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Review

Preparation, characterisation and applications of bone char, a food waste-derived sustainable material: A review

Clara Piccirillo

CNR NANOTEC, Institute of Nanotechnology, Campus Ecoteckne, Via Monteroni, 73100, Lecce, Italy

A R T I C L E I N F O

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A B S T R A C T

The production of increasing quantities of by-products is a key challenge for modern society; their valorisation – turning them into valuable compounds with technological applications – is the way forward, in line with circular economy principles.

In this review, the conversion of bones (by-products of the agro-food industry) into bone char is described. Bone char is obtained with a process of pyrolysis, which converts the organic carbon into an inorganic graphitic one. Differently from standard biochar of plant origin, however, bone char also contains calcium phosphates, the main component of bone (often hydroxyapatite). The combination of calcium phosphate and graphitic carbon makes bone char a unique material, with different possible uses.

Here bone chars' applications in environmental remediation, sustainable agriculture, catalysis and electrochemistry are discussed; several aspects are considered, including the bones used to prepare bone char, the preparation conditions, how these affect the properties of the materials (i.e. porosity, surface area) and its functional properties. The advantages and limitations of bone chars in comparison to traditional biochar are discussed, highlighting the directions the research should take for bone chars' performances to improve. Moreover, an analysis on the sustainability of bone chars' preparation and use is also included.

1. Introduction

In recent decades the growth of the global population led to a significant increase in the use of the available resources, at a non-sustainable long-term level [International Resource Panel, 2019]. This phenomenon was registered in different fields, including raw materials for construction and manufacturing, as well as natural resources (i.e. drinkable water) [Dolan et al., 2021]. Because of this, it is crucial to adopt policies implementing a more sustainable use of resources; the Sustainable Development Goals adopted by the United Nations, for instance, represent a step in this direction [United Nations, 2022].

One way to achieve a more manageable use of natural resources is the valorisation of waste; this term refers to any process aimed at the reuse of waste materials and/or to their conversion into added-value products or energy sources [Wang et al., 2022]. This approach would also help to solve another major problem of our society, i.e. the production and accumulation of increasing amount of wastes, posing risks to both human health and the environment [Wang et al., 2022]. This methodology is also in line with the principles of the circular economy [Zhang et al., 2022b].

Carbon-based materials, in their different forms, have a wide range of important technological applications, from energy production and storage to environmental remediation [Zhang et al., 2022a; Raza et al., 2022]; moreover, if used with other materials in composites, they can make excellent photocatalysts [Rao et al., 2019]. Although materials such as graphene or carbon nanotubes showed great potential, their synthesis is often expensive and it involves the use of toxic chemicals, resulting in a high impact on the environment [Zhang et al., 2022a]. It is, therefore, necessary to develop more sustainable and cheaper carbon-based materials.

Natural sources can be used to prepare carbonaceous materials; carbon, in different organic forms, is in fact the main element of many living organisms, and it can be converted into inorganic graphitic form. Such conversion can be performed with different methods, the most common one being a pyrolysis, i.e. heating in an inert atmosphere (for instance in N_2, with no or very low O_2 concentration). The obtained materials are generally referred to as biochars and have a high carbon content (>70%) [Enaime et al., 2020]. Biochars have been produced from many sources, including wastes from the food industry (one of the sectors which generates more wastes) [Wang et al., 2022] and employed

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E-mail address: clara.piccirillo@nanotec.cnr.it.

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for many different applications [Gupta et al., 2022; Goutham et al., 2022]. Results showed their advanced performance, comparable to those of synthetic carbon materials (not derived from wastes and/or natural sources), due to their high surface area and porosity, as well as the presence of hetero-atoms (i.e. nitrogen) imparting enhanced functionality.

A different type of carbon-containing material is bone char (BC), which is not plant-derived but animal-derived, as it is obtained from animals' bones [Aseem et al., 2022]. Compared to the plant-derived biochars, BCs contain less carbon but a significant amount of calcium phosphate (hereinafter indicated as CaP), as this is the main component in both human and animal bones. More specifically, bones contain hydroxyapatite (HA), a calcium phosphate with formula $Ca_{10}(PO_4)_6(OH)_2$ [Boutinguiza et al., 2012]; the rest of the bones are constituted of proteins, mainly collagen, with the proportion of the CaP and proteins changing according to the animal species, age, the different parts of the body, mineralisation in bones, etc. With the pyrolysis process, the organic carbon from the proteins is converted into amorphous/graphitic C, while nitrogen is also present [Bekiaris et al., 2016].

Both fish and meat industries generate large amount of wastes [Toldrà, 2016], bones being a significant portion of them; it is, therefore, important to investigate their valorisation, with processes aimed at the development of materials with advanced performance and functionalities. Indeed, bones were successfully used to extract valuable compounds, both the inorganic CaPs and the organic proteins and/or their derivates [Boutinguiza et al., 2012, Ferraro et al., 2017]; they could be used in different technological applications, including biomedicine/tissue engineering [Piccirillo et al., 2015], the food industry and environmental remediation.

The conversion of the bones into BC was also studied; indeed, several investigations were performed using bones of different origins, results showing the potential of these materials, and the differences in comparison to the more traditional biochars, as well as the limitations of BC. This review summarises the work performed on the production and uses of BCs of different origins.

1.1. Methodology

In this review, the studies reporting both the preparation and the characteristics of the prepared BC-based materials will be considered, with particular interest in those employed for technological applications.

To focus on the latest development in this field, only the papers published in English, in peer reviewed journals, in the last 15 years, i.e. from 2009 onwards, will be investigated; the databases Scopus and Web of Science will be used as articles sources. The papers that were selected showed a significant degree of novelty, to give some insight into BC potential for a particular applications, or highlight some limitations.

The review will cover BCs applied to environmental remediation (section 2), sustainable agriculture (section 3), catalytic and photocatalytic processes (section 4) and electrochemical applications (section 5). These applications (see Fig. 1 for a summary) were considered as they are the most common ones for this kind of material. Although other review papers were previously published, these generally did not address all the different applications, as they were focused only on a specific one (i.e. environmental remediation or agriculture applications [Azeem et al., 2022; Vassilev et al., 2013]). In this work, a more complete perspective on BC applications will be given; moreover, an assessment on BC production sustainability and environmental impact will also be given (section 6).

1.2. BC preparation: pyrolysis

Before going in detail on the different studies on BC applications, a general introduction on the pyrolysis employed to prepare BC will be given. This will help the understanding and the discussion of the other sections.

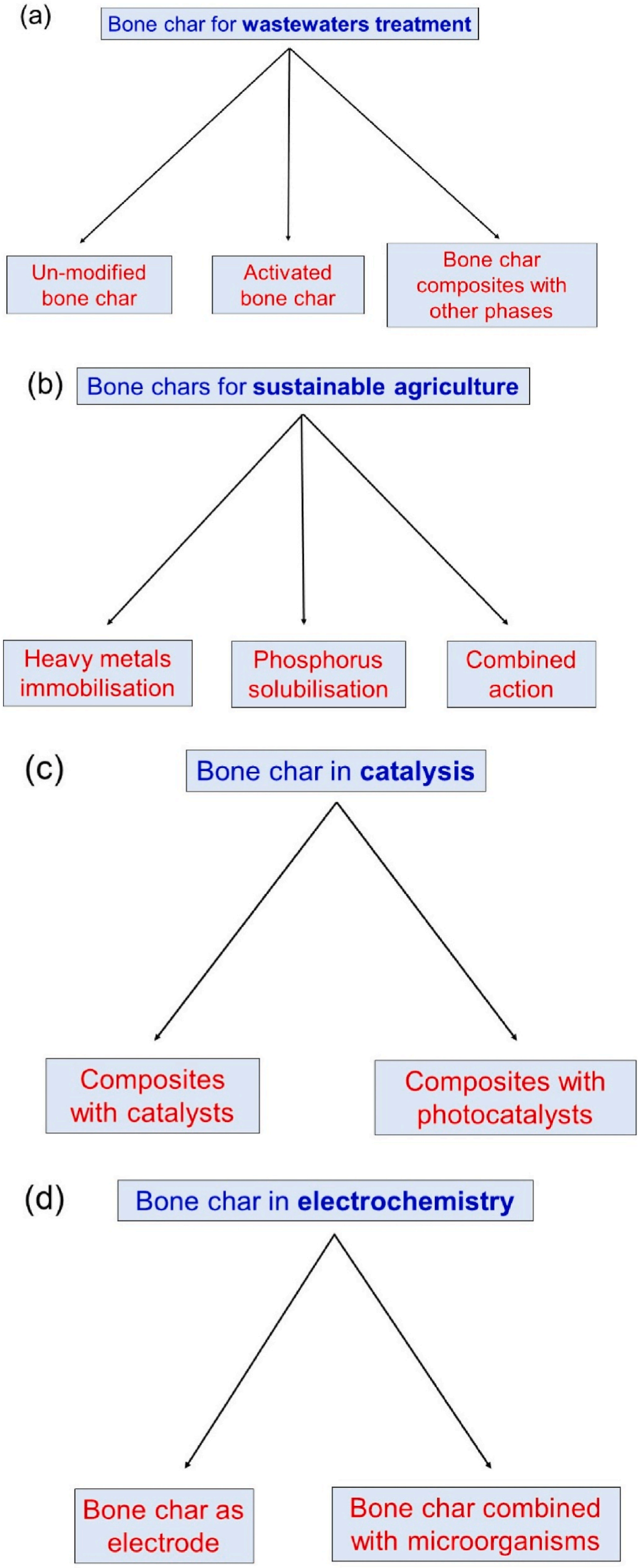

Fig. 1. Scheme of possible BC applications.

As mentioned above, pyrolysis is the process of heating in inert atmosphere; this is different from gasification, which employs selected active gases such as steam, CO_2, etc. And which leads to the production of gases such as carbon monoxide, hydrogen or methane, very useful for industrial processes [Elkhalifa et al., 2019]. When pyrolysis is performed on organic masses (i.e. plants or bones), it results in a change in the form/structure of the carbon, from organic to inorganic. Different reaction mechanisms and steps take place during the process. Initially, a degradation of the (large) organic molecules present in the material occurs; this is due to the effect of the heat/high temperature and leads to the formation of smaller molecules and/or fragments of the original large species. Subsequently, a condensation of such molecules into aromatic benzene rings takes place, leading to the formation of a graphitic structure [Hu et al., 2019]. The extent of conversion from amorphous carbon into more crystalline graphite depends on different parameters, the most significant one being the temperature [Tomczyk et al., 2020]; indeed BCs prepared at lower temperatures tend to have more amorphous and less graphitic carbon, while the opposite is true for higher temperatures [Ai et al., 2020]. The source of the material, however, can also play a role; in fact, the presence of nitrogen can have an effect on the special arrangement of the aromatic rings, leading to different extents of graphitisation [Muretta et al., 2022].

In addition to the solid carbonaceous material, liquid and gaseous molecules are also formed during the pyrolysis; regarding the liquid species, they are generally bio-oils, whose composition depends on the composition of the starting material. As for the gaseous molecules, the most common ones are CO_2, CO, H_2, C_xH_y, although NO_x can also be present, according to the N content of the starting material [Feng et al., 2018]. The pyrolysis conditions can be changed according to target of the process; process with very fast ramps are generally preferred if the objective is the recovery of the gaseous molecules or of the bio-oils; slower ramps, on the other hand, are employed to extract the solid graphitic material [Enaime et al., 2020]. As this review focuses on the solid BC, the formation of liquid and gaseous molecules was not studied in detail, being outside the scope of the work. It has to be highlighted, however, that especially the release of gases have a significant effect on the characteristics of the prepared BC, as this is associated with a significant increase in the surface area. Pyrolysed bones start to release gases at $T > 400$–500 °C, although the values may vary according to the characteristics of the bones and the amount of organic matter present [Kantorek et al., 2019].

Further to the heating ramp, other parameters in the pyrolysis conditions can have a significant effect on the characteristics of the prepared BC materials (see Fig. 2); it is, therefore, important to select the preparation conditions, to obtain materials with the specific properties required for particular applications. Although some features will be discussed in detail according to the BC use, some general aspects will be analysed here.

One of the key parameters is the temperature of the pyrolysis, as higher values lead to BCs with lower carbon content and, consequently, higher CaP proportions; similarly, the amount of nitrogen also decreases with higher temperatures [Piccirillo et al., 2017b]. As stated above, higher temperatures are also associated with higher surface area, due to the release of gaseous molecules; this is in contrast with what is observed in simply calcined bones, where higher temperatures correspond to lower surface areas [Piccirillo, 2017a]. The release of the gas, on the other hand, leads to the formation of pores of different dimensions in the materials, both nanometric or micrometric [Côrtes et al., 2019]. Temperature also has a significant effect on the composition and crystallinity of the BC compounds produced. Generally, like for all materials, higher temperatures correspond to greater crystallinity; indeed, for the CaP phase, pyrolysis performed at higher temperatures gave more crystalline HA [Piccirillo et al., 2017b].

Although high surface area is desirable for most applications, features like the relative amount of carbon and/or its graphitisation can have different effects on materials' behaviour for different features, including their interactions with pollutants, with microorganisms and free radicals. These points will be analysed in depth in the next sections.

2. BC for environmental remediation – adsorption of pollutants

The presence of toxic compounds in the environment is a current serious challenge for our society; indeed, the concentrations of such compounds increased significantly in recent years due to both industrialisation and growth of the world's population [Shaddick et al., 2020]. Pollutants are ubiquitous, i.e. they are in present in air, waters/wastewaters or soils; the nature of such compounds may be different according to the different environments considered. In air, for instance, the most common toxic compounds are Volatile Organic Compounds (VOCs) [Halios et al., 2022], while waters and soils contamination can be due to heavy metals and/or organic pollutants [Choi, 2022]. Within the latter, Persistent Organic Pollutants (POPs) are a cause of growing concern; these are, in fact, molecules which are resistant to traditional wastewater treatment(s) and, because of this, they are progressively accumulating in the environment [Rizzo et al., 2019].

For all these reasons, it is crucial to develop effective and sustainable methods to address this problem; the use of BC to remove the pollutants can be quite effective. In the next sections we will describe how bone chars of different origins were employed in wastewater and/or air; we will first consider simple BCs with no modifications, and successively BCs modified either physically or chemically. Regarding the use in soils,

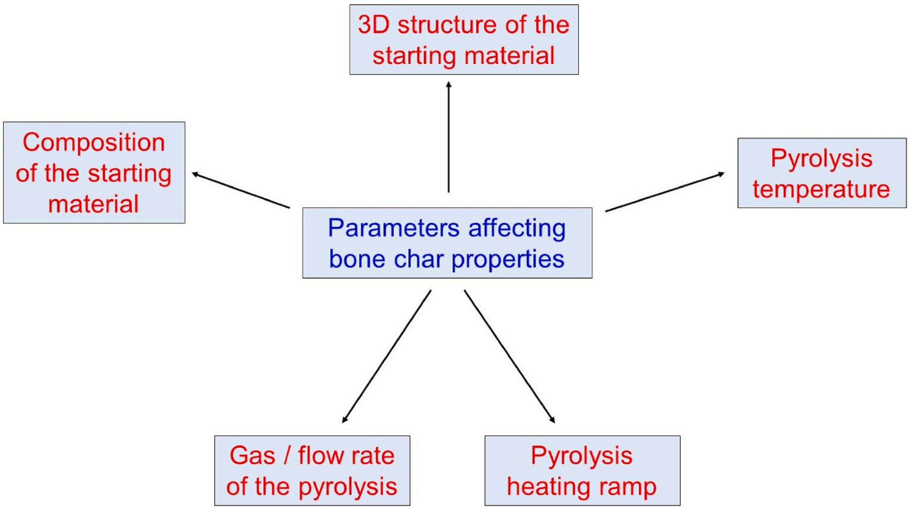

Fig. 2. Parameters affecting the characteristics and functional properties of bone char.

often the environmental remediation was combined with sustainable agricultural practices; hence, this topic will be reported in section 3.

2.1. Un-modified BC

BCs used for environmental remediation is the area in which more studies were reported; in Table 1 a list of the most significant and recent investigations is shown. It can be seen that some studies were performed using commercial materials, while the majority were carried out with non-commercial samples, prepared directly from waste bones. Different kinds of pollutants were targeted, both organic pollutants and heavy metals; indeed, the capability of simultaneously removing with high efficiency different classes of pollutants is one of the most interesting features of BCs, which makes it attractive for this application. This is due to the multiphase composition of BC, which is constituted of both graphitic carbon and CaP.

Graphitic carbon is known for adsorbing organic molecules, with also materials of natural origin showing very good performances (i.e. high values of q_e, amount of pollutant adsorbed for unit of material) [Alkurdi et al., 2019]. The adsorption process involves different mechanisms, including electrostatic interactions, hydrogen bonding and $\pi-\pi$ interactions between organic compounds and the carbon [Jeyasubramanian et al., 2021], see Fig. 3(a). For heavy metals, on the other hand, these materials are much less efficient, indeed the q_e values are much smaller.

CaP and HA in particular show very good performance for heavy metals removal; HA is especially effective with bivalent cations such as Pb(II), Zn(II), Cd(II), etc. The main adsorption mechanism is ion exchange between the metals and the Ca(II) ions of the HA lattice, although other processes also occur (see Fig. 3(b), [Brazdis et al., 2021]).

The combination of these two phases makes BC more versatile for environmental remediation in comparison to simple biochar, containing only the carbonaceous part. Although differences are observed between different BCs, they are all very porous materials, with high values of surface area (see Table 1); this feature makes them very suitable for pollutants adsorption.

As it can be seen from the Table, studies were performed using different starting materials, i.e. using wastes from different animals; this can lead to materials with different composition and characteristics, which can then affect the materials' performance. The reason for such differences is that different bones contain different proportions of inorganic and organic phases [Toppe et al., 2007]. Such differences, after the pyrolysis, will correspond to materials with different amounts of CaP and graphitic carbon; moreover, differences in the value of surface areas and pore dimensions will also be present, due to the different structures of the bones.

On this specific issue, a particularly interesting study was performed by Wang [2020], who compared the performance of BCs obtained by different bovine parts – legs, ribs, scapulae and vertebrae, each part

Table 1
Non-modified bone char materials used for pollutants adsorption from contaminated waters.

Tested Pollutant(s)	Highest surface area (m^2/g)	Pyrolysis conditions and other details of the study	Highest adsorption capacity (mg/g)	Reference
Commercial materials				
Naproxen	74	Carbotecnia (bovine bones)	4.6	Reynel-Ávila, 2015
F^-	104	Fija Flour (bovine bones)	7.7	Medellín-Castillo et al., 2014
Anodising wastewaters (Na^+, K^+, F^-, SO_4^{2-}, PO_4^{3-})	65	Carbones Mexicanos	8.7 for SO_4^{2-}	Acosta-Herrera et al., 2021
Impurities from nickel plating baths (Zn (II))	65	Carbones Mexicanos	29.5	Pérez Jiménez et al., 2021
Anionic dyes (Acid blue 74, Acid blue 25, Reactive blue 4)	113	Carvão Ativado do Brazil	57.4 for Reactive blue 4	Reynel-Ávila, 2016
$Cu(II)^+$, Zn(II)	113	Carvão Ativado do Brazil	42.1 for Cu(II)	Hernández-Hernández (2017)
F^-	139	Bonechar do Brazil (bovine bones)	4.8	Nigri et al., 2017
Calf bones				
Humic acid	112	O_2-limited atmosphere. 400 °C, followed by a rapid treatment at 800 °C	38.1	Moussavi et al., 2022
Chicken bones				
$Cu(II)^+$, Zn(II), Cd(II)	133	N_2 atmosphere, 10 °C/min, 600 °C, 4 hs.	210 for Cu(II)	Park et al., 2015
Pork bones				
Methyl-tert butyl ether	128	500 °C, 1 h.	3.5	Pongkua et al., 2018
Sheep bones				
F^-, As(III), As(V)	120	O_2-limited atmosphere. 10 °C/min, 500–900 °C, holding times 1–2 hs under N_2.		Alkurdi et al., 2020
Inorganic As	120	O_2-limited atmosphere. 10 °C/min, 500–900 °C, holding time 1 h.	9.8 for As(III)	Alkurdi et al., 2021
Bovine bones				
Cu(II)	172	N_2 atmosphere, 10 °C/min, 500 °C.	83.7	Wang et al., 2020
Remazol Brilliant Blue R	137	CO_2 atmosphere, 20 °C/min, 350–850 °C.	20.6	Bedin et al., 2017
Basic red 9	94	Freshwater fish scales also tested. N_2 atmosphere, 10 °C/min, 800 °C.	95.9	Côrtes et al., 2019
F^-	118	N_2 atmosphere, 5–10 °C/min, 550–900 °C, 2–4 hs.	11.8	Asgari et al., 2019
F^-	69	CO_2 atmosphere, 5–10 °C/min, 650–1000 °C.	5.9	Rojas-Mayorga et al., 2015b
Methylene blue	119	O_2-limited atmosphere, 10 °C/min, 400 °C.		Jia et al., 2018a
Other meat-based sources				
Cd(II)	99	Rendering carcass residues. N_2 atmosphere, 250–600 °C	73.5	Park et al., 2021
Fish sources				
F^-, Cd(II)	107	Pleco (*Pterygoplichthys* spp) bones. O_2-limited atmosphere. 500 °C.	213 for Cd(II)	Medellín-Castillo et al., 2020
Diclofenac, fluoxetine, Pb(II)	86	Cod fish bones. N_2 atmosphere, ramp 5 °C/min, 200–1000 °C.	714.2 for Pb(II)	Piccirillo et al., 2017b

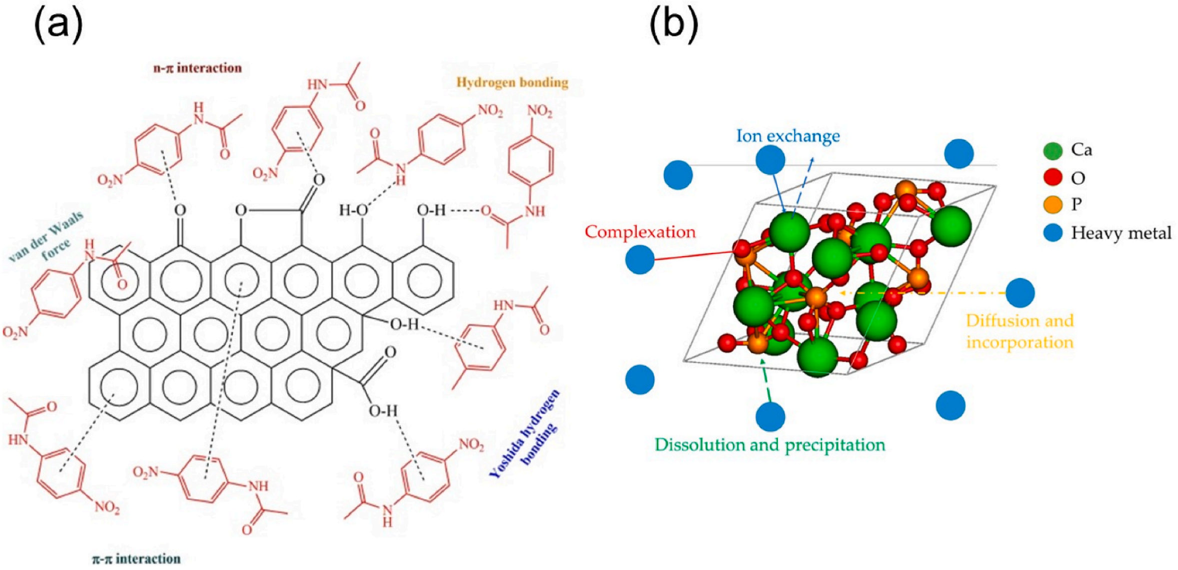

Fig. 3. Mechanisms of (a) interactions between graphitic carbon and organic pollutants [Jeyasubramanian et al., 2021], (b) hydroxyapatite and heavy metals [Brazdis et al., 2021]. Adapted and reproduced with permission, copyrhights of Elsevier and MDPI respectively.

differing in composition from the others. Results showed that the BCs prepared from the different bone parts showed different carbon and CaP content, as well as different surface areas. BC derived from ribs had the highest surface area and, consequently, showed the highest performance in Cu(II) removal. It has to be highlighted, however, that in the study there is not a clear correlation between the features of the starting materials and those of the derived BCs. This shows that the process of converting bones into BC is quite complex, with many parameters affecting it, which makes it difficult to predict BCs' performance.

As already mentioned in section 1.2, the conditions pyrolysis is performed under surely have an effect on the characteristics of the materials; indeed, features such as surface charge, pH, presence of functional groups on the surface of the material, surface area and pore size can change according to the pyrolysis conditions [Alkurdi et al., 2020]. Not all investigations report the details of the pyrolysis process; it is, therefore, difficult to perform a complete comparison. Some general considerations, however, can be made. As can be seen from Table 1, all pyrolyses were performed with standard heating ramps, i.e. between 5 and 10 °C/min; no study with especially slow or fast ramps was published. This is not surprising, as slow ramps are very time and energy-consuming; fast ramps, on the other hand, are generally employed to extract the liquid bio-oil(s) from the carbonaceous residues [Enaime et al., 2020]. No gradient ramp was used either.

Regarding the pyrolysis temperature, a very large range was explored, i.e. between 250 and 1000 °C [Park et al., 2021; Piccirillo et al., 2017b]; moreover, once the pyrolysis temperature was reached, different dwell times were considered, up to 4 h [Asgari et al., 2019]. Contrasting results were reported in the various studies. For Piccirillo et al., [2017b], for instance, the highest surface area was measured for fish bones pyrolysed at 800 °C; in other investigations [Alkurdi et al., 2020; Rojas-Mayorga et al., 2015a], on the contrary, the highest surface area corresponded to lower pyrolysis temperatures – 500 and 650 °C for sheep and bovine bones, respectively.

The optimisation of the pyrolysis of bovine bones was studied using the Taguchi L8 orthogonal array [Asgari et al., 2019]. A more complete and systematic study was performed by Cazetta et al. (2014) on bovine bones; in their investigation they employed the surface response methodology and considered the effects of pyrolysis temperature, time and N_2 flow rate on properties such as surface area, mesopore volume and average pore diameter. Results showed that a temperature of 450 °C, a time of 110 min and a N_2 flow rate of 100 cm^3/g were the most suitable conditions.

The investigations described until now were all performed in inert atmosphere (i.e. N_2, gas flow). In a couple of studies, however, a partially oxidative atmosphere was considered, as CO_2 was used as pyrolysis gas. Bedin et al., [2017] performed an optimisation study of the CO_2 pyrolysis conditions of bovine bone using a methodology similar to that done with N_2 (Cazetta et al., 2014; see above). Results showed that the best experimental conditions were 451 °C, a dwell time of 109 min and a CO_2 flow rate of 130 $cm^3\ min^{-1}$; this led to a slightly higher value of surface area than with N_2 – 137 vs. 128 m^2/g respectively. The improved performance of BC obtained in a CO_2 atmosphere was also confirmed in another study: Rojas-Mayorga et al., [2015b] showed that samples prepared with CO_2 exhibited higher fluoride ion removal in comparison to those prepared in N_2 – about 31% more. The authors do not give a clear explanation of this result; it is likely, however, that the partial oxidative atmosphere led to the presence of additional functional groups on the surface of the material, which favoured the interaction with the fluoride ion.

Overall these two studies confirm that the nature of the gas employed during the pyrolysis can have a significant effect on the properties and performance of the prepared BCs.

It has also to be considered that, according to the targeted pollutant to be adsorbed, the preparation conditions to obtain the best performance may be different. Alkurdi et al. for instance, reported that pyrolysis at 900 °C of sheep bones led to the highest arsenic removal; for fluoride ions, on the contrary, the best performing material was that prepared at 650 °C [Alkurdi et al., 2020]. The authors explained these results considering the different mechanisms involved in the removal of the cations and anions. Similarly, Piccirillo et al., [2017b] reported that cod fish-derived BC showed the highest Pb (II) removal when pyrolysed at 600 °C, while the adsorption of persistent pollutants such as fluoxetine and diclofenac was maximised with BC prepared at 1000 °C. Also in this case, this can be explained considering the different mechanisms of adsorption of the different species. In fact, as heavy metals are removed mainly by the CaP, XRD data of the BC showed that for $T > 600$ °C HA initially present in the bones was converted into a different CaP phase (oxyapatite), less effective for ion exchange. The organic pollutants, on the other hand, were adsorbed by graphite, which was more effective when prepared at higher temperatures. Reduced adsorption due to oxyapatite formation was also reported by Rojas-Mayorga [2013] for fluoride ions.

It has to be highlighted that, although the removal of heavy metals is performed by HA present in BC, the presence of graphite also plays a role. Comparisons were in fact performed of Pb (II) removal by cod fish bones simply calcined or pyrolysed at the same temperatures [Piccirillo et al., 2017b], and the results indicated that the pyrolysed material was much more effective. This can be due to the higher surface areas BCs generally have [Medellín-Castillo et al., 2014], which can favour the adsorption; moreover, graphitic carbon can also remove some heavy metals, even if less effectively than HA – such synergistic effects result in higher efficiency. It is likely that the same effect would be seen for the removal of other ions; however, no direct comparison was performed, and no literature data are available on this.

In the majority of the studies reported here, BC is always employed in wastewaters; only one investigation addresses BC use for adsorption of gaseous volatile compounds [Pongkua et al., 2018]. In this work pork BC is compared to biochars of vegetable origins (i.e. plants or their residues) for methyl tert-buthyl ether removal; results showed that BC was the worst performing material. However, it has to be considered that, in this specific case, the adsorption took place through the interactions between carbonyl groups present on the surface of the materials and the C–O ester bonds of the pollutant; pork BC was shown to have fewer carbonyl groups compared to other biochars, resulting in lower adsorption. Therefore, the potential of BC for gaseous pollutants should be assessed with different kinds of molecules.

Although the majority of the experiments were performed in batch conditions, a few column set-up studies were also performed. A comparison between batch and column experiments was performed by Park et al., [2015] with BC from chicken bones for the simultaneous removal of different heavy metals (Cu(II), Zn(II) and Cd(II)); results showed that the adsorption order was the same for both types of experiments, i.e. Cu > Cd > Zn, indicating that the adsorption mechanisms do not change with the experimental set-up. Moreover, with the column experiments (performed with a PVC cylinder, 25 mm diameter) the efficiency of the adsorption was higher than for the batch ones. Another investigation [Hernández-Hernández, 2017] showed that the column configuration can also be determinant; the authors, in fact, used commercial BC to remove heavy metals in a reverse stratified column, i.e. column with larger particles packed at the bottom of the column and the smaller ones at the top. Results showed that the reverse packing had higher performance than the conventionally stratified columns for Cu(II) and Zn(II) adsorption.

Column experiments were also performed for the removal of organic pollutants, more specifically anionic dyes [Reynel-Ávila, 2016] and the anti-inflammatory drug naproxen [Reynel-Ávila, 2015]. In both cases the efficiency of the adsorption was lower for the column set-up (fixed packed bed columns), the opposite of that observed for the heavy metals removal [Park et al., 2015]; indeed, a very low adsorption was registered for naproxen while results were satisfactory for the anionic dyes. These data confirm the differences between the different classes of pollutants; they also indicate that a batch set-up is more suitable for the removal of organic contaminants.

For all adsorbent materials, their reuse is a key parameter to assess their potential technological applications; ideally, in fact, the adsorption of the pollutants should be reversible, to regenerate the material for use in subsequent cycles. The study from Piccirillo [2017b] with BC from cod fish bones showed that a small fraction of the adsorbed pollutants is released when the material is simply placed in water; indeed, the registered values were 12–15% for organic contaminants and only 2–4% for Pb (II). These data confirm the formation of strong interactions between the BC and the pollutants, especially heavy metals; in these conditions, BC cannot be reused for repeated adsorption processes. Therefore, BC regeneration is essential to address this problem. Literature reports of high temperature treatment to regenerate commercial BC used to adsorb fluoride ions [Nigri et al., 2017]; results showed that, although the adsorption capacity was not fully restored, BC treated at 400 °C kept almost 60% of the original adsorption capacity. Better results were achieved when chemical regeneration was employed. Moussavi et al., [2022] performed a NaOH treatment to regenerate BC derived from calf bones and used to adsorb humic acid; their results show that BC materials retain almost completely the adsorption performance. Overall, it can be stated that more systematic investigations are necessary to understand the regeneration process, and to choose the most suitable conditions leading to a reuse of the materials.

2.2. *Modified BC*

2.2.1. *Activated BC*

To improve the characteristics, and hence the performance, of carbon-based materials of natural origin (biochars), different treatments can be performed after the carbonisation process. This is normally referred to as activation. The objective is to increase the surface area, which can lead to enhanced performance; moreover, the size of the pores and their distribution will also change with the activation.

The activation can be performed either physically or chemically. In the most common physical process, the pyrolysed powder is treated at high temperatures (>700 °C) with gases, the most common one being steam, although CO_2 is also used. These penetrate into the pores of the material and remove residual molecules/contaminants, hence increasing the surface area. Moreover, the interaction between the gas and the surface of the powder may lead to the presence of some additional functional groups such as COOH and OH on the surface itself; this is because both steam and CO_2 behave as oxidative agents [Enaime et al., 2020]. These groups can affect the functional properties of the materials. On the other hand, considering chemical activation, this is performed by treating the pyrolysed material with suitable chemicals (either acids or bases) and successively being washed; as stated for the physical activation, a double action of pore cleaning and surface functionalisation is achieved [Medeiros et al., 2022]. Literature shows chemical activation being more effective than the physical one in terms of increased surface area and enhanced functionality; however, its impact on the environment and cost are generally higher, due to the use of chemicals [Khan et al., 2022].

Considering in particular the activation of BC, the most significant studies in this field are reported in Table 2. No work was performed on materials activated with the traditional physical processes described above. Xiao et al., [2020], however, performed a systematic investigation on the effect of ball milling on BC derived from cow bones. Results show the method being effective, as a significant increase in surface area is observed, with the simultaneous decrease in pore dimensions. In addition to this, XPS analysis also showed changes in the surface composition; indeed, the proportion between different functional groups (i.e. P═O/C═O, C–O/P–O) was affected by the ball milling treatments; a significant increase in surface area and porosity was also registered (from 52 to 313 and 24–193 m^2/g respectively). Enhanced performance in heavy metals adsorption was also observed.

Regarding chemical activation, a complete study on BC was made by Iriarte-Velasco [2014]; the authors used different activating agents, both acid and basic – H_3PO_4, H_2SO_4, NaOH and K_2CO_3. In their protocol, they pyrolysed pork bones at 450 °C; then chemically treated the powder which was successively pyrolysed again at 800 °C. Results showed that the characteristics of the activated materials changed considerably according to the chemical used; with H_2SO_4, in fact, a significant increase in surface area and decrease in pore volume was observed. With H_3PO_4, on the other hand, the opposite behaviour was observed; these data can be due to an interaction of the acid with the phosphate already present in the BC, leading to its partial dissolution, and hence much reduced porosity. This work shows two key points regarding BC use: 1) the peculiarity of BC compared to other carbonaceous materials, due to the HA presence; 2) the importance of selecting the most appropriate experimental conditions in BC preparation/activation, to tailor the properties for specific applications. In a successive work from the same research group, the authors compared the performance of the different

Table 2
Modified bone char materials used for pollutants adsorption from contaminated waters.

Tested Pollutant(s)	Highest surface area (m^2/g)	Pyrolysis conditions and other details of the study	Highest adsorption capacity (mg/g)	Reference
Chemically activated bone chars				
Formaldehyde	118	Cattle and sheep bones, acid activation. 450 °C, 4.5 hs.	291.9	Rezaee et al., 2013
Methylene blue	139	Pork bones, acid activation. N_2 atmosphere, 450 °C, 10 °C/min, followed by further treatment at 800 °C.	1[a]	Iriarte-Velasco et al., 2016
Cd(II)	173	Fish bones, H_2O_2 activation. N_2 atmosphere, 300–900 °C.	228.7	Guo et al., 2022
Toluene	2312	Bovine bones, acid and basic activation, 450 °C, 4hs.	13[a]	Yang et al., 2020
Physically activated bone chars				
Cd(II), Cu(II), Pb(II)	313	Bovine bones, N_2 atmosphere, 300–600 °C, high energy ball milling	558.9 for Pb(II)	Xiao et al., 2020
Nitrogen-modified bone chars				
Cu(II), Pb(II)	–	Commercial material, amino functionalisation	120 for Pb(II)	Liu et al., 2020
Cr(VI), U(VI), methylene blue	470	Pig bones, N_2 atmosphere, 350–650 °C, ball milling with ammonium hydroxide	221.8 for Cr(VI)	Yang et al., 2022
Magnetic bone chars				
Pb(II), Cd(II), Co(II)	162	Camel bones, Fe_3O_4 NPs. N_2 atmosphere, 500 °C.	344.8 for Pb(II)	Alqadami et al., 2018
Rhodamine B, tetracycline	328	Chicken bones, Fe_3O_4 NPs. Thermal activation at 100 °C followed by pyrolysis in O_2-limited atmosphere,. 500 °C.	63.3 for tetracycline	Oladipo et al., 2017
Antibacterial bone chars				
F^-	77	Commercial (Carbones Mexicanos), modification with Ag		Delgadillo-Velasco et al., 2017
Other modifications				
Hg	11	Cattle bones, modification with Au. 450 °C, 4hs.	0.6	Assari et al., 2015
As (V)	4.6	Commercial, modification with Mn	14.5	Liu et al., 2016

[a] Value expressed in mmol/g.

acid-activated BCs for the removal of methylene blue; they show that H_2SO_4 activation corresponds to the BC with the highest adsorption capacity [Iriarte-Velasco et al., 2016].

Acid activation (with acetic acid) was also used by Rezaee [2013] to activate BC derived from either cattle or sheep; the activated materials were tested for formaldehyde adsorption in column experiments. Indeed, the activated materials showed both higher adsorption capacity and longer equilibrium times (95 vs. 75 min, respectively). Yang et al., [2020], on the other hand, performed a comparison between acid and basic activation, in which BC derived from bovine bones was activated with either K_2CO_3 or H_3PO_4. Results confirmed that H_3PO_4 is not a good activating agent to increase surface area and enhance the adsorption performance; K_2CO_3, on the other hand, led to an almost 9-fold increase in toluene adsorption compared to the untreated BC. In this study, the authors also showed that the activated BC can be thermally regenerated; in this case, however, thermal regeneration was successful due to the high volatility of the adsorbed pollutant – toluene.

A different approach was used by Guo [2022], which employed H_2O_2 as the activating agent, which was performed prior to the pyrolysis process. Results showed that the treatment led to a decrease of the organic matter, as well as to an increase of the surface area.

2.2.2. *Chemically modified BC*

Literature reports on chemically modified BC to improve their functional properties. In fact, as already stated in the previous section, chemical modifications can lead to the presence of functional groups on the surface of the BCs, which can enhance their performance. This is because interactions can be established between such functional groups and the targeted species.

With this objective, N-modified BCs were prepared. Yang et al., [2022] reported of BC prepared from pig bones and successively treated with ball milling using ammonium hydroxide. The treatment led to an increase in surface area; this was greater in comparison to the BC ball milled with simple deionised water (470 vs. 313 m^2/g, respectively). A decrease in the pore dimensions and increase in the nitrogen content on the surface of the powder was also registered. The materials exhibited high adsorption capacity for both metals and organic pollutants; more specifically, Cr(VI), U(VI) and methylene blue were tested. This study confirms the importance of particular functional groups on the BC surface to improve its functional properties; in addition, they also showed the potential of BC in removing different types of pollutants, in a more effective way than other adsorbent materials.

Amino functionalisation was performed by Liu et al., [2020]; the BC powder, derived from pig bones, was covered with a layer of SiO_2 functionalised with an amino group. A condensation process was employed to prepare this material. The modified BC was tested for the adsorption of Pb (II) and Cu (II); results showed a significant increase in the removal of both metals, due to the interactions between the charged ions mentioned before (i.e. Pb(II) and Cu(II)) and the $–NH_2$ groups. Moreover, modified BC could be easily regenerated by washing it with acid; the amino groups, in fact, were protonated, hence the metal ions were released from the powder, due to electrostatic repulsion.

Modifications with other elements were also studied to enhance BC performance. Assari et al., [2015] modified BC prepared from cattle bones with Au nanoparticles; the modified powders were employed to adsorb mercury in the vapour phase. It was seen that, optimising Au content, higher Hg (0) adsorption was achieved – more than 40-fold increase was registered for column experiments.

In another work by Liu [2016], commercial BC was coated with a manganese layer; different concentrations were used and the effect on the adsorption of As(V) was assessed. Results showed that Mn presence corresponded to an increase in As(V) adsorption, both in batch and column experiments (glass columns with 2.1 cm diameter and 22 cm length) – a 75-fold increase in the adsorption capacity and a 5-fold increase in the retardation time, respectively, for a Mn concentration of about 47%. Although the Mn presence led to an increase in surface area, it is likely that the improved adsorption was due to a different mechanism; in fact, in desorption experiments only 0.6% of As(V) was released from the Mn-modified BC, vs. about 30% for the unmodified material. These data highlight the importance of considering all the aspects of BC chemical modifications, i.e. although some properties may improve, others could get worse. Because of this, it is essential to investigate all the different features when a modified material is developed.

Chemical modifications were also performed to impart additional properties and make multifunctional BCs. This was achieved by adding other element(s) or other phase(s), which have the desired property.

Literature reports of magnetic BC for pollutants removal; a magnetic material, in fact, could be easily separated from the treated waters, for regeneration and successive reuse. Alqadami et al., [2018] prepared magnetic BC using camel bones; a co-precipitation process was performed to add Fe_3O_4 to the BC powder. The authors showed that the BC amount employed in the co-precipitation had a significant effect on the characteristics of the magnetic material, with the best modified BC obtained with a relatively small amount of BC powder in the reaction vessel. Indeed, the material showed high surface area (162 m^2/g), as well as good magnetic properties (saturation magnetisation of 50.20 emu/g). The magnetic bone char was tested for the removal of heavy metals such as Pb(II), Cd(II) and Co(II) and showed good performance, demonstrating that the presence of another phase does not necessarily compromise the adsorption properties.

Another study was performed by Oladipo et al., [2017], which employed chicken bones and functionalised them using a protocol similar to that described above; compared to the other study, the material presented a higher surface area and magnetisation – 328 m^2/g and 66.5 emu/g, respectively. This modified BC was employed for the adsorption of organic pollutants, such as rhodamine-B and tetracycline.

BC modification with silver was also studied, to prepare an antibacterial material, due to the antibacterial properties of Ag. The contamination of waters with many bacterial strains is in fact a known problem, posing risks to the environment and health, and which needs to be addressed [Thompson et al., 2013]. In the work of Delgadillo-Velasco et al., [2017], commercial BC was modified with Ag using different sources (i.e. colloidal Ag and two commercial products); the adsorption capacity of fluoride ion was assessed, as well as the antibacterial activity towards a Gram-negative strain – *Escherichia coli*. It was seen that the Ag presence in the BC led to an increase in the fluoride ion removal. SEM analysis did not show any significant change in sample morphology due to the Ag presence; moreover, the values of the surface areas were not affected either. Based on this, it is likely that the enhanced performance was due to an interaction between the noble metal and the anions, similar to those reported above for Au and Mn. The Ag-modified materials also showed a significant antibacterial effect. This study shows the potential of BC as a complete a multifunctional material for environmental remediation, for both pollutants and resistant bacterial strains. Indeed, this line of research should be further explored.

3. BC for sustainable and efficient agriculture

3.1. Removal/immobilisation of pollutants

As already mentioned above, soil contamination is a very serious issue, which needs to be addressed; several studies in fact showed that hazardous chemicals in soil will eventually end up in the food chain [Hussain et al., 2022]. Moreover, the presence of contaminants can also have a detrimental effect on plants' growth; this can have implications on the efficiency, and hence sustainability, of some agricultural models. It is, therefore, important to develop materials which could reduce the concentration of pollutants in soil.

In this frame, studies report the use BC for environmental remediation of contaminated soils, similar to what was done for wastewaters. The majority of the studies were performed on heavy metals, due to HAp's immobilisation capacity. Indeed, BC was employed for the removal of different metals, including Pb (II), Cd (II), Zn (II) and Cr(VI), see Table 3 [Azeem, 2021; Aseem et al., 2022 and references therein; Liu et al., 2021].

For BC applications in agriculture, it is important to test not only its effectiveness on the contamination of the soil, but also any possible effect(s) on plants, both in terms of concentration of pollutants in the plants and their growth; indeed, several studies addressed this.

In the work of Mei et al. for instance [2022], the authors use BC from pork to immobilise different contaminants (Pb, Cu, Zn and Cd); they measured the decrease in the concentration of such metals in the soil, and also in plant-in-pot experiments. More specifically, they assessed the concentration in pea plants (*Pisus sativus*), as well as the mass of the plant when BC was applied. Results show that, in addition to lower heavy metals concentration, the use of BC also led to an increase in the plant growth, i.e. higher dry and weight mass (see Fig. 4). A similar study was performed on maize using BC from sheep bones employed on soils contaminated with Cd and Zn [Azeem et al., 2021b]; applying BC, a decrease in Cd and Zn concentration was measured, both in the soil and in the plant (roots and shoots). Moreover, a significant increase in maize roots and shoots length and weight was also registered. The same research group also employed BC derived from chicken bones [Azeem et al., 2021a] in a similar environment; results confirmed the lower heavy metal concentrations in the soil and in the plants and, accordingly, the improved maize growth.

Table 3
Bone char-based materials used for pollutants adsorption in soil.

Tested pollutant(s)	BC source	Details of the study	Reference
Cd (II), Zn (II)	Sheep bones	Test of metal immobilisation in mining-contaminated soil	Azeem, 2021
Cr (VI)	Cattle bones	BC used as substrate for Zero Valent Iron (ZVI)	Liu et al., 2020
Cd (II)	–	Analysis of Cd removal and P dissolution in different soils	Siebers, 2013
Cd (II)	Bones from pig, sheep and cattle	Combination with clay, test on the growth of pak choi plants	Li et al., 2018
Cd (II)	–	Analysis of Cd concentration in lettuce, what and potato	Siebers et al., 2014
Pb (II)	Bovine bones	Combination with FeS and PBS strain	Qu et al., 2022
Herbicides (hexazinone, metribuzin, quinclorac)	Commercial material	Analysis of herbicide concentrations in different soils	Mendes et al., 2019
Petroleum hydrocarbon	Bone meal	Analysis on frozen soil	Karppinen et al., 2017

For soil decontamination, investigations were also performed on the use of BC in combination with other materials, for instance clay [Li et al., 2018]; the composites were shown to be very effective in the immobilisation of Cd, as the concentration in both soil and above-ground *Brassica chinensis* was reduced significantly. However, no quantitative data were given on the effect on the growth of the plant.

Although the majority of studies focused on heavy metals removal, some work was also done with BC is used to remove organic contaminants, such as herbicides [Mendes et al., 2019] or petroleum hydrocarbons [Karppinen et al., 2017]. The promising results presented in these studies show the potential of BC in this field, which surely should be explored more.

3.2. BC as sustainable fertiliser

3.2.1. P solubilisation

In addition to the use of BC for soil decontamination, more recently BC was studied for a different application, that is a material which could be employed as fertilisers and which could improve the characteristics of soils, leading to enhanced growth of crops/plants. This application is due to its composition, as BC contains phosphorus, carbon and nitrogen (as a minor element), all essential elements for agriculture. Nitrogen is a key element in fertilisers; indeed urea is one of the most used ones. BC, however, was not used to release nitrogen, due to its relatively low concentration; moreover, especially for samples prepared at higher temperature, N is present in heterocyclic aromatic rings, hence not very soluble [Feng et al., 2018]. Regarding phosphorus, its importance as fertiliser is well known and, because of this, the global demand for this element has increased significantly in recent years. Several studies showed that, keeping the same level of consumption, P reserves will be depleted, with dramatic effects on worldwide food production [Elser, 2012]. It is, therefore, very important to recover P from all possible sources and use it in an effective manner; the conversion of bones into

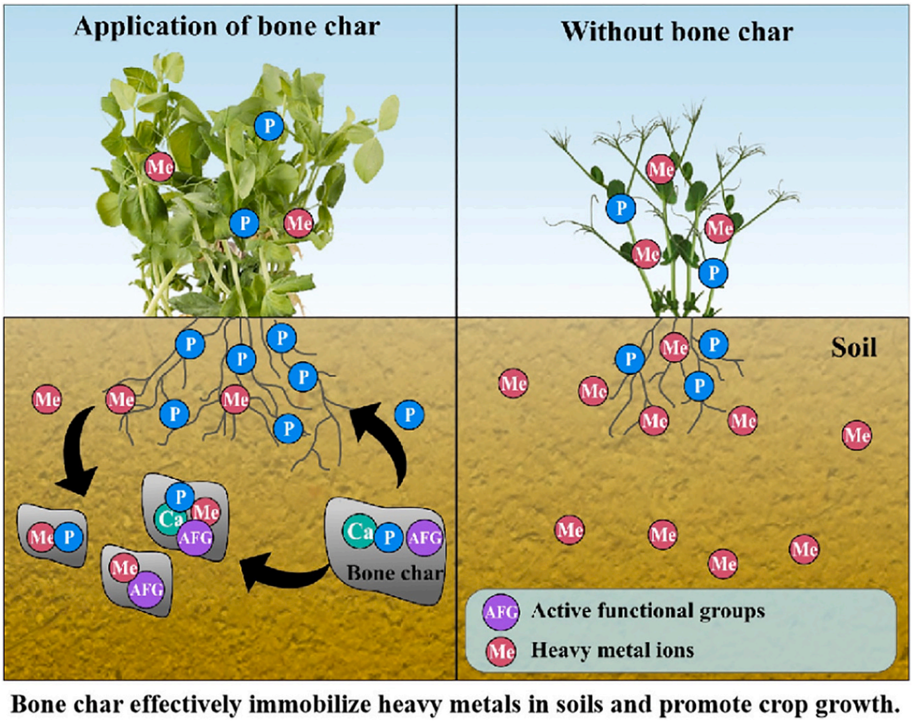

Fig. 4. Effect of BC addition to the soil to the growth pea plant [Mei et al., 2022].

BC and its employment as fertiliser to recover the phosphorus present is in line with this. In this section we report the studies performed in this field, i.e. the use of BC for sustainable agriculture.

As stated above, HAp or other form(s) of calcium phosphate(s) is/are the main BC component(s); hence P is one of the main elements. P content may vary according to the temperature of pyrolysis; with higher temperatures, in fact, the concentration of the carbon fraction decreases, leading to an increase in the CaP proportion and hence higher P content. Piccirillo et al., [2017b], for instance, measured P content from 9 to 14 % wt % for BC from cod fish prepared for temperatures between 200 and 1000 °C. Biswas et al., [2021], on the other hand, reported P concentration from 10 to 15% in BC from muntjac deer in the temperature interval 300–700 °C. These data confirm the potential of BC as a sustainable fertiliser. Its main limitation, however, is the very low HAp solubility [Dorozhkin, 2009], which may make the phosphorus not available, hence affecting BC efficiency as fertilisers.

Several studies investigated the correlation between the conditions of the pyrolysis and P solubilisation; indeed, it was shown that temperature had a significant effect. For BC derived from muntjac, for instance, it was seen that the highest P solubilisation was achieved for a pyrolysis temperature of 300 °C [Biswas et al., 2021]. Similar results were obtained by Ahmed et al., [2021]; they performed a systematic comparison between BC obtained from sheep, chicken and pig bones. Like for Biswas [2021], the highest concentration of soluble P was measured at 300 °C (BC from chicken bones); highest pyrolysis temperatures, on the other hand, converted the phosphorus into insoluble forms, not useful for fertiliser application. In these two studies, standard heating ramps were used to prepare the BCs (5 and 10 °C/min respectively); this parameter, therefore, did not have a significant effect. As already mentioned above, for pyrolysis at 300 °C almost no gases are released, as release takes place for $T > 400–500$ °C; therefore, it can be assumed that the increased surface area associated with the release of gaseous molecules does not have a positive impact on P solubilisation. The effect of the pyrolysis conditions was investigated by Dela Piccolla et al., [2021], which studied not just the effect of the temperature but also the atmosphere – in fact they compared the pyrolysis performed with no gas flow, with N_2 and with N_2 with an additional vapour activation. Results showed that with no gas was flow a higher content of soluble P was obtained at $T = 550$ °C. It has to be highlighted, however, that in both studies soluble P was a small fraction of the total P present in the different BC samples; indeed, maximum values of about 2–3% of soluble P were measured. These data confirm the difficulties for BC use as fertiliser.

Several studies were performed on P dissolution from BC in different kinds of soils, to test their effectiveness; results showed that the amount of available P varied significantly according to the characteristics of the soil. Warren et al., [2009] tested 12 soils with commercial BC from cattle bones; the soils varied significantly in terms of pH, P and C content, origin, etc. Results showed that higher P solubilisation from BC was obtained in acidic soils; in all cases, however, P availability was lower if compared to that of the traditional triple superphosphate fertiliser (TSP). For soils whose pH was below 6.1, however, BC performed better than Gasfa phosphate rock, also commonly employed as fertiliser.

The significant effect of the pH of the soil in P dissolution was also confirmed by Biswas [2021]. In another study, it was shown that the salinity level of the water employed to add BC also played a key role [Amin, 2021a]; experiments with BC from bovine bones were performed comparing the addition with distilled water and irrigation waters from wells, with different ions concentrations. Results showed that the presence of such ions had a positive effect on P solubilisation, as a more than two-fold increase was observed.

A hypothesis to explain the different P solubility with the pyrolysis temperature is the level of crystallinity of the HAp present in the BC. In a study performed on BC made from a mixture of bones, it was shown that for pyrolysis temperature up to 300–500 °C, available P was higher than for the untreated bone meal. For higher temperatures, however, a decrease in the solubilised P was observed. The authors correlated this to HAp crystallinity, identifying an optimal value, corresponding to the highest P solubilisation [Glaesner et al., 2019].

The studies reported above show the potential of BC as fertiliser, as well as the criticalities associated with them; however, they are all focused on the chemical aspects of the process, i.e. in which form(s) P is more soluble and/or which are the best experimental conditions favouring P solubilisation. To better understand BC's potential as a fertiliser, however, experiments on plants should be performed, to quantitatively assess the effect the solubilised phosphorus has on their growth.

Very few studies addressing this issue were published, and the results were not conclusive, as data showed BC to be an effective fertiliser in some cases, but not in others. Siebers et al., [2012] performed pots experiments using BC on wheat, potatoes and onions; results showed no dry mass increase for onions, a decrease in potatoes and an increase only for wheat. There may be several reasons for these contrasting results; indeed, different crops may respond differently to the same stimuli, and different fertilisation protocols may be required.

3.2.2. *BC as fertiliser and for soil amendment*

The use of BC as multifunctional material as both fertiliser and pollutants remover was also explored. Commercial BC was used in a cadmium-containing soil, both Cd immobilisation and P solubilisation were monitored [Siebers, 2013]. Results showed that BC was more effective than TSP to immobilise the heavy metal; P release in the soil, however, was higher for the traditional TSP. In a successive work by the same research group, the authors investigated the effect of the combined action (fertiliser/soil amender) on lettuce, wheat and potato in pot experiments [Siebers et al., 2014]; they tested soils with different initial P concentrations (sufficient and deficient) and Cd contamination (moderate and severe), monitoring the effect on the crops' growth (yield of the dry matter). Results showed that BC led to better plant growth than TSP and diammonium phosphate (DAP) in soil with high Cd contamination and sufficient P concentration; in other cases, on the other hand, both TSP and DAP were more effective than BC. This study is very interesting, as it shows the experimental conditions in which BC use is more effective than other traditional P-based fertilisers, to fully exploit its potential.

3.2.3. *BC combined with microorganisms*

The use of selected microorganisms with specific functionalities is another route widely explored in advanced and sustainable agriculture [Das et al., 2022]. A complete analysis of this topic is not within the scope of this work; in this section we discuss the studies involving BC in which microorganisms were also employed.

An interesting investigation was reported by Zwetsloot et al., [2016], which employed BC in combination with root hairs and with the inoculation of arbuscular mycorrhizae (AM); these are microorganisms generally present in the soil, living in symbiosis with the crops. This combination gave good results on maize experiments in pot, as both P accumulation in the plants and dry mass increase were comparable to that obtained with traditional TSP. This study shows a promising element for the full exploitation of BC as fertiliser, which surely is worth investigating more.

Another route worth exploring is BC use combined with Phosphorus Solubilising Bacteria (PSB). These are strains capable of solubilising phosphorus, through the production of organic acids; indeed PBS have been employed to solubilise HAp, as studies were performed focused on both solubilisation and effect on plant growth [Santana et al., 2019; Li et al., 2020]. Regarding the combination with BC, a composite made of carboxymethyl cellulose, BC and FeS was prepared and used as substrate for PBS immobilisation (*Enterobacter sp.*) [Qu et al., 2022]; such a composite was employed in lead-containing soil, for decontamination. Results showed that the use of the substrate led to an enhanced P solubilisation which, in turn, resulted in an increased lead removal; this was because solubilised phosphate ions interact with lead, to form insoluble lead phosphates. This study, although interesting, only considers the application for environmental remediation, and does not investigate the possible effects on plant growth induced by the higher P availability.

Other strains employed in agriculture are Sulphur Oxidising Bacteria (SOB); these are microorganisms which oxidise elemental sulphur into sulphates, leading to an acidification of the soil [Rezvana Boroujeni et al., 2021]. This, in turn, can increase the amount of soluble P and, hence, have a beneficial effect on crops' growth. Potentially SOB can be employed in combination with BC, as different studies showed that BCs also contain sulphur, in concentrations up to 1.5 mg/g [Wang et al., 2020; Zwetsloot et al., 2016]. In the work from Amin et al., [2021b], BC from bovine bones was tested with *Thiobacillus spp*. In solubilisation experiments in calcareous soils; results indicated a significant increase in the available P in comparison to the control samples. No data were shown assessing the actual effect on plant characteristics.

Overall, it can be stated that BC combined with microorganisms has great potential for enhanced P solubilisation and efficiency as sustainable fertilisers; this is a topic which should be investigated more, particularly in experiments with plants.

4. BC in composites with catalysts and/or photocatalysts

The use of efficient catalysts is crucial for many processes, including production of essential chemicals or their conversion into more valuable ones, and catalytic degradation of emerging pollutants. In many cases, the combination of different compounds with specific functionalities can lead to enhanced catalytic properties. Carbon-based materials, for instance, have been reported to improve the catalytic performances of other species; this feature was also observed for biochars. Indeed, in literature there are several reports on the use of biochar-containing catalysts [Saquing et al., 2016]; more specifically, biochars can be employed for redox reactions, as they can transfer electrons, thanks to some functional groups and/or electron-conducting structure present on their surface [Yu et al., 2015]. The functional groups include carbonyl and heteroatoms like B, N, S, and P, as well as redox-active metals such as Cu and Fe [Haoyu et al., 2022]; graphite particles on the surface of the materials, on the other hand, show some electron-conductive properties [Jiang et al., 2022].

Focusing in particular on BC, for its characteristics it is an ideal material for this kind of application; as mentioned above, BC contains graphitic carbon, and both functional groups and N heteroatoms derived from the pyrolysis of the protein. It also contains P as a major element, due to the HAp presence; moreover, in animal bones other ions like Cu and Fe are present, their concentration depending on the source of the bones (animal, its age, etc.) [Nasrollahzadeh et al., 2020]. Despite the potential, only a few studies were performed on BCs employed as catalysts, with most investigations focusing on the degradation of toxic molecules (see Table 4).

Asgari et al., [2013] reported on the catalytic degradation of ozone (O_3) by BC derived from cattle bones; both acid and basic pH conditions were tested, as it is known that OH^- ions promote O_3 degradation. It could be seen that BC presence led to a 6-fold increase in the degradation rate under both conditions; the active sites for the reaction were found to be the Ca–OH and P–OH groups. In agreement with this, it was seen that in basic conditions the efficiency was higher due to the additional –OH groups on BC surface. In another study, the same group investigated the degradation of humic acid by BC in the presence of O_3 by BC derived from cattle and sheep [Mortazavi et al., 2010]; also in this case, the best degradation was observed at basic pH. BC presence, however, results in enhanced performance in all conditions.

BC was employed in combination with green rust (GR) for the catalytic dechlorination of the chlorinated solvent trichloroethylene [Ai et al., 2020 and 2022]. The authors performed a complete a systematic study using BC derived from bone meal, to assess the best conditions for the solvent degradation; in these studies BC acted as a mediator for electron transfer between the Fe(II) and Fe(III) ions constituting the GR. Results showed that the efficiency of the system was affected by different parameters, the most significant being the pyrolysis temperature of the bone meal; indeed, the Electron Accepting Capacity (AEC) reached its maximum for pyrolysis at 850 °C and was about 4 times higher than for biochar of plant origin i.e. only carbonaceous, with no CaP phase. Other factor(s) affecting the catalyst performance were the GR/BC ratio and

Table 4
Bone char-based materials employed in catalytic and photocatalytic processes.

Reaction system studied	Surface area (m^2/g)	BC source and pyrolysis conditions	Performance of BC material	Reference
Ozone degradation	92	Cattle bones. N_2 atmosphere, 5 °C/min, 800 °C.	Degradation rate 0.07 min^{-1}.	Asgari et al., 2013
Humic acid degradation with ozone	122	Cattle/sheep bones. 600 °C, 4 hs.	1.6-fold increase due to BC presence	Mortazavi et al., 2010
Persulphate degradation of 2,4-dichlorophenol	1024	Pork bones. N_2 atmosphere, 450 °C followed by further treatment at 900 °C	Degradation rate 0.07 min^{-1}.	Zhou, 2020
Persulphate degradation of acetaminophen	1024	Pork bones. N_2 atmosphere, 450 °C followed by further treatment at 900 °C	Degradation rate 0.31 min^{-1}.	Zhou, 2020
Dechlorination of trichloroethylene by green rust	1200	Bone meal. N_2 atmosphere, 450–1050 °C.	Degradation rate 2 h^{-1}.	Ai et al., 2020
Dechlorination of trichloroethylene by green rust	1200	Bone meal. N_2 atmosphere, 150 °C/h, 950 °C.	Degradation rate 0.2 h^{-1}.	Ai et al., 2021
Dechlorination of trichloroethylene by green rust	–	Bone meal. 950 °C, 1 h.	2.4-fold increase due to BC presence.	Ai et al., 2022
Glucose isomerisation	98	Cattle bones. O_2-limited atmosphere, 1200 °C.	Conversion 27% selectivity 55%	Matsagar et al., 2018
Photocatalytic degradation of methylene blue with bone char/ZnO	100	Bone granules. 400 °C, 2 hs.	Degradation of 60%.	Jia et al., 2018b
Photocatalytic degradation of formaldehyde with bone char/ZnO	90	Cattle-sheep bones. 450 °C, 4.5 hs.	Degradation of 73%.	Rezaee et al., 2014

the species/functional groups present on the surface of the material. The higher efficiency of BC in comparison to traditional biochar was also confirmed in a successive study performed by the same authors [Ai et al., 2021].

Zhou et al. studied BC effect on the persulphate system (PS) for the degradation of 2,4- dichlorophenol [2020a], employing BC derived from pork bones and with a very high surface area (>1000 m^2/g). Experimental data showed the high efficiency of the combined BC/PS, as the molecule gets completely degraded in less than 3 h (while almost no degradation was observed by just PS or BC alone). Authors also investigated the mechanisms of the degradation reaction, showing that radical species ($\cdot OH$, $SO_4^{\cdot -}$, $O_2^{\cdot -}$) were involved in the reaction. The same research group also investigated the degradation of acetaminophen with the same BC/PS combination [Zhou et al., 2020a, b]; in this case, although radical formation was observed, non-radical electron-transfer mechanisms were dominant. The comparison of these two studies illustrates that BC materials can work with different paths according to the molecule to degrade, hence showing their potential and versatility.

BC was also employed as catalyst for the conversion of glucose into 5-hydroxymethlylfurfural [Matsagar et al., 2018]; cattle bones were employed to prepare BC, while a Brønsted acidic ionic liquid (BAIL) was used in the conversion reaction. Hydroxymethlylfurfural (HMF) is generally prepared from fructose, a more expensive and less available sugar. In this study BC catalysed first the reaction from glucose to fructose and successively, with BAIL, its conversion to HMF. Both the yield and the selectivity changed according to the solvent used, the best performing one being water; this added a sustainability element to the whole process, considering the non-toxicity of water in comparison with other solvents (i.e. DMF, DMSO, etc.). Authors also showed that BC was stable in the reaction conditions and could be successfully reused; this is a key point when assessing the performance of a catalyst.

The studies described here show that BC is very suitable to be employed as a catalyst, in combination with other species/chemicals; to fully exploit its potential, more investigations are necessary, both to assess its activity for other chemical reactions, and to have a better understanding of the reaction mechanism(s), with possible preferential path(s).

Photocatalysts are another class of catalysts widely employed for environmental remediation (degradation of pollutants) [Ikrari et al., 2022] and hydrogen generation (i.e. use of alternative fuels) [Lijarani et al., 2022]. Literature reports that combining photocatalysts with graphitic carbon can have a beneficial effect on the photocatalytic activity [Yue et al., 2009]; indeed, several combinations with biochars were also studied [Fito et al., 2022]. To the best of our knowledge, however, only two studies were reported on photocatalysts combined with BCs. More specifically, BC derived from either bone granules or cattle-sheep bones was combined with ZnO to degrade methylene blue and formaldehyde, respectively [Jia et al., 2018b; Rezaee et al., 2014]. For methylene blue degradation, the composite was obtained by co-precipitating ZnO on a BC suspension; photodegradation experiments showed a significant effect of the pH of the solution. In the work from Rezaee, on the other hand, ZnO nanoparticles were mixed with BC powder; a comparison with simple ZnO showed an enhanced performance due to the BC presence. In both cases, a significant element is the high surface area of the composites, which favours the pollutants' adsorption and their successive degradation; this feature was already observed for HAp-based photocatalysts [Piccirillo, 2017a], and it plays an even major role here.

Overall, these composite materials did not show excellent performance; it has to be highlighted, however, that HAp combined with other photocatalysts, especially TiO_2, showed very high activity [Piccirillo, 2017a]. Considering this, it is worth investigating BC combined with other photocatalytic phases.

5. BC as material for electrochemical processes

As already stated above, carbon-based materials are widely used to prepare materials employed in electrochemistry; indeed, literature reports numerous studies in this field, including the use of natural sources/wastes to fabricate electrodes or capacitors [Chen et al., 2020]. Some investigations were also published on the use of bone-derived materials for these applications [Niu et al., 2019]. It has to be highlighted, however, that in the majority of cases, after the bones' pyrolysis, the BC formed underwent an acid treatment whose objective was to remove the calcium phosphate phase present in the material [Ai et al., 2017; Cazetta et al., 2018]. Although some of these materials show interesting performance, without the calcium phosphate they cannot be really defined as BCs; indeed, they are more similar to other plant-derived biochars, consisting mainly of graphitic carbon. The presence of the bones in the original material can impart some characteristics to the materials, such as the porosity or the 3D structure [Shan et al., 2018]; from chemical point of view, however, the materials cannot be classified as BCs anymore. In addition to this, it has to be highlighted that such strong acid treatment makes the production of material a less green process, with a higher impact on the environment, and hence less sustainable.

Some studies involving BC use (i.e. with no acid HAp dissolution) were published; Deng et al., [2021], for instance, reported on pig-bones derived BC used for electrocatalytic hydrogen evolution reaction. The prepared BC is employed to make an electrode; results showed that the pyrolysis temperature had a significant effect. Moreover, N presence in the BC was also a key element. In another investigation, BC derived from devil fish bones is used as an electrode material in combination with microorganisms, i.e. in microbial fuel cells [Flores et al., 2021]; the system was used to biodegrade carbamazepine, a pharmaceutical/persistent pollutant. Although a good carbamazepine degradation was measured (>75%), the system showed worse performance in comparison to other materials previously studied, which were not derived from animals.

On the whole, it could be stated that for this specific application BCs do not seem the most promising materials, as the presence of the inorganic calcium phosphate phase does not add enhanced functionalities and/or lead to improved properties. A possible reason for this unsatisfactory performance is the lower electrical conductivity of BC in comparison to traditional biochar– a value of about 160 μs cm^{-1} was measured for BC from bovine bones, vs. > 7500 μs cm^{-1} for plant-derived biochars [Paul, 2022].

6. Environmental impact and sustainability

For materials derived from wastes to be worth being used in technological applications from a sustainability point of view, their impact on the environment should be assessed. Such impact, in fact, should be as low as possible, and definitively lower than that of synthetic materials (i.e. prepared with chemical reactions, not derived from wastes) employed for the same function. To assess this, studies such as Life Cycle Assessment (LCA) of the production process are generally performed. Literature reports several investigations regarding pyrolysis processes to obtain biochars [Gaur et al., 2022]; considering in particular food industry residues as biochar sources, studies showed that the impact can be affected by several parameters, the main one being the nature of the wastes, but also the pyrolysis equipment employed and its heating ramps and temperatures [Zhu et al., 2022]. These last two factors, in particular, can have an effect also on the production costs and on the greenhouse gas emissions. Overall, however, such residues resulted to have a good carbon reduction potential.

Very few studies were performed specifically on BCs. Yami et al., [2015] investigated the LCA of several materials for arsenic removal from waters, including BC; the results indicated that BC had the lowest impact on the environment if compared to other materials such as alumina or wood-derived biochar. One of the reasons for the lower impact was BC's high efficiency in As adsorption, as less material is required to remove the same amount of pollutant.

Ramanan et al., [2022] performed a more detailed study on BC LCA for the same application (i.e. As removal). They assessed the emissions of both greenhouse gases and other possible polluting gases (for instance CO and SO_2) associated with each step of the production; their results indicated that one key element to assess the impact and sustainability is the transport of the bones to the industrial facility. This is indeed a point which should always be taken into account when waste valorisation processes are considered. In this study, the authors also analysed the effective cost for the BC production, in relation to its retail price. Although this study explored many aspects of the topic, no comparison was performed with other materials commonly used to adsorb As, which would allow one to understand if BC use more sustainable is or not than other synthetic materials.

Such comparison, on the other hand, is reported in the study by Xiong et al., [2022]; they investigated the use of a composite of ferrous hydroxide and BC to dechlorinate chlorinated ethylene. In addition to the study of the efficiency of the dichlorination of the composites, the authors also assessed their LCA in comparison to standard iron-based materials; results showed that the BC-based compounds were much more sustainable than the traditional ones.

An investigation more oriented towards the estimation of greenhouse gases emissions was performed by Cascarosa et al., [2013]; the authors, in fact, assess the potential reductions in such emissions associated with the pyrolysis of meat bone meal. Results showed that a significant decrease in greenhouse emissions was achieved (about 600–1000 kg CO_2-eq per ton of treated waste). This study is indeed very interesting, and it shows the potential benefit to the environment linked to the use of BC; however, it has to be highlighted that the main focus of the investigation was the production of bio-oil, to be used as fuels. Therefore, most of the savings in CO_2 emissions were due to the reduced use of fossil fuels, and not to the (various) technological uses of BC.

These studies show the different features to consider when assessing the sustainability of BC-based materials; it is a topic which should be investigated much more, ideally taking into account the specific characteristics of each application.

7. Summary and conclusions

In the present review the most significant studies on the preparation and use of BCs were reported; it can be seen that there are different fields of application, from environmental remediation to sustainable agriculture and catalysis. The research performed on these materials confirms BCs' potential; in the majority of cases their enhanced performances are due to their composition. The presence of both hydroxyapatite and graphitic carbon, in fact, gives BC additional functionalities and improved properties in comparison to the standard plant-derived biochar; moreover, BCs often have a porous and 3D-ordered structure, derived from the bones used to prepare them, as well as high surface areas – these elements also play a role.

Considering more specifically the possible different applications, despite the potential already shown in the studies, additional features should be investigated more in detail.

For BCs' use in environmental remediation, for instance, one more possible application worth considering, and almost not investigated at all, is the simultaneous removal of different kind of pollutants, organics and heavy metals. Due to its composition, in fact, BC is the ideal material for this; the preparation conditions, however, should be tailored and adjusted for this double functionality. The optimised material would fully exploit the BC's unique potential.

For the applications in agriculture, on the other hand, BCs use together with appropriate microorganisms, either P- or S-solubilising bacteria, has not been fully explored; their use could significantly enhance P availability in the soil and, hence, show comparable performance to the traditional (non-renewable) fertilisers. For BCs to be used as photocatalysts, the combination with more efficient materials (i.e. TiO_2) should be considered.

Overall, BCs are a promising and sustainable class of materials, whose use is in line with the principles of circular economy, and which should be studied further, to better understand them, widen their potential applications and assess their sustainability.

Declaration of competing interest

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

Data availability

No data was used for the research described in the article.

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