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Original

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1 **Impact of biochar on anaerobic digestion: meta-analysis and economic evaluation**

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11

12 **Abstract**

13 The growing global energy demand encourages the request for renewable sources, as biomethane from the
14 anaerobic digestion (AD) of waste biomass. Biochar (BC) can effectively increase methane production when
15 supplemented to AD, depending on BC physico-chemical properties. This study was developed in two phases.
16 Firstly, a systematic meta-analysis of current literature was performed to correlate AD performance with BC
17 properties, aiming to define their optimal range. The obtained results prove that BC enhances and accelerates
18 biomethane production. Considering 408 experimental conditions of 76 studies in batch mode, biomethane
19 yield and maximum production rate were significantly increased by BC addition. From the results of the
20 subgroups meta-analysis, an optimal range of BC physico-chemical properties may be suggested as follows:
21 high ash ($\geq 20\%$) and low C contents ($< 50\%$), high O/C molar ratios (≥ 0.3), high contents of O ($\geq 20\%$) and
22 N ($\geq 0.6\%$), acidic pH (< 7.0), low surface area ($< 10 \text{ m}^2 \text{ g}^{-1}$). Secondly, an economic analysis aimed at
23 assessing the economic profitability of BC addition to an existing AD plant suggest avoiding a dose above
24 $0.45\text{-}0.76 \text{ g}_{\text{BC}} \text{ g}_{\text{VS}}^{-1}$, independently of the specific AD operating conditions. In conclusion, BC application in
25 full-scale digesters is able to maximize biomethane production and economically feasible.

26

27

28 **Highlights**

- 29 – Biochar addition enhances and accelerates biomethane production from waste biomass
- 30 – An inventory of literature on biochar addition to anaerobic digestion is provided
- 31 – Meta-analysis defined the optimal range of biochar properties for this application
- 32 – An economic analysis suggested to use biochar doses below 0.45-0.76 g_{BC} g_{VS}⁻¹

33

34 **Keywords**

35 anaerobic digestion; biochar; economic analysis; methane; meta-analysis.

36

37 **Abbreviations**

38 AD: anaerobic digestion; ANOVA: analysis of variance; BC: biochar; BET: Brunauer–Emmett–Teller; C: total
39 carbon; CEC: cationic exchange capacity; CI: confidence intervals; CHP: combined heat and power; COD:
40 chemical oxygen demand; DF: degrees of freedom; EAC: electron accepting capacity; EC: electrical
41 conductivity; EDC: electron donating capacity; FC: fixed carbon; GHG: green-house gas; H: herbaceous;
42 HRT: hydraulic retention time; HSD: honestly significant difference; λ : lag-phase; LR: lignocellulosic
43 residue/crops; M: Manure or animal-based residue; NLR: Non-lignocellulosic residue; OFMSW: organic
44 fraction of municipal solid waste; OLR: organic loading rate; P: biomethane potential; PI: prediction interval;
45 R_{max}: methane production rate; S: sludge; SA: specific surface area; SD: standard deviation; SE: standard error;
46 SMD: standardized mean difference; SRT: sludge retention time; SS: suspended solids; TKN: total Kjeldahl
47 nitrogen; TP: total phosphorous; TS: total solids; UM: unit of measure; VFA: volatile fatty acids; W: wood;
48 VM: volatile matter; VS: volatile solids.

49

50 **1. Introduction**

51 Biochar (BC) is the solid carbonaceous material derived from the thermo-chemical treatment of biomass in
52 absence or with limited air [1]. BC can be produced using a wide array of feedstocks, including wood,
53 herbaceous crops, agro-industrial residues, animal manure, and biosolids [2,3], through different technologies

54 (pyrolysis, hydro-thermal carbonization, torrefaction, gasification, or partial combustion) [4,5]. By controlling
55 biomass feedstock, thermo-chemical operating conditions, and further activation, BC can be produced with a
56 wide-range of physico-chemical features [5,6] and tailor-made for specific applications. Initially, a strong
57 interest about BC was focused on its agronomical application, due to its potential benefits on soil quality
58 coupled with the significant effects on the reduction of greenhouse gases (GHG) emissions through carbon
59 sequestration. Recently, other BC applications have been proposed [7–9], as adsorbent for contaminants in
60 soil, water, and gaseous streams, precursor for BC-based materials or for energy storage applications, and
61 additive for anaerobic digestion (AD) or composting.

62 In recent years, the production of renewable energy, including bioenergy from waste biomass, has increased
63 considerably to reduce GHG emissions from fossil fuels. AD could reduce the global GHG emissions by 3.29
64 to 4.36 Gt CO₂ eq. [10], representing the 7-9% of the world's current GHG emissions (equal to about 50 Gt
65 CO₂ eq) [11]. AD is a biological process used to convert organic wastes and wastewater in absence of oxygen
66 to biogas and digestate. Biogas, mainly formed by CH₄ and CO₂, can be burnt in cogeneration units to produce
67 heat and electricity or upgraded to renewable natural gas. Digestate can be recycled into value-added products
68 as bio-fertilizer, solid biofuel, or carbon-based materials [12]. Despite AD is a mature technology some critical
69 issues persist; process instabilities due to volatile fatty acids (VFAs) accumulation may occur in case of high
70 organic loads of easily degradable biomass [13]. Other issues include low biogas yield due to a slow hydrolysis
71 of recalcitrant substrates [14,15], as well as the presence of AD inhibitors [16,17].

72 The application of BC as additive in AD process has shown the potential of enhancing and accelerating
73 methane production from different substrates [18]. Several review studies analysed the complex mechanisms
74 of BC intervention in AD [18–27]: mitigation of potential inhibitions and process instabilities through the
75 adsorption of inhibitors, increased AD buffering capacity, immobilization of microbial cells on BC,
76 acceleration of metabolic activities by BC, and transfer of electrons and/or other metabolites among the
77 microorganisms involved in hydrolysis, acidogenesis, acetogenesis, and methanogenesis. Further, BC can
78 remove CO₂ and impurities from biogas. BC doesn't need to be separated from the digestate due to the
79 enhanced agronomic quality in terms of carbon sequestration, nutrients and water retention in soils, reduction
80 of nutrients and contaminants run-off, and reduction of GHG emissions from soils [18,19,28]. However,
81 despite a good agreement in literature regarding the benefits of BC intervention, a proper understanding of the

82 role of the BC physico-chemical properties in these mechanisms and their correlation with AD performance
83 still needs to be reached. As a result, the addition of BCs with a wide variation of physico-chemical properties
84 had proven effects on AD, often positive, but, sometimes, also detrimental. Therefore, it is necessary to define
85 the optimal range of BC physico-chemical properties specifically targeting AD. Despite the wide range of
86 experimental data, few attempts have been performed to correlate BC properties with AD performances
87 [27,29,30]. In a previous study [29] we explored the correlation of each BC feature with AD performances
88 through Principal Component Analysis on experimental data achieved from our batch tests, suggesting some
89 key BC properties and underlying the need of more experimental data to draw additional conclusions. In this
90 direction, Khashaba et al. [30] identified strong correlations between the physico-chemical BC features and
91 AD performances through an artificial neural network based on literature data focusing just on sewage sludge
92 substrate.

93 Meta-analysis is a statistical approach allowing to analyse the results of multiple complex studies, and to
94 critically compare the results of different studies to identify patterns and relationships, leading to robust
95 conclusions. Meta-analysis has been carried out to investigate the effects of BC on GHG emissions from soil
96 [31–33], on plants growth and productivity [34,35], and on soil properties [36–38]. Recently, Xiao et al. (2021)
97 [27] conducted a meta-analysis on 27 publications and 156 datasets to assess the impact of BC properties (dose,
98 pH, size, specific surface area, feedstock, pyrolysis temperature) on AD performance, concluding that BC
99 enhances methane production, and suggesting that BC features able to control methane production were
100 feedstock type, pyrolysis temperature and BC dose, without defining their optimal range values. Compared to
101 Xiao et al. (2021) [27], submitted in March 2021, this work presents the results of a meta-analysis that has
102 elements of novelty as follows. In details, just 6 months later (in September 2021) over 50 additional studies
103 exploring BC influence on AD have been published. Therefore, compared to the ones considered in Xiao et al.
104 (2021) [27], more datasets can be accounted in a meta-analysis, considering a wider number of BC physico-
105 chemical features (conductivity, pH, specific surface area, particle size, dose), BC production characteristics
106 (feedstocks and pyrolysis temperature), and also AD operating conditions and substrates. To our knowledge,
107 a key knowledge gap of the available literature is that it only suggests what are BC desirable properties, without
108 defining the optimal range values of the single BC physico-chemical properties, their correlation with AD
109 performance, nor the AD substrates and operating conditions that are more likely improved by BC

110 supplementation. It is reasonable to expect different effects of BC on AD performance depending on the
111 specific combinations of AD temperature and feeding conditions (substrate, organic load); for instance, BC
112 was acknowledged as a stabilizing agent [21] in case of high organic loads of a highly degradable substrate as
113 the organic fraction of municipal solid waste (OFMSW), and also as an accelerating agent of metabolic
114 activities [24,26], in case of a relatively refractory substrate as sewage sludge. Another knowledge gap of
115 current literature on the topic is the uncertainty related to the full-scale application of BC to AD is the
116 comparison between BC input cost and output revenues deriving from the energy production, since few studies
117 have investigated the economic feasibility of BC supplementation in AD [39–41] Therefore, compared to
118 existing literature, the present work has two key elements of novelty, as follows. Firstly, it compiled a
119 systematic inventory (in the form of a database) of the literature selected according to specific strict
120 requirements, which was further used to conduct a meta-analysis on the effects of BC and its properties on AD
121 performance, considering not only BC features but also the ones of AD process. Secondly, it evaluated the
122 economic feasibility of BC addition to existing full-scale AD installations.

123 This study was focused on the application of BC as additive in AD, with a specific interest for the optimization
124 of BC physico-chemical properties and their correlation with AD performance. In details, this study had the
125 following aims: (1) assessment of the overall effect of BC on AD performance derived from batch and semi-
126 continuous studies; (2) analysis of the impact of BC physico-chemical properties, BC production conditions,
127 and AD operating conditions on the global effects of BC on AD products yields and quality; and (3) assessment
128 of the economic feasibility of BC addition on AD at full-scale, based on data from published semi-continuous
129 AD studies.

130

131 **2. Material and methods**

132 **2.1. Data collection and selection**

133 A systematic bibliographic search was conducted using Scopus and Google Scholar, and compared with the
134 list of references of a previous review paper [18]. The collection of references was completed in September
135 2021. The size of the selected datasets was significant (613 paired experimental conditions of BC amended
136 “treatment” and “control”), and the following phases of data extraction and database compilation applied to

137 such a huge amount of information required several months before starting data analysis. The compiled
138 systematic database was made available as Appendix A to other researchers with the specific aim of providing
139 a common dataset that can be updated and expanded with new studies for future analyses. The literature survey
140 was based on the following combination of keywords: (methane or CH₄ or biogas or anaerobic digestion) and
141 (biochar or bio-char). Around 450 references were collected and imported to Mendeley.

142 A first filtering phase (data extraction) was conducted checking abstract and title, and based on the following
143 criteria: (1) use of English language; (2) access provided to the full text; (3) only primary sources (no review
144 studies); (4) studies with “treatment” group (reactors with biochar supplementation) and “control” group
145 (reactors without biochar), where other experimental conditions were identical; (5) studies reporting methane
146 and/or biogas production; (6) no combined use of biochar and other additives. As a result, 112 studies with
147 613 paired measurements of “treatment” vs “control” met the inclusion criteria.

148 A second screening phase (data selection) was conducted to select the studies eligible for a meta-analysis,
149 according to the following additional criteria: (7) provided number of experimental replicates > 1; (8) provided
150 mean and uncertainty of methane production, as standard deviation (SD) or standard error (SE). Therefore, 76
151 studies with 408 experimental conditions in batch mode and 18 studies with 83 conditions in semi-continuous
152 mode were included in the meta-analysis performed in this work.

153

154 **2.2. Categorization of the selected literature data**

155 The full text and the supplementary materials of the studies passing the first screening phase were subjected
156 to data extraction, and an inventory of the selected literature (613 paired measurements of “treatment” vs
157 “control”) was compiled as a database in Excel (Appendix A). The data were grouped based on variables
158 affecting methane production, considering the most frequent to include enough results within each category.
159 These include BC production conditions (feedstock, pyrolysis temperature), physico-chemical properties of
160 BC (surface area, ash, pH, contents of C, H, N, and O, H/C and O/C molar ratios), substrates for AD, operating
161 conditions of AD (temperature, batch vs semi-continuous, dose of BC). Data included in the database were
162 obtained directly from tables and text, or extracted using Web Plot Digitizer (Pacifica, US) [42] if only
163 presented in figures. Primary data about methane production were extracted: in case of batch AD tests, CH₄

164 yield and kinetic parameters (CH_4 production rate R_{\max} and lag-phase λ) when the modified Gompertz model
165 was applied; in case of semi-continuous AD tests, CH_4 production rate and CH_4 content. For all parameters,
166 mean, SD, number of replicates (n), and unit of measures (UM) were extracted. When only SE was provided,
167 SD was estimated as $SD = \sqrt{n} SE$. Further relevant data and information were extracted: (1) article
168 information, (2) operating conditions of BC production, (3) physico-chemical properties of BC, (4) operating
169 conditions of AD tests, (5) physico-chemical features of substrate and inoculum for AD. UMs were converted
170 to standardized units for comparison; for instance, the dose of biochar provided in different UMs ($\text{g g}_{\text{TS}}^{-1}$, g
171 $\text{g}_{\text{VS}}^{-1}$, g L^{-1} , % bw) was standardized to g L^{-1} or derived from the available data. When it was not possible, it
172 was recorded in the database, but excluded from the statistical analysis. Further, when the oxygen content of
173 the BC was not available, it was determined by difference as $O = 100 - \text{Ash} (\%) - C (\%) - H (\%) - N (\%) - S$
174 $(\%)$, according to standard method ASTM D3176. H/C and O/C molar ratios were derived from H, C, and O
175 element contents (%), and their corresponding atomic weights (H: 1.008; C: 12.011; O: 15.999). Table B1 in
176 Appendix B summarises the data presented in the inventory (Appendix A): data category, data type, unit of
177 measure, notes, and assumptions for data extraction. Table B2 in Appendix B contains the selected research
178 studies included in the inventory, published from 2012 to September 3rd, 2021.

179 **2.3. General overview of the selected literature data**

180 The systematic bibliographic research firstly selected around 450 studies, reduced to 112 after the first
181 screening. 613 paired experimental conditions of BC amended “treatment” and “control” were included in the
182 inventory from the 112 selected studies. The detailed inventory (Appendix A) represents a systematic overview
183 of current state of the research, while summarized data are in Tables B1 and B2 of Appendix B. These include
184 article information, conditions of BC synthesis, BC physico-chemical properties, operating conditions of AD
185 tests, and physico-chemical features of substrate and inoculum for AD. More data than the information used
186 for the meta-analysis was collected in the inventory (Appendix A) to provide a common basis for further
187 research exploring BC role in AD.

188 The 613 paired case-studies were grouped based on parameters possibly affecting methane production (Table
189 1). Most AD tests were conducted in batch (82%) than in semi-continuous mode (17%). A clear imbalance
190 was observed between tests in AD mesophilic (77%) and thermophilic (22%) conditions. Regarding the

191 substrates for AD, in case of mono-digestion (81%) the most frequent substrates were sewage sludge (20%),
 192 food waste or OFMSW (18%), and manure (10%), along with agro-industrial wastes, wastewater and simple
 193 substrates. In case of co-digestion (19%), food waste (with manure or sewage sludge) was the most abundant
 194 substrate (11%). BC was obtained from several feedstocks, as wood (49%), herbaceous materials (19%),
 195 sludge (13%), crops and lignocellulosic residues (9%), manure (6%), and non-lignocellulosic residues (5%).
 196 Different temperatures were employed during the thermo-chemical production of BCs, from less than 400 °C
 197 (11%), to typical pyrolysis temperatures ranging between 400 and 700 °C in most cases (64%), up to more
 198 than 700 °C in the gasification range (25%). Because of the varying feedstocks and temperature of pyrolysis,
 199 a large spectrum of physico-chemical properties of BCs was observed, as discussed in section 3.1. Despite a
 200 wide range of BC doses reported (0.2-100 g L⁻¹), in most cases it was below 20 g L⁻¹ (75%), and 10 g L⁻¹ was
 201 the most adopted.

202

203 Table 1. Classification of case studies in the inventory: variables and corresponding levels, relative and
 204 absolute (between brackets) frequency of studies in each level.

Variable [unit of measure]	Levels/subgroups	Definition	Relative frequency (number of occurrence)
Feedstocks for biochar production (classification adapted from [31,37])	Wood (W)	Oak, pine, willow, unidentified wood mixtures, spruce, pine trimmings, coppiced woodlands, orchard pruning, pristine wood, bamboo, sawdust, vineyard pruning, holm oak, Ash juniper, white oak, shrub, cotton wood, douglas fir	48.5% (289)
	Herbaceous (H)	Green waste, straws, and corn stover, maize stover, wheat straws, miscanthus straw, rice straw, water hyacinth, fucus serratus, corn straw, switchgrass, reed straw	18.5% (110)
	Manure or animal-based residue (M)	Manure or manure-based materials (bone, meat, blood, etc.) sheep manure, dairy manure, cattle bone, poultry waste, cow dung, cow manure, chicken manure	6.2% (37)
	Sludge (S)	Any sludge, usually obtained from wastewater treatment but also including brewery sludge biosolids	12.8% (76)
	Crops/Lignocellulosic residue (LR)	Walnut shells, peanut shells, maize cobs, furfural from corn cobs, rice husk, nuts shells, paper mill waste, coconut endocarp or shell, coffee grounds, almond shells	9.2% (55)

	Non-lignocellulosic residue (NLR)	Fruit peels, seeds, beet-root chips, spent brewer's grains or draff, wheat bran, citrus peel, oil seed rape, canola meal, corn stalk, whiskey draff	4.9% (29)	
Pyrolysis temperature [°C]	< 400	$t < 400\text{ °C}$	10.5% (58)	
	400 - 550	$400\text{ °C} \leq t < 550\text{ °C}$	32.9% (182)	
	550-700	$550\text{ °C} \leq t < 700\text{ °C}$	31.3% (173)	
	≥ 700	$t \geq 700\text{ °C}$	25.3% (140)	
Biochar dose [g L ⁻¹]	< 5	Dose < 5 g L ⁻¹	18.7% (108)	
	5-10	$5\text{ g L}^{-1} \leq \text{dose} < 10\text{ g L}^{-1}$	16.1% (93)	
	10-15	$10\text{ g L}^{-1} \leq \text{dose} < 15\text{ g L}^{-1}$	26.5% (153)	
	15-20	$15\text{ g L}^{-1} \leq \text{dose} < 20\text{ g L}^{-1}$	13.5% (78)	
	≥ 20	Dose $\geq 20\text{ g L}^{-1}$	25.3% (146)	
Feeding mode of anaerobic digestion	Batch		82.1% (503)	
	Fed-batch		1.0% (6)	
	Semi-continuous		17.0% (104)	
Substrate for anaerobic digestion	Agro-industrial	Draff, brewer's spent grain, citrus peel, cardboard, dried sorghum, orange peel, corn-straw, wheat straw, sugar beet pulp, water hyacinth, corn stalk, beer lees, whiskey draff, broadleaf cattails	15.4% (93)	
	Other co-digestion	Other co-digestion (no food waste with manure/sewage sludge): orange peel + sewage sludge, cassava wastewater + poultry litter, straw + cow manure, wheat husk + mixed sludge, swine manure + sewage sludge, corn stover + chicken manure, cow dung + sewage sludge pyrolysis liquid, algal biomass + food waste, corn straw + cow dung, wheat straw + cow dung, rice straw + cow dung	7.9% (48)	
	Co-digestion of food waste + manure/sludge	Co-digestion of food waste with manure or sewage sludge	10.8% (65)	
	Manure	Swine manure, poultry litter, chicken litter, dairy manure, chicken manure, swine waste, pig manure	9.8% (59)	
	OFMSW/food waste	Fruit waste, food waste, organic fraction of the municipal solid waste (OFMSW), rice, chicken, vegetables, kitchen waste, cooked rice/egg white/lard	17.9% (108)	
	Sewage sludge	Mixed sludge, sewage sludge, wastewater sludge, waste activated sludge, primary sludge	19.7% (119)	
	Simple substrate	Phenol solution, glucose solution, oil, mixed volatile fatty acids, ethanol, glucose, cellulose, acetate, propionate, butyrate, valerate, oily sludge + starch + naphthalene	10.4% (63)	
	Wastewater	Wastewater, synthetic wastewater, piggery wastewater, food waste fermentate, cassava wastewater, soured anaerobic digestate, Aqueous phase of bio-oil generated via Hydrothermal Liquefaction of Algae, synthetic blackwater, cread fermentate, synthetic dairy wastewater	8.1% (49)	
	Temperature for anaerobic digestion [°C]	Low temperature	$t < 25\text{ °C}$	0.8% (5)
		Mesophilic	$25\text{ °C} \leq t < 40\text{ °C}$	77.3% (469)
Thermophilic		$t \geq 40\text{ °C}$	21.9% (133)	

Ash [% wt]	< 5.0	Ash < 5%	15.5% (51)
	5-10	5% ≤ ash < 10%	29.0% (95)
	10-20	10% ≤ ash < 20%	20.4% (67)
	20-50	20% ≤ ash < 50%	25.6% (84)
	≥ 50	Ash ≥ 50%	9.5% (31)
Surface area [m ² g ⁻¹]	< 10	surface area < 10 m ² g ⁻¹	12.8% (56)
	10-20	10 m ² g ⁻¹ ≤ surface area < 20 m ² g ⁻¹	16.7% (73)
	20-100	20 m ² g ⁻¹ ≤ surface area < 100 m ² g ⁻¹	21.2% (93)
	≥ 100	surface area ≥ 100 m ² g ⁻¹	49.3% (216)
pH [-]	< 7	pH < 7	12.9% (48)
	7-8	7 ≤ pH < 8	11.8% (44)
	8-9	8 ≤ pH < 9	23.9% (89)
	9-10	9 ≤ pH < 10	34.3% (128)
	≥ 10	pH ≥ 10	17.2% (64)
Total C [% wt]	< 50	Total C < 50%	17.7% (76)
	50-70	50% ≤ total C < 70%	29.1% (125)
	70-80	70% ≤ total C < 80%	26.3% (113)
	≥ 80	Total C ≥ 80%	26.8% (215)
H [% wt]	< 1	H content < 1 %	23.5% (81)
	1-2	1% ≤ H content < 2%	21.2% (73)
	2-3	2% ≤ H content < 3%	20.3% (70)
	3-4	3% ≤ H content < 4%	13.9% (48)
	≥ 4	H content ≥ 4%	21.2% (73)
O [% wt]	< 5	O content < 5%	12.0% (42)
	5-10	5% ≤ O content < 10%	17.7% (62)
	10-15	10% ≤ O content < 15%	25.7% (90)
	15-20	15% ≤ O content < 20%	16.6% (58)
	≥ 20	O content ≥ 20%	28.0% (98)
N [% wt]	< 0.3	N content < 0.3%	15.1% (54)
	0.3-0.6	0.3% ≤ N content < 0.6%	37.5% (134)
	0.6-1.2	0.6% ≤ N content < 1.2%	16.5% (59)
	≥ 1.2	N content ≥ 1.2%	30.8% (110)
H/C molar ratio [-]	< 0.1	H/C molar ratio < 0.3%	16.7% (56)
	0.1-0.3	0.1% ≤ H/C molar ratio < 0.3%	22.4% (75)
	0.3-0.5	0.3% ≤ H/C molar ratio < 0.5%	24.5% (82)
	0.5-1.0	0.5% ≤ H/C molar ratio < 1.0%	22.4% (75)
	≥ 1.0	H/C molar ratio ≥ 1.0%	14.0% (47)
O/C molar ratio [-]	< 0.075	O/C molar ratio < 0.075%	26.0% (92)
	0.075-0.15	0.075% ≤ O/C molar ratio < 0.15%	29.1 (103)
	0.15-0.3	0.15% ≤ O/C molar ratio < 0.3%	23.4 (83)
	≥ 0.3	O/C molar ratio ≥ 0.3%	21.5 (76)

205

206 3. Theory and calculations

207 3.1. Analysis of variance (ANOVA)

208 The influence of feedstocks for BC production and pyrolysis temperature on the physico-chemical properties
 209 of BCs was assessed. All BCs included in the inventory were considered. Feedstocks and pyrolysis

210 temperatures were grouped according to the criteria described in Table 1. One-way analysis of variance
 211 (ANOVA) at $\alpha = 0.05$ was used to compare different feedstocks and temperatures. Then, significant differences
 212 between individual subgroups were identified through Tukey's honestly significant difference (HSD) post-hoc
 213 test ($\alpha = 0.05$). Boxplot was used to identify the distribution of each BC property. All statistical analyses were
 214 conducted using R language [43].

215

216 3.2. Meta-analysis

217 Meta-analysis was carried out using the Metafor package v. 3.0-2 [44] in R. Firstly, data were divided into two
 218 subsets (batch and semi-continuous AD tests). Then, three separate meta-analysis were carried out to
 219 investigate the effect of BC supplementation on CH₄ yield, CH₄ production rate (R_{\max}) and lag-phase (λ) from
 220 the modified Gompertz model fitting. In case of the semi-continuous subset, an additional meta-analysis was
 221 conducted to assess the effect of BC on CH₄ production rate. In each case, the paired results of the "treatment"
 222 and "control" groups were computed to obtain the effect size (ES), being the extent of the treatment effect (BC
 223 addition), or, in other words, the magnitude of the difference between the "treatment" and the "control" means
 224 [45]. The Hedges's standardized mean difference (g) [46] was adopted as ES index, suitable for small sample
 225 sizes (2 or 3 replicates in most studies) and for comparing means with different units of measure across studies
 226 (see Table B1, Appendix B). Thus, for each variable the Hedges's g was calculated according to eq. 1:

$$227 \quad g = J \frac{\bar{X}_1 - \bar{X}_2}{S_p} \quad (1)$$

228 where

$$229 \quad S_p = \sqrt{\frac{(n_1-1)S_1^2 + (n_2-1)S_2^2}{n_1+n_2-2}} \quad (2)$$

$$230 \quad J = 1 - \frac{3}{4df-1} \quad (3)$$

231 \bar{X}_i , n_i , and S_i are the variable mean, the standard deviation, and the number of samples of the treatment (1) and
 232 the control groups (2), respectively; S_p is the pooled standard deviation across groups (eq.2); J is a correction
 233 factor accounting for small sample sizes (eq.3); df is the degrees of freedom ($n_1 + n_2 - 2$ for two independent
 234 groups). The second term of eq.1 is the standardized mean difference (SMD). Thus, considering for example

235 the CH₄ yield, a positive value of g means that BC addition enhances CH₄ yield compared to the control group.

236 The variance V_g of g was determined according to eq. 4:

$$237 \quad V_g = J^2 \frac{n_1+n_2}{n_1 n_2} \frac{g^2}{2(n_1+n_2)} \quad (4)$$

238 Once determined the ESs and associated variances for the individual measures, the summary effects of BC
239 addition on CH₄ yield, R_{\max} , λ , and CH₄ production rate were determined. The summary effect was estimated
240 using a random-effects model instead of a fixed-effects model, which assumes a common or fixed effect for
241 all the tests, since the true effects likely varied across studies due to the different experimental conditions [47].

242 DerSimonian and Laird method was used to estimate the between-studies variance (T^2) [48]. The confidence
243 intervals (CIs) for the summary effect were determined by using the Knapp-Hartung adjustment [49].

244 Therefore, the summary ES was considered significantly different from zero whether the 95% CIs did not
245 overlap zero, i.e., supplementation of BC had a significant impact. Significance of heterogeneity in true effects
246 was tested by Q-value, degrees of freedom (df) and corresponding p-value. I^2 statistic (%) was also reported to
247 express the fraction of observed variance due to heterogeneity rather than random error [46].

248 Subgroup analyses were carried out to investigate how different variables could influence the effects of BC
249 supplementation on CH₄ yield, R_{\max} , and λ . Each moderator variable was classified in two or more subgroups.

250 Then, a mixed-effects model was used to estimate the effect of each variable, assuming that variation across
251 tests within each subgroup was due to random error, while variation between subgroups was fixed [47]. The
252 summary effect within each subgroup was computed using a random-effect model with a separate estimate of
253 T^2 , whereas the summary effect across subgroups was calculated with a fixed-effects model. An omnibus test
254 (Q_M) was used for testing the significance of each moderator variable: if a Q_M was significant, this meant that
255 on average the ES differed between subgroups (p-value < 0.05); consequently, each pair of effects of moderator
256 subgroups was considered significantly different if their 95% CIs did not overlap [47].

257 All results of meta-analysis were condensed on a forest plot. The vertical axis (g equal to zero) is known as
258 line of no effect [45]: a positive ES on the right constitutes an increase of CH₄ yield, R_{\max} , λ , or CH₄ production
259 rate due to BC supplementation, whereas a negative value on the left indicates a decrease. A square represented
260 the mean ES of a given subgroup together with its 95% CIs, being significantly different from zero when CIs
261 did not intercept the vertical axis. The summary effect was represented by a diamond, where its location

262 identified the mean, and its vertices depicted the lower and upper 95% CIs. Further, the 95% prediction
263 intervals (PIs) were shown by a dashed line in case of a random-effects model.

264

265 **3.3. Economic analysis**

266 To assess the economic feasibility of BC supplementation to AD at full-scale, an economic analysis was carried
267 out using input data of semi-continuous AD tests at lab or pilot scale (Table B3 in Appendix B). Input data
268 (ΔCH_4 , VS , d_{BC}) were directly extracted or derived from the available information. Some studies were excluded
269 due to lack of useful data, and 14 studies with 57 experimental conditions in semi-continuous mode were
270 included in the economic analysis. Since the specific objective of the analysis was to assess the economic
271 feasibility of biochar addition to AD, the main assumption was to consider BC addition to an operating full-
272 scale digester, compared to AD without BC as baseline, similarly to another recent techno-economic analysis
273 [50] related to the addition of various BCs to the AD of food waste. Therefore, capital costs and operation and
274 maintenance costs of the digester were not considered, as in the mentioned study [50].

275 The economic analysis estimated the maximum sustainable unit cost of BC ($C_{BC,max}$) [41], which equals the
276 higher (compared to AD without BC) revenues from the enhanced AD. The $C_{BC,max}$ was compared with the
277 current BC market price. The revenues consisted of the extra thermal energy and electricity from the
278 combustion of biogas in a combined heat and power (CHP) unit due to the enhanced CH_4 production respect
279 to the baseline scenario without BC supplementation. The revenues from thermal (R_{TH}) and electrical (R_{EL})
280 energy were derived as in eq. 5:

$$281 \quad R_i = \Delta CH_4 \cdot VS \cdot LHV_{CH_4} \cdot \eta_{CHP,i} \cdot C_{EN,i} \quad (5)$$

282 where ΔCH_4 is the difference between CH_4 production rates ($m^3 \text{ kg}_{VS}^{-1}$) of BC and CTRL conditions of each
283 study, and VS ($\text{kg}_{VS} \text{ m}^{-3}$) is the concentration of VS of the substrate fed to digester of each study (Table B3,
284 Appendix B); LHV_{CH_4} is the lower heating value of CH_4 , $\eta_{CHP,i}$ is the thermal or electrical energy efficiency
285 of CHP unit, $C_{EN,i}$ is the average EU-27 thermal or electrical energy price for non-household consumers (Table
286 B4, Appendix B). The operational cost of BC supply depends on the dose and unit cost of BC. Other capital
287 costs related to the equipment for BC storage, handling, and dosing in the AD substrate can be reasonably
288 neglected [50]. Therefore, by equalling the revenues and the cost of BC supply for each experimental condition,
289 the maximum BC unit cost ($\$ \text{ ton}_{BC}^{-1}$) was estimated as in eq. 6:

290
$$C_{BC,max} = \frac{R_{TH}+R_{EL}}{d_{BC}} \cdot ER \cdot 1000 \quad (6)$$

291 where R_{TH} and R_{EL} are the revenues from thermal and electrical energy determined according to eq. 5; d_{BC} is
292 the dose of BC (kg m^{-3}) supplemented to digester of each study (Table B3 in Appendix B), ER is the average
293 exchange ratio USD-euro (Table B4 in Appendix B).

294

295 **4. Results and discussion**

296 **4.1. Effects of feedstocks and pyrolysis temperature on biochar properties**

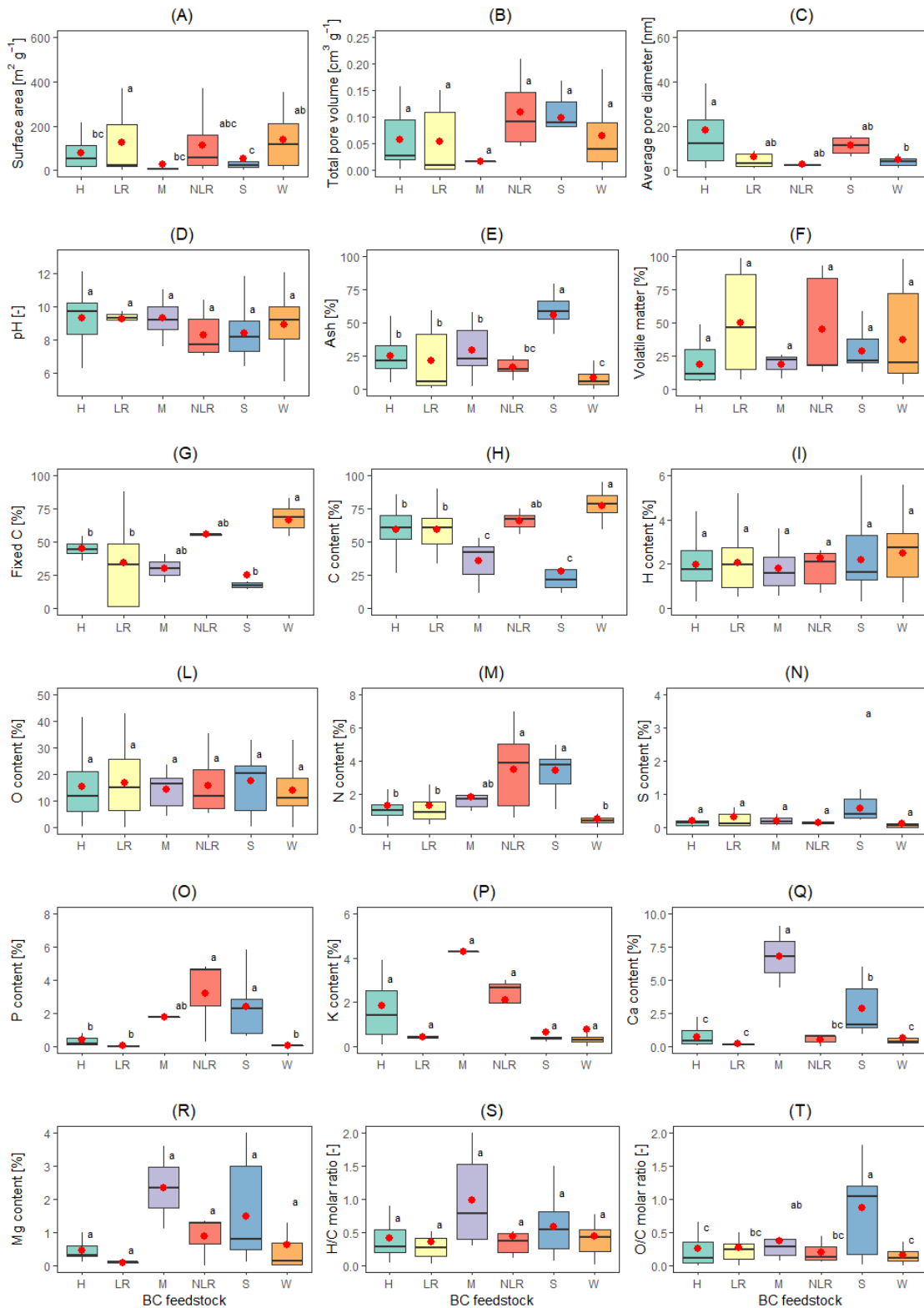
297 The distinctive properties of BC parent biomass and the operating conditions of its production (temperature,
298 heating rate, duration) determine the physico-chemical properties of BC. Figure 1 shows the influence of the
299 different feedstock categories on BC physico-chemical properties. The chemical composition of BC parent
300 biomass vary significantly: wood or lignocellulosic residues are mainly composed by cellulose, hemicellulose,
301 and lignin and have a low ash content [5]; herbaceous precursors have a similar composition, but they are
302 richer in ashes [51]; bio-solids (manure or sewage sludge) are a mixture of moisture, micro-organisms,
303 organics, and abundant ashes rich in nutrients and metals [52,53]. Regarding BC physical features,
304 lignocellulosic residue BCs (LR) and wood BCs (W) exhibited a larger specific surface area (SA) (Figure 1A),
305 while W and non-lignocellulosic residue BCs (NLR) a lower average pore diameter compared to other
306 feedstock-based BCs (Figure 1C). Feedstocks with high contents of volatile matter and lignin (W and LR) tend
307 to develop a dense porous structure through the release of volatile compounds during pyrolysis, which is
308 preserved by the thermal stability of lignin [54], resulting in a larger porosity and SA. Conversely, BCs from
309 sludge (S) and manure (M) showed a relatively low SA, probably due to the large ash content (Figure 1E)
310 inducing structural cracking or micropore blockage during pyrolysis [55]. Further, BCs from sludge (S) and
311 manure (M) presented scarce contents of fixed carbon (FC) and total carbon (C) (Figure 1G-H). W clearly
312 differed from other feedstock-based BCs for a lower ash content and higher FC and C. Concerning the contents
313 of nutrients and alkali metals, a larger content of N and P was observed for BCs from S, M, and NLR (Figure
314 1M-O), while M exhibited a high content of Ca (Figure 1Q). Finally, the O/C molar ratio (Figure 1T) was
315 higher for S and M BCs, suggesting a higher polarity and presence of O-containing functionalities than for
316 herbaceous (H) and W BCs. The cation exchange capacity (CEC) is the amount of exchangeable cations that

317 BC is able of holding [6]. Manure- and sludge-derived BCs have greater CEC compared to other BCs due to
318 an increased ash content [55]. Unfortunately, differences between feedstocks relative to the other BC properties
319 were not significant.

320 The variation of pyrolysis temperature also affects BC physico-chemical properties, as clearly shown in Figure
321 2. The specific surface area increased with pyrolysis temperature (Figure 2A). At low temperatures (up to 200
322 °C) the evaporation of moisture and light volatiles occurs with breakage of bonds; from 200 to 500 °C
323 hemicellulose and cellulose devolatilize and decompose at faster rate; over 500 °C lignin and other organic
324 matter with stronger chemical bonds tend to degrade [56]. The progressive release of gas, water, micro-organic
325 compounds during pyrolysis can create more voids within the BC matrix [56].

326 A rising processing temperature increased the pH (Figure 2D), likely due to the release of acidic functional
327 groups as carboxyl, hydroxyl, or formyl groups [6] and the formation of Ca-, Mg-, Na-, and K-bearing oxide,
328 hydroxide, and carbonate phases [55], and decreased the volatile matter (VM) (Figure 2F). Since elements as
329 H, O, N tend to volatilize with increasing temperatures (Figure 2I-L-M), the contents of FC and C concentrated
330 significantly within the BCs (Figure 2G-H). Both H/C and O/C molar ratios were reduced significantly by an
331 increased pyrolysis temperature (Figure 2S-T). BCs produced at low temperatures are mostly polar and
332 hydrophilic due to the abundance of oxygen-containing functional groups; BCs produced at high temperatures
333 contain less functional groups and more aromatic structures, with hydrophobic functional groups and a higher
334 stability [5,57]. CEC of BCs tends to decrease with rising temperatures due to the removal of surface functional
335 groups providing negative charges [56].

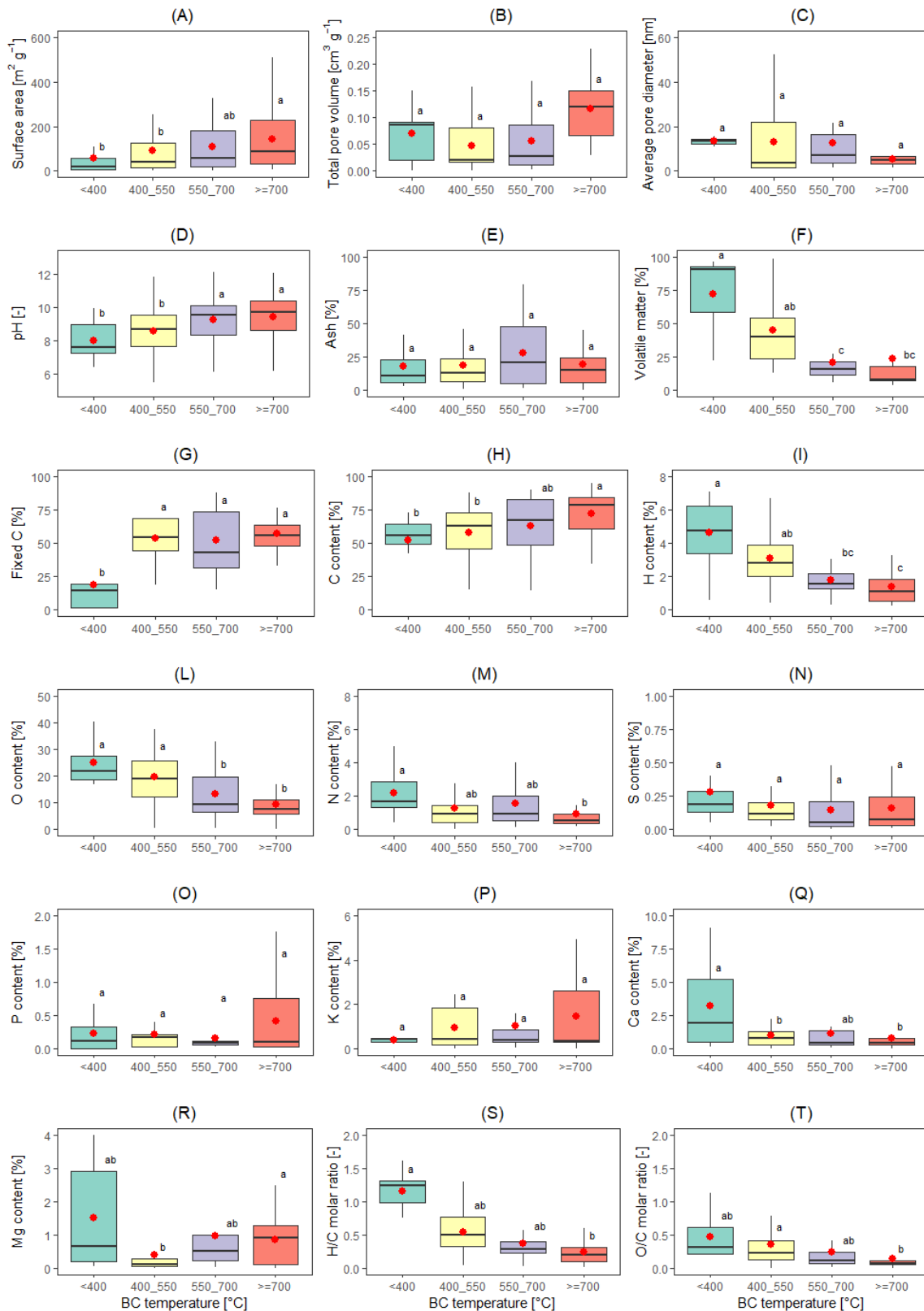
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337

338 Figure 1. Effect of the different parent biomass on the physico-chemical properties of biochar: (A) Surface
 339 area; (B) Total pore volume; (C) Average pore diameter; (D) pH; (E) Ash; (F) Volatile matter; (G) Fixed C;
 340 (H) C content; (I) H content; (L) O content; (M) N content; (N) S content; (O) P content; (P) K content; (Q)
 341 Ca content; (R) Mg content; (S) H/C molar ratio; (T) O/C molar ratio. Parent biomass: H: herbaceous; LR:

342 lignocellulosic residue; M: manure; NLR: non-lignocellulosic residue; S: sludge; W: wood. A significant
343 difference ($p < 0.05$) within a boxplot is identified by different letters. Red dots: average values; lines: median
344 values.



345
346

347 Figure 2. Influence of pyrolysis temperature of the physico-chemical properties of biochar: (A) Surface area;
348 (B) Total pore volume; (C) Average pore diameter; (D) pH; (E) Ash; (F) Volatile matter; (G) Fixed C; (H) C
349 content; (I) H content; (L) O content; (M) N content; (N) S content; (O) P content; (P) K content; (Q) Ca

350 content; (R) Mg content; (S) H/C molar ratio; (T) O/C molar ratio. A significant difference ($p < 0.05$) within
351 a boxplot is identified by different letters. Red dots: average values; lines: median values.

352 **4.2. Meta-analysis**

353 *4.2.1. Overall effects of biochar addition on anaerobic digestion performance*

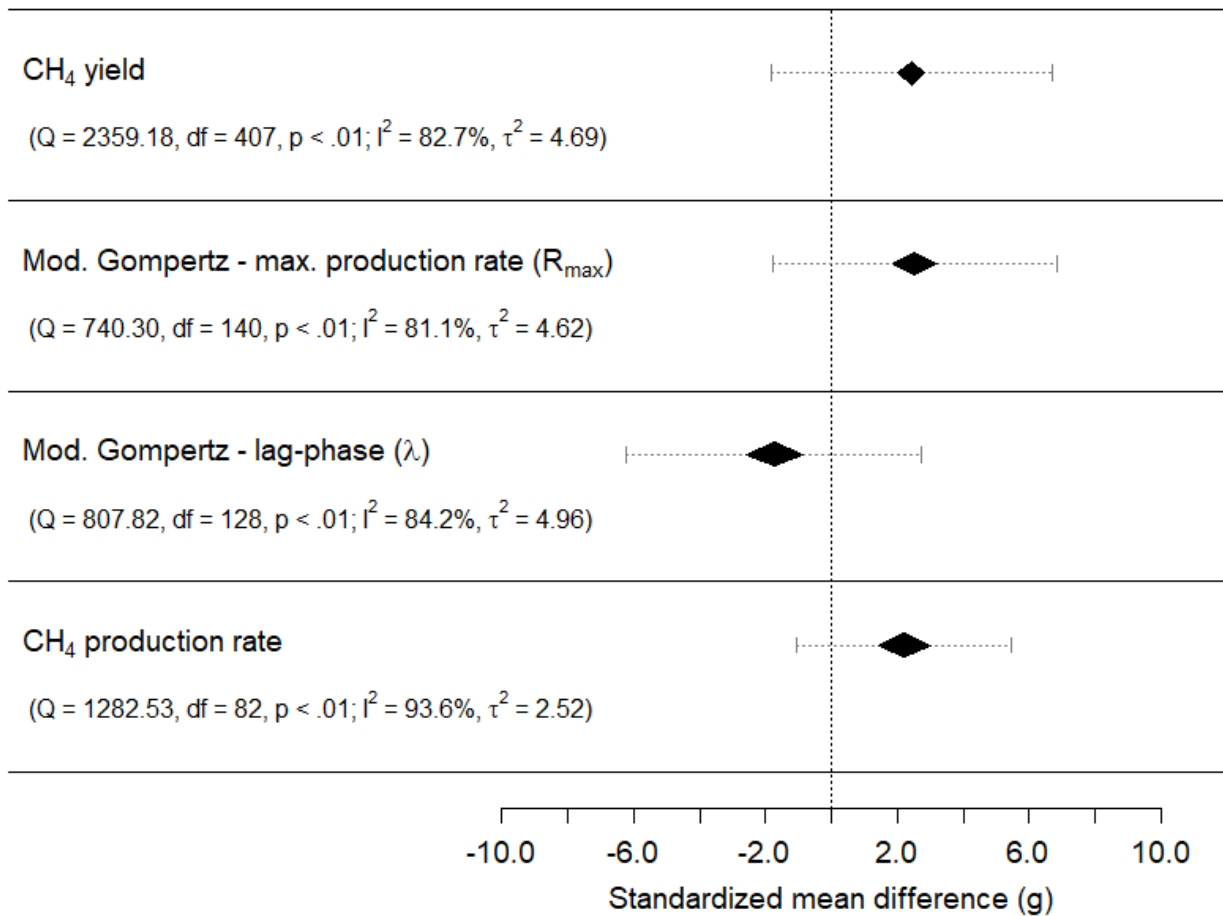
354 A meta-analysis was used to assess the overall (“summary”) effect of BC addition on AD performance using
355 408 experimental conditions of 76 studies in batch mode and 83 conditions of 18 studies in semi-continuous
356 mode (Figure 3). Considering the subset of batch tests, the effect of BC addition was investigated on three
357 variables: CH₄ yield, maximum CH₄ production rate (R_{\max}) and lag-phase (λ) from the modified Gompertz
358 model fitting of experimental data. On the one hand, the overall effect of BC on CH₄ yield was significant (g
359 = 2.43, 95% CIs = 2.02-2.84), clearly indicating that BC enhances bio-methane production, consistently with
360 the previous meta-analysis by Xiao et al. (2021) [27]. The PIs overlap the line of null effect, indicating also
361 adverse impacts of BC in a reduced number of studies, likely due to inhibitory effects on the methanogenic
362 active toity related excessive doses of BC [58,59]. Not surprisingly, there was a significant heterogeneity
363 among different studies ($Q = 2359$, $p < 0.01$, $I^2 = 82.7\%$), given the high variability of the physico-chemical
364 properties of BCs (section 3.1), AD substrates and AD operating conditions tested (Table 1). Therefore, the
365 role of different moderator variables was further investigated by subgroups analysis. On the other hand, the
366 supplementation of BC accelerated methane production, as indicated by the significant enhancement of R_{\max}
367 ($g = 2.54$, 95% CIs = 1.86-3.22). Besides, the duration of lag-phase was shortened by the presence of BC ($g =$
368 -1.74 , 95% CIs = $-2.60/-0.88$) able to reduce biomass adaption period to new AD conditions. Therefore, both
369 the start-up phase of a digester or the adaption to substrate changes may be accelerated through BC
370 supplementation. For both R_{\max} and λ , there was a significant heterogeneity among studies ($Q = 740$, $p < 0.01$,
371 $I^2 = 81.1\%$ and $Q = 807$, $p < 0.01$, $I^2 = 84.2\%$, respectively). Overall, BC supplementation can significantly
372 enhance and accelerate methane production during AD in batch mode.

373 In case of semi-continuous AD studies, an additional meta-analysis was conducted to assess the effect of BC
374 on CH₄ production rate. Consistently with previous results, BC supplementation significantly improved CH₄
375 production rate ($g = 2.21$, 95% CIs = 1.42-3.01). Therefore, the overall positive impact of BC addition on AD
376 performance was confirmed in feeding conditions closer to the full-scale than batch tests.

377

378

Summary effects



379

380 Figure 3. Forest plot of the summary effects (*g* - Hedges' standardized mean difference): influence of biochar
 381 addition on CH₄ yield, maximum CH₄ production rate (R_{max}) and lag-phase (λ) from modified Gompertz fitting
 382 during batch AD tests; influence of biochar addition on CH₄ production rate during semi-continuous AD tests.
 383 The position of a diamond stays for the average effect size and its vertices for the lower and upper 95%
 384 confidence intervals; the dashed line identifies the 95% prediction intervals.

385

386 4.2.2. Effect of moderator variables on CH₄ yield, maximum CH₄ production rate and lag-phase

387 As just described, in the meta-analysis with 408 paired conditions in batch mode and 83 conditions in semi-
 388 continuous mode, BC supplementation to AD significantly enhanced CH₄ yield, R_{max}, and CH₄ production rate,
 389 while reduced λ. Therefore, globally, BC addition to AD can enhance and accelerate methane production.
 390 However, the summary effects exhibited a significant heterogeneity (Figure 3) due to the high variability of

391 the employed BCs and AD conditions. In this section, the influence of different moderator variables was
392 assessed and discussed through subgroups analysis. The variables considered were as follows: AD temperature
393 and substrates, BC dose, temperature and feedstocks for BC production, and BC physico-chemical properties
394 (ash, pH, surface area, O/C and H/C molar ratios, C, H, O, N contents). The impact of the moderator variables
395 on CH₄ yield is discussed as follows, while their effect on R_{max} and λ is exposed in Appendix B (Figure B1
396 and B2).

397

398 *4.2.3. Impact on methane yield*

399 In Figure 4, the results of the subgroups analysis on CH₄ yield are presented, focusing on the influence of
400 different moderator variables. Considering AD temperature (Figure 4A), both mesophilic and thermophilic
401 conditions showed positive effects of BC on CH₄ yield, slightly larger at mesophilic temperatures. Further, a
402 positive effect of BC addition was observed for most AD substrates except in the case of wastewater, which
403 did not exhibit a statistically significant difference from the null effect. The lowest effect was for simple
404 substrates, the largest for co-digestion. The relatively large CIs of co-digestion and wastewater may be
405 attributed to the limited sample size, but also to the highly heterogeneous nature of these substrates. The effect
406 for sewage sludge was slightly lower than for other substrates, as for OFMSW, manure, food waste with
407 manure or sludge.

408 Overall, the BC dose resulted in significantly positive effects on CH₄ yield, except for the range 15-20 g L⁻¹.
409 The largest effect was observed for the lowest doses (<5 g L⁻¹), followed by the range 5-10 g L⁻¹. Conversely,
410 a previous meta-analysis by Xiao et al. (2021) [27] found a larger impact on methane production for BC doses
411 exceeding 10 g L⁻¹. Excessive BC doses can inhibit methane production, possibly due to the non-selective
412 adsorption of gases or intermediate AD products, or to the presence of excessive concentrations of metals in
413 the BC [40,60–62]. The optimization of BC dose is a crucial step to apply BC to AD at full-scale.

414 The impact of BC production temperature on CH₄ yield was positive but remarkably diversified. BCs produced
415 at temperatures below 400 °C displayed the highest improvements, followed by BCs produced at 550 - 700
416 °C. BCs synthesized at temperatures above 700 °C had the lowest impact. The diverse response can be linked
417 to the variation of BC physico-chemical properties controlled by changes in pyrolysis temperature (section
418 3.1).

419 Overall, all feedstocks for BC production exhibited positive impacts on CH₄ yield. The largest effects were
420 slightly different from each other: sludge>manure>lignocellulosic residues>herbaceous. Instead, BCs from
421 wood feedstocks presented the lowest *g*, significantly lower than the others. Again, the diverse influence of
422 feedstocks can be related to the different physico-chemical properties of BCs (section 3.1).

423 Considering the BC physico-chemical properties, larger ash contents resulted in the highest *g*, equal to 4.10
424 for ash contents in the range 20-50% and 5.12 for ash contents beyond 50%. Significantly lower *g*, around
425 0.82-1.94, were found for ash contents below 20%. These findings are consistent with previous observations:
426 wood-based BCs with the lowest ash contents (Figure 1C) had the lowest impact compared to other BC
427 feedstocks; instead, BCs from sludge and manure characterized by large ash contents showed the largest *g*
428 values. BCs having large O/C molar ratios, over 0.3, exhibited the largest *g* of 3.71 (Figure 4B), significantly
429 different from BCs having O/C below 0.15. Therefore, BCs with more polar O-containing functional groups
430 and hydrophilicity may be more favourable for methane production. Again, consistently with previous
431 findings, sludge-based BCs showed the largest impacts on CH₄ yield having larger O/C molar ratios than wood-
432 based BCs (Figure 1T). Furthermore, pyrolysis temperatures below 400 °C resulted in the highest *g* values,
433 having large O/C which tends to decrease with rising temperatures (Figure 2T). Except for the BCs having
434 H/C molar ratios ranging 0.5-1, other BCs led to significant positive effects on CH₄ yield without net
435 differences between categories.

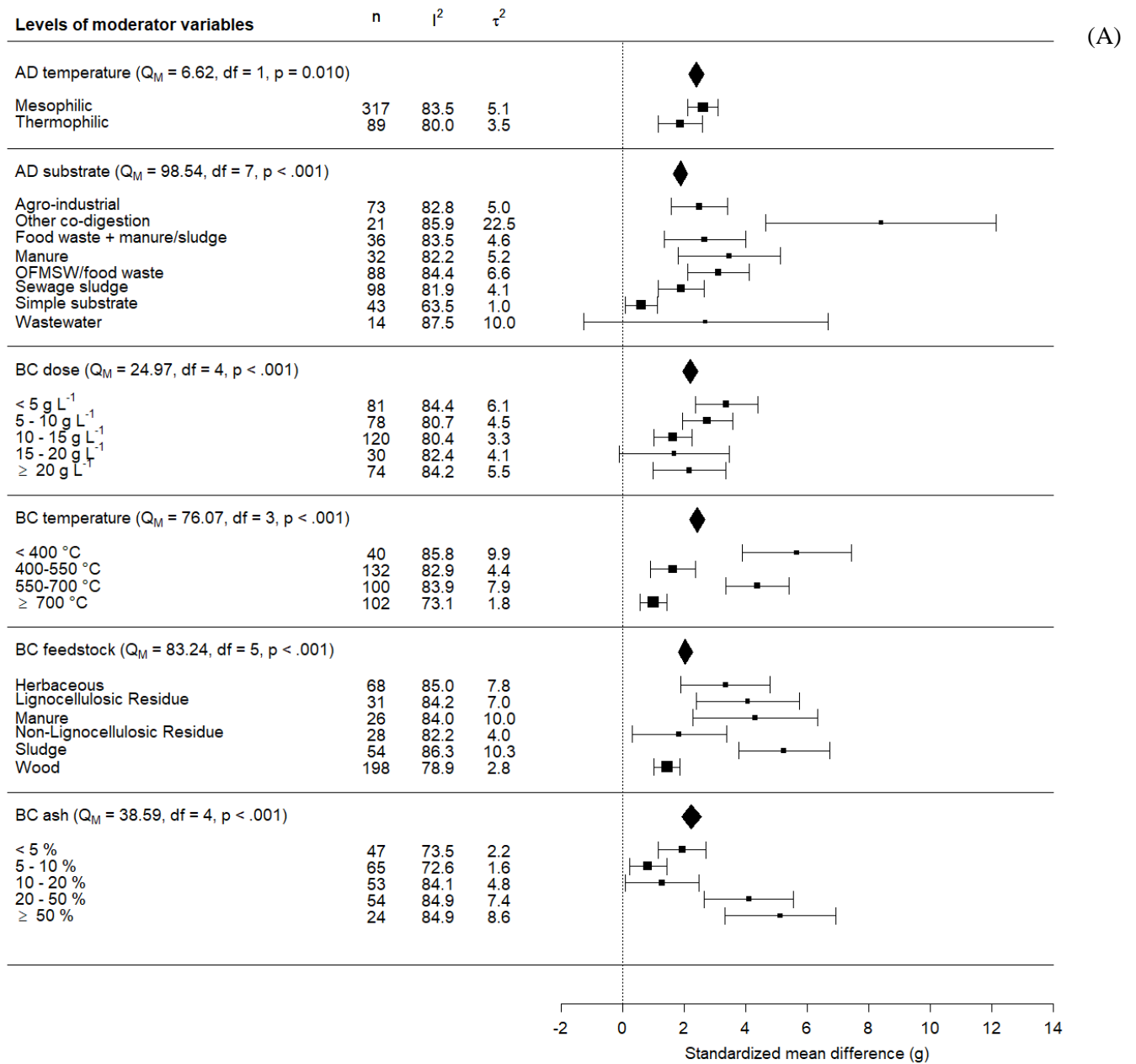
436 Regarding the C content, the largest impact on CH₄ was found for BCs having C <50%, consistently with ash
437 content (as sludge-based BCs, Figure 1H). The lowest effect was for C >80% (as wood-based BCs, Figure
438 1H). BCs with higher content of O (≥20%) and N (≥0.6%) exhibited larger improvements of CH₄ yield. An
439 increasing content of H resulted in improvements up to a range of 2-3%, though higher H contents exhibited
440 insignificant (H 3-4%) or low *g* (H ≥ 4%). Regarding the role of pH on CH₄ yield, the largest *g* value was
441 observed for BCs with an acidic pH, while BCs having pH 8.0 - 9.0 resulted in the lowest *g*, which became
442 slightly larger for BCs having a pH ≥9. Therefore, there is no evidence that BCs with basic pH can determine
443 better enhancements of CH₄ yield when compared to acidic BCs.

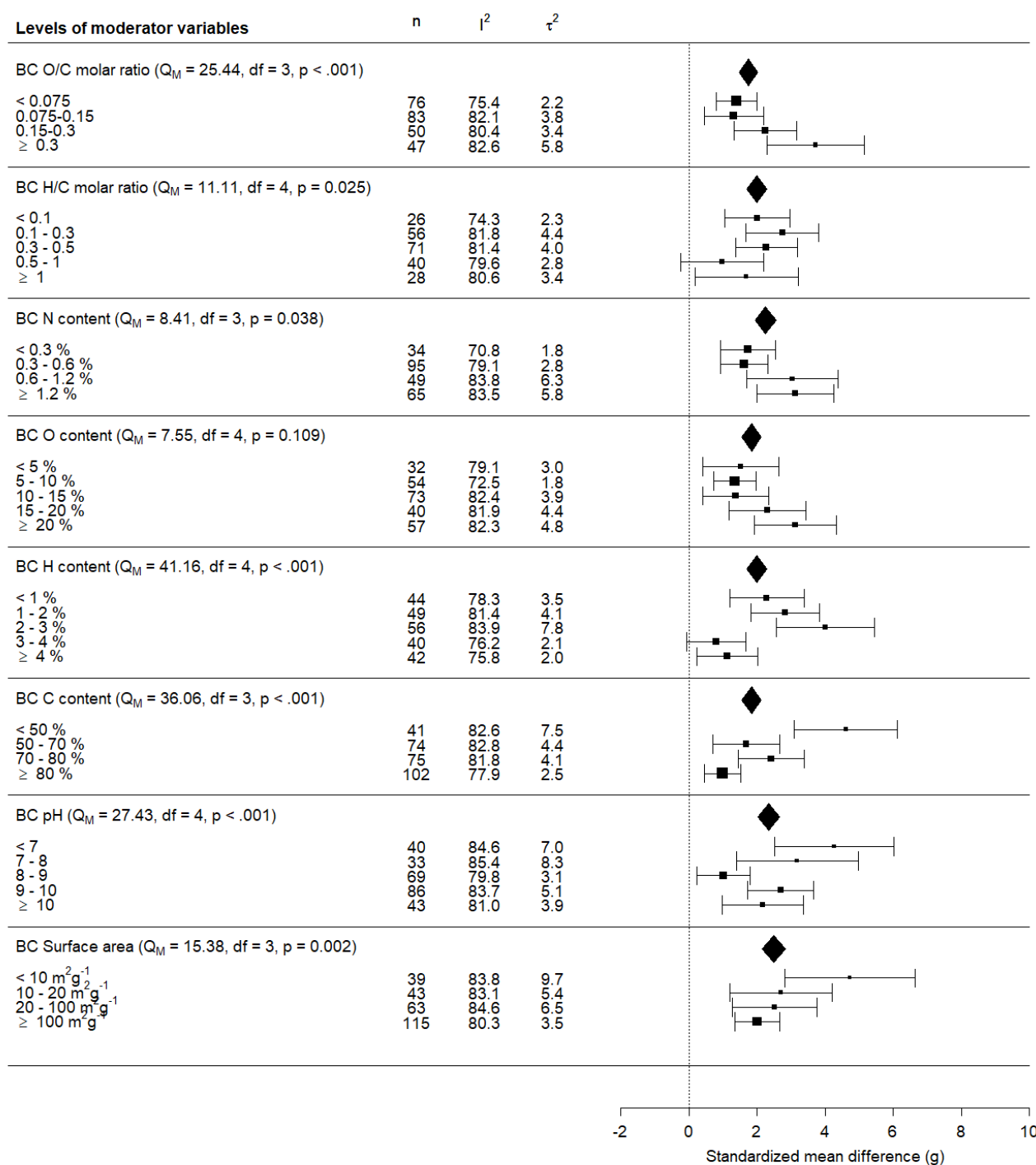
444 All specific surface area categories resulted into positive effects of BC. The largest effect was for BCs with
445 SA <10 m² g⁻¹, the lowest for BCs with SA >100 m² g⁻¹, consistently with previous observations about
446 feedstocks and pyrolysis temperature. Wood-based BCs having large SA resulted in the lowest effects among

447 different feedstocks. BCs from low pyrolysis temperature exhibited the highest impacts as for BCs having low
448 SA, likely because an increasing temperature tends to enhance SA of BCs (Figure 2A). Despite large SA was
449 reported to enhance the BC absorption capacity and favour microbial attachment on BC [18], there is no
450 evidence of an enhancement of BC effect on CH₄ yield due to high SA.

451 Overall, based on the subgroups meta-analysis, the optimal range of BC physico-chemical properties to
452 enhance CH₄ yields may be as follows: high ash contents ($\geq 20\%$) and low C contents ($< 50\%$), high O/C molar
453 ratios (≥ 0.3), high contents of O ($\geq 20\%$) and N ($\geq 0.6\%$), acidic pH (< 7.0), and modest values of SA ($< 10 \text{ m}^2$
454 g^{-1}). To synthesize BCs having these range of properties, the selection of feedstocks as manure or sludge may
455 be recommended, as well as the use of moderate pyrolysis temperatures (see section 3.1). The results of the
456 subgroups meta-analysis of BC feedstocks and pyrolysis temperature support these observations. Our findings
457 are in partial agreements with the literature. Masebinu et al. (2019) [21] suggested that BCs with diverse
458 functional groups, increased SA and porosity, abundance of micropores over other pore sizes, high electrical
459 conductivity, and large alkalinity can favour the AD stability and enhance CH₄ content. A meta-analysis by
460 Xiao et al. (2021) [27] declared more favourable for AD the BCs having low electrical conductivity (< 450
461 $\mu\text{S}/\text{cm}$), synthesized at temperatures $< 700 \text{ }^\circ\text{C}$. Conversely, they did not find any statistically significant impact
462 on the global effect of BC of properties as pH, SA, and particle size; this may be attributed to the restricted
463 sample size compared to the present study.

464





(B)

467

468 Figure 4. Forest plot with results of subgroups analysis on CH₄ yield, influence of different moderator
 469 variables: (A) AD temperature and substrates, BC dose and production temperature, BC feedstocks and ash
 470 content; (B) BC characteristics: O/C and H/C molar ratios, C, H, O, N contents, pH, surface area. For each
 471 subgroup the mean effect size and 95% confidence intervals are reported. The summary effect for each
 472 moderator variable is indicated by a diamond. The results of testing the significance of each moderator variable
 473 are reported between brackets: Q_M , degrees of freedom and p-value (significant when <0.05).

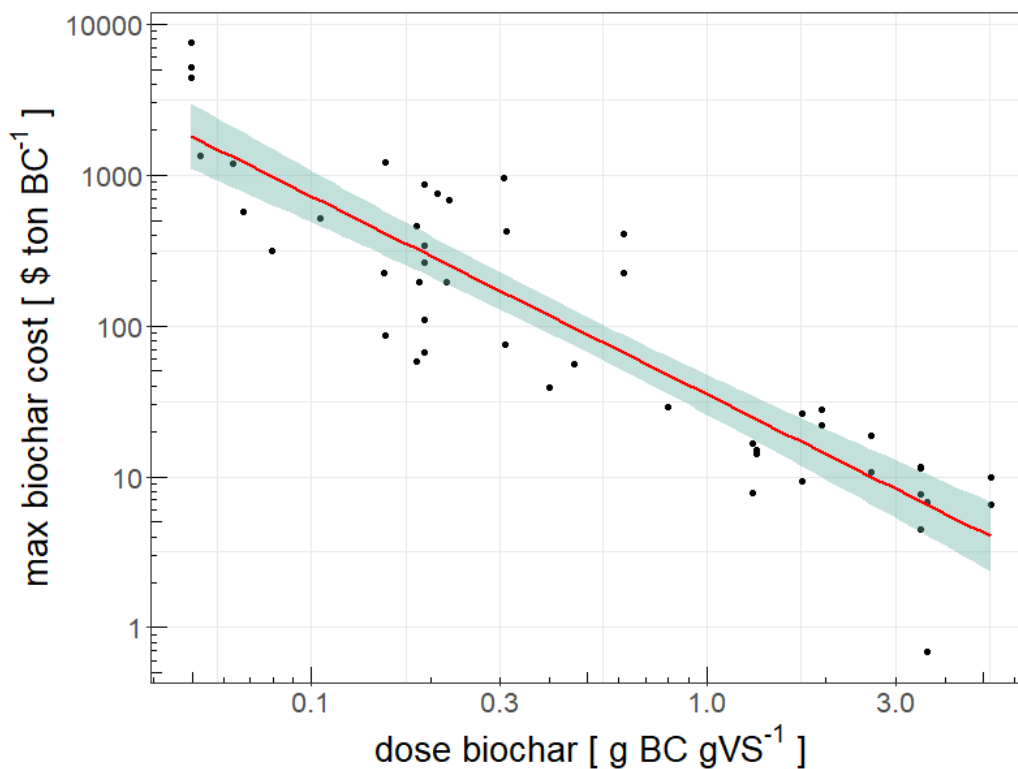
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475 **4.3. Economic analysis**

476 An economic analysis was carried out using input data of semi-continuous AD tests at lab or pilot scale from
477 recent studies (Table B3 in Appendix B) to compare the estimated maximum sustainable BC unit cost ($C_{BC,max}$)
478 with BC market price. Assuming to add BC to an operating digester, the higher revenues due to the extra
479 thermal and electrical energy from the enhanced methane production respect to the scenario without BC
480 supplementation were accounted. As a result, a wide range of $C_{BC,max}$ (0 – 7,500 USD ton_{BC}^{-1}) was found.
481 Compared with a typical range cost for BC of 50-500 USD ton_{BC}^{-1} [18,41], 49% of $C_{BC,max}$ was below the lower
482 limit, 30% between 50 and 500 USD ton_{BC}^{-1} , and 21% above the upper limit. Therefore, under certain
483 conditions BC addition to AD may be economically feasible. On the one hand, the choice of the proper BC is
484 also crucial from the cost-effectiveness perspective, since BC market price may vary significantly based on
485 feedstock, production process, and location. For instance, lower pyrolysis temperature could reduce BC market
486 price due to a larger BC yield [63,64]. According to our meta-analysis, the largest improvements of CH_4 yield
487 and CH_4 production rate was found for the addition of BCs produced at temperatures <400 °C (Figure 4 and
488 Figure B1 in Appendix B). The feedstock, and specifically its availability and BC yield, and the availability of
489 waste biomass with tipping fees [24,63], may also affect the BC market price. For instance, sludge-based BC
490 was found to have the lowest cost compared to wood- and crop-based BCs due to the higher BC yield [63].
491 Indeed, in the next few years it is expected that a rapid rise of BC supply will likely decline its market price.
492 The market volume of BC was around 1.6 billion USD in 2020 and is expected to reach 4.0-6.3 billion USD
493 by 2026-2031 [65,66].

494 Going further, the role of the main influencing variables on $C_{BC,max}$ was investigated (e.g., the organic load
495 (VS) of AD substrate and the dose of BC) to identify the conditions of cost-effectiveness. A single linear
496 regression was used to predict $C_{BC,max}$ based on BC dose as $g_{BC} g_{VS}^{-1}$ (Figure 5). A significant regression
497 equation was found ($F(1,47) = 197.3$, p-value: $< 2.2 e-16$) with $R^2 0.8076$. The prediction equation was $\log(y)$
498 $= 1.545 - 1.313 \log(x)$, where y is the maximum BC cost as USD ton_{BC}^{-1} , x is the dose of BC as $g_{BC} g_{VS}^{-1}$. A
499 reduction of BC dose can significantly favour the economic effectiveness of BC addition, where an upper
500 threshold of 0.45-0.76 $g_{BC} g_{VS}^{-1}$ may be suggested based on the regression equation (corresponding to 100-50
501 USD ton_{BC}^{-1}). In any case, BC doses above these limits should be avoided despite the choices of BC and AD

502 conditions. Therefore, the optimization of BC dose is a crucial step to maximize methane production (section
503 3.2.2) and to reach the economic feasibility, towards a practical application of BC to full-scale digesters.
504



505
506 Figure 5. Influence of the dose of biochar on the maximum sustainable cost of biochar (both variables were
507 log-transformed). The red line is the regression equation ($F(1,47) = 197.3$, p-value: $< 2.2 \times 10^{-16}$), with R^2 equal
508 to 0.8076, with 95% confidence intervals. The prediction equation is $\log(y) = 1.545 - 1.313 \log(x)$, where y is
509 the maximum BC cost in USD $\text{ton}_{\text{BC}}^{-1}$, x is BC dose in $\text{g BC g}_{\text{VS}}^{-1}$.

510

511 5. Conclusions

512 This study presents a systematic analysis (organized in an inventory) of the current literature on BC addition
513 to AD processes. The selected and categorized literature underwent a meta-analysis, aimed at correlating AD
514 performance with BC physico-chemical properties, to define their optimal range. Overall, the findings of the
515 meta-analysis proved that BC enhances and accelerates biomethane production. Considering 408 experimental
516 conditions from 76 studies in batch mode, methane yield and maximum production rate were significantly
517 enhanced by BC supplementation. Besides, the duration of the lag-phase was shortened by the presence of BC.

518 Further, 83 experimental conditions from 18 studies in semi-continuous mode proved that BC supplementation
519 significantly improved CH₄ production rate. Based on the subgroups meta-analysis, an optimal range of BC
520 physico-chemical properties favouring AD performance may be suggested as follows: high ash contents
521 (≥20%) and low C contents (<50%), high O/C molar ratios (≥0.3), high contents of O (≥20%) and N (≥0.6%),
522 acidic pH (<7.0), and modest values of SA (<10 m² g⁻¹). Consequently, the choice of feedstocks as manure or
523 sludge for BC production, and the use of moderate pyrolysis temperatures can be recommended. The economic
524 analysis, aimed at evaluating the economic viability of adding BC to an existing full-scale AD plant, identified
525 a wide range of maximum unit cost of BC (0 - 7,500 USD ton_{BC}⁻¹) that equals the revenues from energy
526 production. The minimization of BC dose is crucial to reach the economic feasibility of BC application to AD.
527 A dose of BC above 0.45-0.76 g_{BC} g_{VS}⁻¹ should be avoided despite the features of BC properties and AD
528 conditions. In conclusion, the overall findings of this work can provide guidance to further studies exploring
529 the underlying mechanisms of BC addition to AD processes, and support BC practical application in full-scale
530 digesters.

531

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534 contributions: methodology, data elaboration, original draft writing: M. Chiappero; conceptualization,
535 methodology, supervision, manuscript revision: S. Fiore; conceptualization and manuscript revision: F.
536 Berruti.

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