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Review Article

Amalia Lara Bursztyn Fuentes, Claire Dazon, Alain Celzard, and Vanessa Fierro*

Non-conventional approaches to producing biochars for environmental and energy applications

https://doi.org/10.1515/rams-2025-0116 received September 27, 2024; accepted May 16, 2025

Abstract: As demand for biochar and its suitability for a wide range of applications increases worldwide, biochar production is subject to continuous optimization. However, unlike the cutting-edge technologies in specialized industries, old-fashioned practices with low-cost devices are the main techniques used for biochar production in developing countries and other vulnerable contexts. Until recently, these traditional biochar syntheses had not received much academic attention, as they were considered inefficient, non-reproducible, and polluting. However, recent studies have demonstrated that unconventional pyrolysis techniques, including artisanal plants, microwave-assisted pyrolysis, and solarassisted pyrolysis, can be optimized to reduce emissions and environmental impacts and can produce materials with yields and performance similar to conventional and commercial materials in several environmental and energy applications, which is of particular interest for developing countries. This review compiles the latest advances in sustainable biochar production with unconventional pyrolysis approaches and highlights some environmental and energy applications.

Keywords: sustainable biochar production, non-conventional techniques, environment and energy

Claire Dazon: Universidade Federal dos Vales de Jequitinhonha e Mucuri. Minas Gerais. Brazil

Alain Celzard: Université de Lorraine, CNRS, IJL, F-88000, Epinal, France; Institut Universitaire de France (IUF), F-75231, Paris, France

1 Introduction

Porous carbonaceous materials – namely activated carbon. charcoal, and biochar - are interesting materials that have long been the subject of research. Currently, biochar has regained importance in the academic field due to its versatility in a variety of novel technological applications [1–3] and, at the same time, it is becoming a prime material for the bioeconomy linked to carbon capture, among others [4,5]. Comprehensive reviews and papers are constantly being published, showing updates on the synthesis and characterization of biochar and porous carbon at laboratory and industrial scale, in several fields [6-9]. A quick search on the Scopus database clearly evidences that biochar and carbonaceous materials in general have been a hot topic since 2010. With the keyword "biochar" alone, just 127 documents were reported in 2010, whereas this number reached almost 30,000 documents in December 2023 (Figure 1a).

It is also interesting to analyze the fields that deal with biochar-related topics (Figure 1b). In 2010, half of the studies were oriented toward environmental sciences, followed by agricultural and biological sciences. After 7 years, biochar is addressed in three times as many fields (grouped in the "others" category), even if the aforementioned areas remain the most reported fields. A more detailed bibliometric survey carried out between 2006 and 2019 based on the Web of Science Core Collection database highlights the growing interest in biochar world-wide [10].

Biochar materials are now well established in scientific studies and in multidisciplinary areas, and the trend is unlikely to abate in the coming years, given the amazing applications they can have in the environment and energy fields. More than 50 uses for biochar have been identified, as shown in part in Figure 1b, and demand for and interest in biochar is growing rapidly worldwide [11–13]. In addition to its historical interest in agriculture and energy, biochar is seen as a potential solution for climate change

^{*} Corresponding author: Vanessa Fierro, Université de Lorraine, CNRS, IJL, F-88000, Epinal, France, e-mail: vanessa.fierro@univ-lorraine.fr

Amalia Lara Bursztyn Fuentes: Instituto de Ciencias Polares, Ambiente y Recursos Naturales (ICPA), Universidad Nacional de Tierra del Fuego, Ushuaia (9410), Tierra del Fuego, Argentina; Centro Austral de Investigaciones Científicas, Consejo Nacional de Investigaciones
Científicas y Técnicas (CADIC-CONICET), Ushuaia (9410), Tierra del Fuego, Argentina

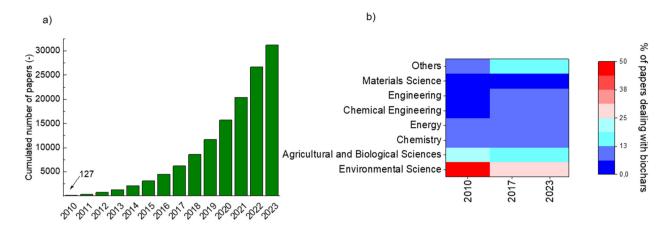


Figure 1: (a) Cumulated number of papers published over 13 years containing the term "biochars" in the title, abstract, and/or keywords (source Scopus, surveyed to December 2023). (b) Scientific fields dealing with the subject of biochar in 2010, 2017, and 2023. "Others" include social sciences, medicine, earth and planetary sciences, and multidisciplinary articles.

mitigation [14,15]. Many institutions around the world have proposed initiatives for the study of biochar, such as the International Biochar Initiative (www.biochar-international.org) and the European Biochar Research Network (http://cost.european-biochar.org). These are newly created international networks aimed at coordinating research and cooperation activities in the field of biochar and, additionally, at articulating biochar producers, consumers, and policymakers with environmental and ethical standards. Indeed, the European Biochar Certificate has been developed to become a voluntary standard in Europe [15]. North America [16], Africa, South America, and Asia [17,18] have also launched several programs and studies to enable lowincome populations to access the biochar economy. In this sense, many NGOs and field experts have focused their activities on the production of biochar for heating and cooking purposes and, more recently, for water treatment.

The development of sustainable, accessible, and lowcost biochar production methods is a key and challenging aspect. Biochar can be produced using a variety of technologies adapted to different scales, most of them based on pyrolysis approaches. However, cutting-edge technologies remain beyond the reach of developing countries, which need low-cost methods, and traditional pyrolysis practices are preserved as part of a cultural heritage. While some traditional techniques are inefficient and produce harmful substances that affect both human health and the environment [19,20], others have been judiciously optimized to achieve local objectives with improved efficiencies and reduced emissions [21]. Today, academia and industry are working on original and smart non-conventional pyrolytic devices to produce biochar for various applications, promoting environment-friendly processes, some of them

inspired by ancestral approaches. What are these non-conventional pyrolysis methods? Are they cost-effective, and do they produce efficient and eco-friendly materials? This review compiles the latest advances on this subject, compares some of the available results with those obtained with conventional techniques, and explores potential applications and gaps in knowledge. The novelty of this work lies in the fact that it highlights techniques and traditional practices that have been long used to produce biochar outside the academy but are widely used in many vulnerable contexts and analyzes and compares them in the light of more modern, cutting-edge technologies.

In this review, the term 'non-conventional' refers to new pyrolysis techniques that are emerging in academia (i.e., solar-assisted and microwave-assisted pyrolysis), but also to non-mainstream techniques, i.e., devices and processes that are becoming the focus of academia, despite having been used for decades, mainly in developing countries (i.e., ancient, artisanal, and traditional practices). In contrast, the term 'conventional' refers to well-established biochar production techniques, including temperature-controlled reactors.

2 Biochar: Overview of definition and chemical process

Biochar is the term used to define "the solid product remaining after biomass has been heated to temperatures typically between 300 and 700°C under oxygen-deprived conditions, a process known as pyrolysis" [15]. Unlike

biomass, the carbon in biochar material is in aromatic form, giving it a number of interesting properties. Thermochemical technologies for converting biomass into energy or chemical compounds include combustion, pyrolysis, liquefaction, gasification, torrefaction, and hydrothermal carbonization [22]. Generally speaking, during these processes, lignin, cellulose, hemicellulose, starch, and other biomolecules in the feedstock are thermally treated. leading to three main products: char or biochar (solid fraction), oils or bio-oil (partially condensed volatile compounds), and syngas or non-condensable gases [23]. While biochar is the main product of pyrolysis, with a typical yield of 35%, the gaseous fraction accounts in most cases for 70-80% of the co-products [24].

Bio-oil can be used as an energy source or as a precursor for further refining. Similarly, due to incomplete combustion during the thermochemical conversion of biomass, syngas can be produced, which is a mixture of non-condensable gases such as CO, CO2, CH4, and H2. The composition of this volatile fraction is complex, and reforming or conversion into fuels is not very efficient with current technologies. The gaseous fraction is then commonly used directly as a source to generate heat in boilers, electricity in turbines, or, with some adjustments for specific applications, hydrocarbons via Fischer-Tropsch processes and ethanol via biological conversion [25]. Figure 2 summarizes the general processes involved in obtaining biochar and its useful by-products. Finally, the main solid product of pyrolysis is a porous, carbon-rich solid with distinct chemical and physical properties [13]. To generalize our analysis of the biochar literature as much as possible, we group together under the term biochar all the porous, carbonaceous materials derived from pyrolysis described above, irrespective of feedstock, carbon content, or application, and the term activated carbon (AC) will be used for carbonaceous materials that have undergone further activation.

As previously mentioned, biochars can be manufactured from different feedstocks, the latter being one of the main parameters influencing the final properties of the biochar [27–29]. With regard to the type of feedstock, industrial waste and by-products, forestry, and agricultural residues, municipal solid wastes and numerous non-conventional materials have been studied [30,31]. Several organic residues have been used to produce biochar, including bamboo [32], grape seeds [33], coconut shells [34], corn residues [35], rice straw [36], and various types of wood such as eucalyptus [37], poplar and spruce [38], and oak [39]. In addition, several non-conventional waste materials have been studied, such as algae [40], plastics [41], tires [42], sewage sludge [43], food waste [44], and bones [45]. In short, feedstocks for biochar production

are available in abundance, and the main concern is how to prepare biomass feedstocks or combine biomass to obtain quality biochar with interesting properties. Some of these properties are obtained through an activation process, which we discuss below.

3 Conventional pyrolysis techniques

This section briefly reviews conventional pyrolysis techniques for comparison purposes only, as exhaustive reviews on such techniques have been published [46,47]. Conventional thermochemical techniques for biochar production are shown in Figure 2b; of all these, pyrolysis is the most commonly used method [47]. Biochar synthesis involves heating biomass in the presence of little or no oxygen. To achieve this, biomass pyrolysis can be carried out in a variety of reactors or devices and at different scales: from small-scale devices for fundamental research such as laser pyrolyzer, plasma pyrolysis reactor, tube pyrolyzer, and fluidized bed types to pilot-scale pyrolysis reactors such as fixed bed, vacuum furnace, and auger reactor [48]. Many of these reactors are similar in terms of operating principles but differ in their configuration, gas system, and heating programs. Thus, in addition to the various reactor types, different process parameters can be controlled, and these parameters will determine the physical, chemical, and mechanical properties of the final product [49,50]. For this reason, one of the main objectives of research into the thermochemical conversion of biomass is to optimize pyrolysis to reduce the amount of unwanted by-products and promote the desired properties of the biochar [48].

Operating conditions (final temperature, heating rate, and residence time) and the type of feedstock are parameters frequently studied. These parameters are subject to optimization based on the desired properties of the final product, which are directly related to the application [51]. There are several exhaustive reviews on this topic [52,53]. In brief, to give a general idea, the maximum pyrolysis temperature has an impact on the volatilization of different compounds, resulting in different C, H, N, and O contents, as well as variable O/C and H/C ratios that are directly correlated to the aromaticity, biodegradability, and polarity of the resulting material [54]. Moreover, pyrolysis temperature can have an impact on textural parameters such as specific surface area and porosity of biochar, important properties for air and water treatment [53].

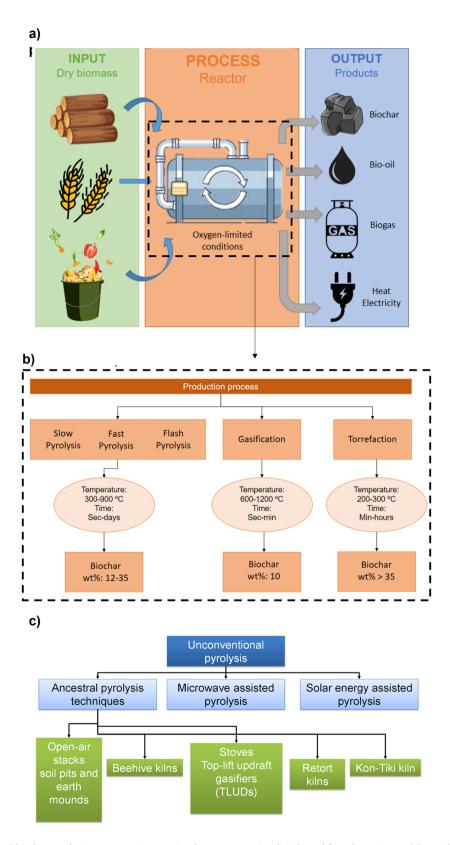


Figure 2: (a) Conceptual biochar production process (conventional or unconventional) (adapted from https://rotarykilnsupplier.com/activated-carbon-info/about-biochar-process/). (b) Classification of common thermochemical techniques used for biochar production [26]. (c) Main groups of unconventional pyrolysis techniques for biochar production addressed in this article, based on sustainability and affordability criteria.

Depending on operating conditions, in particular heating rate and residence time, biomass pyrolysis techniques can be classified into conventional slow pyrolysis (carbonization), fast pyrolysis, and flash pyrolysis [26] (Figure 2b). Slow pyrolysis is a process in which biomass is thermally treated at a low heating rate (0.1–1°C·s⁻¹) and reaches final temperatures ranging from 350 to 700°C, allowing sufficient residence time for biomass pyrolysis (300–550 s). In contrast, fast pyrolysis and flash pyrolysis involve high heating rates (10–200 and >1,000°C·s⁻¹, respectively) to 600-1,000°C and low residence times (0.5-10 and <0.5 s, respectively). While fast pyrolysis tends to produce higher proportions of oils, low heating rates and long residence times tend to produce higher proportions of the solid fraction [24]. In addition to these three parameters, other operating parameters, including particle size, the presence of a catalyst, and the pyrolysis atmosphere can have an impact on the quality and yield of the final product [8].

Evaluation of the data has shown that, of all the operating parameters, heat-treatment temperature is an important parameter influencing all the resultant chemical and textural properties of biochar [55,56]. In general, it has been reported that higher pyrolysis temperatures increase biochar's carbon content, pH, ash content, surface area, and stability, while H/C and O/C ratios decrease [56,57]. However, the recent meta-analysis by Hassan et al. [6] showed that these relationships varied according to the type of feedstock analyzed.

To extend the application of biochars and/or improve their performance for a given application (e.g., to maximize adsorption capacity), various engineering methods have been developed [58]. Thus, in addition to pyrolysis, a step of activation or modification of the pristine biochar is usually required, which can be performed before (biomass treatment), during, or after the carbonization step. The aims of the various modifications or treatments may be: (a) to activate the material - which may involve increasing its specific surface area by creating an interconnected network of micropores, as well as modifying and/or increasing the material's surface properties (e.g., functional groups) - and (b) to use the surface as a support for another material (or microorganism) that possesses properties beneficial to a certain application.

The most common approaches involve physical or chemical activation. On the one hand, physical activation is based on controlled material gasification with an oxidizing gas, such as steam [59,60], CO₂ [61–63], air, or mixtures. This is generally a two-step process: first, carbonization of the precursor takes place at 700-900°C, followed by physical activation at 350-1,000°C, depending on the activating agent [64]. The final textural and chemical properties of the resultant activated

carbons (ACs) depend largely on the activation time and temperature. In general, a higher degree of activation leads to greater porosity development and a broadening of the pore size distribution [65]. On the other hand, chemical activation is carried out using acids, bases, or salts such as KOH [42,66], NaOH [34,67], H₃PO₄ [32], or ZnCl₂ [68,69], among others. The precursor is mixed with the activating agent - various techniques can be used – and then subjected to carbonization at temperatures between 400 and 900°C in a single thermal step [64]. After chemical activation, an extensive washing step with water and sometimes HCl is needed [70]. Chemical activation improves textural properties, such as higher surface area, porosity and broadened pore size distribution, but they also dependent on activation time, temperature, and activating agent/char ratio [42]. In addition, surface functional groups are introduced.

Although conventional pyrolysis techniques have been widely studied, and biochar and/or activated carbon produced by these methods have been successfully tested for several applications at laboratory scale, the main limitations are the availability of temperature-controlled reactors and a stable energy supply (mainly in rural areas or low-income populations), as well as the volumes of material produced. Therefore, alternatives need to be considered to overcome these problems.

4 Nonconventional pyrolysis techniques

This section discusses the latest advances in non-conventional pyrolysis techniques for biochar production. Feedstocks and activation steps have not been considered here. From the results presented so far, it is clear that the study of the synthesis, characterization, activation, and application of carbonaceous materials at the laboratory scale benefits from bibliographic support, providing a strong basis for establishing industrial production parameters through non-conventional approaches, as we will to show below. Indeed, even if pyrolysis cannot be avoided to produce biochars, sustainable and low-cost approaches are now expected for sustainable and massive biochars synthesis.

On the one hand, there are other types of pyrolysis techniques and devices, based on the knowledge of ancient civilizations, which enable biochar to be produced using local and low-cost materials. Carbonization is as old as civilization itself, and the use of charcoal has a long history [71]. The fact that many of these methods are still used in many contemporary small communities proves that these technologies are easy to reproduce and affordable, enabling the production of large quantities of material without the aid of sophisticated technology. On the other hand, intensive research over the last 3 years has focused on adapting pyrolytic devices or designing innovative pyrolysis techniques, even leading to bench-top installations for biochar production, an interesting approach to stimulate laboratory research.

Then, based on our literature review, we will divide non-conventional pyrolysis into three groups: ancestral approaches, microwave-assisted pyrolysis, and solar-assisted pyrolysis. Figure 1c summarizes these different families of non-conventional pyrolysis techniques for biochar production addressed in this article. It should be noted that this classification includes non-conventional methods that we consider affordable, readily available, and sustainable. Consequently, plasma-assisted pyrolysis will not be included in this analysis based on this cost and environmental criterion.

The group of ancestral techniques includes devices such as open-air burning stacks [72,73], artisanal earth pits or earth mound kilns [74], beehive kilns [75–77], steel retort systems [19,78,79], stoves [80,81] and top-lit updraft gasifiers (TLUDs) [25,82], as well as flame curtain or Kon-Tiki kilns [83]. Devices vary in design, but in general, kilns or gasifiers are ovens made from clay or metal that produce enough heat to complete biomass pyrolysis in a matter of days, or even hours. In addition to being easy-to-operate and inexpensive technologies that enable local residues to be reused, these improved systems offer other advantages: they enable large amounts of carbonaceous material to be produced in situ with minimal infrastructure and, usually, without external energy supply, and with low greenhouse gas emissions [81,84]. The selection of the technology is related to socio-cultural and economic factors. These include the desired application, the production scale, the available feedstock, and the resources required to adopt a certain technology. In addition, the cultural component plays a major role in some communities, where ancient practices are reproduced from generation to generation.

Microwave-assisted pyrolysis is fairly recent for the specific production of biochar [85,86]. The first studies reporting biochar production with microwave irradiation as a heat source for pyrolysis were launched in 2014. Microwave-assisted pyrolysis involves the use of electromagnetic energy to heat the feedstock. It offers several advantages over conventional pyrolysis heating steps: fast and uniform heating of the feedstock with low thermal inertia, high power conversion efficiency and very low energy consumption, higher yields, and biochars produced with excellent textural properties [85].

Solar-powered pyrolysis is attracting increasing interest [87,88]. Indeed, the use of solar energy for

pyrolysis implies a greener approach to biochar production since the energy source is renewable. However, the main challenge is to adapt the optical installation to that of pyrolysis, which requires several upgrades and improvements in materials, and to obtain a homogeneous product [89].

In the following sections, we detail the most recent studies concerning the aforementioned unconventional pyrolysis devices and comment on their sustainability in relation to climatic and economic challenges.

4.1 Open-air stacks, soil pits, and earth mounds

Open-air stacks are an artisanal burning technique for heating – the principle is that of a campfire – and for charcoal production in rural areas. They are perhaps the simplest technology for charcoal production. It is still widely practiced in many developing countries because it is cheap and easy to implement. The main problems associated with this approach are the inefficient conversion into biochar and the high amount of emissions, which contribute to air pollution [72]. To improve carbonization and efficiency, the earth has been used as insulation. This is a very affordable approach: the earth is readily available, incombustible, and an excellent candidate as a sealant to enclose the charring wood and reduce heat loss.

There are two methods: underground carbonization (soil pits, Figure 3a and b) or surface carbonization (earth mounds) [90]. The former method involves digging a pit the most labor-intensive part of the process - placing the feedstock in it and lighting the fire [91]. Covering the hole with excavated earth to seal and insulate the chamber, or including a steel shield protecting the pit (Figure 3b), are some of the frequent variations. In contrast, the earth mound consists of building a pile of feedstock – usually wood – on the ground and covering it with earth, sand, and leaves, to form a gas-tight layer behind which charring takes place [92]. Earth mounds are usually large, and large pieces of wood are used, but these kilns can also be built small and are therefore suitable for household production. Once lit, the kiln requires continuous attention for 3–15 days, depending on its size. Yields range from 8 to 12% and, given that parameters such as feedstock moisture, kiln size, and process control play an important role, a significant gain can be achieved with improved technology [93]. To improve yields, pit kilns can be equipped with a chimney (Casamance kiln) [90]. These techniques can produce good-quality biochar, but with great variability due to

ventilation, which can be difficult to control and leads to heterogeneous heating of the feedstock. For domestic use, this is not a serious problem, mainly affecting overall yield [90].

4.2 Beehive kilns

During the 19th century, most earth kilns were replaced by so-called beehive kilns, in which other materials such as concrete or bricks take over the insulating function of the earth. There are different designs, but the main difference between them is the construction material: mud and brick kilns are the best known [74]. One of the main improvements of these devices was to recover condensable gases that were normally lost in pits and mounds, making them more efficient and reducing emissions [90]. However, these devices are stationary and can only be used in areas where

the supply of feedstock is easy. Beehive kilns have yields of up to 20–30%, depending on their size [77], and like earth mounds, are generally large, and large pieces of wood are indeed normally used. The most notable designs are the Brazilian beehive kiln (Figure 3c) and the Argentine half-orange kiln (Figure 3d) [90]. Carbonization cycles last 9 and 14 days, respectively. Large-scale kilns, with a diameter of about 6 m, can produce up to 15 tons of biochar per month and last up to approximately 6 years. However, small-scale devices can be built locally for household production.

4.3 Stoves and TLUDs

Stoves, which are not strictly pyrolysis devices for biochar production but gasifiers, have the advantage of simultaneously producing syngas and biochar as by-products (Figure 4a). One particular type of reactor is called TLUD,



Figure 3: (a) Conical soil pit [84], (b) steel-shielded soil pit [84], (c) Brazilian beehive kiln [77], and (d) Argentinian half-orange kiln [77].

commonly referred to as TLUD (Figure 4b). Like stoves, they were developed for cooking, but they can be used to produce biochar. Hence, most published work on stoves and TLUDs has focused on applications for cooking purposes in developing countries [80]. Kumar *et al.* [81]

presented a detailed historical review of cookstove development and improvement, and it was pointed out that with appropriate design and optimization, stoves generate low pollutant emissions [94,95] with relatively high yields [25]. Several designs have been reported [96] and, in many

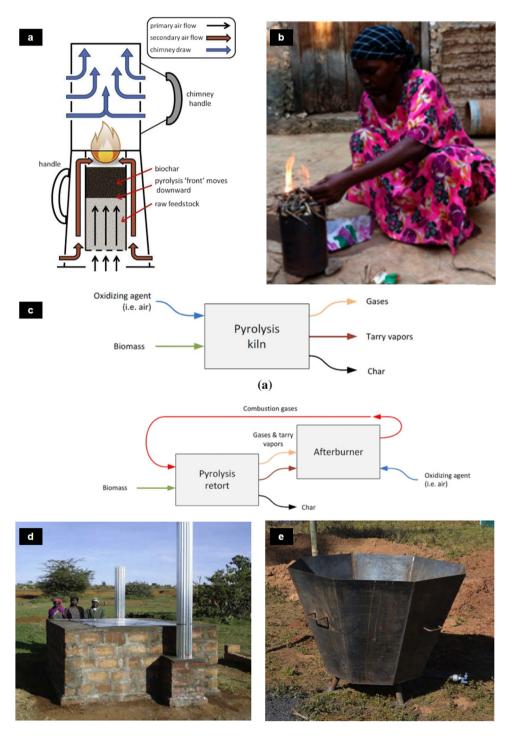


Figure 4: (a) General diagram of a TLUD (reproduced with permission from Elsevier from Vaughn *et al.* [82]), (b) gasifier ignited at the top [95], (c) schematic comparison between non-retort kilns and retort kilns (reproduced with permission from Springer Nature from Panwar *et a.* [100]), (d) field installation of the Adam retort kiln (reproduced with permission from Elsevier from Adam [78]), and (e) flame curtain metal octagonal kiln [84].

cases, stoves and TLUDs have been easily adapted and built with recycled materials such as stacked empty steel drums [97] or tin cans [98], depending on scale. Research has shown that small- and medium-scale reactors regularly reach high temperatures (650-950°C).

Unlike the above-mentioned techniques, which generally use large logs or wood pieces, these devices can be easily operated with agricultural and forestry residues and by-products, and are ideally suited for small-grain, chipped, or pelletized biomass fuels. With these homemade devices, biochar yields are 10-20%, but the quantity produced per batch is low. An important efficiency factor is that the user must quench the biochar before it starts to gasify and turn into ash. It is therefore up to the user to stop the gasification of the char, which he does by shutting off the air sources. If the TLUD has converted all the wood to char, there will be very little smoke, if any at all [98]. Another advantage is that biochar production is rather rapid. The potential of TLUDs, particularly for biochar production, has recently been recognized and, indeed, some NGOs have introduced them in several countries.

4.4 Retort kilns

Retort kilns are improved systems, where the volatile fraction can be recovered, reducing emissions considerably (Figure 4c). This technology is an important method of industrial charcoal production in many countries, but due to high investment costs, it is not viable for traditional charcoal manufacturers in rural areas [74]. This is why the improved charcoal production system or "Adam-retort" - a more efficient and affordable design - has recently been introduced to transfer and adapt this retort technology to rural areas (Figure 4d). The Adam retort kiln involves a two-phase operation based on the use of a separate firebox to generate combustion heat and an open pyrolysis chamber where the pyrolysis process takes place. The advantage is that the retort flue gases are directed into a system of channels, exchanging their heat with the feedstock to sustain the pyrolysis process before being released [79]. As with TLUDs, recycled materials can be used [99].

The mobile retort kiln or drum retort kiln was designed as an improvement of earlier drum-based concepts, with the incorporation of gas recovery. The material is fed directly into the drums, and a lid seals the pyrolysis chamber. In retort mode, the gases are directed into the space beneath the drums, where they ignite and fuel the pyrolysis process [79]. The retort mode can be applied to batch or continuous systems [100], and it operates at yields of about 30–35%. The production cycle is completed in 12–30 h (including cooling) in medium-sized systems or in about 5 h for the small-sized ones [19]. Sparrevik et al. [79] reported that biochar yield varied between 24.9 and 37.4% without being significantly different between systems with and without retort. However, they found that the average emission factors for retort kilns were lower than those of non-retort kilns, with significant differences for carbon monoxide, non-methane volatile organic components, products of incomplete combustion, and nitrogen oxides (NO_x).

4.5 Flame curtain kilns

The Kon-Tiki flame curtain kiln follows the principle of pyrolysis of biomass layer by layer in an open conical metal kiln (Figure 4e) [83,101]. With yields ranging from 20 to 30%, a 2-m³ batch device can produce 500 kg of biochar and almost 2 MWh of heat from various biomass residues such as shrubs, husks, straw, and prunings and requires just one worker to maintain and control the process. Jayakumar et al. [102] achieved a biochar yield (28 wt%) similar to that produced from the same feedstock (i.e., hardwood woodchips) in a continuous-scale pyrolysis unit. Pyrolysis temperatures can reach 680-750°C, and the process is much faster than in most traditional and retort kilns, taking just a few hours depending on the size of the unit. Its application for biochar production in farming contexts has reached over 50 countries.

Interestingly, due to its conical configuration, there is virtually no smoke during carbonization. This is because a curtain of flames is formed in the combustion zone, which protects the underlying biochar from oxygen and completely burns off all smoke and pyrolysis gases as they pass through the fire front. Indeed, Cornelissen et al. [79] reported that average emission factors for carbon monoxide and nitrogen oxides for flame curtain kilns were significantly lower than for retort and other kilns, and even reported almost no methane emissions for dry feedstock [21]. As with other devices, training is required to operate and optimize biochar production. In particular, it is important to regulate the timing of the spreading of each new layer of biomass, which will be determined by monitoring the flame, smoke, and ash formation. Another important aspect is regulating the rate and amount of biomass added, as excess or lack of feedstock can smother the flame, enabling access to oxygen and, consequently, ash formation and smoke production [83].

4.6 Comparison of ancestral pyrolysis devices

Figure 5 compares the gases and aerosols released by the various artisanal approaches, in terms of released factor (g·kg⁻¹). All ancestral technologies produce a quite low rate of aerosols (total suspended particulates), and the most released gas remains CO_2 , with over 1,000 g·kg⁻¹ released. No data are available for the TLUD technology, but it can be supposed that based on the kiln description, CO_2 should also be released at a significant rate. Beyond the concern of large amounts of CO_2 released, toxic gases (NO_x , VOCs) and more harmful greenhouse gas such as CH_4 are emitted at low levels.

As far as the properties of biochar obtained by ancestral approaches are concerned, they are quite similar to those produced by conventional techniques. Many experiments with non-conventional devices are not developed as part of an academic research project, so there are few reports on the properties of materials obtained with these techniques. Table S1 summarizes some of the main

characteristics of each technique/device, and Table S2 compiles some of the reported physicochemical properties of biochars produced using non-conventional devices and their intended applications.

As expected, the physicochemical properties of biochars obtained with non-conventional methods vary according to feedstock and device - and hence pyrolysis parameters - just like biochars derived from modern, more conventional techniques. Comparison with the abundant literature on conventional techniques shows that, in many cases, properties are similar to those of pristine biochar obtained with conventional devices in terms of biochar yield and composition [8,56]. Biochar yield and textural properties, such as surface area, are variable, ranging from 14 to 34.5% and from 4 to 473 m²·g⁻¹, respectively, but are well known to depend on feedstock type and pyrolysis parameters, even in conventional syntheses. For instance, wood-based biochars tend to have higher surface area and total pore volume, while biochars derived from crop waste, other grasses, and biosolids or manure tend to have lower

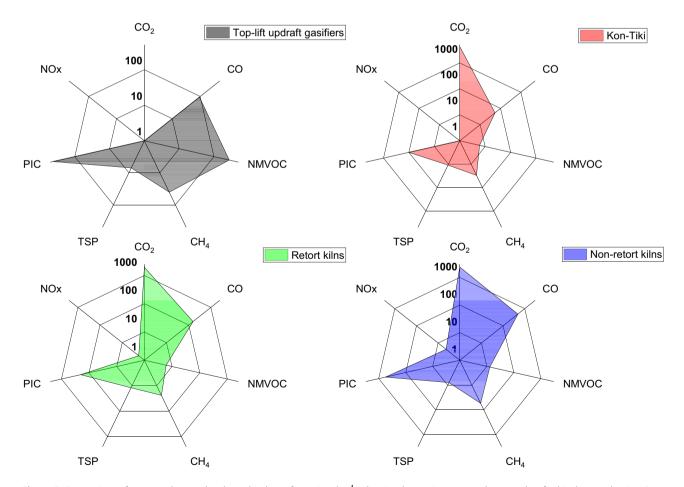


Figure 5: Comparison of gases and aerosols released (release factor in $g \cdot kg^{-1}$) when implementing ancestral approaches for biochar production. Data taken from refs. [79,84,103]. No data were available for CO₂ and TLUD technology.

surface area and total pore volume. This could be related to the chemical composition (i.e., lignin content) and cellular structure of the initial biomass, as well as to the occurrence of deformation, cracking, or blockage of micropores due to volatilization of water and gases during pyrolysis [27].

In contrast to the three operating parameters controlled in conventional pyrolysis (heating rate, temperature, and residence time), in non-conventional devices, visual indicators are used to define process steps (e.g., presence/absence of smoke or ash and changes in char or flame color). When parameters can be defined, duration or exposure time is frequently studied to control the properties of the biochar obtained [71]. Consequently, one of the main concerns of non-conventional devices is the impossibility of controlling many, if not all, operating variables during pyrolysis and the fact that the feedstock may not be heated uniformly. This would lead to a non-reproducible biochar.

Some authors have addressed this issue. Cornelissen et al. [84] reported that biochar yields were indeed variable (13-32%) and linked this to variations in operating conditions and weather conditions while stressing that this represented realistic production conditions. Kearns et al. [75] also observed variability in the physicochemical properties of the biochars produced, leading to variable adsorption capacities of organic compounds. Thus, although it is difficult to effectively reduce variability within and between batches in non-conventional techniques, mainly for applications such as water treatment where consistency of physicochemical properties is often required, several simple strategies can be adopted and investigated for optimization: operator training, storage of feedstock so that it dries out in advance, and use of pooled samples. Moreover, the fact that production costs are low means that dose optimization may not be a relevant issue, depending on the desired application.

Interestingly, their low cost means that they can be used widely in low-resource countries, but also in developed countries, as they are easy to use. As recently demonstrated in six model countries [17] and in Tanzania [18], ancestral methods can induce a cost-benefit with a reduced environmental impact, provided that educational programs are launched to convince populations to change their habits. Sufficient feedstock is available, and certain adaptations to kiln designs are proposed for large-scale dissemination.

5 Microwave-assisted pyrolysis

The basic set-up for microwave-assisted pyrolysis is shown in Figure 6a. It consists of putting biomass or other feedstock samples in a quartz flask and then placing the whole assembly in a microwave chamber. An inert gas (usually N₂) is passed through the microwave chamber to carry the volatile compounds to a condensation system. At the end of pyrolysis, the carbonaceous residue obtained (char) and the condensable fraction (bio-oil) are weighed. The feedstock used is very often combined with a microwave absorber before pyrolysis to enhance the energy absorbed. Indeed, even if the biomass contains water and inorganic matter, the target wavelength to be absorbed is not systematically reached, necessitating the addition of typical metalbased microwave absorbers (pure metal such as Fe or Al. carbonate (K₂CO₃, CaCO₃), metal oxide (NiO, MgO), hydroxide (KOH), chloride (ZnCl2, FeCl3), or carbon-based absorbers (AC, graphite, silicon carbide or coke) [104]. Typical heating rates range from 0.1 to 1,000°C·s⁻¹, reducing total synthesis time in some cases (flash pyrolysis concept), and the temperature reached ranges from 100 to 850°C [104]. As with conventional pyrolysis, feedstock profiles and the pyrolysis times and temperatures obtained are the main parameters influencing biochar yields and final materials properties.

However, in the case of microwave-assisted technology, the absorbers can also greatly influence the final product obtained. Nevertheless, new type of feedstock, notably organic municipal solid waste, could limit the use of microwave absorbers in the process in the future, due to the higher water content inside the waste compared with basic biomass feedstock, as demonstrated by Beneroso et al. [105]. However, another advantage of microwave-assisted pyrolysis concerns biochar yields and final product quality. Indeed, in Figure 6b, a ternary diagram by Li et al. [104] demonstrates the added value of using microwave as a heating source for pyrolysis for given biomass feedstock (coffee hulls, glycerol) or waste (sewage sludge) to achieve good biochars and by-products yields. Solid yields are around 50%, and even higher in the case of waste (sewage sludge), confirming the need for further research into the production of biochars from feedstock derived from human waste. Product quality refers to the chemical composition and textural properties obtained for various applications. Here, microwave-assisted pyrolysis offers adequate control of aromatization rate, hydrogen and oxygen contents, specific surface area, and pore profiles (meso and micropores), as recently demonstrated in a comparative study [106].

Figure 6c compares the pros and cons of microwaveassisted pyrolysis with conventional heating. From the point of view of energy consumption, it is clear that microwave irradiation is much better than conventional heating, and that it corresponds to the environmental and energy concerns highlighted for biochar production. Both microwave-assisted and conventional pyrolysis are limited in the use of solvents, and the absorbers added in the case of the microwave approach are available and cheap most of the time. However, this can add another step to the process, which is not desirable when scaling up. Microwave scale-up for biochar production is indeed being questioned today, mainly due to the relatively difficult temperature measurement, the uncertain economics of process scale-up, limitations related to the availability of microwave technologies, and the difficulties of the large-scale process [85].

In the context of energy resilience, microwave-assisted pyrolysis should be a relevant solution to put forward for the synthesis of biochars and their derivatives [108]. Provided that intensive research is carried out on large-scale synthesis, microwave-assisted pyrolysis of biomass and human waste has the potential to establish itself as a sustainable, low-cost biochar production technique.

6 Solar-assisted pyrolysis

Solar energy is of great interest in the process of mitigating climate change and generally decarbonizing the planet. The use of the sun as an energy source to initiate pyrolysis first required achievements on the sunlight collection system before being used in installations that are more complex. In the case of pyrolysis using solar energy for the specific production of biochar, we detected several important papers accurately describing the challenges involved in implementing such a concept [87–89,109].

The first step in a solar-assisted pyrolysis is to collect the solar rays and concentrate them on the pyrolysis reactor to increase the solar flux density and start the reaction. With this in mind, several improvements have been made to optical concentration devices. These fall

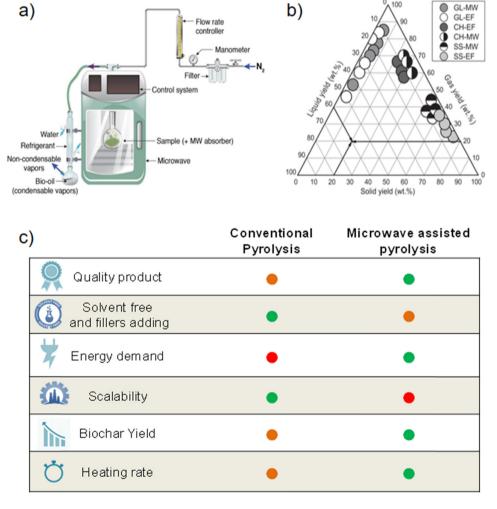
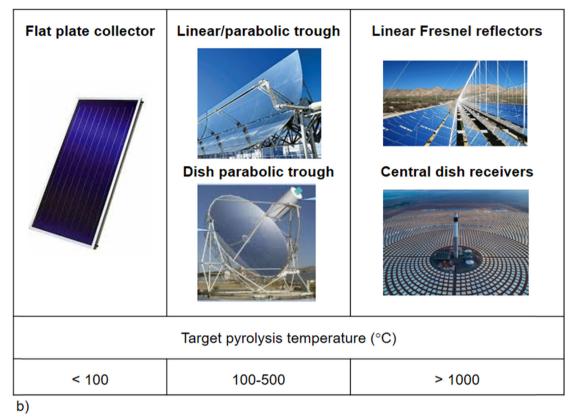


Figure 6: (a) General configuration that could be encountered for microwave-assisted pyrolysis to produce biochars [107]. (b) Comparison of biochar yields with conventional pyrolysis using electric furnace (EF) and microwave pyrolysis (MW) and for different feedstocks: glycerol (GL), coffee hulls (CH), and sewage sludge (SS) (reproduced with permission from Elsevier from Li *et al.* [104]). (c) Comparison of conventional and microwave-assisted pyrolysis for biochar production in terms of advantages and disadvantages.

a)



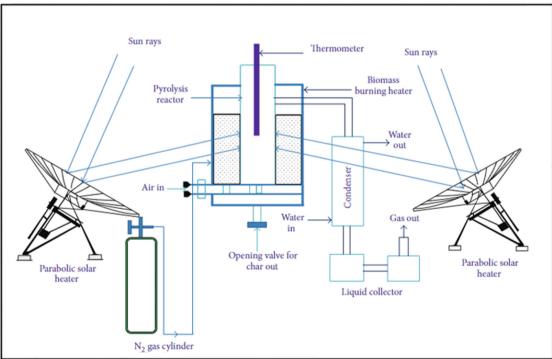


Figure 7: (a) Different types of optical devices for collecting solar rays. (b) Typical vertical solar-assisted pyrolysis for biochar production [110].

into three categories, depending on the pyrolysis target temperature. Figure 7a shows the various concentrators capable of reaching temperatures from 100 to 1,000°C. The group of flat plate collectors is a modified solar hot water panel, here with an optocaloric utility to initiate a chemical reaction, whereas the classic trough family or complex Fresnel reflectors are inspired by astronomical instruments to concentrate the various wavelengths of the light spectrum. Despite their complexity, all these systems have great potential for scaling up pyrolysis to produce biochars, thanks to the large areas they can cover.

Typical systems used in research laboratories are linear or curved mirrors, but a given system and its strategic development require precise adaptation of the orientation of the collection device, in line with the design of the pyrolysis reactor. The type of light source must be included in the final system design. For instance, three alignments of collection and concentration devices can be used: (a) vertically oriented continuous-heating reactors (with a natural light source), (b) vertically oriented continuous-heating reactors.

The concept of vertically oriented optical devices is illustrated in Figure 7b. The optical devices are installed away from a borosilicate reactor. This appears to be the easiest configuration for collecting natural light rays. A lens system can be used to focus the rays on the reactor walls. The design of the pyrolysis reactor is quite similar to that of conventional heating, with a thermometer inserted to manage temperature. In the case of simulators using light as an energy source, the main difference relies in the nature of the light (typically a xenon lamp), offering the possibility of precisely controlling light intensity, with discontinuous focusing on the reactors. The horizontal configuration is rarer and involves heating under the reactor using a parabolic concentrator or a trough.

Another major challenge of solar pyrolysis is the heterogeneity of the biochar obtained. Lobato-Peralta *et al.* [89] compared the production of carbon materials from agave bagasse using a tube furnace and a solar furnace at 700°C under similar operating conditions and obtained, for the latter, a biochar with a lower surface area and less uniform porosity development. However, for the solar-produced biochar, they found out that the use of fibers rather than powders resulted in carbons with a porosity that is more homogeneous, and that the carbons produced with the solar furnace were reproducible.

Despite the fact that yields are similar to those of conventional pyrolysis [111], when it comes to sustainability and low-cost biochar production, solar-assisted technology is a complex balancing act. Indeed, on the one hand, the

production of the solar energy system is not totally decarbonized. On the other hand, the use of solar energy limits the sources of carbon fuels for producing biochars by pyrolysis. The overall cost of solar energy system is decreasing, but it varies according to the energy policies of different countries [112]. However, developing countries represent the sunniest regions of the world most of the time, and could benefit from solar energy for biochar production. The adaptation of optical devices could be of great interest, particularly in desert areas from this point of view.

7 Applications: Toward a sustainable biochar concept

Biochars and activated carbons have multiple applications, including soil conditioning, carbon stabilization and sequestration, energy storage, and environmental restoration, such as soil and water treatment to mitigate pollution [1,113–116]. For materials derived from non-conventional syntheses, the end-use option chosen depends on the objectives of the biochar producer: they have historically been produced for energy use in households (*i.e.*, heating and cooking) [80,81,117] and as a soil amendment in farming [118]. Table S2 shows this clear preference for these applications, while water treatment has received little attention. This section highlights the main application areas benefiting from the use of biochars, with greater emphasis on materials synthesized using unconventional pyrolysis techniques.

7.1 Soil-related applications

Biochar has always been used as soil amendment: "Terra Preta" is a well-known ancient practice in which soils in the Amazon basin were improved by the addition of biochar. This practice has been considered a model of sustainable agriculture in the humid tropics [119]. Among the main functions, biochar has been used primarily to improve the physical and chemical fertility of the substrate and, ultimately, to enhance crop yields [53,120], as shown in Figure 8a with the application of biochar to Australian soil [121].

Biochar can improve water and nutrient retention [123,124] and chemical fertility and fertilizer use efficiency [125] and develop soil structure [126,127], among others. The performance of biochars is strongly determined by the type of feedstock and pyrolysis temperature, as well

as by the type of activation process [128]. In this sense, many commercially activated carbons are produced and can be purchased for incorporation into the soil to improve fertility, at relatively high prices. However, many NGOs and the International Biochar Initiative promote the local production of biochar for farmers, and there are even many open-source guidelines for doing so (https://biochar-international.org/open-source-biochar-technologies/). Significant agronomic benefits have been reported with the incorporation of biochar doses compatible with household production, an important factor for farmers in developing countries. For instance, in degraded soils in Kenva, Lehmann and Rondon [129] reported productivity improvements from 20 to 220% with the application of low doses of biochar, and Kimetu et al. [130] reported that maize yields doubled after the application of 8 tons C·ha⁻¹. With regard to biochar pre-treatment and fertilizer use efficiency, Pandit et al. [131] studied the effect of nonconventional biochar on maize biomass production and found that different nutrient-enrichment treatments and doses of biochar resulted in significant increases in aboveground biomass production. Schmidt and Taylor [83] synthesized biochar with the Kon-Tiki kiln and obtained premium quality biochar (according to the European Biochar Certificate). Studying the effect on pumpkin yield, they obtained that a low-dose application to the root zone of urine-enriched biochar led to substantial yield increases in fertile loam soil [118]. Pristine biochar and

activated carbons have also been used for soil remediation [132,133], particularly in areas with high concentrations of heavy metals, since biochar can reduce the bioavailability of metals in soils by immobilizing them in stable forms [134]. Remediation of sediments polluted by heavy metals is also viable with non-conventional biochar. An acute toxicity test carried out in the germination phase of *Lactuca sativa* seedlings, widely used as a bioindicator, showed that additions of 20% Kon-Tiki-derived biochar significantly improved the germination index of lettuce in sediments contaminated with multiple heavy metals and that performance was comparable to that of commercial activated carbon [135].

7.2 Water treatment

Another environmental application for biochar is water treatment. Biochars and activated carbons have been extensively tested for several pollutants [136,137], including heavy metals [51,52] and organic compounds [1,22,138,139]. Likewise, biochars produced using non-conventional devices have been tested in water treatment, notably for fluoride, arsenic, and organic compounds such as agrochemicals and antibiotics [140–144].

Kearns *et al.* [75] compared the performance of various types of chars (lab-made chars, traditional kiln charcoals

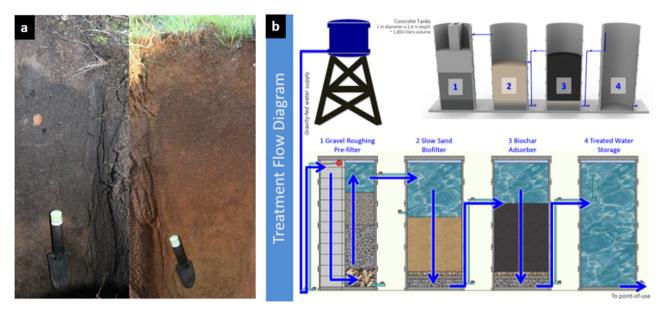


Figure 8: (a) Soil profile of a typical "Terra Preta" in Australia (site TPA2) characterized by a predominant sandy soil. On the left, the soil treated with biochar is more fertile than the right-hand side of the photo (the natural sandy soil). The clear difference in color is a characteristic of Terra Preta profiles. The trowel shown is 20 cm high (reproduced with permission from Elsevier from Downie *et al.* [121]). (b) Filtration system integrating biochar in multi-barrier treatment trains for decentralized communities [122].

collected in Asian villages, and commercial activated carbons) and observed that one of the field-collected char had a comparable 2,4-dichlorophenoxyacetic acid (2,4-D) uptake than an activated carbon. Studying the effect of temperature and feedstock on 2,4-D adsorption capacity, the authors observed that increasing pyrolysis temperature or duration improved herbicide removal, whereas feedstock did not affect adsorption capacity for the materials studied. These authors related this increased sorption capacity to lower H:C and O:C ratios and higher surface area (>500 m²·g⁻¹). A similar herbicide adsorption capacity was observed for furnace chars and kiln charcoals generated using similar thermal profiles [75]. Other studies have shown enhanced adsorption by biochars generated from TLUDs under conditions of simultaneous pyrolysis and activation with air for the removal of herbicides 2,4-D and simazine [143]. Additionally, wood-based biochar produced at around 850°C and with 400 m²·g⁻¹ surface area gave similar results to commercial powdered activated carbon in the removal of the antibiotic sulfamethoxazole at environmentally relevant concentrations [145]. With regard to the pollutant removal mechanisms involved, few authors carried out in-depth characterizations to explain the underlying mechanisms. In most cases, mechanisms analogous to those of conventional biochars were reported [73].

Moreover, activation and modification of pristine biochar derived from non-conventional techniques can be performed to improve the sorption capacity of the material. For instance, CO_2 activation of Kon-Tiki kiln-derived biochar had a positive effect on the development of porosity and surface area, while preserving the structure of pristine biochar [146]. These types of textural modifications are desirable in applications such as water treatment as they can broaden the range of pollutants to be removed, and can even improve the removal of a specific pollutant. In particular, the removal of paracetamol – an emerging pollutant – with activated carbon derived from the Kon-Tiki kiln achieved sorption capacities comparable to those of other materials reported in the literature [146].

While conventional biochar and activated carbons have shown high sorption capacities for various pollutants, arsenic removal remains a challenge. For this reason, composites based on carbon and magnetic particles have recently been proposed as materials capable of removing various pollutants in the liquid phase [147–149], in particular arsenic [32,150]. This is because iron oxide particles have shown a high affinity for arsenic, and their incorporation into a porous support material avoids the problems of agglomeration and oxidation of isolated magnetic particles. Furthermore, the magnetic response that these particles confer on carbon facilitates the separation of the

material from the liquid phase after water treatment, whatever the pollutant analyzed [151].

Similarly, the porous structure of biochars obtained by non-conventional syntheses can also be used as a support material for impregnation with iron nanoparticles in situ to enhance arsenic removal from water. The removal efficiency of Kon-Tiki-derived magnetic biochar was superior to that of a commercial magnetic activated carbon using the same synthesis procedure to form the iron particles (Bursztyn Fuentes et al. [152]). Eikelboom [97] studied the arsenic sorption capacity of chars derived from a TLUD and soaked in FeCl₃ and obtained sorption capacities of 55 µg·g⁻¹, which allowed the treatment of 35 L of water for a household of five people. In this context, pristine and modified biochars produced using non-conventional devices can be integrated into low-cost, multi-barrier treatment systems for on-site water treatment to solve local problems (Figure 8b).

Finally, it is worth mentioning that despite current advances in the application of biochars to water treatment, such as photocatalytic applications of composites for water purification [3,153], biochars produced with unconventional techniques have not yet been tried, resulting in unexplored avenues for future research.

7.3 Energy-related applications

As far as energy applications are concerned, biochar is a high-calorific solid fuel commonly used in kilns and boilers [22]. Depending on the pyrolysis temperature, the energy content of biochar can even be comparable to that of raw anthracite [56]. For energy storage in particular, numerous technologies have been developed, such as solar and fuel cells, high-performance batteries, and supercapacitors [114]. For instance, with regard to the latter, the use of porous carbon materials as electrodes has increased over the years, as these materials have desirable properties such as high chemical and thermal stability, good conductivity, the possibility of adjusting their textural properties, and industrial processes for electrode synthesis and production already established [67].

Although biochars produced with non-conventional devices have historically been used for cooking and heating, their energy contents have been little studied and quantified. With regard to heating potential, Bursztyn Fuentes *et al.* [146] obtained an average heat of combustion of eucalyptus-derived biochar of 27.3 MJ·kg⁻¹ produced with a Kon-Tiki kiln. Vaughn *et al.* [82] obtained between 17.9 and 19.0 MJ·kg⁻¹ for other wood-derived biochar produced in

TLUDs, and Zhou et al. [72] produced biochars from corn residues in a soil pit and obtained values of 19.8 MJ·kg⁻¹. These values were consistent with what has been reported in the literature for various residues with conventional carbonization procedures [154–157], indicating that biochars derived from non-conventional devices can be successfully used as fuel.

With regard to energy storage, Huggins et al. [158] tested biochars made in TLUDs from pine wood chips and compressed milling residues as electrode materials in microbial fuel cells, and both materials showed satisfactory power density, comparable to commercial activated carbon electrodes. However, for these value-added applications, compulsory post-pyrolysis treatments are usually required such as activation, impregnation, and several acid-washing steps to remove ashes. Consequently, energy storage is perhaps a rather difficult application for low-cost biochars, but further research must be conducted to fully explore their potential in this important and growing field.

7.4 Climate change mitigation

Rather than being used as fuel in energy-related applications, biochar has recently been proposed as a strategy for climate change mitigation [159]. Climate change mitigation, among other global challenges, has been explicitly reinforced in the international agenda through the UN Sustainable Development Goals [160]. Several approaches have been identified in the literature concerning this application: emissions reduction, carbon stabilization, and carbon sequestration [161].

On the one hand, as biochar is not easily degradable compared with raw feedstock, a biochar-based soil amendment could act as a long-term carbon sink in soils and sediments, stabilizing carbon for thousands of years. In this sense, from a carbon point of view, the most important issue is the difference between the decomposition rate of the original feedstock without pyrolysis and that of biochar [162]. Consequently, the combination of biochar production in smokeless pyrolysis devices from residual biomass and its incorporation into the soil has been proposed as a promising strategy for global carbon stabilization and storage [14]. Another environmental benefit derived from biochar incorporation is the reduction of non-CO2 greenhouse gas emissions from soils [163]. On the other hand, many authors have focused on the carbon sequestration capacities of different types of activated carbon by measuring CO₂ capture. These values are usually measured with manometric instruments used for textural characterization of

solids at 0°C, but this is not a realistic operating temperature of post-combustion, and CO₂ adsorption is highly temperature-dependent [164]. However, this application needs to be approached critically, as the reported carbon sequestration capacities are interesting for comparison purposes, but not very representative of reality.

8 Conclusions and perspectives

Biochar is a promising material for use in many environmental and energy applications. Despite the extensive knowledge of biochar synthesis, characterization, and application, robust studies on non-conventional pyrolysis techniques are scarce. Data collected on biochar produced from artisanal devices suggest its potential for many applications, including water treatment; biochar can be integrated into low-cost, multi-barrier treatment schemes for on-site water treatment to solve concrete local problems. In this respect, further research is required to identify the broad spectrum of pollutants that can be effectively removed with non-conventional biochars, the synthesis of biochar-based composites, and the mechanisms involved.

Biochars obtained using non-conventional pyrolysis techniques and devices can be used, like conventional activated carbons, in many novel applications, particularly in the energy sector. However, there is no experience with unconventional biochar for energy storage, which opens up potential future research avenues for biochar-derived carbon materials for batteries or fuel cell components. In this sense, open-source technologies could be a good starting point for many research projects.

The selection and implementation of alternative, nonconventional pyrolysis methods for sustainable biochar production faces challenges. As regards selection, there are no clear guidelines as to the most suitable technique for each application, given that the choice, especially in developing countries, depends on many sociocultural and economic factors, leading to non-systematic experiences and studies. As for implementation, variable control and reproducibility raise concerns. However, our study shows that different biochars from non-conventional devices (raw or modified) are comparable to those from conventional methods in many fields, offering a sustainable and cost-effective solution. Effective use of these biochars, particularly in technical applications, requires consistent quality. Strategies like operator training and pooled sampling can help, but more academic research is needed to optimize and standardize production with non-conventional methods.

Environmental impacts of biochar production, especially in developing countries, also raise concerns, mainly greenhouse gas emissions and health hazards. Advances in device efficiency and emissions reduction are promising. The choice of feedstock also significantly affects biochar quality, with human waste showing potential. Therefore, academic and industrial sectors should focus on studying biochar production using both conventional and non-conventional pyrolysis techniques with human waste feedstocks, while contributing to circular economy by reusing waste material.

Funding information: This study was partly supported by the French PIA project "Lorraine Université d'Excellence," reference ANR-15-IDEX-04-LUE and the TALISMAN project funded by ERDF. Amalia Lara Bursztyn Fuentes acknowledges financial support from Universidad Nacional de Tierra del Fuego (UNTDF) through the project PIDUNTDF B 12/2023.

Author contributions: All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Conflict of interest: Vanessa Fierro, who is the co-author of this article, is a current Editorial Board member of *Reviews on Advanced Materials Science*. This fact did not affect the peer-review process. The authors state no other conflict of interest.

Data availability statement: Data sharing is not applicable to this article as no datasets were generated or analyzed during the current study.

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