuming biodegradation processes (Schuler and Conrad, 1990; Kightley et al., 1995; Boeckx et al., 1997; Hanson and Hanson, 1996; Gebert and Gröngröft, 2006; Huber-Humer et al., 2008; Sadasivam and Reddy, 2014; Kjeldsen and Scheutz, 2018; Randazzo et al., 2024) and (ii) CO_2, besides being less reactive, represented the metabolite of CH_4 oxidation (Pratt et al., 2013; Randazzo et al., 2024). The temporal evolutions of CH_4/CO_2 ratio at the column bottoms (Fig. 3a) roughly followed the trends for the generated biogases in the digester headspaces (Fig. 2a), while the temporal evolutions of O_2/N_2 ratio (Fig. 3b) were partially specular to the CH_4/CO_2 ones, confirming the opposite relationship between O_2 levels and CH_4 aerobic biodegradation. The resulting H_2/CH_4 ratios at the column bottoms (Fig. 3c) resembled the temporal evolutions by the biogases from the digesters (Fig. 2b), showing values similar to or slightly higher than those within the respective digester headspaces. The higher H_2/CH_4 ratios and related fluctuations observed in Fig. 3c might be attributed to the consumption, to a higher extent, of CH_4 than H_2 by methanotrophs and *Knallgas bacteria* (i.e., a bacterial group using O_2 as final electron acceptor to oxidize H_2), respectively (Mohammadi et al., 2017; Piché-Choquette and Constant, 2019).

The occurrence of both air dilution and bacterial biodegradation at the column bottoms was further suggested by the δ^{13}C-CO_2 and δ^{13}C-CH_4 values (Fig. 3d,e), resulting slightly higher than those for the biogases from the digesters (Fig. 2c,d). The carbon isotopic signature of CO_2 from the column bottoms showed less marked differences compared to those of the digesters, while the δ^{13}C-CH_4 values displayed more noticeable fluctuations: this could be attributed to mixing with ^{12}C-enriched CO_2 derived from CH_4 oxidation (Bogner et al., 1996; Hackley et al., 1996; Liptay et al., 1998; Randazzo et al., 2020) that probably masked the increase in ^{13}C-enriched CO_2 due to CO_2 fixation process within soil (Šantrůčková et al., 2005; Nel and Cramer, 2019).

4.2. *Biodegradation processes of main AD-related species and VOCs along the soil columns*

Biodegradation processes of the main AD-related species and VOC functional groups along the soil columns were considered and quantified using the initial index I_d, calculated according to equation (3). The resulting I_d values (Table 3) provided an estimation of the effects of biochemical processes on the biogas composition within the tested soil columns, based on the following distinctive partitions: (i) $I_d < 1$, the species – functional group was biodegraded within the soil column (with

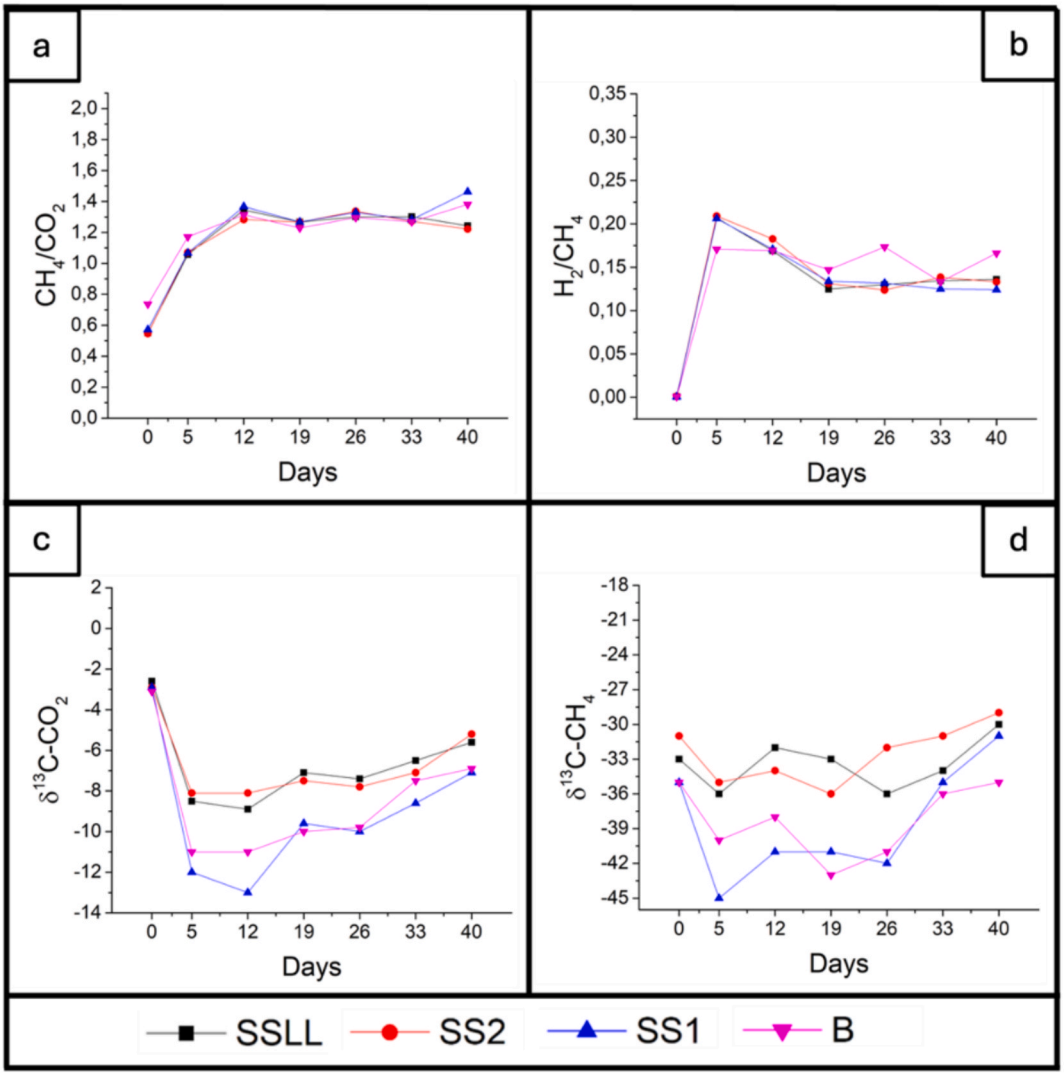

Fig. 2. CH_4/CO_2 (a) H_2/CH_4 (b), $\delta^{13}C$-CO_2 (c), and $\delta^{13}C$-CH_4 (d) from headspaces of digesters (identified by pertaining columns: SSLL, SS1, SS2, and B). The isotopic signatures are expressed in ‰ vs V-PDB.

lower limit of $I_d = 0$ indicating the condition of species even not detected in the soil gas); (ii) $I_d = 1$, the species – functional group was not affected by biochemical processes within the soil column; and (iii) $I_d > 1$, the species – functional group was bioproduced within the soil column.

As expected, species and functional groups that were known in the technical-scientific literature to be affected by biodegradation processes promoted by methanotrophs and VOCs-degrading bacteria, i.e., CH_4, H_2, alkanes, alkenes, aromatics, cyclics, and terpenes (Schuler and Conrad, 1990; Kightley et al., 1995; Boeckx et al., 1997; Hanson and Hanson, 1996; Gebert and Gröngröft, 2006; Huber-Humer et al., 2008; Pratt et al., 2013; Sadasivam and Reddy, 2014; Pandey et al., 2014; Kjeldsen and Scheutz, 2018; Jabłońska and Tawfik, 2019; Randazzo et al., 2024), were overall characterized in Table 3 by $I_d < 1$ (except for sporadic samples on H_2 and alkenes). Moreover, the I_d values of these species and functional groups depicted decreasing trends towards the tops of the columns, remarking increasing biodegradation promoted by aerobic bacterial consortia occurring in the shallower portions of the columns, where oxygen availability increased. Complementarily, the I_d values of O-substituted compounds resulted > 1 (Table 3), with a sharp increase upwards along the columns; since they are expected metabolites of CH_4 and VOC degradation (Pratt et al., 2013; Randazzo et al., 2024), their spatial and temporal trends supported the enhancement of bacterial consortia activity upwards along the columns and over time (Rojo, 2009; Tassi et al., 2009; Tan et al., 2017). The I_d values of CO_2 were sporadically > 1 or predominantly close to 1 at the column bottoms (Table 3), suggesting that, besides being assimilated by heterotrophic microorganisms within the soil to be used in aerobic metabolism, especially with anaerobic reactions (Šantrůčková et al., 2005; Scheutz et al., 2009; Nel and Cramer, 2019), this species was also produced by CH_4 biodegradation. Upwards along the columns, the I_d values of CO_2 were characterized by lower values, probably indicating how the soil microbial anaerobic CO_2 fixation efficiently counteracted the CO_2 bioproduction due to CH_4 degradation. The I_d values of halogenated compounds at column bottoms were sporadically > 1 or close to 1, while upwards in the columns their I_d displayed values < 1 (Table 3). This evidence suggested that these compounds were affected to a certain extent by biodegradation within soil under aerobic conditions, despite their recalcitrant nature (Scheutz and Kjeldsen, 2005; Tassi et al., 2009). Finally, the S-substituted compounds, mainly represented by DMSO, showed generally I_d values > 1 at the column bottoms (from day 5 onwards: Table 3) due to the expected production of this compound from dimethylsulfide (DMS) oxidation (Jonkers et al., 1996; Lomans et al., 2002), the latter dominating the S-substituted compounds in biogases from the digester headspaces (Table S1, Supplementary material).

The isotopic signatures of C in CH_4 and CO_2 (Table S1, Supplementary material) remarked the biodegradation processes

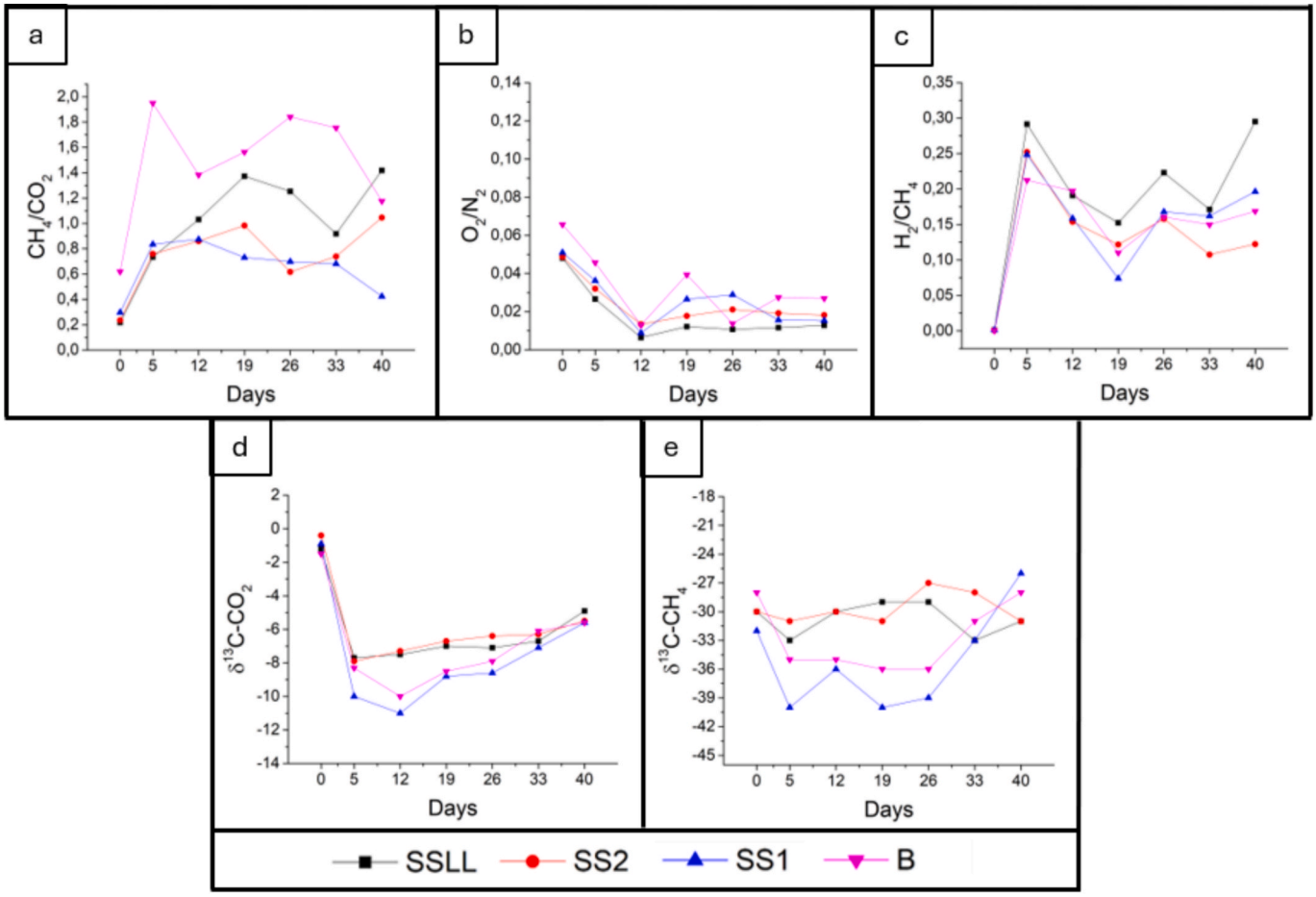

Fig. 3. CH_4/CO_2 (a), O_2/N_2 (b), H_2/CH_4 (c), $\delta^{13}C$-CO_2 (d), and $\delta^{13}C$-CH_4 (e) from sampling points located at the bottom (75 cm below the top) of the columns SSLL, SS1, SS2, and B. The isotopic signatures are expressed in ‰ vs V-PDB.

affecting these species. The $\delta^{13}C$-CO_2 values slightly increased both along the soil columns and over time, indicating how this species was biodegraded within the soils causing a ^{13}C-enrichment within the residual CO_2. Gases from the column tops showed $\delta^{13}C$-CO_2 temporal trends characterized by smoothed fluctuations, possibly due to atmospheric air mixing that masked the enrichment in ^{13}C-CO_2 in residual CO_2 derived from its biodegradation. The $\delta^{13}C$-CH_4 depicted a pronounced increase along the columns, compared to $\delta^{13}C$-CO_2. Moreover, the temporal trends of $\delta^{13}C$-CH_4 were quite variable, being dependent on the balance between (i) CH_4 consumption, causing a general ^{13}C increase in the residual CH_4, and (ii) addition of CH_4 (with more negative isotopic signature) migrating upward from the digesters. Gases from the column tops showed less marked temporal fluctuation of $\delta^{13}C$-CH_4 values, likely due to the addition of atmospheric CH_4 masking the enrichment in $\delta^{13}C$-CH_4 in residual CH_4 derived from its biodegradation, as detected for CO_2.

4.3. Assessment of soil treatment effects on biogas abatement

The benefits on biogas abatement (in terms of main Ad-related species and VOC functional groups) provided by the investigated biocovers were finally represented by the complementary, novel index I_{ba}, calculated according to equation (4). The resulting I_{ba} values (Table 4) provided the following distinctive partitions: (i) $I_{ba} > 1$, indicating relatively high bioproduction and/or low biodegradation of the species – functional group in treated soil relative to untreated one; (ii) $I_{ba} < 1$, indicating relatively high biodegradation of the species – functional group in treated soil relative to untreated one; and (iii) $I_{ba} = 1$, indicating no evident effect of the soil treatments.

On the whole, the soil treatments tested as novel biocovers promoted a general improvement in the abatement action on CH_4, alkanes, alkenes, aromatics, cyclics, halogenated compounds, and terpenes, as shown by the resulting I_{ba} values of SSLL, SS1, and SS2 globally or predominantly < 1 (Table 4, Fig. 4). However, each soil treatment provided benefits in biogas biodegradation with distinctive temporal and spatial patterns. Benefits provided by LL emerged in the first 12 days of the experiment (Table 4) when CH_4 and alkanes were predominantly characterized in SSLL by the lowest I_{ba} values. From day 19 onwards, the SS amended columns outperformed the SSLL column in terms of CH_4 and alkane degradation efficiency (Table 4). Additionally, the SSLL biodegradation of H_2 was mostly lower than in other treated columns (Table 4), even relative to untreated soil B (due to some I_{ba} values > 1). These results suggested that the addition of LL (expected to be rich in nutrients) to the soil (i) favored microbial activity producing H_2 and other VOC species as by-products, and (ii) decreased the availability of O_2 that was also used to degrade organic matter dissolved in LL itself (Kjeldsen et al., 2002; Williams, 2005; Pichè-Choquette and Constant, 2019; Li et al., 2021). The fate of CO_2 was regulated by the mentioned contrasting effects of (i) CO_2 fixation and (ii) bioproduction from CH_4 oxidation, the latter prevailing in the deepest parts of the treated columns as shown by I_{ba} values resulting predominantly > 1 (Table 4, Fig. 4), while CO_2 fixation dominated in the shallower parts of the treated columns (due to I_{ba} values resulting predominantly < 1: Table 4, Fig. 4). At the SSLL column top, the I_{ba} values for CO_2 were sporadically > 1, indicating a worse fixation of this species compared to other columns (Table 4, Fig. 4). Biodegradation of alkenes, aromatics, cyclics,

Table 3

Resulting I_d values for main AD-related species and VOC functional groups within treated (SSLL, SS1, SS2) and untreated (B) soil columns. Numerical rows refer sequentially to gas samples at points from lower (1) to upper (4) along each column (see Fig. S1). Shades of red and blue represent biodegradation and bioproduction, respectively (bright color correspond to high bacterial activity, faded color correspond to low bacterial activity; bright red values under 0.33, medium red values between 0.33 and 0.66, faded red values between 0.66 and 1, white values equal to 1, faded blue values between 1 and 25, medium blue values between 25 and 50, bright blue values over 50).

Sampling Points	Index I_d	Day 0				Day 5				Day 12				Day 19				Day 26				Day 33				Day 40			
		B	SSLL	SS1	SS2	B	SSLL	SS1	SS2	B	SSLL	SS1	SS2	B	SSLL	SS1	SS2	B	SSLL	SS1	SS2	B	SSLL	SS1	SS2	B	SSLL	SS1	SS2
4	H_2	0.00	0.00	0.00	0.00	0.03	0.03	0.03	0.00	0.03	0.10	0.06	0.00	0.11	0.05	0.05	0.00	0.22	0.49	0.05	0.00	0.20	0.04	0.05	0.00	0.00	0.12	0.00	0.00
3		0.00	0.00	0.00	0.00	0.08	0.06	0.02	0.02	0.07	0.20	0.08	0.08	0.27	0.18	0.03	0.04	0.71	0.64	0.11	0.00	0.47	0.11	0.04	0.00	0.13	0.51	0.00	0.00
2		0.00	0.00	0.00	0.00	0.69	0.73	0.64	0.60	0.67	0.50	0.41	0.36	0.95	0.36	0.46	0.69	0.99	0.42	0.34	0.61	0.66	0.33	0.15	0.48	0.71	0.31	0.10	
1		0.24	0.52	0.75	0.38	1.12	0.88	0.80	0.75	1.00	0.75	0.62	0.52	0.73	0.95	0.35	0.61	1.24	1.23	0.77	0.65	1.01	0.77	0.70	0.38	0.93	1.03	0.50	0.35
4	CO_2	0.21	0.21	0.14	0.15	0.30	0.24	0.21	0.09	0.47	0.52	0.30	0.07	0.71	0.45	0.27	0.13	0.50	0.53	0.37	0.08	0.30	0.44	0.26	0.04	0.36	0.35	0.17	0.04
3		0.47	0.40	0.33	0.39	0.65	0.59	0.54	0.37	0.78	0.58	0.83	0.47	0.93	0.71	0.72	0.59	0.78	0.57	0.71	0.56	0.71	0.55	0.66	0.34	0.64	0.51	0.56	0.31
2		0.62	0.63	0.64	0.60	0.68	0.65	0.70	0.68	0.73	0.80	0.90	0.75	0.78	0.69	0.87	0.77	0.70	0.63	0.98	0.76	0.69	0.50	0.83	0.69	0.81	0.49	0.90	0.49
1		0.97	0.94	0.76	0.94	0.54	0.90	0.85	0.87	0.82	0.87	1.05	0.92	0.76	0.72	1.11	0.85	0.70	0.75	1.15	1.09	0.65	0.85	1.01	0.85	0.85	0.41	1.10	0.44
4	CH_4	0.18	0.05	0.08	0.07	0.10	0.01	0.01	0.01	0.23	0.06	0.07	0.03	0.22	0.13	0.07	0.04	0.15	0.06	0.04	0.02	0.12	0.03	0.01	0.01	0.06	0.02	0.01	0.01
3		0.44	0.17	0.22	0.17	0.24	0.10	0.11	0.08	0.37	0.21	0.23	0.16	0.37	0.28	0.26	0.26	0.36	0.22	0.18	0.15	0.28	0.21	0.17	0.16	0.21	0.08	0.02	0.02
2		0.45	0.26	0.30	0.24	0.67	0.47	0.50	0.43	0.80	0.52	0.55	0.48	0.83	0.57	0.57	0.55	0.91	0.53	0.48	0.43	0.78	0.41	0.34	0.30	0.60	0.28	0.20	0.20
1		0.81	0.37	0.39	0.41	0.90	0.62	0.67	0.62	0.86	0.67	0.67	0.61	0.97	0.78	0.64	0.66	0.99	0.72	0.61	0.51	0.90	0.60	0.54	0.50	0.72	0.47	0.32	0.38
4	Alkanes	0.17	0.05	0.09	0.06	0.10	0.01	0.01	0.01	0.24	0.06	0.09	0.03	0.22	0.12	0.10	0.03	0.15	0.06	0.07	0.02	0.12	0.03	0.01	0.01	0.06	0.02	0.00	0.00
3		0.45	0.17	0.22	0.16	0.24	0.10	0.10	0.08	0.38	0.22	0.23	0.16	0.35	0.27	0.25	0.24	0.34	0.21	0.16	0.14	0.26	0.19	0.15	0.15	0.19	0.08	0.03	0.01
2		0.47	0.24	0.32	0.23	0.67	0.46	0.47	0.43	0.81	0.52	0.54	0.49	0.81	0.55	0.53	0.54	0.90	0.50	0.44	0.40	0.78	0.39	0.30	0.28	0.57	0.26	0.18	0.18
1		0.82	0.36	0.41	0.39	0.90	0.64	0.66	0.61	0.87	0.67	0.65	0.60	0.97	0.75	0.60	0.63	0.99	0.70	0.56	0.49	0.89	0.57	0.49	0.46	0.71	0.45	0.29	0.34
4	Alkenes	0.16	0.00	0.00	0.00	0.04	0.00	0.00	0.00	0.21	0.00	0.00	0.00	0.19	0.04	0.00	0.00	0.07	0.00	0.00	0.00	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3		0.25	0.10	0.04	0.05	0.22	0.04	0.00	0.00	0.32	0.07	0.00	0.04	0.35	0.15	0.00	0.00	0.32	0.04	0.00	0.00	0.23	0.06	0.00	0.00	0.07	0.00	0.00	0.00
2		0.50	0.17	0.07	0.18	0.62	0.44	0.24	0.28	0.69	0.41	0.08	0.15	1.32	0.34	0.11	0.14	0.74	0.29	0.04	0.04	0.74	0.18	0.02	0.03	0.55	0.05	0.00	0.00
1		0.91	0.32	0.26	0.32	0.80	0.64	0.38	0.42	0.82	0.59	0.26	0.35	1.05	0.39	0.13	0.17	0.99	0.44	0.14	0.16	0.91	0.41	0.07	0.11	0.62	0.24	0.04	0.06
4	Aromatics	0.15	0.04	0.03	0.03	0.09	0.00	0.00	0.00	0.18	0.05	0.02	0.02	0.15	0.10	0.03	0.02	0.13	0.05	0.02	0.02	0.10	0.02	0.00	0.00	0.05	0.01	0.00	0.00
3		0.40	0.17	0.10	0.09	0.22	0.09	0.04	0.05	0.30	0.16	0.10	0.11	0.25	0.17	0.09	0.10	0.27	0.16	0.09	0.10	0.21	0.15	0.09	0.10	0.15	0.06	0.01	0.01
2		0.43	0.26	0.16	0.15	0.69	0.37	0.23	0.26	0.60	0.40	0.25	0.30	0.66	0.37	0.22	0.29	0.73	0.42	0.24	0.29	0.65	0.29	0.14	0.16	0.49	0.22	0.09	0.11
1		0.83	0.28	0.33	0.25	0.91	0.56	0.38	0.42	0.67	0.58	0.43	0.47	0.68	0.43	0.25	0.31	0.73	0.80	0.32	0.43	0.67	0.41	0.26	0.32	0.54	0.34	0.17	0.25
4	Cyclics	0.19	0.06	0.03	0.03	0.14	0.00	0.00	0.00	0.27	0.05	0.01	0.01	0.22	0.10	0.00	0.00	0.11	0.03	0.00	0.00	0.10	0.01	0.00	0.00	0.04	0.01	0.00	0.00
3		0.50	0.15	0.10	0.09	0.28	0.12	0.04	0.04	0.42	0.24	0.08	0.08	0.36	0.24	0.06	0.07	0.28	0.14	0.03	0.04	0.19	0.13	0.05	0.05	0.17	0.04	0.00	0.00
2		0.48	0.20	0.17	0.17	0.86	0.48	0.28	0.28	0.89	0.46	0.27	0.28	0.69	0.44	0.18	0.21	0.66	0.31	0.11	0.14	0.59	0.25	0.12	0.12	0.53	0.16	0.07	0.08
1		0.89	0.33	0.29	0.26	1.08	0.63	0.40	0.42	0.97	0.65	0.31	0.35	0.87	0.53	0.24	0.35	0.75	0.41	0.16	0.21	0.74	0.35	0.16	0.17	0.67	0.29	0.12	0.15
4	Halogenated compounds	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.22	0.00	0.23	0.00	0.00	0.18	0.00	0.00	0.25	0.00	0.00	0.00	0.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3		0.37	0.00	0.00	0.00	0.31	0.00	0.00	0.00	0.55	0.33	0.47	0.40	0.47	0.50	0.44	0.00	0.56	0.20	0.25	0.00	0.51	0.38	0.37	0.29	0.44	0.00	0.00	0.00
2		0.39	0.00	0.00	0.00	0.67	0.46	0.46	0.44	0.88	0.55	0.58	0.56	1.24	0.65	0.59	0.27	1.20	0.79	0.64	0.44	1.03	0.55	0.47	0.40	0.51	0.40	0.00	0.00
1		0.74	0.19	0.00	0.21	0.92	0.59	0.63	0.63	0.94	0.62	0.77	0.63	1.14	0.75	0.74	0.72	1.03	1.23	0.60	0.54	0.83	0.69	0.54	0.56	0.58	0.59	0.29	0.41
4	S-substituted compounds	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.57	0.71	0.00	0.00	0.60	0.56	0.00	0.00	0.72	0.00	0.00	0.00	0.64	0.00	0.00	0.00	0.76	0.00	0.00	0.00
2		0.00	0.00	0.00	0.00	0.70	0.88	0.00	0.00	1.22	1.07	0.00	0.00	1.05	0.58	0.00	0.00	1.30	0.73	0.00	0.00	1.19	0.00	0.00	0.00	1.01	0.78	0.00	0.00
1		0.00	0.00	0.00	0.00	1.47	1.44	0.97	1.28	1.01	1.59	1.71	1.97	1.16	1.30	1.68	2.39	1.27	1.65	1.75	2.21	1.12	1.87	2.10	2.37	1.07	2.36	2.23	2.79
4	Terpenes	0.12	0.01	0.00	0.01	0.06	0.01	0.01	0.00	0.13	0.03	0.01	0.02	0.11	0.08	0.02	0.02	0.15	0.03	0.01	0.01	0.08	0.03	0.01	0.01	0.03	0.01	0.00	0.00
3		0.19	0.02	0.02	0.03	0.15	0.05	0.02	0.03	0.21	0.06	0.03	0.04	0.16	0.11	0.03	0.04	0.20	0.07	0.03	0.03	0.13	0.06	0.02	0.02	0.10	0.04	0.01	0.01
2		0.25	0.02	0.02	0.02	0.37	0.13	0.11	0.13	0.36	0.12	0.06	0.06	0.56	0.17	0.07	0.07	0.58	0.15	0.05	0.07	0.37	0.13	0.05	0.06	0.21	0.13	0.03	0.03
1		0.32	0.08	0.09	0.09	0.51	0.21	0.15	0.15	0.45	0.18	0.08	0.09	0.58	0.21	0.08	0.11	0.59	0.23	0.09	0.11	0.46	0.18	0.07	0.09	0.35	0.21	0.06	0.07
4	O-substituted compounds	11.07	79.59	57.74	66.80	19.41	36.81	60.39	72.01	24.68	33.54	69.41	67.18	21.96	52.70	55.41	69.33	24.45	40.97	62.73	74.65	16.75	39.83	54.04	61.14	21.30	47.08	52.26	54.88
3		8.20	53.51	20.65	49.49	10.66	26.26	38.83	38.15	14.98	25.28	40.48	39.81	14.58	27.42	35.22	38.80	12.55	30.45	39.26	46.63	9.39	23.58	32.12	39.50	13.74	30.59	35.08	37.38
2		7.11	33.32	9.39	36.61	5.14	15.64	19.36	21.90	6.60	13.62	24.60	22.65	6.55	14.62	17.11	21.07	6.43	15.96	17.65	23.40	4.68	16.12	20.44	23.69	7.25	20.56	19.66	24.14
1		4.00	5.36	6.48	5.15	2.41	8.24	10.25	10.56	3.74	7.45	10.38	10.57	2.99	8.43	9.04	12.14	3.56	6.39	9.03	10.85	2.75	5.99	5.17	11.13	4.06	9.43	10.99	14.33

Table 4

Resulting I_{ba} values for main AD-related species and VOC functional groups within treated soil columns (SSLL, SS1, SS2) compared to untreated one (B). Numerical rows refer sequentially to gas samples at points from lower (1) to upper (4) along each column (see Fig. S1). Shades of red and blue reflect the following ranges of values: bright red values under 0.33, medium red values between 0.33 and 0.66, faded red values between 0.66 and 1, white values equal to 1, faded blue values between 1 and 2.5, medium blue values between 2.5 and 5, bright blue values over 5. Empty spaces indicate that species/functional groups were not detected within treated columns.

Sampling Points	Index I_{ba}	Day 0			Day 5			Day 12			Day 19			Day 26			Day 33			Day 40		
		SSLL	SS1	SS2	SSLL	SS1	SS2	SSLL	SS1	SS2	SSLL	SS1	SS2	SSLL	SS1	SS2	SSLL	SS1	SS2	SSLL	SS1	SS2
4	H_2				0.94	0.95		3.00	2.01		0.43	0.45		2.13	0.21		0.20	0.24				
3					0.82	0.28	0.29	2.71	1.08	1.12	0.67	0.13	0.15	0.90	0.16		0.22	0.08		3.83		
2					1.06	0.93	0.87	0.75	0.62	0.53	1.69	0.65	0.81	1.42	0.60	0.50	1.08	0.54	0.25	1.48	0.66	0.22
1		2.17	3.11	1.57	0.78	0.72	0.67	0.75	0.62	0.52	1.31	0.49	0.84	0.99	0.62	0.52	0.76	0.69	0.38	1.10	0.54	0.37
4	CO_2	0.99	0.65	0.73	0.81	0.71	0.29	1.11	0.64	0.15	0.63	0.39	0.19	1.05	0.74	0.17	1.49	0.88	0.14	0.96	0.48	0.10
3		0.85	0.72	0.84	0.90	0.84	0.57	0.74	1.07	0.61	0.77	0.78	0.63	0.73	0.91	0.72	0.78	0.93	0.48	0.80	0.88	0.49
2		1.01	1.02	0.97	0.95	1.02	0.99	1.10	1.24	1.02	0.89	1.11	0.98	0.90	1.39	1.09	0.72	1.21	1.00	0.60	1.10	0.60
1		0.97	0.78	0.98	1.66	1.58	1.61	1.06	1.28	1.12	0.94	1.46	1.11	1.07	1.65	1.57	1.31	1.56	1.31	0.49	1.30	0.52
4	CH_4	0.29	0.46	0.36	0.05	0.14	0.05	0.26	0.30	0.12	0.58	0.30	0.16	0.42	0.29	0.14	0.23	0.11	0.06	0.27	0.09	0.10
3		0.40	0.51	0.37	0.40	0.45	0.33	0.57	0.63	0.43	0.75	0.71	0.70	0.61	0.52	0.43	0.75	0.61	0.58	0.40	0.10	0.08
2		0.58	0.67	0.52	0.70	0.74	0.64	0.65	0.68	0.61	0.69	0.69	0.66	0.59	0.52	0.48	0.53	0.44	0.38	0.47	0.33	0.34
1		0.46	0.48	0.50	0.69	0.74	0.69	0.77	0.78	0.71	0.80	0.66	0.68	0.73	0.61	0.51	0.67	0.60	0.55	0.66	0.44	0.53
4	Alkanes	0.30	0.52	0.36	0.06	0.12	0.05	0.25	0.39	0.12	0.57	0.45	0.14	0.40	0.48	0.13	0.22	0.10	0.05	0.25	0.03	0.01
3		0.37	0.49	0.36	0.41	0.43	0.33	0.58	0.59	0.41	0.75	0.71	0.68	0.61	0.47	0.41	0.74	0.58	0.56	0.40	0.14	0.07
2		0.52	0.67	0.48	0.68	0.71	0.64	0.64	0.67	0.61	0.68	0.66	0.67	0.55	0.49	0.44	0.50	0.38	0.36	0.46	0.31	0.31
1		0.44	0.50	0.48	0.71	0.73	0.68	0.77	0.75	0.69	0.77	0.62	0.65	0.71	0.57	0.49	0.65	0.55	0.52	0.64	0.40	0.48
4	Alkenes										0.23											
3		0.38	0.16	0.18	0.19			0.23		0.13	0.43			0.13			0.26					
2		0.33	0.15	0.35	0.71	0.39	0.45	0.59	0.12	0.22	0.25	0.08	0.10	0.38	0.06	0.06	0.24	0.03	0.04	0.10		
1		0.36	0.29	0.36	0.79	0.47	0.52	0.72	0.32	0.43	0.38	0.12	0.16	0.44	0.14	0.16	0.45	0.08	0.12	0.38	0.06	0.09
4	Aromatics	0.28	0.20	0.17	0.03	0.03	0.02	0.26	0.11	0.08	0.67	0.19	0.16	0.37	0.16	0.13	0.22	0.03	0.03	0.28	0.01	0.01
3		0.43	0.26	0.24	0.41	0.19	0.21	0.53	0.33	0.37	0.70	0.38	0.41	0.61	0.33	0.36	0.73	0.43	0.48	0.44	0.06	0.07
2		0.60	0.38	0.36	0.54	0.34	0.38	0.67	0.42	0.50	0.56	0.34	0.44	0.58	0.33	0.40	0.45	0.21	0.24	0.45	0.18	0.22
1		0.33	0.41	0.31	0.62	0.42	0.46	0.87	0.64	0.71	0.63	0.37	0.46	1.10	0.45	0.59	0.60	0.39	0.48	0.63	0.32	0.46
4	Cyclics	0.29	0.18	0.17	0.03	0.01	0.02	0.18	0.04	0.05	0.48	0.02	0.02	0.32	0.02	0.02	0.14	0.00	0.00	0.18	0.00	0.00
3		0.30	0.20	0.19	0.43	0.14	0.13	0.57	0.20	0.19	0.66	0.16	0.20	0.50	0.10	0.13	0.69	0.26	0.27	0.26	0.01	0.01
2		0.41	0.35	0.36	0.56	0.33	0.33	0.52	0.31	0.31	0.64	0.27	0.31	0.48	0.16	0.21	0.41	0.21	0.20	0.30	0.12	0.14
1		0.37	0.33	0.29	0.59	0.37	0.39	0.67	0.32	0.36	0.61	0.28	0.40	0.55	0.21	0.29	0.47	0.22	0.22	0.43	0.18	0.23
4	Halogenated compounds								1.04													
3								0.59	0.85	0.71	1.06	0.94	0.00	0.36	0.45	0.00	0.74	0.73	0.56			
2					0.69	0.69	0.65	0.63	0.65	0.64	0.53	0.48	0.22	0.65	0.53	0.36	0.54	0.46	0.39	0.80		
1		0.25		0.29	0.64	0.68	0.68	0.66	0.82	0.67	0.66	0.65	0.63	1.20	0.59	0.52	0.83	0.65	0.67	1.02	0.50	0.71
4	S-substituted compounds																					
3								1.24			0.94											
2					1.25			0.88			0.55			0.56						0.77		
1					0.98	0.66	0.87	1.57	1.69	1.95	1.12	1.45	2.06	1.29	1.38	1.73	1.68	1.88	2.13	2.20	2.08	2.60
4	Terpenes	0.05	0.03	0.06	0.12	0.10	0.09	0.24	0.09	0.11	0.79	0.14	0.22	0.21	0.07	0.09	0.36	0.07	0.09	0.35	0.08	0.08
3		0.11	0.12	0.14	0.32	0.16	0.19	0.29	0.16	0.19	0.66	0.17	0.21	0.33	0.13	0.17	0.42	0.13	0.17	0.40	0.07	0.05
2		0.09	0.08	0.09	0.48	0.29	0.36	0.34	0.17	0.18	0.31	0.12	0.13	0.26	0.09	0.12	0.36	0.14	0.15	0.60	0.13	0.16
1		0.25	0.28	0.28	0.41	0.29	0.30	0.39	0.18	0.21	0.37	0.13	0.18	0.38	0.15	0.18	0.39	0.16	0.21	0.61	0.16	0.19
4	O-substituted compounds	7.19	5.22	6.03	1.90	3.11	3.71	1.36	2.81	2.72	2.40	2.52	3.16	1.68	2.57	3.05	2.38	3.23	3.65	2.21	2.45	2.58
3		6.53	2.52	6.04	2.46	3.64	3.58	1.69	2.70	2.66	1.88	2.42	2.66	2.43	3.13	3.72	2.51	3.42	4.21	2.23	2.55	2.72
2		4.69	1.32	5.15	3.05	3.77	4.26	2.06	3.73	3.43	2.23	2.61	3.22	2.48	2.75	3.64	3.45	4.37	5.06	2.84	2.71	3.33
1		1.34	1.62	1.29	3.42	4.25	4.38	1.99	2.77	2.82	2.82	3.02	4.05	1.79	2.54	3.05	2.18	1.88	4.04	2.32	2.70	3.52

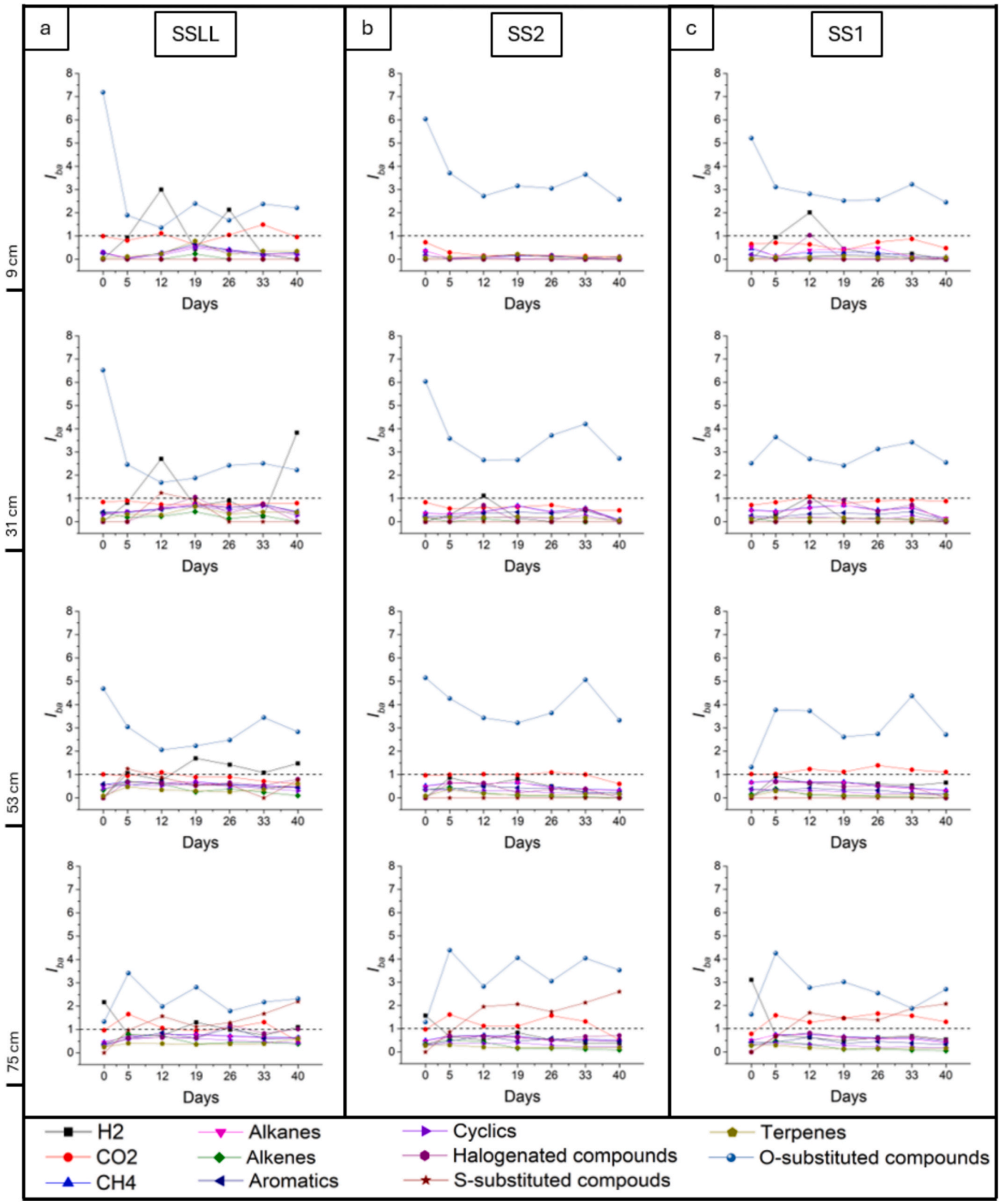

Fig. 4. Temporal and vertical evolutions of resulting I_{ba} values for main AD-related species and VOC functional groups within SSLL (a), SS2 (b), and SS1 (c) columns. Dashed lines (y = 1) indicate the reference efficiency of untreated soil (B column), species – functional group above the line were characterized by higher bio-production or lower biodegradation relative to untreated soil, whereas below the line were characterized by higher biodegradation relative to untreated soil. On left, depths (from top) of each sampling point (see Fig. S1).

and terpenes was largely enhanced by the addition of SS, as demonstrated by lower I_{ba} values among the SS amended columns relative to the SSLL one (Table 4). Halogenated compounds showed I_{ba} values < 1 within the SS amended columns (except for one value slightly higher than 1 for SS1), and particularly the SS2 column showed a stronger biodegradation upwards (Table 4). From day 19 onwards, the I_{ba} values of halogenated compounds within SSLL column were generally the highest, sporadically even > 1 (Table 4, Fig. 4a). This lower efficiency in SSLL column was presumably related to the relatively high amount of chloride expected in LL (Kjeldsen et al., 2002; Williams, 2005), which promoted halogenation reactions (van Pée and Unversucht, 2003) masking the slight degradation of these recalcitrant VOCs. S-substituted compounds, represented largely by DMSO produced at the bottom of the treated columns (with corresponding I_{ba} values predominantly > 1), were no longer detected upwards in SS1 and SS2 columns (with corresponding empty spaces in Table 4). Differently, DMSO was still detected in gases from intermediate depths of the SSLL column, suggesting that this combined soil treatment was less efficient than the other ones for DMSO biodegradation (Table 4, Fig. 4). O-substituted compounds had a larger production within the columns hosting the treated soils compared to the untreated one, as shown by the resulting I_{ba} values > 1 (Table 4, Fig. 4), highlighting the efficiency in aerobic biodegradation processes within the tested treated soils. The I_{ba} values relative to O-substituted bioproduction resulting generally lower within the SSLL column (from day 5 onwards: Table 4, Fig. 4a), associated with a lower biodegradation of aliphatic compounds, were likely due to the LL addition. Indeed, the biodegradation of dissolved organic matter expected to be apported by LL likely outcompeted for O_2. The SS2 column (Fig. 4b) generally showed slightly higher I_{ba} values than those of SS1 (Fig. 4c) related to bioproduction of O-substituted compounds, suggesting that under aerobic conditions a lesser amount of SS used in soil treatment favored the biodegradation of aliphatic compounds, corresponding to an increase in the bioproduction of O-substituted compounds.

The results of the present study highlighted how the investigated novel biocovers, particularly involving the addition of anaerobically stabilized and dewatered sewage sludge, efficiently minimized the emissions of odorous, environmental impacting, and toxic gas species expected in the LFG into the atmosphere. Noteworthy, despite being produced as end-product of CH_4 oxidation, CO_2 was influenced by possible biodegradation processes within the investigated novel biocovers, resulting in an abatement of CO_2 emissions derived from degrading waste. The relatively low amounts of SS and LL used in the investigated soil treatments encourages a potential implementation at full scale in real landfill sites, minimizing the transportation, management, and treatment costs. While the adoption of SS, typically derived from a wastewater treatment plant, could be configured as a beneficial reuse option in the perspective of sustainable circularity in integrated wastewater and waste management, the contextual application of LL, with an impacting chemical composition, in soil treatment requires pertaining mitigation strategies: to avoid a detrimental dispersion of LL in the surrounding areas, the landfill site should be properly equipped with a LL drainage and recovery system to collect residual liquids from the landfill biocover, with a possible, expected increase in the overall management costs.

5. Conclusions

The performed experimental run, corroborated by data elaborations finalized with the original bacterial activity index I_{ba} (in fuctional combination with the initial index I_d), indicated the potential of the tested novel biocovers, specifically treated with LL spraying and/or anaerobically stabilized and dewatered SS addition, in the abatement of main species and VOC functional groups of passing biogases that reliably simulated diffuse LFG emissions. Soil columns treated with anaerobically stabilized and dewatered SS only displayed the highest biodegradation activity on CH_4, H_2, and VOC functional groups (alkanes, alkenes, aromatics, cyclics, halogenated compounds, S-substituted compounds, and terpenes) compared to soil column additionally treated with LL spraying. More in detail, the soil amended with a lower quantity of SS (specifically, in a soil:SS mass ratio of 20:1) appeared to be slightly more efficient relative to that one characterized by a soil:SS mass ratio of 10:1.

The original bacterial index I_{ba} proposed in this study, elucidated, at proper experimental scales, the benefits and disadvantages in the mitigation efficiency of biogas species within a specific biocover relative to a conventional landfill cover, proving to be a useful tool in the development and testing of novel and existing landfill biocovers. Furthermore, since the biogas fugitive emissions from landfill site are influenced by several factors (i.e., landfill geometry, waste type and age, efficiency of LFG collector and recovery systems), the index could be applied to LFG fugitive emission monitoring from landfill sites, to discern between the variations in concentrations of LFG species apported by the application of the biocover from a variety of ambient and engineering factors, to underline criticalities in specific LFG species mitigation and find a solution in a short period of time.

Considering the high variability of LL chemical composition, influenced by waste type and age and landfill operation, further investigations are to be carried out for testing different types of LL derived from several landfill sites, to understand the advantages and disadvantages of LFG abatement considering several LL chemical compositions. Future investigations could be coupled with periodical moisture and microbiological measurements, to highlight the moisture retention capabilities of the soil and sludge-soil mixtures, and the bacterial proliferation rates expected from nutrient availability. Moreover, to discern the moisture contribution from the nutrient supply in promoting bacterial proliferation, and consequently the biogas biodegradation capacities, a new prototype, implemented with the soil column treated with water spraying only, may be used.

Finally, complementary experimental works, with the prototypes implemented in this study, could be also focused on varying other operating column conditions (e.g., soil type, thickness, and compaction), digesting different organic waste categories (i.e., those commonly disposed of at landfills) as degrading substrates, and treating soil with additional amendment as the aerobically stabilized and dewatered SS.

CRediT authorship contribution statement

Gregorio Viti: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Stefania Venturi:** Validation, Supervision, Conceptualization. **Antonio Randazzo:** Supervision. **Fabio Tatàno:** Supervision. **Franco Tassi:** Validation, Supervision, Project administration, Formal analysis, 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2026.115686.

Data availability

All data are contained in the Supplementary Material

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