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The properties and potential utilization in agriculture of fly ash from a typical biomass-fired power plant

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ABSTRACT: The rapid expansion of biomass power plants has led to the generation of enormous amounts of ash, which urgently requires appropriate treatment. In this study, the fly ash from a typical actual biomass power plant was sampled and characterized. The major/trace element composition and leaching characteristics of fly ash were analyzed; the environmental risk was assessed by the risk assessment code (RAC), and the fertilizer availability of fly ash was discussed. The results showed that the fly ash was rich in nutrients and accumulation of harmful trace elements, which may be related to the volatility of elements during combustion. Sequential chemical extraction was employed to reveal the chemical speciation of trace elements. Except for residue fraction, Cd, Zn, and Pb in fly ash mainly exist in carbonate fraction, As was found in Fe-Mn oxide fraction, and Cr, Ni, and Cu were mainly in organic matter fraction. The result of RAC demonstrated that Cd in fly ash needs special attention during utilization. The samples have a high leaching amount of K, Na, and Ca but low leaching amounts of Cr, Ni, Cu, Zn, Cd, As, and Pb. This indicates that fly ash can be used as fertilizer. The concentrations of trace elements were far below the upper limit prescribed by the standard, which indicates that the fly ash was suitable for direct use in agriculture and forestry due to the low contents of toxic elements. The results can provide reference for the management and utilization of fly ash from biomass.

KEYWORDS: biomass power plants, fly ash, trace elements, chemical speciation, potential ecological risks

INTRODUCTION

In recent years, the energy crisis and environmental pollution have become serious increasingly, and the development of renewable energy has attracted worldwide attention [1]. Compared with traditional fossil fuel power generation, biomass energy is favored by many countries for its renewable and green, and accounting for 11% of the world's energy consumption. By 2050, biomass can meet 33% to 50% of the world's current major energy consumption [2]. China is rich in biomass energy resources, which provides enormous development potential for the biomass power generation industry, and the biomass power plants in China will continue to maintain rapid development [3].

During the power generation process of biomass power plants, biomass is converted into eco-friendly energy through pathways such as combustion, gasification, pyrolysis, and incineration. With the significant increase in biomass power plants and the rapid development of biomass energy production, the amount of ash by-products generated has significantly increased and has now become a serious global problem [4]. It was estimated that the biomass burned is 7 billion tons and the average ash yield calculated on a dry basis is 6.8%, then the world may produce approximately 476 million tons of biomass ash annually [5]. Therefore, the disposal or comprehensive utilization of ash has become an important consideration for the daily operation of biomass-fired power plants.

Biomass direct combustion power generation is the main way of utilizing biomass in the world. Biomass power plants that burn biomass as raw materials generate ash including bottom ash and fly ash. Extensive research has been conducted worldwide to develop different methods for utilizing biomass ash appropriately and effectively [6, 7]. In some developed countries such as Germany, Japan, Denmark, etc., the resource utilization rate of biomass ash is between 70% and 90% [8]. Biomass ash has a wide range of applications, including agricultural soil improvement, road construction, cement, ceramic materials, and catalytic applications [7, 9, 10]. From the perspective of sustainable utilization of biomass energy, applying biomass ash to agricultural and forestry soils was generally considered the best approach [11–13]. The ash of biomass power plant was mostly alkaline, containing nutrients (such as P, K, Si, Ca, Mg, etc.); it also contained various harmful elements (As, Cd, Cr, Cu, Pb, Ni, Zn, etc.) [14]. Therefore, it is necessary to evaluate the environmental impact of biomass ash before its application to agricultural and forestry soils [15]. The bottom ash contains more nutrients such as K, Mg, Ca, P, etc., while the content of trace elements is relatively low and can generally be directly applied in agriculture and forestry [16]. Although the fly ash also contains nutrients, due to its fine particles, harmful trace elements released during biomass combustion are easily enriched in fly ash [17]. In order to prevent ash accumulation, slagging, sintering, and corrosion, except for straw, the actual fuel of biomass direct-fired power plants in China is mixed with wooden biomass such as waste wood. Compared with straw, wood has a high content of trace elements, which leads to the fly ash formed containing a relatively high level of trace elements. Trace elements have potential impact on the environment and limit the application in agriculture and forestry of fly ash. It is necessary to evaluate the nutritional value and environmental impact characteristics of biomass ash before its application to agricultural and forestry soils [18]. The leaching characteristics of nutrients are generally used to represent the fertilizer value of fly ash, while the leaching characteristics of trace elements indicate the harmfulness of fly ash [19]. The environmental behavior and biological toxicity are not only related to the total amount and mineral composition but also closely related to the fractionations of the trace elements.

The most commonly used analysis methods for trace element forms include the Tessier's five-step continuous extraction method [20] and the BCR method [21, 22]. The occurrence forms of trace elements are divided into exchangeable forms (F1), carbonate-bound fraction (F2), Fe-Mn oxide-bound fraction (F3), organic-bound fraction (F4), and residual fraction (F5) based on the former method. Exchangeable fractions are very sensitive to environmental changes, are easy to migrate and transform, and can be absorbed by plants. Carbonate-bound fraction refers to some trace elements bound by carbonate precipitation, which is easy to release and enter the environment when the pH value drops. Fe-Mn oxide-bound fraction is generally formed by the presence of mineral outer capsules and fine dispersed particles, active iron manganese oxides with a large specific surface area, and adsorption or co-precipitation of anions. Organic-bound fraction refers to the entry or encapsulation of trace elements in particulate matter in different forms onto organic matter particles, forming sulfides or chelation with organic matter. Residual fraction is not easily released under normal natural conditions and is difficult for plants to absorb [23]. Many studies have demonstrated that the characteristics of biomass ash in China mainly focus on the impact of fuel and combustion processes [24–26], but there is little research on the occurrence forms of trace elements in fly ash. Therefore, necessary analysis and evaluation of the composition and characteristics of fly ash is a prerequisite for its application [27].

Despite the significant environmental impacts of trace elements in fly ash, it is imperative to analyze the components of fly ash and to predict the potential environmental impacts related with trace elements in fly ash. The main objectives of this study are to (1) determine the physical and chemical characteristics of fly ash; (2) analyze chemical composition, leaching characteristics, and chemical morphological characteristics of trace elements in fly ash; and (3) assess the potential utilization in agriculture of fly ash. The results will provide significant guidance for the management and application of fly ash from biomass-fired power plants.

MATERIALS AND METHODS

Biomass fly ash samples were collected from a typical biomass combustion power plant in Huainan city, Anhui province, which adopts direct combustion power generation technology, with an annual power generation capacity of 2.1×10^8 kwh/a. Burning a mixture of straw and waste wood as fuel (wheat straw:corn straw:forestry waste = 4:4:2), with fuel consumption of approximately 260,000 tons per year. The annual fly ash production of the power plant is 5.67×10^3 tons. All collected samples were stored in sealed bags and sent to the laboratory, dried at 105 °C for 3 h, grounded, and passed through a 100-mesh sieve for application.

The microstructure of biomass ash was tested using Quanta200 scanning electron microscopy, and the crystal structure was analyzed by X-ray diffraction (XRD, D8 Advance, Bruker, Germany). The total organic carbon (TOC) was measured in accordance with the standard of soil improvers and growing media – Determination of organic matter content and ash (EN 13039-2000). The pH and conductivity are measured with references to the EU standard of EN 13037-2000 and EN 13038-2000 method, respectively. The measurement of elemental composition is based on the EU standards for the determination of major elements (EN15290-2006) and trace elements (EN 15297-2006) in biomass fuel. The sample was digested with mixed acid H_2O_2-HNO_3-HF. Chemical speciation is divided into 5 forms, and the sequential extraction method [21] was modified and improved to analyze the different speciation of the trace elements (Table 1). The contents of nutrients and trace elements were measured by inductively coupled plasma optical emission spectrometry (ICP-OES) and inductively coupled plasma mass spectrometry (ICP-MS), respectively.

The leaching characteristics of major nutrients and trace elements were tested with reference to the Chinese standard "Toxic leaching method for solid waste leaching: Horizontal oscillation method" (HJ557-2009). After mixing the sample and deionized water in a ratio of 1:10, shaking for 8 h, and then stewing for 16 h, the obtained leachate was filtered with a 0.45 µm filtration, and the concentration of elements were determined by ICP-MS. Standard addition recovery, blank test, and parallel sample measurement were employed for quality control. And the recovery of the standard solution is between 85% and 115.6%.

The risk assessment code (RAC) method [28] was adopted to evaluate the potential risk and mobility based on the different occurrence fractions of trace elements. According to the results of sequential extraction method, the RAC method considers the exchangeable and carbonate-bound fractions of trace elements as effective states, which are easily migrated and transformed, absorbed and utilized by plants, and have high

Table 1 Sequential extraction procedure for metal fractionation analysis.

Sequence	Fractionation	Extractant	Extractant condition
i	Exchangeable fraction (F1)	1.0 mol/l $MgCl_2$ (pH = 7.0) solution	25 °C oscillation for 1 h
ii	Carbonate fraction (F2)	1 mol/l CH_3COONa (pH = 5.0)	25 °C agitated continuously for 5 h
iii	Fe-Mn oxide fraction (F3)	0.04 mol/l $NH_2OH \cdot HCl$ solution	Water bath at 96 °C for 6 h, 3500 r/min centrifugation for 20 min
iv	Organic matter fraction (F4)	0.02 mol/l HNO_3+30% (v/v) H_2O_2 (pH = 2.0) solution	Water bath at 85 °C for 2 h
v	Residual fraction (F5)	$HCl+HNO_3+HClO_4$	Oscillation in 100 °C water bath at 250 r/min for 2 h

bioavailability [29]. The formula was calculated as follows:

$$\mathrm{RAC} = \frac{\mathrm{F1}+\mathrm{F2}}{\mathrm{C_t}} \times 100\%,$$

where F1 is the concentration of trace element in exchangeable fraction, F2 is the concentration of element in carbonate fraction, and C_t is total concentration of trace element in 5 fractions. Mobility and availability were then classified on a scale of 1 to 5 based on the RAC value: (1) RAC < 1%, no risk; (2) 1% < RAC ⩽ 10%, low risk; (3) 10% < RAC ⩽ 30%, medium risk; (4) 30% < RAC ⩽ 50%, high risk; and (5) RAC > 50%, very high risk.

RESULTS AND DISCUSSION

The properties of ash samples are presented in Table 2. Moisture, ash content, TOC, pH, and conductivity have mean value of 2.87%, 82.74%, 17.90%, 8.96, and 24.38 mS/cm, respectively. TOC reflects the combustion status of fuel in the boiler, and the higher it is, the lower the burnout rate of fly ash particles. The pH reflects the concentration of acid and alkali ions in fly ash, while the value of conductivity reflects the concentration of metal ions (such as Na^+, K^+, Ca^{2+}, Mg^{2+}, etc.) and anions (such as Cl^-, SO_4^{2-}, HCO_3^-, etc.). Generally, the higher the salt concentration in fly ash, the higher the conductivity value of the leaching solution. The results showed that the fly ash has strong alkalinity, as indicated by its pH value. As the main component of fly ash, the residual carbon has various characteristics similar to biochar such as high-temperature combustion, good stability, and adsorption ability [30]. It can adsorb trace elements and organic pollutants in soil, improving soil quality. In addition, it can also supplement the loss of organic carbon in the soil, thereby improving soil fertility and increasing crop yield [31]. When biomass combustion ash is used in agriculture and forest soil, the pH value and conductivity value are used together to characterize the activity of the ash. High pH and conductivity values mean that the salt in the ash dissolves quickly, which may cause harm to plants. It is generally realized that fly ash cannot be applied when pH > 13, while pH < 13, the dosage of ash application is further determined based on electrical conductivity [32]. The pH value results of this study indicated that it can be used in agriculture and forestry, and it can adjust the pH of soil and slow down soil acidification [12].

The SEM images of the fly ash samples with different magnifications are shown in Fig. 1. It demonstrated that the shape was extremely irregular and uneven. The shapes of the particles varied significantly. It can also be seen in the SEM image of high magnanimity that the surfaces of all particles are uneven, the small particles are attached to the large particles, and there are some gaps between the convex part and the sphere.

The typical XRD patterns of the as-prepared samples are displayed in Fig. 2. As shown in Fig. 2, the characteristic peaks of SiO_2 phase can be obviously observed at $2\theta = 25°\sim30°$. On the one hand, silicon is the main component element in biomass fuel [33]; on the other hand, biomass fuel is often mixed with sand and stone during the combustion process. Therefore, some sand and stone substances remain in the fly ash. As a result, the characteristic peak of the SiO_2 phase in the ash is clearly observed. The fly ash of the biomass fuel also contains albite minerals, which may be generated by the reaction of Na with Al_2O_3 and SiO_2 in the fuel at high temperatures [34].

The concentrations of major elements in fly ash samples are presented in Table 3. Fly ash is mainly composed of major elements (Mg, K, and Ca) and trace elements (Cr, Ni, Cu, Zn, Cd, As, and Pb). The high contents of Si, K, and Cl in ash reflect the typical characteristics of rice straw biomass ash. Si is the main ash forming element in rice husks and straw, while K, Na, Ca, Mg, and P are the essential nutrients for plant growth. The high contents of Si, Ca, and Mg reflect the

Table 2 Properties of fly ash samples.

Item	Moisture (t%, ar)	Ash content (%, db)	TOC (%, db)	pH value	Conductivity (ms/cm)
Minimum	2.39	81.62	17.34	8.48	23.40
Maximum	3.26	83.64	18.39	9.60	25.30
Mean	2.87	82.74	17.90	8.96	24.38
Std. Deviation	0.28	0.69	0.30	0.40	0.75

Note: ar, as received basis and db, dry basis.

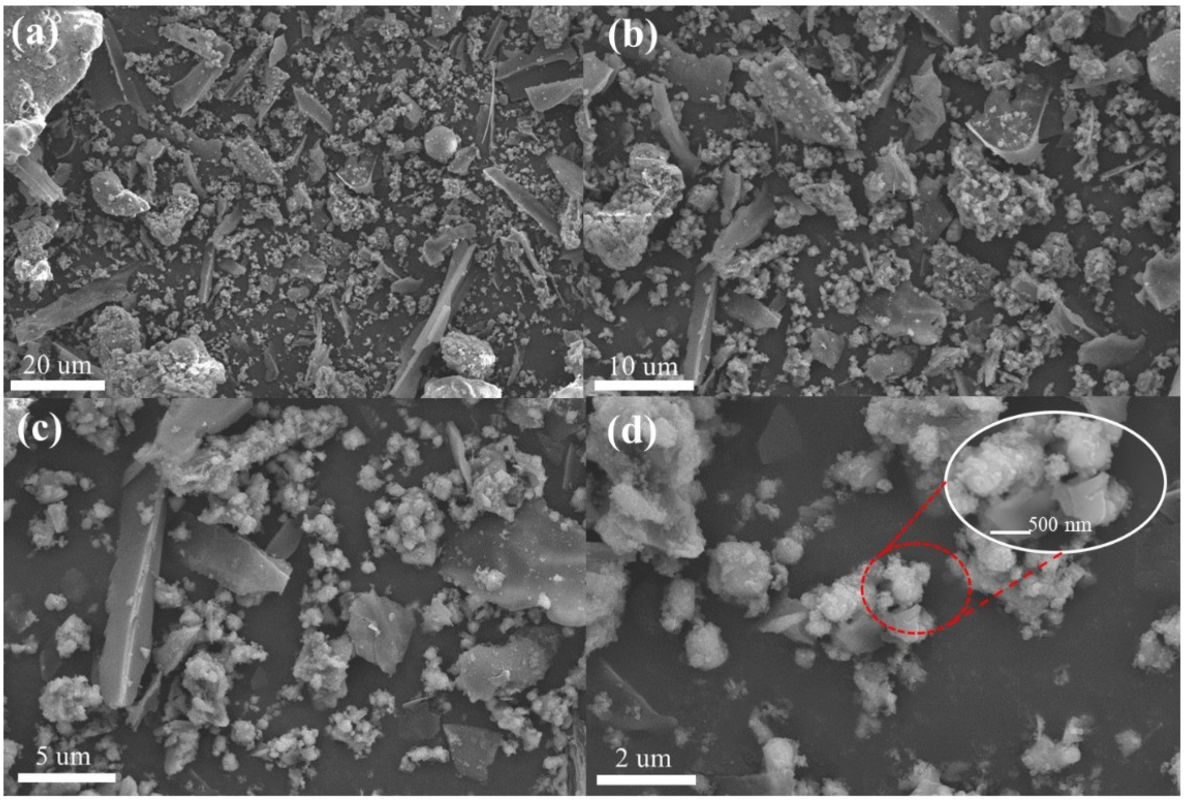

Fig. 1 Scanning electron micrograph of biomass fly ash.

Table 3 Contents of elements and leaching amount (mg/kg, db).

Item	Element	Biomass fuel	Fly ash		Leaching solution		
			Content	Enrichment factor	Leaching amount	GB8978 -1996	Leaching rate
Major element	P	–	4864		7.73		0.16%
	Si	–	89471		16.28		0.02%
	Mg	–	92364		539		0.58%
	Ca	–	58329		6580		11.28%
	Al	–	6327		2.47		0.04%
	Na	–	4765		611		12.83%
	K	–	59826		25492		42.61%
Trace element	Zn	188.92	362.18	1.92	0.006	2	0.00%
	Cu	19.23	51.62	2.68	0.006	0.5	0.01%
	Pb	21.36	50.36	2.36	0.180	1	0.36%
	Cd	0.52	2.24	4.31	0.001	0.1	0.04%
	Ni	78.93	80.62	1.02	0.049	1	0.06%
	Cr	2.74	52.65	19.22	0.072	1.5	0.14%
	As	4.59	20.15	4.39	0.053	0.5	0.26%

Note: db, dry basis.

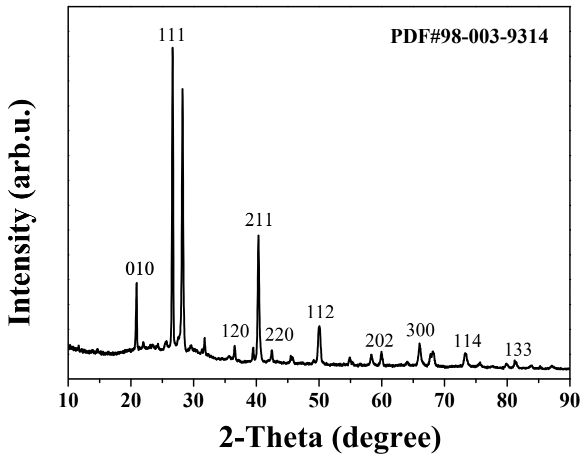

Fig. 2 X-ray diffraction spectrum of biomass fly ash.

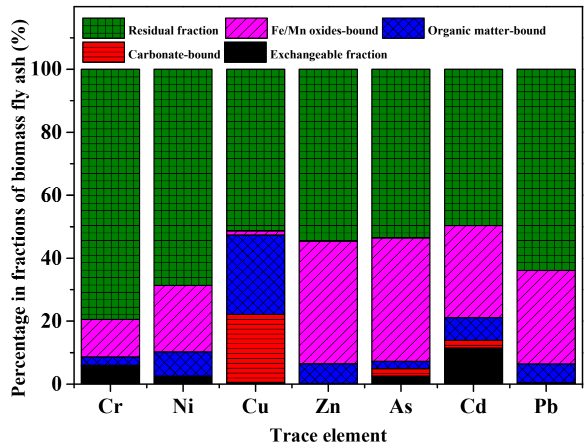

Fig. 3 Percentage of trace elements in different fractions in biomass fly ash.

characteristics of woody biomass ash. They have a high content in woody biomass but low volatility, which may be due to the fine wood fuel particles carried by the flue gas being crushed during the combustion process to form small-sized fly ash. The fuel in the power plant contains waste wood, resulting in a high content. P, K, and Na are volatile elements, so they are easily volatilized into the flue gas during combustion. During the cooling process of the flue gas, fine fly ash particles are formed or deposited on the surface of particles, resulting in a high content in the fine fly ash [35]. The content of Al in biomass fuels is generally low, and the higher Al content in fly ash may be related to the addition of impurities such as clay during the harvesting, processing, transportation, and treatment of the fuel in the furnace. High content of K and other elements lead to high conductivity, while high Ca content leads to higher pH values. Despite some differences, the contents of K, Na, Ca, Mg, and P in ash were relatively high, and the application of fly ash in agricultural and forestry vegetation or soil is very valuable.

The contents of trace elements in fly ash are also demonstrated in Table 3. The concentration of trace elements in fly ash was low, and their maximum values were far lower than the Chinese standard (GB/T23349-2009). The enrichment factor (EF) was characterized by the element content in biomass ash to that in biomass fuel. The EF values are shown in Table 3, and the descending order is: Cr > As > Cd > Cu > Pb > Zn > Ni. Xu et al [29] have demonstrated that Pb, Cd, As, and Zn are more enriched in fly ash than in slag, and it is speculated that they evaporate during the combustion process and condensed on the surface of fine particles.

The leaching characteristics of elements were further tested and also presented in Table 3. From the perspective of the leaching amount of each nutrient element, the order of element leaching amount is K > Na > Ca > Mg > P > Al > Si. The main elements in the leaching solution are high, while the content of trace elements is extremely low, so the fly ash can be directly applied as fertilizer in agricultural and forestry soils, which is consistent with the conclusions drawn from previous studies [36, 37]. The leaching characteristics of nutrients and trace elements can also provide a basis for the resourceful utilization and harmless treatment of fly ash.

All the leaching amount of trace elements is lower than 1%. The order of leaching amounts for different trace elements is Cr > Zn > As > Pb > Ni > Cd > Cu. As shown in Table 3, the results show that the leaching solution is below the contents required in the limit standard of the trace elements in the Integrated wastewater discharge (GB8979-1996). The biomass fly ash can be classified as the first type of industrial solid waste. The leaching experiment of harmful trace elements in biomass fly ash shows that the leaching amount of these elements is within the standard limit, which has the feasibility of being applied to agricultural and forestry soils.

Determination of chemical partitioning is of great significance, which can reflect the transferability of trace elements in ash and predict the possibility of potential environmental impact. Various fractionation phases of trace elements have different toxicity, bioavailability, and environmental behaviors [21]. The trace element content in various fractionation phases of fly ash is shown in Fig. 3. The trace elements mostly exist in the form of residues and in the lattice of silicates, primary, or secondary minerals [22]. The residual fraction is considered to be in an inactive state and difficult to release under natural conditions.

All trace elements—Cr, Ni, Cu, Zn, As, Cd, and Pb—were found to be highest in the Residual fraction. The fraction which contained the second highest

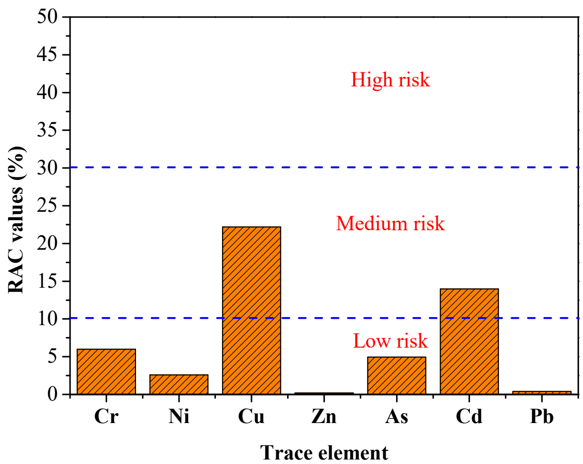

Fig. 4 Environmental impacts of trace elements in biomass fly ash.

amount of trace elements was the Fe-Mn oxide-bound fraction with the exception of Cu which was found in high amounts in the organic matter and carbonate-bound fractions. The exchangeable fraction contained mostly low amounts of trace elements (Fig. 3).

The proportion of carbonate-bound states in Cu was higher than that of other elements, and the carbonate-bound state is easily released with acidic conditions. Cd was found in a higher proportion of exchangeable states compared to other elements, and elements in this state are more prone to release. If the content is high, there may be certain environmental risks. The content of trace elements in fly ash is relatively low, but there is a possibility of precipitation in the natural environment. It is inferred that the amount of precipitation poses a certain pollution risk to the environment and requires a potential risk assessment.

Based on the content of exchangeable and carbonate-bound fraction, the RAC code method was used to assess the potential ecological risk. The higher the value of RAC, the greater the ecological risk, and vice versa [29]. As presented in Fig. 4, the RAC values followed in the order of Cu (22.20%) > Cd (13.97%) > Cr (6.00%) > As (4.94%) > Mn (2.78%) > Pb (0.40%) > Zn (0.19%). The results demonstrated that Cu and Cd pose a medium ecological risk (10%∼30%), while other elements belong to low risk (1%∼10%). Therefore, attention should be paid to the environmental risks of Cu during the usage of biomass ash, which was consistent with the results reported by Xu et al [29].

The main nutrients in fly ash are K and P, which have certain fertilizer availability. According to the leaching test results, K was relatively easy to leach out, so it can be used as K fertilizer. Although P in biomass fly ash is difficult to dissolve, its application to agricultural and forestry can promote crop absorption of P and increase the content of soluble P in the soil. Fly ash contains almost no N that promotes plant growth. When applied to mineral soils with low N content, a certain amount of N fertilizer needs to be applied at the same time. When fly ash is applied as fertilizer, certain treatments are required, such as adding appropriate amounts of potassium, phosphorus, and nitrogen fertilizers, processing them into coarse particles, and reducing chlorine content.

Table 4 Limiting concentrations of trace elements of China's agricultural fly ash contamination control standard (mg/kg).

Element	Maximum allowable content	
	pH < 6.5	(pH ⩾ 6.5)
Cd	5	10
Ni	200	300
Cr	250	500
As	75	75
Cu	250	500
Pb	250	500
Zn	–	–

Table 5 Limiting concentrations of trace elements in biomass ashes used for agricultural land and forest land (mg/kg).

Element	Denmark	Norway	Sweden	Finland	Austria
Cd	5/15	30	30	1.5/7.5	5/8
Ni	30	70	70	100/750	100
Cr	100	100	100	300	250
As	–	30	30	25/30	–
Cu	–	400	400	600/700	250
Pb	120	300	300	100/150	250/100
Zn	–		7000	4500	1000/1500

Denmark: left value for straw ash/right value for wood ash; Norway: limit values applicable to forestry; Sweden: limits valid only for the utilization on forest soils. Finland and Austria: left limits for utilization on agricultural soils/right limits for utilization on forest soils.

The main impact of fly ash on the environment is caused by trace elements, so the content of trace elements is an important factor in evaluating the utilization value of fly ash, and it is also a limiting factor in evaluating the fertilizer availability of fly ash. At present, there are no relevant standards or specifications for the application of biomass ash (including agricultural and forestry) in China. This study refers to the upper limit values for the contents of major heavy metals in China for the application of fly ash to agricultural and forestry soils (GB 8173-1987) (Table 4), as well as those in major European countries [37] (Table 5).

As shown in Table 4 and Table 5, the contents of trace elements are within the limit standard set by China and other countries. The contents of harmful trace elements and the leaching amount of trace elements in fly ash were relatively low, indicating that only a small portion of trace elements has mobility and bioavailability when applied to agricultural and forestry soils. Therefore, the possibility of causing pollution to such soils was relatively low [38]. Using an appropriate amount of biomass ash will not significantly increase the available content of trace elements in the soil but will instead increase the soil pH and has a passivation effect on trace elements in the soil [39, 40]. After alkaline biomass ash is applied to soil, it can change the occurrence form of trace elements in the soil, reduce the migration ability of trace elements, and thereby reduce the available trace element contents of plants, achieving the remediation of trace elements contaminated soil [41]. The leaching characteristics of nutrients showed that fly ash has fertilizer value, and the leaching amount of harmful trace elements is within the standard limit. Moreover, the content of harmful elements in biomass fly ash is very low, and its application in agricultural and forestry soil will not significantly increase the effective content of soil trace elements [42]. Therefore, attention should focus more on the amount of the utilization of biomass fly ash.

CONCLUSION

The results of sequential chemical extraction showed that the trace elements mostly exist in the form of residues, and the proportion of carbonate-bound states in Cu was higher than that of other elements, and Cd had a higher proportion of exchangeable states compared to other elements. The leaching characteristics of nutrients show that fly ash has certain fertilizer value, and the leaching amount of trace elements is all within the standard limit. Biomass fly ash can be applied to soil. The results of RAC demonstrated that Cu and Cd pose a medium ecological risk, while other elements belong to low risk. The trace elements in biomass fly ash were below the limit value in standard and can be used directly in soil improvement for agricultural and forestry fields. Therefore, attention should be paid to the utilization of biomass fly ash. Further research is needed on the ash generated by different biomass and combustion technologies.

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