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Review

Fabrication and environmental applications of metal-containing solid waste/biochar composites: A review

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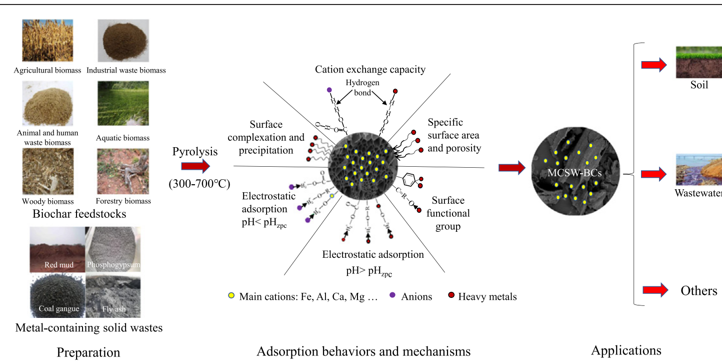
HIGHLIGHTS

- Utilization status of metal-containing solid wastes is reviewed.
- Adsorption behaviors and mechanisms of MCSW-BCs on pollutants are summarized.
- The applications of MCSW-BCs for soil amendment and environmental remediation are discussed.
- Suggestions and future perspectives are proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

The resource utilization of industrial solid waste has become a hot issue worldwide. Composites of biochar with metal-containing solid wastes (MCSWs) can not only improve the adsorption performance, but also reduce the cost of modification and promote the recycling of waste resources. Thus, the synthesis and applications of biochar composites modified by MCSWs have been attracting increasing attention. However, different MCSWs may result in metal-containing solid waste/biochar composites (MCSW-BCs) with various physicochemical properties and adsorption performance, causing distinct adsorption mechanisms and applications. Although a lot of researches have been carried out, it is still in infancy. In particular, the explanation on the adsorption mechanisms and influencing factors of pollutant onto MCSW-BCs are not comprehensive and clear enough. Therefore, a systematic review on fabrication and potential environmental applications of different MCSW-BCs is highly needed. Here we summarize the recent advances on the utilization of typical metal-containing solid wastes, preparation of MCSW-BCs, adsorption mechanisms and influencing factors of pollutants by MCSW-BCs as well as their environmental applications. Finally, comments and perspectives for future studies are proposed.

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Contents

1. Introduction

The continuous push to improve agricultural and industrial output as well as socio-economic conditions has raised concerns about water and soil pollution (Chen et al., 2020). Polluted soils can be cleaned up through phytoremediation (Verma et al., 2017) in conjunction with microbial (Yin et al., 2019), chemical, and physical methods (Dhaliwal et al., 2020; Yadav et al., 2019). Some of these approaches, however, are time-consuming, costly, or not very effective (Morillo and Villaverde, 2017; Verma et al., 2017). Similarly, polluted water may be treated by physical (Jorge et al., 2017), chemical, physicochemical (Hua et al., 2014; Wu et al., 2014), biological (Lares et al., 2018), and biochemical methods (He et al., 2017; Nolte et al., 2020), or a combination of the above (Fu and Wang, 2011). Here again, some of these methods are costly, ineffective, and pollutant-specific. On the other hand, adsorption to a suitable solid material is an effective, inexpensive, and relatively simple method of removing pollutants (Li et al., 2019; Wang et al., 2021). In this respect, biochar has received much attention in recent years (Wang et al., 2020).

Biochar is produced by the decomposition of biomass from plant or animal waste such as lignin (Han et al., 2021), invasive plants (Feng et al., 2021), animal manure (Hassan et al., 2020), etc. under limited oxygen conditions with 300–700 °C (Lehmann and Joseph, 2015). It is a structurally stable solid material, with a high degree of aromatization and anti-decomposition ability. Because of its large specific surface area (SSA) and abundance of oxygen-containing functional groups, biochar is well suited to remediating polluted environments (Ali et al., 2020; Bashir et al., 2018; Wan et al., 2020; Wang et al., 2018a; Yu et al., 2019).

The adsorption capacity of (pristine) biochar may be improved by several means. More specifically, biochar can be modified by chemical treatment (Mahdi et al., 2019; Wang et al., 2018), amination (Oladele et al., 2019; Zhang et al., 2019), impregnation (Li et al., 2014; Yoon et al., 2019a), steam activation and magnetization (Rajapaksha et al., 2016), metal salts ($FeCl_3$, $MnSO_4$) (Wu et al., 2016; Xiang et al., 2020), and clay minerals (Lahijani et al., 2018; Liu et al., 2020; Wang et al., 2017; Xu et al., 2019; Yu et al., 2019), etc. In an attempt at improving the adsorption capacity of biochar, reducing modification costs, and promoting the rational use of resources, many studies have synthesized biochar composites with solid wastes, including agricultural and forestry wastes, animal manure, and activated sludge. There are relatively few reports, however, about modifying biochar with solid wastes of industrial origin, such as phosphogypsum, fly ash, red mud, and coal gangue, etc. (Gao et al., 2019; Huang et al., 2017; Ni et al., 2019; Shaheen et al., 2018; Wang and Wang, 2019). The research trends on biochar and MCSW-BCs from 2010 to 2020 are shown in Fig. S1, which indicates that MCSW-BCs have been gradually attracted attention due to its low cost and high adsorption capacity.

The resource utilization of solid wastes is a major task that needs to be tackled in an environmentally rational manner (Guerrero et al., 2020). Countries rich in mineral resources (e.g., China, Poland, USA) tend to accumulate solid wastes, notably phosphogypsum, red mud, fly ash and coal gangue (Shahba et al., 2017). A large amount of solid wastes not only occupy a large area of land but also damage the ecosystem (Belyaeva and Haynes, 2011; Xia et al., 2019). Therefore, reasonable treatment and resource utilization of industrial solid wastes are in highly need. Due to the special physicochemical properties such as containing rich metal elements (Ca, Al, Fe, etc.), higher pH, large SSA (fly ash), when these solid wastes are pyrolyzed with biomass, the SSA and surface functional group content of the biochar component may be so altered as to enhance their capacity for adsorbing pollutants (Wang et al., 2020a; Zhou et al., 2017).

A variety of metal-containing solid wastes (MCSWs) have been used to modify biochar (Fig. 1), which is called metal-containing solid waste biochar composites (MCSW-BCs). At present, the research of MCSW-BCs has received wide attention due to its low modification cost and high adsorption capacity. However, with different metal element composition and physicochemical properties of MCSWs, the adsorption mechanisms, influencing factors and environmental applications of MCSW-BCs are not fully understood. Therefore, it is necessary to

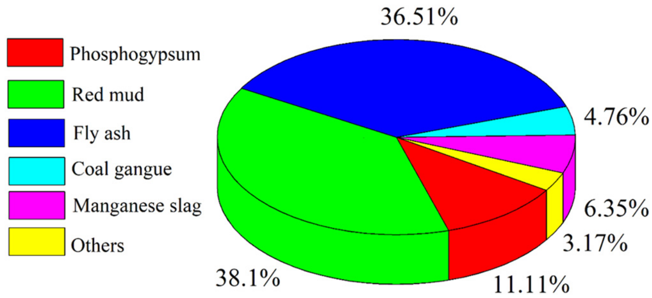

Fig. 1. Percentage of literatures on various biochar composites modified by different MCSWs (data from 63 literatures in Web of Science).

systematically summarize recent advances in pollutants adsorption by MCSW-BCs to provide a reference for future study. In this study, some mainstream academic databases (e.g. Web of Science) were used to search for relevant papers on MCSW-BCs. With "phosphogypsum+biochar", "red mud+biochar", "fly ash+biochar", "coal gangue+biochar" and "solid waste+biochar" as the keywords, respectively, we totally obtained 277 papers. Based on the title and abstract, we judged their relevance to the subject of this work and obtained more than 100 papers closely related to the subject.

Here we summarize the recent advances on the utilization of typical MCSWs, the preparation methods of MCSW-BCs, adsorption mechanisms and influencing factors of different pollutants adsorption by MCSW-BCs as well as their applications. Finally, comments and perspectives for future studies are proposed.

2. Utilization status of metal-containing solid wastes

Being enriched in metallic elements, industrial solid wastes are also referred to MCSWs (Cho et al., 2019; Yoon et al., 2019b). The global production of MCSWs is estimated at 70–100 billion tons per annum (Tan et al., 2016). The haphazard treatment of MCSWs is wasteful of resources as well as damaging to the ecological environment. The rational use of MCSWs as a secondary resource, and following the principles of cleaner production, would minimize environmental damage. Indeed, the recycling of such MCSWs as phosphorous slag, fly ash, coal gangue, and red mud has already brought certain economic benefits. Table 1 summarizes the current status, disadvantages, and environmental risks of recycling MCSWs.

Phosphogypsum is a solid waste of phosphate fertilizer plants, which is produced by the reaction of sulfuric acid with phosphate rock. Most of the global phosphogypsum production of 100–280 tons per annum is dumped on land or discharged into sea and rivers without prior treatment. In China about 40 million tons are discharged in this manner every year, making phosphogypsum become one of the largest industrial pollutants (Bouargane et al., 2019; Al-Hwaiti et al., 2010). Phosphogypsum is mainly composed of $CaSO_4 \cdot 2H_2O$ but also contains Si and various radioactive elements (P.M et al., 1994). It is used in the construction industry, and as a soil amendment after purification (Campos et al., 2017; Kwang-hyun, 2003; Soo et al., 2003). Containing Si and Ca, phosphogypsum is also used to forge glass-ceramics. The recovery rate of phosphogypsum, however, is low relative to its overall production. The secondary application of phosphogypsum also poses a risk to the environment as the material contains heavy metals. The heavy metals contained in phosphogypsum, especially Hg, Pb, Cd, and As, are the most common toxic elements and may pose threat to human health through agricultural products (Peng et al., 2020). Liu et al. advocated the large-scale use of phosphogypsum as an embedded filler which, however, raised questions of technique and expenditure (Liu et al., 2019). The applications of phosphogypsum in agriculture still face strict restrictions and problems. Therefore, reasonable disposal of phosphogypsum and the ability to minimize its toxicity to the environment during secondary use are the key to the problems that need to be resolved.

Red mud (bauxite residue) is an industrial solid waste generated in alumina refining from bauxite (Liu et al., 2014) containing a variety of transition metal oxides (Yoon et al., 2020). Due to the high alkaline, red mud is highly corrosive and harmful to the environment (Ozden et al., 2018). The global reserves were estimated to exceed 4 billion tons by 2015 (Wang et al., 2020). Red mud has given a serious threat to water, soil, and the ecosystem. How to deal with red mud, and sensibly utilize this waste product, has therefore become a hot issue. Red mud can recover Al_2O_3 after magnetic separation of iron-rich components. After de-alkalinization, red mud can be turned into bricks, pipes, and flooring material. It can also be used for the production of construction materials and glass ceramics (Hulya et al., 2005; Joseph et al., 2019). It is difficult and costly, however, to extract valuable elements from red mud. In addition, due to its alkalinity and large SSA, red mud has been proved to remove heavy metals from soil or water (Zou et al., 2017). Although the utilization rate has increased, there is still a large amount of red mud accumulated in the landfill.

Fly ash, produced by burning coal in power stations (Kutchko and Kim, 2006), is composed of clay minerals (60–80%), quartz, and pyrite (Gao and Goldfarb, 2019; Li et al., 2017). The main chemical components of fly ash are oxides of Si, Al, and Fe. There are several options for the recovery and treatment of fly ash: 1) Disposal in a landfill after stabilization/solidification. 2) Reuse as part of raw material in a cement kiln. By mixing with cement, fly ash is used as a stabilizer to compress soil blocks or as building materials (Islam et al., 2020). Since a large amount of gas is emitted during the treatment process, the method of recycling as a cement raw material may lead to environmental pollution (Huang et al., 2017b). 3) Reuse as part of aggregate in bricks. Having fine-grained particles, a light texture, and a high water-holding capacity, fly ash is the main cementitious material in dry-mixed mortars. 4) Reuse as alkali by the Waelz process (including granulation, solidification, kiln heating, zinc oxide collection, and slag treatment). Waelz process had the least harm to the environment but cost more. The technology was benefit from the production of zinc oxide, protects the ecosystem, reduced the impact of climate change, and avoids zinc mining. Although there have been some treatment methods for fly ash, there is still a lack of a method with low treatment cost and environmental harm.

Table 1
The current status of MCSWs recycling.

MCSWs	Contained elements/main components	Utilization status	Disadvantages	References
Phosphogypsum	Si, Ca, rich in $CaSO_4$ H_2O	Construction industry	Low recovery rate	(Campos et al., 2017; Soo et al., 2003)
Phosphogypsum	Si, Ca, rich in $CaSO_4$ H_2O	Used for soil amendment	Potential environmental risks	(Kwang-hyun, 2003)
Red mud	Rich in Fe and Al	Extracting Fe and recovering Al_2O_3	High technical difficulty and high cost	(Kutchko and Kim, 2006)
Red mud	Rich in Fe and Al	Construction material	Potential environmental risks	(Joseph et al., 2019)
Fly ash	Si_xO_y	construction material	Pollute the air	(Islam et al., 2020)
Fly ash	Si_xO_y	Stabilizer	Contain toxic elements	(Islam et al., 2020)
Coal gangue	SiO_2, Al_2O_3	Concrete aggregate	Contains harmful ions	(Luo et al., 2020)
Coal gangue	SiO_2, Al_2O_3	Preparation of lightweight aggregate	Contains harmful ions	(Luo et al., 2020)
Coal gangue	SiO_2, Al_2O_3	Improve the performance of cement	Potential environmental risks	(Wang et al., 2009)

Coal gangue, generated during coal mining and washing, is not only encroaches on a huge number of land resources, but also brings potential risks to the ecosystem. Coal gangue is used in power generation as well as in the preparation of sintered bricks and lightweight aggregates (Luo et al., 2020). Coal gangue contains many useful metallic and nonmetallic elements, such as SiO_2 and Al_2O_3 (Wang et al., 2009). Its use in concrete production not only improves cement performance but also increases economic efficiency. When coal gangue is left in the open for a long time, its surface weathers, releasing heavy metal ions (such as As, Pb, Cr), which harm the ecological environment (Sun et al., 2020; Tan et al., 2016).

Although MCSWs can be recycled by various means, the recycled proportion is low in comparison with the amount produced. Since (raw) MCSWs may be harmful to humans, we need to find environmentally friendly and cost-effective ways of recycling these materials. In this regard, an attractive option is to incorporate MCSWs into biochar (Wang et al., 2020a; Wang et al., 2020b). The resultant MCSW-BCs not only greatly reduce the bioavailability of the toxic and harmful elements in MCSWs, but also have been shown to be superior to pristine biochar in adsorbing heavy metals and organic pollutants, and in improving soil conditions (Cho et al., 2019; Yoon et al., 2019b).

3. Preparation methods of MCSW-BCs

MCSW-BCs can be obtained by two means: (1) Mixing the biochar feedstock with MCSW and pyrolyzing the mixture at a certain temperature; (2) Pyrolyzing the biomass and MCSW separately, and mixing the two powders at a certain ratio. The composites prepared by the first method have a larger SSA, porosity, and oxygen functional group content than the pristine raw materials. The yield of modified biochar by the second method mainly depends on the adsorption capacity of biochar for different metal elements. However, the mineral elements may undergo certain changes in morphology during pyrolysis. It was observed that the hematite in the red mud began to convert to magnetite (Fe_3O_4, $2\theta = 30.2$, 35.5, 37.1, 43.3. 57.2 and 62.7°) as the temperature increased (>500 °C), and further transformed into zero-valent iron (Fe^0, $2\theta = 44.7$ and 62.1°) when the temperature increased to >700 °C (Yoon et al., 2019a). So the effect of remixing after pyrolysis may not be as good as that after direct mixing. The method of preparing MCSW-BCs can profoundly influence the physicochemical properties of the composite materials, and hence influence their adsorptive behavior towards different pollutants. Table 2 lists some physicochemical properties of different MCSW-BCs composites. Fig. 2 shows the methods of preparing MCSW-BCs, and Fig. S2 shows the SEM images of the surface morphology of biochar and MCSW-BCs.

4. Adsorption mechanisms

The various mechanisms controlling the adsorption of pollutants to MCSW-BCs are summarized in Fig. 3. (1) SSA and porosity. Rich SSA and pores enable MCSW-BCs to adsorb more pollutants. And with the increase of pyrolysis temperature, the surface porosity of composite materials is increasing. (2) Surface complexation and precipitation. With rich SSA, more pollutants are adsorbed on MCSW-BCs through surface precipitation or complexation, thereby removing them from the environment. The mineral ash that accumulates on the surface of MCSW-BCs is involved in the complexation and precipitation of inorganic pollutants (especially metal cations) and insoluble salts (Sizmur et al., 2017). (3) Cation exchange capacity. Heavy metals in solution can exchange for cations associated with functional groups (carboxyl, hydroxyl groups) on the surface of MCSW-BCs. Since the process is stoichiometric, adsorption efficiency depends on pH. (4) Electrostatic adsorption. It was found that conductive ions were generated after the phosphogypsum modified the distillers grains biochar (PG-BC), which caused an increase in EC. The positively charged PG-BC can adsorb more phosphate by electrostatic interactions (Lian et al., 2019; Wang et al., 2020a). When $pH > pH_{zpc}$, protonation of functional groups on the surface of biochar leads to an increase in surface positive charge, which promotes the absorption by MCSW-BCs of negatively charged pollutant ions such as phosphate. Conversely, when $pH < pH_{zpc}$, the increased anions on the surface of MCSW-BCs promote their electrostatic attraction to cations (most heavy metal ions, ammonium, etc.). (5) Surface functional groups. In the adsorption process based on chemical adsorption, the amount of adsorption mainly depends on the surface functional groups of MCSW-BCs. After MCSWs are mixed with biochar and pyrolyzed, it brings a large number of oxygen-containing functional groups such as hydroxyl, carboxyl and ketone to the composite material. These functional groups can selectively absorb pollutants in the environment through bond interactions. Organic functional groups on the surface of MCSW-BCs can take up pollutants selectively through π-π

Table 2
The physicochemical properties of MCSW-BCs.

Adsorbent	EC ($\mu S\ cm^{-1}$)	pH	Elemental contents								Ash (%)	References
			TC ($g\ kg^{-1}$)	TH ($g\ kg^{-1}$)	TN ($g\ kg^{-1}$)	TS ($g\ kg^{-1}$)	C/N ratio	Ca ($g\ kg^{-1}$)	Fe ($g\ kg^{-1}$)	Al ($g\ kg^{-1}$)		
PG-BC	2950	10.10	40.22	1.54	2.97	5.62	13.56	–	–	–	36.51	(Lian et al., 2019)
PG-BC	–	5.30	28.40	2.70	0.80	8.60	35.50	10.20	–	–	–	(Karim et al., 2018)
RM-SD	–	6.30	–	–	–	–	–	5.40	26.30	5.60	7.50	(Ghanim et al., 2020)
RM-OP	–	5.80	40.80	–	–	–	–	0.20	3.00	2.60	–	(Yoon et al., 2020)
RM-BL350	–	2.0	34.20	3.50	–	–	–	–	12.30	–	–	(Wang et al., 2020d)
RM-BL650	–	2.0	26.00	1.70	–	–	–	–	26.10	–	–	(Wang et al., 2020d)
RM-BL850	–	2.0	23.20	0.80	–	–	–	–	31.50	–	–	(Wang et al., 2020d)
WA-SS	470	9.71	27.48	2.17	2.97	0.93	9.25	–	–	–	62.25	(Mumme et al., 2018)
WA-SS	4040	11.31	13.84	0.99	0.74	1.11	18.70	–	–	–	83.66	(Mumme et al., 2018)
WA-CLO	2280	9.74	20.36	1.18	1.13	1.34	18.02	–	–	–	72.43	(Mumme et al., 2018)
WA-CLO	5710	11.76	11.79	0.67	0.37	1.16	31.86	–	–	–	87.63	(Mumme et al., 2018)
H_2SO_4-FA-BB	5380	7.49	13.86	0.049	0.20	–	–	–	–	–	–	(Mehr et al., 2020)
HCl-FA-BB	9950	3.46	13.16	–	0.25	–	52.64	–	–	–	–	(Mehr et al., 2020)
FA-DP	–	8.92	1.19	0.13	0.87	–	1.37	9.28	26.8	7.55	–	(Lei et al., 2020)
PPG	–	7.72	220.48	–	23.06	16.02	9.56	48.61	46.24	7.70	–	(Yuan et al., 2018)
PPG + DCD	–	7.72	221.59	–	22.49	–	9.85	48.42	47.33	7.80	–	(Yuan et al., 2018)
FA-GW	1470	10.40	276.00	–	9.10	–	30.33	108.60	4.90	2.20	–	(Belyaeva and Haynes, 2011)

PG-BC: biochar modified by phosphogypsum; RM-SD: sawdust biochar modified by red mud; RM-OP: orange peer biochar modified by red mud; RM-BL350: black liquor biochar modified by red mud at 350 °C; RM-BL650: black liquor biochar modified by red mud at 650 °C; RM-BL850: black liquor biochar modified by red mud at 850 °C; WA-SS: wood ash-sewage sludge biochar pyrolysis at 550 °C; WA-CLO: wood ash- compost-like output biochar pyrolysis at 550 °C; H_2SO_4-FA-BB: fly ash-bamboo biochar washed by H_2SO_4; HCl-FA-BB: fly ash-bamboo biochar washed by HCl; FA-DP: dead pig biochar modified by fly ash; FA-GW: green waste compost biochar modified by fly ash; PPG: sewage sludge cornstalks biochar modified by phosphogypsum; PPG + DCD: sewage sludge cornstalks biochar modified by phosphogypsum and dicyandiamide. TC: total carbon content; TH: total hydrogen content; TN: total nitrogen content; TS: total sulfur content.

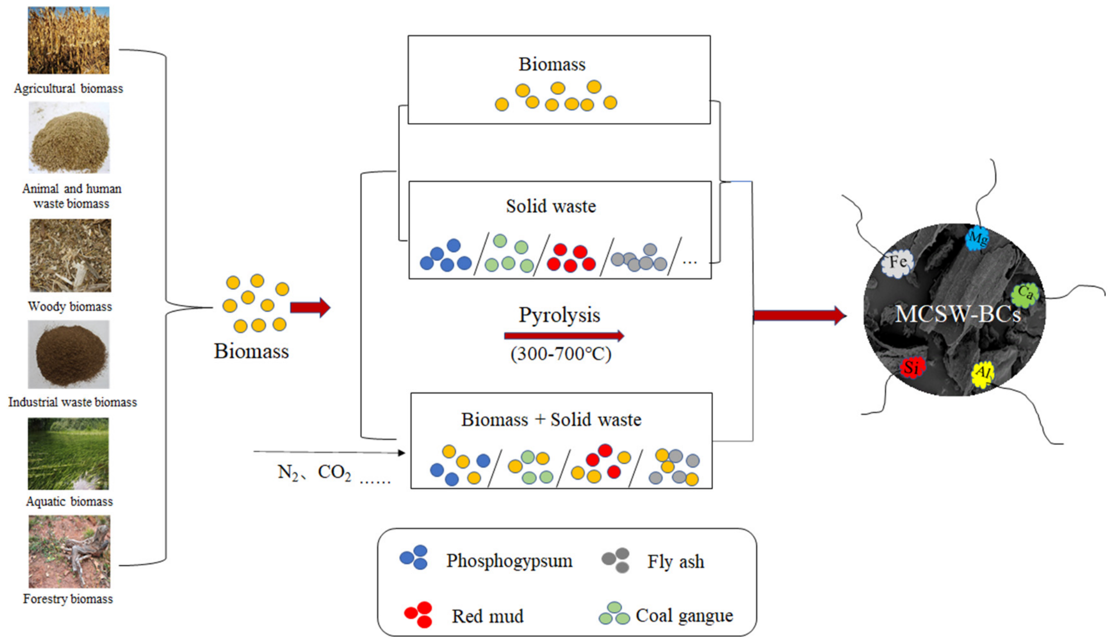

Fig. 2. Preparation methods of MCSW-BCs.

interactions. However, due to the vastly different physicochemical properties of MCSWs, the mechanisms of MCSW-BCs to adsorb pollutants are also different. Therefore, the adsorption mechanisms of different MCSW-BCs to various pollutants are discussed respectively as follows:

4.1. Phosphogypsum

Phosphogypsum was found that co-pyrolyzing with biomass can adsorb anionic pollutants (Wang et al., 2020a). The possible mechanisms for PG-BC to adsorb pollutants are mainly as follows: 1) Cation exchange capacity. Based on its rich elements such as Ca, the introduction of phosphogypsum increases the H/C of biochar, which brings rich oxygen-containing functional and calcium-containing groups to biochar. These functional groups play an important role in adsorption (Chen et al., 2019). According to the FTIR spectra of PG-BC, there is a characteristic peak at 698 cm^{-1}, indicating that Ca is successfully embedded on the surface of PG-BC. By comparing the distillers grains biochar and PG-BC in the FTIR spectra, it is found that PG-BC has an obvious peak at a wave-number of 1065 cm^{-1}, indicating that the modification of phosphogypsum adds C—O functional groups to the biochar (Lian et al., 2019). 2) Electrostatic adsorption. Many biochars are negatively charged, which is not conducive to adsorbing pollutants in the state of anions or oxygen anions. The addition of phosphogypsum also affects the conductive ions on the surface of biochar, which greatly improves the conductivity, promotes cation exchange on the surface, and provides conditions for the adsorption and transfer of heavy metals (Chen et al., 2019). Under acidic conditions, PG-BC is positively charged, which

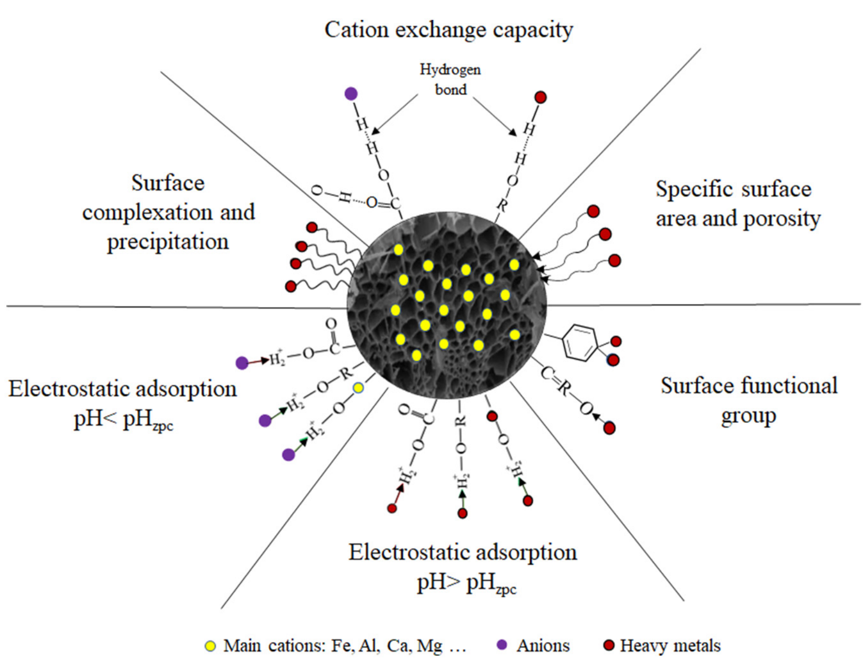

Fig. 3. Various mechanisms involved in the adsorption of pollutants by MCSW-BCs.

promotes the adsorption of negatively charged PO_3^{2-}, $Cr_2O_7^{2-}$ and $HCrO_4^-$, etc. (Lian et al., 2019; Wang et al., 2020a).

4.2. Red mud

Red mud (bauxite residue) is an insoluble fine powdery waste product generated during the production of alumina, which is mainly composed of Si, Al, Fe, and Ti (Ghanim et al., 2020). When a mixture of red mud and biomass is pyrolyzed, the surface of the resultant RM-BC becomes enriched in Fe, Al (Guo et al., 2021) and oxygen functional groups (such as 2-methoxyphenol and 4-ethyl-2-methoxyphenol), which can improve the adsorption capacity of anions, promote carbonization, and act as a catalyst for the degradation of non-persistent organic pollutants (Hassan et al., 2020). The surface enrichment by Fe, Al, and other elements also enhanced the capacity of the composite to adsorb heavy metals through cation exchange. For instance, Cho et al. (2019) used red mud and lignin to prepare multifunctional metal-biochar (MM-BC) composites and evaluated their ability to take up various pollutants. They found that the iron and other substances in the red mud can catalyze the conversion of lignin, thereby forming organic compounds such as 2-methoxyphenol and cresol on the surface of the biochar. These compounds can be adsorbed by complexing with Cr(VI) and reduced to Cr(III) (Cho et al., 2019). In addition, the high alkalinity of RM-BC promotes the adsorption and precipitation of heavy metals, while larger SSA provides more adsorption sites for pollutants. Ghanim et al. pyrolyzed a mixture of dry red mud and wood chips at 700 °C to obtain a RM-BC composite capable of adsorbing V(V) in sewage. They found that the porous, dendritic structure of RM-BC facilitated access of V(V) to surface sites (Ghanim et al., 2020). The abundance of Fe_2O_3, Al_2O_3 is conducive to heavy metal complexation. Wu et al. found that As(V) adsorption of RM-BC was closely related to the Fe_2O_3 and Al_2O_3 content of the composite, indicating that the process was one of surface complexation and electrostatic interaction (Wu et al., 2017). Therefore, RM-BC is a good heavy metal adsorbent.

4.3. Fly ash

Fly ash is a byproduct from the combustion of pulverized coal in electric power plants. This powdery material has excellent fluidity and is rich in Al, Fe, Ca, and Si (Belyaeva and Haynes, 2011). Among them, Si is a major inorganic phase involved in metal co-precipitation. Si can also protect organic phases against thermal decomposition during pyrolysis. Therefore, the Si within MCSW-BCs acts as a co-precipitator of adsorbed heavy metals (Hassan et al., 2020). In addition, because of its low bulk density, high water-holding capacity, and alkaline pH, fly ash is a potentially useful soil amendment (Ram and Masto, 2014). Because of its large SSA and high porosity, FL-BC can control the availability, and reduce the mobility, of heavy metals in soil (Munda et al., 2016). Since fly ash is alkaline and biochar is rich in K and P, the composite material would increase soil pH (Lei et al., 2020), and provide K and P for plant growth when FL-BC is added to the soil. In addition, the aluminosilicate particles in fly ash can immobilize heavy metals.

Several studies have been conducted to study the adsorption mechanisms of different pollutants onto FL-BC composite. Qiu et al. (2019) modified biochar (from pine sawdust) with fly ash, and used the FL-BC composite to remove phosphate from solution. Raising soil pH above 6 through addition of FL-BC, has the effect of reducing the mobility of metal ions, such as Cd(II), in soil under rice (Lei et al., 2020). Yousaf et al. (2017) found that the FL-BC composite, obtained by pyrolysis at 700 °C, had the smallest content of polar functional groups and the lowest H/C and O/C ratios, indicative of a highly hydrophobic and aromatic material. The composite prepared by co-pyrolysis and other methods mixed with biochar is rich in elements such as Si, which changes the functional groups on the surface of the biochar. Therefore, it can immobilize heavy metals and adsorb pollutants. In addition, the alkaline environment of fly ash changes the pH of FL-BC. After being added to the soil, it greatly changes the species of pollutants.

4.4. Other solid wastes

In addition to the phosphogypsum, red mud, and fly ash mentioned above, which have been widely studied, there are also some solid wastes such as steel slag and coal gangue (Wang et al., 2020b; Wang et al., 2020) used to modify biochar.

Research has found that after co-pyrolysis of steel slag and biochar (SS-BC), metal elements such as Ca, Mg, and Fe can adhere to the surface of biochar, and form rich oxygen-containing functional groups. At the same time, due to containing metal salts and Fe_2O_3, etc., adding SS-BC to the soil would significantly increase the pH of the acid soil. The increasing pH of soil changes the composition, structure and content of methanogens of the microbial community, which can reduce the emission of CH_4 in the soil (Wang et al., 2020f).

About biochar modified with coal gangue (CG-BC), due to much SiO_2 with coal gangue, CG-BC could increase the content of Si and Fe on the surface, thereby forming Fe-OP or other oxygen-containing functional groups on the biochar (Wang et al., 2020b). Besides, some alkaline cations such as Mg(II), Ca(II), and K(I) in CG-BC may rise the solution pH. Because of its increased alkalinity and higher decomposition rate, the added inorganic and organic modifiers significantly improve the electrical conductivity, pH, organic carbon content, and surface functional groups, leading to the leaching of heavy metals, increasing precipitation and adsorption capacity of heavy metals (Munir et al., 2020).

5. Factors influencing the adsorption capacity of MCSW-BCs

Many factors are affecting the adsorption capacity of MCSW-BCs. Fig. 4 shows the proportion of factors affecting different MCSW-BCs in environmental applications. According to Fig. 4, adsorption capacity is affected by feedstocks of biochar, modifiers, pyrolysis temperature, ambient pH, dosage, initial concentration of pollutants, and ambient temperature etc. Several main influencing factors are discussed as follows.

5.1. Feedstocks

The feedstocks of biochar can be divided into agricultural waste, animal manure, wood, industrial waste, and aquatic biomass, etc. (Fig. 2). When different feedstocks are used to produce biochar under different pyrolysis conditions, the physicochemical properties of the obtained biochar are usually different, which can affect the agricultural and environmental performance of biochar in practical applications. Algal biochar has higher nitrogen and extractable inorganic nutrients, including

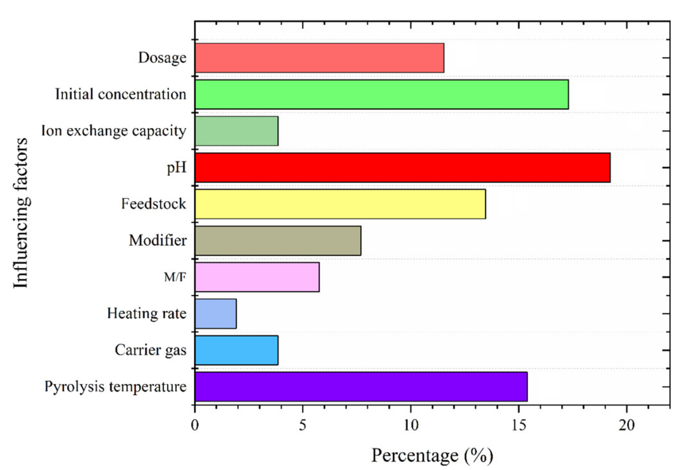

Fig. 4. Proportion of factors affecting different MCSW-BCs in environmental applications (data from 10 literatures) (M/F: Weight ratio of modifier to feedstock).

P, K, Ca and Mg, which provides nutrients for soil and crop productivity (Ok et al., 2020). Li et al. have found that at the same pyrolysis temperature, the pH of agricultural waste biochar is the highest, while that of wood and herbal raw materials biochar is neutral to slightly acidic (Li et al., 2019a). When biochar is used to regulate acid soil, wood biomass could be selected as raw materials. Therefore, suitable feedstocks should be selected according to different application purposes.

5.2. Pyrolysis process

The pyrolysis process is the dominant factor influencing the physicochemical properties of biochar, including SSA, functional groups, hydrophobicity, stability, Zeta potential, and pH (Amen et al., 2020; Hassan et al., 2020; Xia et al., 2019), which determines the adsorptive performance of modified biochar. Table S1 summarizes the different conditions of preparing MCSW-BCs using different raw materials and modifiers, and the adsorption capacity for different pollutants of the MCSWs.

5.2.1. Modifiers

Different MCSWs as modifiers modified biochar can determine their environmental applications. RM-BC rich in Fe and Al has a good adsorption effect on heavy metals such as As(V) (Yoon et al., 2020). The high pH of FL-BC determines that it can reduce the toxicity of heavy metals in the soil and amend acid soil well, and it can also reduce soil nutrients loss (Buss et al., 2019; Ukwattage et al., 2020). Phosphogypsum brings cations to the surface of biochar, which can promote the adsorption of negatively charged ions such as phosphate (Wang et al., 2020a).

5.2.2. Pyrolysis temperature

Pyrolysis temperature is the most significant factor that affects the aromaticity and aromatic condensation, and determines the stability of biochar (Amen et al., 2020; Wang et al., 2016), so do MCSW-BCs. With increasing pyrolysis temperature, the carbon content, aromaticity, pH, ash content, SSA, stability, and pore size increase, while biochar yield, hydrogen content, oxygen content, H/C, and O/C ratios and cation exchange capacity (CEC) decrease (Amen et al., 2020; Hassan et al., 2020; S. Li et al., 2019; Tan et al., 2020). The pyrolysis temperature could affect the adsorption capacity of different pollutants by changing the stability and surface structure of MCSW-BCs. It is found that as the pyrolysis temperature increases, the content of stable fused aromatic ring structures increases, while that of the labile non-aromatic structure decreases in dimension and quantity (Leng and Huang, 2018), which increases adsorption capacity of MCSW-BCs. However, the adsorption capacity of MCSW-BCs is not linearly related to pyrolysis temperature. Wang et al. (2019) used hematite modified pinewood biochar to adsorb Cu(II) and Cd(II), and the optimal pyrolysis temperature was 300 °C (Wang et al., 2019), while Lian et al. (2019) chose phosphogypsum modified biochar (PG-BC) to adsorb Cr(VI) at 600 °C (Lian et al., 2019). In generally, MCSW-BCs prepared under low pyrolysis temperature have more functional groups, which can improve the chemical adsorption, while at high pyrolysis temperature have large SSA, which can promote the physical adsorption. Therefore, when MCSW-BCs are applied to a contaminated environment, the optimal pyrolysis temperature of MCSW-BCs should be selected according to the environmental characteristics to achieve the best remediation effect.

5.2.3. Carrier gas

The surface morphology of biochar under different carrier gas has certain differences in the pyrolysis process. Yoon et al. (2019a) observed the surface morphology of red mud-lignin biochar (RM-BC) pyrolyzed under different gas conditions (N_2 and CO_2). They found that although the average pore diameter of RM-BC pyrolyzed under the two gas atmospheres was not much different, the SSA was significantly different: the measured SSAs of RM-BC produced under N_2 and CO_2 were 134.8 and 184.1 m^2/g, respectively (Yoon et al., 2019a). In addition, the CO produced by CO_2 reduced the formation of H_2, which was more conducive to the thermal cracking of raw materials. CO can act as a reducing agent in the pyrolysis process, reducing the iron in the modifier to zero valence and promoting adsorption (Yoon et al., 2019b).

5.3. Adsorption process and conditions

5.3.1. Ambient pH

Ambient pH not only influences the chemical form of ions but also alters the variable charge on biochar surfaces via protonation and deprotonation of surface functional groups, thus affecting adsorption (Zhang et al., 2020). Truong et al. (2020) found that when pH was 3, FL-BC had the smaller pore structure, thus the adsorption capacity increased. In addition, low pH may increased the Zeta potential of the composite material, resulting in less agglomeration of the adsorbent at pH = 3, and obtained a larger SSA (Truong et al., 2020). Qiu et al. (2019) indicated that the solution pH had a great influence on the speciation and surface charge of the metal ions. At lower pH values, hydrogen ion competition was high, resulting weak removal rate of some metal ions such as Cu(II) (Qiu et al., 2019). Yoon et al. (2020) proved that, at low pH, the dissolution of magnetite/Fe^0 eliminated the adsorption sites of As(V) in RM-BC, thereby limiting its adsorption of As (V) (Yoon et al., 2020). In general, an acidic environment increases the Zeta potential of MCSW-BCs, which in turn increases the cations on the surface. Therefore, the acidic environment increases the adsorption of pollutant ions in the form of anions. However, when the pH is too low, too much hydrogen ions increase the competitive adsorption of some cations (such as heavy metal cations). Similarly, an alkaline environment increases the adsorption of cationic pollutants.

5.3.2. Dosage of MCSW-BCs

As the dosage of MCSW-BCs increases, the adsorption capacity of pollutants increases, on the contrary, the adsorption efficiency decreases, and after reaching a certain value, the increase/decrease trend tends to be flat (Wang et al., 2020a). Therefore, one should not only blindly pursue a higher adsorption capacity and use a high dose of MCSW-BCs, but also find a dosage at the equilibrium point of adsorption capacity and adsorption efficiency as the optimal dosage.

5.3.3. Initial concentration and ambient temperature

The initial concentration of pollutants and ambient temperature could affect the adsorption efficiency. In most papers studying the adsorption of pollutants by MCSW-BCs in water, the influence on the adsorption effect of initial concentration of pollutants and ambient temperature was discussed (Fig. 4). Generally, there is a positive correlation between environmental temperature and adsorption capacity. The higher the ambient temperature becomes, the larger the adsorption capacity is (Wang et al., 2020b). For the concentration of pollutants, at the beginning, the adsorption capacity increased rapidly with the increase of the concentration of pollutants. However, after reaching a certain concentration, MCSW-BCs reached the maximum adsorption capacity, which is almost saturated (Wu et al., 2017).

5.4. Ion exchange capacity

In the process of multi-factor adsorption, different ions could form competitive adsorption. Some ions compete for the adsorption sites on the surface of MCSW-BCs during co-adsorption, resulting in a decrease in adsorption capacity. On the contrary, when ions are adsorbed on the surface of biochar, they would generate new adsorption sites, increasing the adsorption capacity. Yoon et al. (2020) found that the adsorption amount of As(V) increased with the increase of Ni(II) concentration, indicating that Ni(II) provided new adsorption sites for As(V) after being adsorbed on the Fe-biochar composite material (Yoon et al., 2020).

6. Environmental applications of MCSW-BCs

The range and variety of applications of MCSW-BCs are shown in Fig. 5, and Fig. 6 mainly counts the main application ratios of RM-BC (a) and FL-BC (b). The principal applications are in soil remediation and amendment, heavy metal adsorption, and removal of organic pollutants in water.

6.1. Soil amendment and remediation

Because of the favorable general characteristics in terms of pH, CEC, porosity, and SSA, MCSW-BCs can remediate degraded and low-fertility soils by providing or retaining plant nutrients. MCSW-BCs can also adsorb or passivate pollutants in the soil such as heavy metals. In addition, it can amend acidic soil by changing pH.

The ability of MCSW-BCs in sequestering carbon and amending soil is highly dependent on the quality of the biochar and the condition of the soil/land (Fan et al., 2020b). RM-BC prepared by pyrolysis at elevated temperatures can effectively neutralized soil acidity and promote soil nutrient retention (Yoon et al., 2019a), while FL-BC is a good source of plant nutrients and can stabilize organic carbon in soil (Mumme et al., 2018). The addition of FL-BC to soil enhances the physicochemical properties, electrical conductivity, carbon sequestration capacity, and organic matter content (Merino et al., 2017), as well as the size and activity of soil microbial communities (Belyaeva and Haynes, 2011). Adding FL-BC to acidic soils would also raise soil pH, and hence reduce the toxicity of heavy metals such as Al(III) and Cr(VI) (Buss et al., 2019). On the other hand, phosphate and other anions may combine with Ca and Mg in fly ash, and precipitate out. As a result, the vertical movement of P in the soil profile is inhibited, while P retention in the topsoil is enhanced (Hong et al., 2018; Ukwattage et al., 2020). Addition of FL-BC to soil can also reduce demand for nitrogen fertilizers (Munda et al., 2016), increase water-holding capacity of the soil, and moderate the expansion and contraction of clay (Lu et al., 2014). FL-BC application can also change the texture of sandy soils, increase carbon sequestration, improve nutrient retention, and increase soil fertility by promoting ion exchange (Major et al., 2010; Masto et al., 2013).

Addition of MCSW-BCs to the soil reduces the migration rate of heavy metals. Some modified biochar can adsorb heavy metals by a cation exchange process. Zou et al. reported that RM-BC application promoted the microbial reduction of Fe_2O_3, and significantly reduced the concentration of As(V) extractable by $NaHCO_3$ and HCl, causing TOC to increase. RM-BC addition also increased the relative abundance of proteus, while retarding the release of Fe(III) and As(V) (Zou et al., 2017). Raising the pyrolysis temperature to 250 °C has the effect of decreasing the utilization rate of boron (B) in the liquid phase to about 20 mg/kg, and the soluble content of B by 66% (Fan et al., 2018). The application of MCSW-BCs to soil would, therefore, reduce the concentration of boron in soil and its harmful effect on human health (Fan et al., 2018). When applied to soil FL-BC can also reduce the mobility of Cd(II) and its uptake by rice (Lei et al., 2020). Similarly, FL-BC amendment of soil can reduce the risk of Cr(VI) toxicity, salt stress, and potassium leaching in soil (Buss et al., 2019). Some studies did not need to mix biochar and MCSWs into the soil to adsorb heavy metals. Mehmood et al. (2018) added biochar, slag, and ferromanganese slag to the soil, and found that biochar effectively reduced the toxicity of heavy metals, enhanced the tolerance of sesame plants, and increased plant development and biomass yield (Mehmood et al., 2018). In short, FL-BC was applied to soil contaminated by coal mining to reduce the mobility of heavy metals in the soil to stabilize the soil. In addition, it could also be used, while serving as a source of nutrients for plant growth (Mehr et al., 2020). Peng et al. (2020) found that adding a mixture of phosphogypsum and biochar in the soil as a soil amendment is compared to adding only phosphogypsum, the added mixture of soil can reduce the toxicity of As, F and Pb.

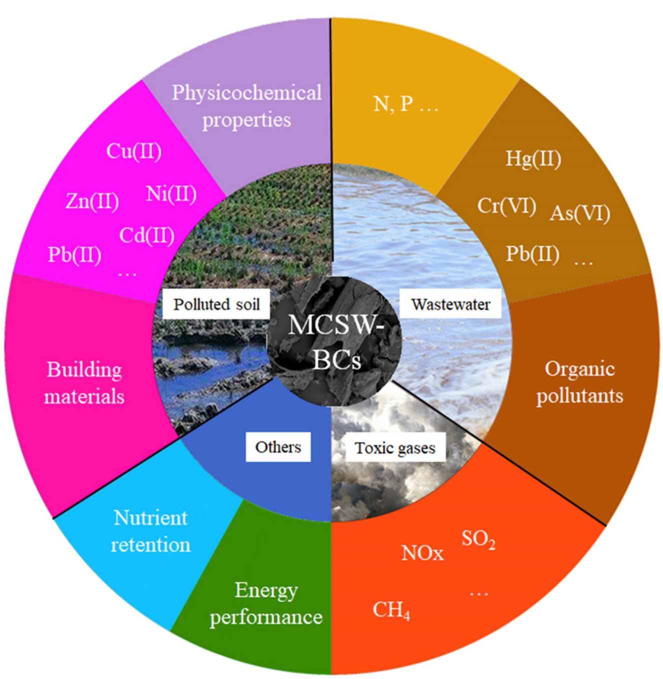

Fig. 5. Environmental applications of MCSW-BCs.

6.2. Wastewater treatment

Compared with conventional biochar, MCSW-BCs show a better adsorption effect on pollutants in water (Du et al., 2019; Ghanim et al., 2020; Qiu and Duan, 2019; Wang et al., 2020a; Wang et al., 2020b). Fig. 8 shows the proportion of papers on MCSW-BCs adsorbing different pollutants in water. It can be seen that MCSW-BCs has been widely studied to adsorb heavy metals, while other pollutants such as organic pollutants are less. Being an efficient adsorbent of phosphate, FL-BC can alleviate the phosphate pollution crisis (Qiu and Duan, 2019). Lian et al. (2019) used PG-BC composite for Cr(VI) adsorption and obtained a maximum adsorption capacity of 157.9 mg/g, which was more than twice that observed for distillers grains biochar. Similarly, RM-BC has a large capacity for removing heavy metals, such as V(V) and Cu(II), from wastewater (Ghanim et al., 2020; Qiu et al., 2019). Du et al. (2019) reported that a composite of corn straw loaded with zero-valent iron and red mud (ZVI@GRM) could adsorb up to 149.42 mg/g Pb(II) and 37.14 mg/g Cr(VI) from wastewater. The amount of heavy metals adsorbed by different modifiers on biochar is also different. Many experiments prove that compared with the adsorption capacity of unmodified biochar and some other modifiers such as nano-zero-valent iron (NZVI) to Pb(II) and Cr(VI), MCSW-BCs show higher adsorption capacity. Fig. 7a and b compare the maximum adsorption capacity of Cr(VI) and Pb(II) with different MCSW-BCs. In general, MCSW-BCs are more efficient than unmodified biochar in adsorbing heavy metals (e.g., Pb(II) and Cr(VI)). In this regard, PG-BC and RM-BC are the most effective, showing a maximum capacity of 157.90 and 223.14 mg/g, respectively. As a result, MCSW-BC has been widely studied in the treatment of pollutants in wastewater, especially heavy metals.

6.3. Other applications

Besides being effective in soil amendment and wastewater treatment, MCSW-BCs are good catalysts for the synthesis of biodiesel. Compared with conventional catalysts RM-BC can promote biodiesel synthesis at greatly reduced temperatures, while increasing product yield, and reducing production cost (Yoon et al., 2019b). MCSW-BCs have good catalytic performance for the reforming of biomass tar (Guo et al., 2019). Input the alkaline or acidic biochar into alkaline

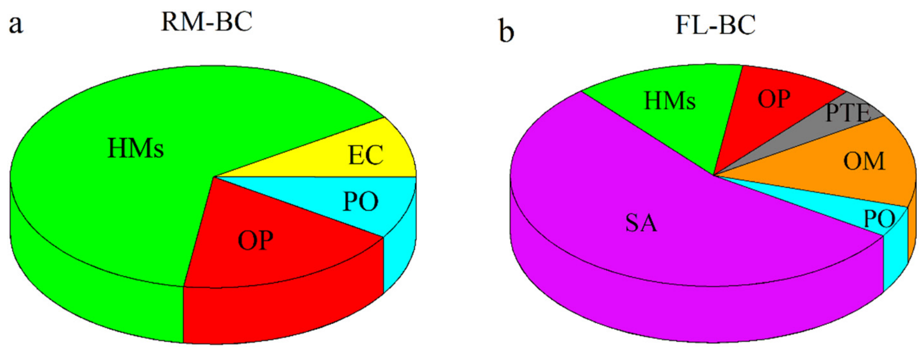

Fig. 6. The application ratios of RM-BC (a) and FL-BC (b) (data from 46 literatures in Web of Science).
PO: adsorption of phosphate; SA: soil amendment; HMs: adsorption of heavy metals; OP: adsorption of organic pollutions; EC: effective catalyst for biodiesel synthesis; PTE: reduction of the volatilization potential of PTE; OM: other materials or catalysts.

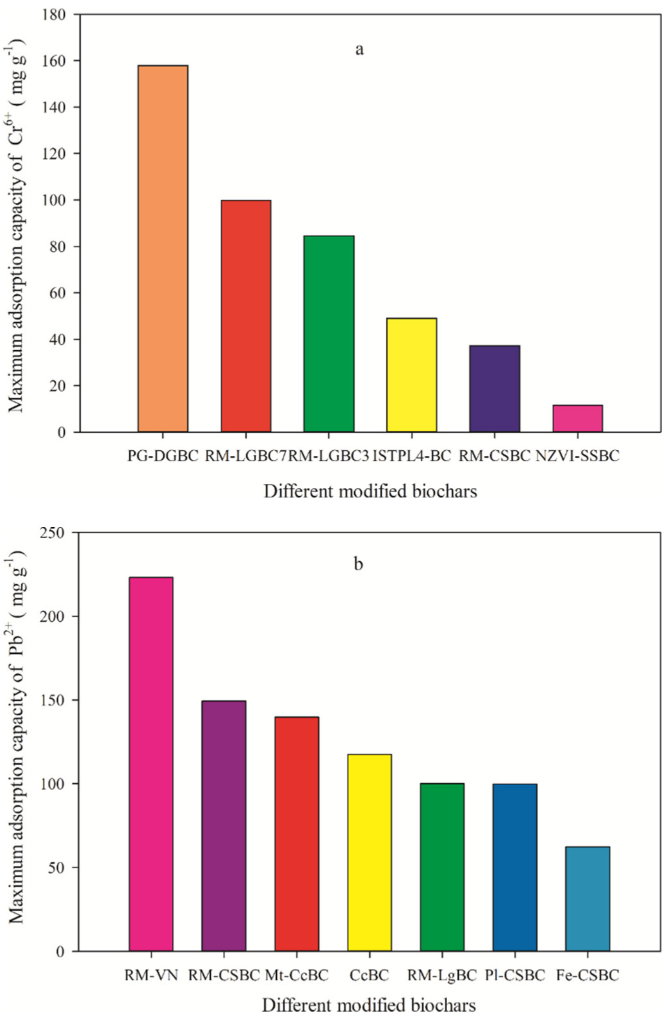

Fig. 7. The maximum adsorption capacity of biochar modified by different substances for Cr(VI) (a) (Cho et al., 2019; Du et al., 2019; Lian et al., 2019; Liu et al., 2020; Liu et al., 2020; Mishra et al., 2020) and Pb(II) (b) (Chen et al., 2020; Cho et al., 2019; Du et al., 2019; Fan et al., 2020; Fu et al., 2020; Kazak and Tor, 2020).
PG-DGBC: distillers grains biochar modified by phosphogypsum; RM-LGBC3: lignin biochar modified by red mud at 380 °C; RM-LGBC7: lignin biochar modified by red mud at 700 °C; RM-CSBC: corn stalks biochar modified by red mud; NZVI-SSBC: sewage sludge biochar modified by NZVI; ISTPL4-BC: switchgrass biochar modified by *Actinobacterium Zhihengliuella* sp. ISTPL4; RM-VN: vinasse biochar modified by red mud; PM-CSBC: corn stalks biochar modified by red mud; Mt-CcBC: corncob biochar modified by montmorillonite; CcBC: corncob biochar; RM-LgBC: lignin biochar modified by red mud; Pl-CSBC: corn stalks and stems biochar modified by polyethylene; Fe-CSBC: crab shell biochar modified by Fe_3O_4 nanoparticles.

bauxite slag sand significantly changed the effectiveness of nitrogen in bauxite slag sand (Rezaei Rashti et al., 2019a, 2019b; Rezaei Rashti et al., 2019c). The modified biochar can also promote the conversion of biomass into electricity for making microbial fuel cells (Jia et al., 2018). By improving combustion performance during burning, fly ash, peanut husks and other crop wastes can greatly reduce the volatility of potentially toxic elements (PTE) such as As, Ba, Bi, Cd, Cr, Ni, Pb, Sb, Sn, and Zn, and hence the risk of environmental contamination by PTE. The combustion of FL-BC can potentially provide clean energy by reducing the emission of gases that produce acid rain (Yousaf et al., 2017). The addition of biochar to a mixture of cement and fly ash can enhance CO_2 adsorption by cement, and hence increase the strength of the cement (Praneeth et al., 2020). Further researches into the synthesis and properties of MCSW-BCs would provide valuable information on the ability of these composite materials to improve soil fertility, take up heavy metals, and generate energy.

7. Conclusions and future perspectives

This review represents a systematic summary of the preparation methods, physicochemical properties, influencing factors and capacity for adsorbing pollutants of MCSW-BCs together with their applications in remediating polluted soil and water. The use of MCSWs to modify biochar is an attractive way of turning solid wastes into an environmentally sustainable resource. To realize the large-scale applications of MCSW-BCs, however, the following issues need to be resolved.

Firstly, biochar has been detected with toxic organic compounds at a lower pyrolysis temperature (Lyu et al., 2016), which may cause potential environmental toxicity risk (Godlewska et al., 2021; Shi et al., 2021). After co-pyrolysis of MCSWs with biomass, the toxicity risk of MCSW-BCs is still unclear. Further researches should focus on reducing the environmental risks of MCSW-BCs applications. For example, Wang et al. suggested coating composites with calcium alginate before application to reduce the risk of pollutants being released into the environment (Wang et al., 2018b; Wang et al., 2018c).

The second issue relates to the stability of the biochar component in that its structure is subject to slow biological degradation and abiotic oxidation. As a result, the physicochemical properties of aged biochar would differ from those of its pristine counterpart (Wang et al., 2020; Wang et al., 2021). Thus, the long-term stabilities of MCSW-BCs need to be checked.

The third issue concerns the removal of biochar-adsorbed pollutants. For example, FL-BC can reduce the utilization of heavy metals in the soil system, but ultimately cannot successfully separate its heavy metals from the soil. Some literatures have reported that the applications of FL-BC to soils polluted by coal mining can effectively reduce the mobility of heavy metals in soil (Mehr et al., 2020). It is not clear, however, whether the immobilized heavy metals can be released when both

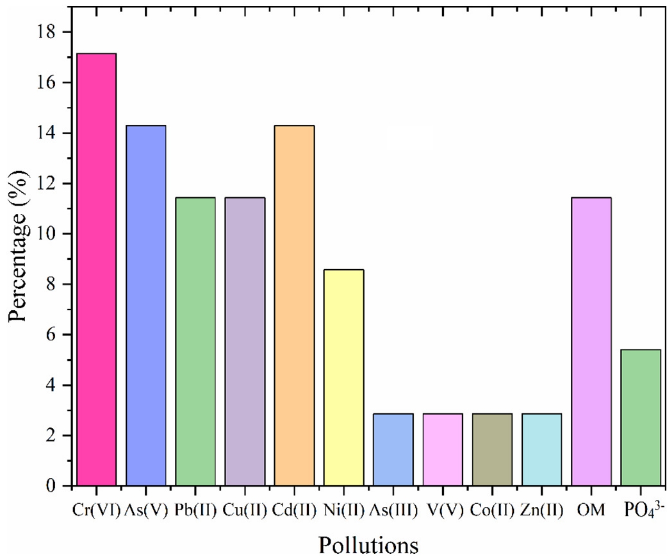

Fig. 8. Proportion of papers on MCSW-BCs adsorbing different pollutants in water (data from 46 literatures) (OP: Organic pollutants).

soil pH and the structure of modified biochar change over time. The retention of heavy metals, adsorbed to modified biochar, merits further investigation.

Statistics of 46 literatures on MCSWs modified biochar found that most of the studies focused on how to adsorb heavy metals in wastewater and the soil amendment (Fig. 8). The applications in the adsorption of organic pollutants, especially emerging pollutants such as antibiotics, are very little. Therefore, which MCSWs modified biochar can obtain higher adsorption capacity for emerging pollutants should be the next research direction.

It also seems worthwhile to examine the formation and properties of biochar modified by coal gangue, iron slag, manganese slag, steel slag, and chromium slag in relation to the potential of MCSW-BCs for recycling as well as the suitability of such composites for environmental remediation, soil amendment, and sewage treatment.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.149295.

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