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REMOVAL OF NITROGEN FROM WATER USING BIOCHAR BASED PASSIVE SYSTEM

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Abstract

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Abstract

Biochar is a stable carbon-rich product formed from thermochemical conversion of biomass in an anoxic condition. It has a great potential in adsorbing nutrients such as nitrogen and other inorganic matters. Working as an effective adsorbent, biochar has received a lot of attention. Its special chemical structure enables biochar to have good adsorption capacity, which can absorb nutrients (like nitrogen and phosphorus) and inorganic ions (Cu^{2+}, Pb^{2+} and Hg^{2+}, etc.) in water, soil or sediment, as well as polar or non-polar organic compounds. Thus, biochar has a strong adsorption capacity.

The theoretical part of the thesis is based on the existing literature review, focusing on the influencing factors of biochar production methods and factors which will affect adsorption capacity. Two types of biochar (H5 and UEF) were used to test the effect of removing nutrients from mine water and landfill water in the column on a laboratory scale. Under the monitoring by piezometer, biochar were filled in filtration columns during the experiments and the results were obtained by measuring the content of nutrients and assessing the performance of biochar.

Keywords

biochar, adsorption, nitrogen

CONTENTS

1 INTRODUCTION

Nowadays, because of the great demand of farming and increase use of fertilizers, eutrophication of groundwater and surface water has become a serious environmental problem. Excessive amounts of nitrogen and phosphorus in the environment has seriously affected the environment and human health.

Biochar is a stable carbon-rich product formed by thermal decomposition of biomass such as agricultural and forestry wastes under anoxic conditions. It has a certain removal effect on ammonia nitrogen in water with high removal efficiency, high flexibility, and good economic and environmental benefits, but its adsorption capacity for high ammonia nitrogen wastewater is limited.

Biochar has received widespread attention as an adsorbent that efficiently adsorbs and removes nutrients. The surface adsorption mechanism is the main mechanism of nitrogen adsorption. In the process of adsorbing nitrogen, biochar is affected by many factors, such as the added amount of biochar, the concentration of nitrogen to be adsorbed, pH, and specific surface area and volume of biochar, contact time and reaction temperature. Therefore, these factors should be noted in the adsorption experiment of biochar to achieve better adsorption (Dai et al. 2020).

2 BIOCHAR ADSORPTION MECHANISM

Nowadays, due to the augment of discharges of nutrients like nitrate into the natural environment from industrial, agricultural, and household wastewater all over the world, the problem of nutrient enrichment and eutrophication has aroused a worldwide serious environmental concern. Eutrophication may stimulate the rapid growth of organisms, especially algae, leading to the depletion of dissolved oxygen and the deterioration of the aquatic environment. Besides, high levels of NO_3^- in water may affect human health. Therefore, the removal of excessive NO_3^- from wastewater has important ecological and social significance (Zhou et al. 2019).

Usually, there are three kinds of technologies used for removing NO_3^- from wastewater, which are based on physical, chemical and biological aspects. Considering the cost of processing and the generation of other pollutants, physical treatment is the most common choice at the moment. Physical treatment includes methods such as electrodialysis, reverse osmosis and adsorption (Zhou et al. 2019). Compared with electrodialysis and reverse osmosis, adsorption is a widely accepted and useful method for dealing with wastewater in situ because of its relatively low cost, high efficiency and smaller possibility of secondary pollution (Bhatnagar and Sillanpää 2011). Therefore, it is of great potential value to study cost-effective adsorbents to remove NO_3^- from wastewater.

Biochar is a substance with a high aromatization structure and its outer layer is an oxygen-rich structure formed by different carboxyl and phenolic functional groups, biochar has high biochemical and thermal stability (Zhou, Wu and Wu 2015). The specific surface area and porosity of biochar are large, the pore size varies from nanometre to micrometre and carboxyl group, phenolic hydroxyl group, carbonyl group, lactone, pyrone, acid anhydride, etc. are the major groups of biochar. The presence of groups makes biochar have good adsorption capacity, which can absorb water, the inorganic ions in soil or sediment (Cu^{2+}, Pb^{2+} and Hg^{2+}, etc.) and polar or non-polar organic compounds. Therefore, biochar has a strong adsorption of pollutants, not only can adsorb heavy metals in soil and water, but also can adsorb organic and inorganic pollutants in soil and water. In terms of chemical properties, biochar has a high degree of aromatization and carboxylic acid esterification. In addition to containing organic carbon such as polycyclic aromatic hydrocarbons and aliphatic compounds, there are minerals such as calcium and magnesium and inorganic carbonates in biochar. Therefore, biochar has strong thermal stability and anti-biodegradability. Biochar is difficult to dissolve in water, and at the same time it has strong adsorption capacity, strong biological stability, anti-oxidation and other characteristics, making it potential material for soil improvement, reduction of greenhouse gas emissions and environmental pollution restoration. Biochar is widely used in industrial, agriculture, energy, environment and other fields as well. (Li 2019, 142.)

2.1 Mechanism of N adsorption

Biochar's ability to widely and efficiently adsorb NO_3^- in the environment is mainly attributed to its characteristics. The high specific surface area, porosity, polar and non-polar surface parts of biochar are the main reasons why biochar can physically adsorb nitrate. Besides, in the experiment of adsorbing nitrate by modified activated carbon, it was found that the modified granular activated carbon had increased ion exchange capacity when the pH was in the range of 3 to 6 and the adsorption capacity decreased when pH increased. When pH was greater than 6, ion exchange was the main adsorption mechanism, because as the pH value increased, the negative charge concentration increased, the electrostatic adsorption effect decreased. The adsorption isotherm analysis at pH = 6.5 shows that the adsorption of NO_3^- by the adsorbent is mainly through ion exchange (Yadav et al. 2019). Some scientists also used the biochar-loaded nano-zero-valent iron combined system to remove NO_3^-. It was found that the modified biochar can promote the reduction of nitrate by catalytic reduction and conversion, and produce N_2 and NH_4^+, thereby removing nitrate (Wu et al. 2019).

During the process of dehydroxylation/dehydrogenation and aromatization, many functional groups (e.g., carbonylic, carboxylic, hydroxylic and phenolic hydroxyl groups) are formed on biochar (Li et al. 2019). These functional groups play essential roles in nitrate adsorption because ion exchange occurs between NH_4^+/ NO_3^- and functional groups (Liu et al. 2010). The removal efficiency of NH_4^+ and NO_3^- is positively correlated with the number of acidic and basic functional groups, respectively (Wang et al. 2015). Some scientists analyzed the zeta potential of the adsorbed biochar and found that the zeta potential is negative, indicating that the biochar surface is negatively charged, and its potential is about 0.82–1.19 mmol/g (Shaaban et al. 2014). Such a structure is easily combined with NH_4^+ ions. Meanwhile, NH_4^+ ions can be fixed on biochar by the oxygen-containing functional groups such as hydroxylic group, carboxylic group and sulfonic acid group on its surface through the following process:

$$- OH + NH_4^+ \rightarrow - O - NH_4$$
$$- SO_3H + NH_4^+ \rightarrow - SO_3 - NH_4$$
$$- COOH + NH_4^+ \rightarrow - COO - NH_4$$

The adsorption mechanism of ammonia nitrogen is mainly due to cation exchange, surface complexation and the formation of magnesium ammonium phosphate precipitation (Shang 2019, 9). In addition, there are some positively charged ions on the surface of the biochar, such as Na^+, Mg^{2+} ions, which can remove NH_4^+ ions from the environment (Dai et al. 2020).

3 FACTORS AFFECTING BIOCHAR ADSORPTION CAPACITY

The surface area of biochar, the properties and initial concentration of adsorbates, the pH value of the solution, the temperature, the interfering substances, and the properties and quantity of adsorbents are among the factors that will influence the adsorption rate. Usually, the adsorption is proportional to the surface area available for adsorption, but the magnitude varies (Naeem, Westerhoff and Mustafa 2007). As a result, materials with a more porous and larger surface area will generally adsorb more because of an enhancement to its mass transfer mechanism, but other factors, for instance, the chemical composition, size and morphology of the adsorbed material are related as well.

The physicochemical nature of the adsorbent strongly impacts on adsorption rate and adsorption capacity. Fine powder adsorbents have higher adsorption capacity than those consisting of large particles due to the increase in surface area (Kara et al. 2007). The presence of surface functional groups promotes chemical interactions, and thus the adsorption capacity can be enhanced by increasing the concentration of appropriate functional groups, however, this may influence negatively based on the composition of the functional group. The presence of cations or anions on the surface of the adsorbent may enhance its ion-exchange capacity, which is an important mechanism in the removal of heavy metals (Premarathna et al. 2019).

Since adsorption occurs in the pores of the adsorbent, the molecular size of the adsorbate is also related to the adsorption rate and capacity. Therefore, the rate is controlled by intra-particle transport. As the molecular weight of the adsorbate molecule decreases, the adsorption rate will increase accordingly (Zhang and Huang 2007).

Moreover, pH has a significant impact on the adsorption process, because it directly affects the chemical form and surface charge of the pollutant, as well as the ionization or morphology of the adsorbate (Li et al. 2017). Incorporation of a material in the preparation of the composite material may affect the change in the pH_{pzc} (Ismadji et al. 2016).

The adsorption reaction is an exothermic reaction; therefore, the adsorption depends on temperature and usually increases with temperature (Premarathna et al. 2019). Biochar pyrolysis temperature can affect the type and amount of functional groups as well as the capacity of biochar for ion exchange (Cheng et al. 2006). An increase in pyrolysis temperature decreases the quantity of acidic functional groups (Nguyen and Lehmann 2009; Zeng et al. 2013). Specifically, compared with biochar derived at a high pyrolysis temperature, biochar obtained at a low pyrolysis temperature has more negatively charged acidic groups (carboxyl and hydroxyl) and shows better complexation with NH_4^+ (Hollister et al. 2012; Zhang et al. 2014). In addition, adsorption may be influenced positively or negatively by the presence of interfering organic or inorganic compounds (Premarathna et al. 2019). Meanwhile, the presence of natural organic matter may have an impact on the adsorption, relying on the nature of the adsorbate, the effect may be positive or negative. Furthermore, the thesis published in CNKI (Shang 2019, 9) describes that the presence of Na^+ and Ca^{2+} inhibits the adsorption of ammonia nitrogen by cow dung biochar. When the concentration of Na+ and Ca^{2+} in water is the same, the order of influence on the adsorption of ammonia nitrogen is $Na^+ > Ca^{2+}$.

3.1 Unmodified biochar

The amount of biochar used takes an essential part in the adsorption of nitrogen by biochar. Many studies have shown that increasing the amount of biochar can improve the removal rate of ammonia nitrogen (Xie 2015), but the dosage of biochar should be controlled within a certain range as well (Dai et al. 2020).

Nitrogen concentration is also an influencing factor of nitrogen adsorption by biochar (Dai et al. 2020). In general, within a certain range of ammonia nitrogen concentration and the same amount of biochar, as the ammonia nitrogen concentration decreases, the ammonia nitrogen recovery rate gradually increases. The limiting factor of biochar's adsorption of ammonia nitrogen is that its adsorption capacity is too low, so it’s adsorption capacity for high ammonia nitrogen wastewater is limited (Xie 2015).

Solution’s pH value also plays a role in the experimental process of biochar adsorption of nitrogen (Antunes et al. 2018; Jiang et al. 2015). The initial pH value of the solution has a significant influence during the process of adsorbing nitrogen. According to the Ma (2018), researchers using corn stalk biochar to adsorb NH_4^+ observed the effect of pH on the adsorption efficiency. It was found that the initial pH value of the solution had a great influence on the adsorption of ammonia nitrogen by the biochar. When the pH value was less than 7, it had little effect on the adsorption efficiency of ammonia nitrogen; when the pH value of the solution was 9, the adsorption efficiency of ammonia nitrogen was significantly increased, which reached 39%; when the pH value was greater than 9.0, the adsorption efficiency quickly decreased, which was related to the nature of the corn stalk biochar adsorbent itself, and also due to the existence of ammonia N in the solution (Dai et al. 2020; Ma 2018).

In addition to the amount of biochar added during the adsorption process, the initial concentration of nitrogen and phosphate, the influence of pH, the specific surface area and volume of the biochar, the contact time and the reaction temperature also have an effect (Dai et al. 2020). The specific surface area and volume of biochar have an influence on its adsorption capacity. Scientists have found that as the surface area and pore volume of biochar increase, NH_3 volatilization increases (Mandal et al. 2018). Nevertheless, not all biochar adsorption capacity increases with the increase in surface area and volume. Other researchers found that the adsorption capacity of biochar made up of wood and rice husk decreased as the particle size of biochar increased (Kizito et al. 2015). Further research is still needed in this area.

3.2 Modified biochar

The adsorption capacity of modified biochar for nitrogen and phosphorus is considerably higher than pristine biochar, because the modified biochar has larger specific surface area, more reaction activity and more surface functional groups. (Xiang et al. 2020).

Taking magnetic biochar as an example, the main factors influencing the adsorption efficiency of magnetic biochar include pyrolysis temperature, material type, modifier type, dosage of adsorbent, pH value, reaction temperature, contact time, initial pollutant concentration and competitive anion (see Fig. 1) (Li et al. 2019).

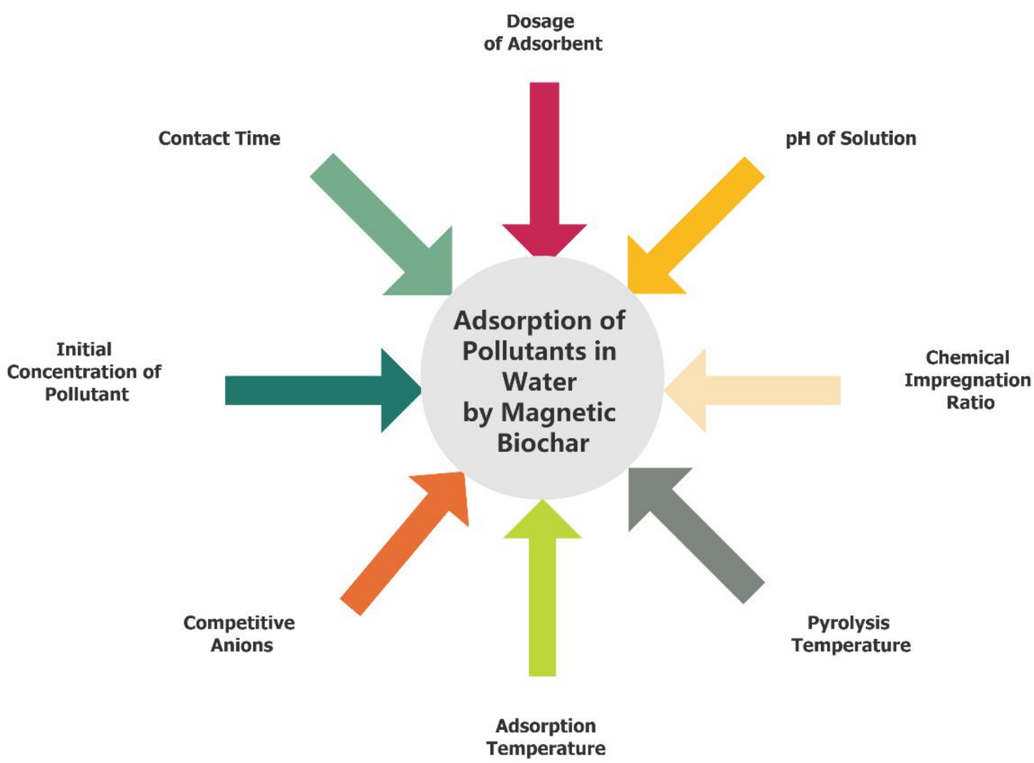

FIGURE 1. Factors affecting the adsorption of pollutants by magnetic biochar (Li et al. 2019.)

The yield and adsorption capacity of carbon will be influenced by the ratio of raw materials to magnetic materials during the impregnation process. After evaluating the adsorption of heavy metals by magnetic biochar under different impregnation ratios of raw materials to magnetic materials, Yap et al. obtained the result: as the impregnation ratio of raw materials to magnetic materials increased from 0.25 to 0.50, the carbon yield increased from 67.37% to 88.98% (Yap et al. 2017). However, once the impregnation rate was higher than 0.50, the generation of carbon reduced. When the ratio of biochar to $FeCl_3$ was 0.5, the best adsorption capacity of magnetic biochar and the highest yield of activated biochar derived from coconut shell could be obtained (Li et al. 2019).

The optimum ratio of biochar to $FeCl_3$ can also promote the development of pores, which can lead to a significant increase in specific surface area and pore volume. The influence of chemical impregnation rate on the removal of water pollutants by magnetic biochar is complicated, and each magnetic carbon sample needs to be analyzed separately (Li et al. 2019).

Moreover, Yin et al. (2018) suggested that both physical and chemical sorption were mechanisms for ammonium adsorption. A study by Wang et al. (2020) had proved that biochar modified with $FeCl_3$ and a mixture of $FeCl_3$ and HCl could improve ammonium adsorption in aqueous solutions not only by increasing the surface area and the abundance of oxygen functional groups of biochar but also by promoting electron transfer in the adsorption process. It was indicated that the modification methods enhanced biochar's adsorption capacity for ammonium by at least 14%, due to increased abundance of C-OH and O-C=O functional groups and larger specific surface area, and the development of electron shuttle with the Fe^{3+}/Fe^{2+} redox coupling as well (Feng et al. 2019). Consequently, it can be concluded that chemical modification of wheat straw biochar using $FeCl_3$ and HCl increased the effectiveness of biochar for the treatment of ammonium-contaminated wastewater (Wang et al. 2020).

3.3 Other influencing issues

In addition to the amount of biochar added during the adsorption process, the initial concentration of nitrogen and the influence of pH, the specific surface area and volume of biochar, contact time and reaction temperature also have its own effect (Dai et al. 2020). The specific surface area and volume of biochar make a difference to its adsorption capacity. Some researchers found that ammonia volatilization increased while the surface area and pore volume of biochar increased (Mandal et al. 2018), whereas not all the adsorption capacity of biochar increases with its surface area and volume. Other scientists found that the adsorption capacity of biochar made up of wood and rice husk decreased as the particle size of the biochar increased (Kizito et al. 2015). Therefore, further research is needed in this area.

Another factor during the process of biochar adsorption of nitrogen is contact time. The influence of contact time on the adsorption process mainly relies on the reaction mechanism (Dai et al. 2020).

The reaction temperature also plays a role in the process of nitrogen adsorption in biochar. A large number of studies have found that within a certain range, the denitrification effect increases with the rising of temperature (Dai et al. 2020).

In most cases, especially for actual wastewater, there are not merely target ions but many other ions, that is, coexisting ions, in the adsorption solution. Therefore, for the practical application of biochar, it is crucial to determine whether and how coexisting ions affect the adsorption of target ions. Several studies have been carried out on the use of biochar to remove nitrogen from model wastewater (a synthetic solution in which target ions and other ions coexist) or actual wastewater (Yin et al. 2017).

In short, there are many factors that affect the adsorption of nitrogen by biochar, but there are still some factors that have not been explored or fully studied (such as coexisting ions) and are expected to be improved in future experiments (Dai et al. 2020).

4 REMOVAL OF NUTRIENTS FROM WATER USING BIOCHAR

Biochar can also adsorb nutrients such as nitrogen and phosphorus in the aqueous phase (Zhang et al. 2012; Yao et al. 2013; Zhang and Gao 2013). Ammonium, nitrate, and phosphate are common forms of reactive nitrogen and phosphorus in wastewater, which can bring about eutrophication (Yao et al. 2012; Xu et al. 2018).

Pre-treatment of biochar feedstock have a positive effect on the adsorption of nutrients. For example, the digested sugar beet tailing biochar showed the highest phosphate removal ability with a removal rate around 73% (Yao et al. 2011). The pretreatment can be carried out during plant growth. Additionally, biochar produced from wood waste pretreated with magnesium oxides (Mg-biochar) was used to recover ammonium and phosphate (Xu et al. 2018). The struvite precipitation on the surface of biochar is the major mechanism for the removal of ammonium and phosphate. Other reports have also shown that modified biochar can remove nitrate (NO_3^-), total Kjeldahl nitrogen (TKN), total nitrogen (TN), total phosphates (TP), and phosphate (PO_4^{3-}) from aqueous solutions (Mohan et al. 2014; Usman et al. 2016; Sun et al. 2018; Vikrant et al. 2017). A general conclusion is that these modifications change the surface chemistry of biochar, thereby resulting in enhanced nutrients sorption capacity compared with the original biochar (Xiang et al. 2020).

Different from the pre-treatment of biochar feedstock which shows a significant effect on phosphorus adsorption, biochar's post-treatment has a remarkable effect on ammonium adsorption. Oxidized maple wood biochar is regarded to have higher ammonium adsorption capacity than maple wood biochar (Wang et al. 2016). Besides, the pyrolysis temperature affects the adsorption of ammonium. Due to the higher H/C and O/C ratios and the presence of more functional groups on its surface, biochar produced from pine wood chips at 300°C is regarded as the highest NH_4^+ adsorption capacity (Yang et al. 2017). This study shows that the chemical bond and polar interaction between NH_4^+ and surface functional groups may be the mechanism to enhance NH_4^+ adsorption (Xiang et al. 2020).

4.1 Adsorption mechanism of nutrients

In the process of nitrogen and phosphorus adsorption, biochar is mainly influenced by the distribution mechanism, surface adsorption mechanism, combined action mechanism and other microscopic mechanisms. The surface adsorption mechanism is the primary mechanism of nitrogen and phosphorus adsorption (Dai et al. 2020). In the process of absorbing nitrogen and phosphorus, biochar is affected by many factors, such as the amount of biochar added, and the adsorption concentration of nitrogen and phosphorus, pH, specific surface area and volume, contact time, reaction temperature (Xie 2015; Mandal et al. 2018; Antunes et al. 2018).

Biochar can be divided into carbonized parts and uncarbonized parts after preparation. When biochar interacts with pollutants, the interaction between the uncarbonized part and the pollutant belongs to distribution, while the carbonized part belongs to surface adsorption, such as ion exchange, electrostatic adsorption, pores, H-bond, hydrophobic interaction (see Fig. 2) (Dai et al. 2020). Surface adsorption refers to the adsorption process formed by intermolecular attraction (physical adsorption) or chemical bonding (chemical adsorption) between the adsorbed surface and the adsorbed substance (Dai et al. 2020). Studies have shown that the adsorption of nitrate and phosphate by modified biochar not only meets the physical adsorption characteristics, but also conforms to the second-order kinetic reaction equation, the secondary or quasi-secondary kinetic reaction. Therefore, the process of biochar adsorption of nitrate and phosphate is chemical adsorption. (Dai et al. al.2020.) Therefore, these factors should be paid attention to in the adsorption experiment of biochar in order to achieve a better adsorption effect.

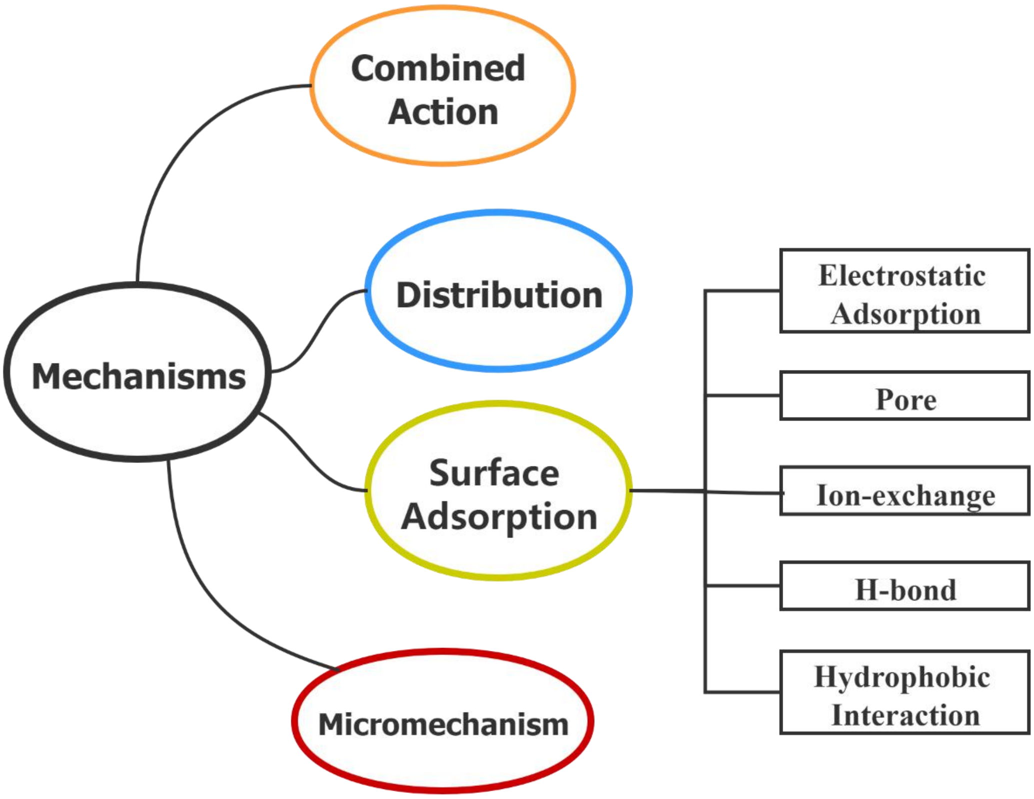

FIGURE 2. Mechanism of removing N and P from biochar in environment (Dai et al. 2020.)

Many scientists have also conducted experiments to explore the mechanism by which biochar removes N and P from water. The mechanism was ion exchange by removing the nitrate from the heterotrophic denitrification system via redox-active biochar (Wu et al. 2019). The adsorption mechanism of phosphate by biochar is different from the adsorption mechanism of nitrogen, which is mainly related to the content of Ca, Mg, Fe, Al and other substances on the carbon material itself. All kinds of cations on the surface of biochar combine with phosphate to form insoluble phosphate precipitate, thus realizing phosphorus removal. Studies have shown that Fe and Al are more likely to react with phosphate under acidic conditions, while Ca and Mg are more important under alkaline conditions (Xie 2015).

4.2 Results about nutrients removal

In order to explore the internal reasons for the difference in the adsorption performance of different feedstocks of biochar to nitrogen and phosphorus, the chemical composition and surface properties of three kinds of biochar were analyzed by scientists. The component properties of the three kinds of biochar were shown in Table 1.

It can be seen from Table 1 that bamboo biochar has a greater difference in chemical composition from the other two types of biochar. Bamboo biochar has the lowest nitrogen content, that is, the background value of the N element in bamboo biochar is the lowest (Lu, Tian and Wang 2019).

TABLE 1. Component properties of 3 kinds of biochar (Lu et al. 2019.)

Feedstocks	Ash Content /%	pH	N Content (*w*)/%
Reed	13.74	7.97	0.98±0.045
Wheat	12.59	7.73	1.20±0.052
Bamboo	7.12	7.53	0.52±0.031

Derived from experimental data, overall, the removal rate of NH_4^+-N by the three types of biochar is maintained below 4.00%, and the removal rate of PO_4^{3-}-P by the three types of biochar is 10.00%~20.00%. The removal effect of P is better than that of NH_4^+-N; secondly, bamboo biochar has the best adsorption effect for NH_4^+-N among the three types of biochar, with a 6-hour removal rate of 3.59%, followed by wheat biochar (Lu et al. 2019).

Bamboo biochar has the best adsorption effect on NH_4^+-N, mainly because the background value of nitrogen in bamboo biochar is relatively lower, and its low absolute value of zeta potential and pH value are both conducive to the adsorption of ammonia nitrogen (Lu et al. 2019).

Moreover, the adsorption capacity of biochar prepared from four raw materials (straw, reed, sawdust and eggshell) at different pyrolysis temperatures for ammonia nitrogen was analysed by Xu et al. (2019), and the results showed that when the pyrolysis temperature of straw and sawdust biochar was 500 °C, the adsorption performance of ammonia nitrogen was the best, reaching 4.2 mg/g and 3.3 mg/g respectively. Hale et al. (2013) used cocoa husks and corncobs as raw materials to prepare biochar at a lower pyrolysis temperature of 300-350 °C and found that it also had a significant adsorption effect on ammonia nitrogen, but the adsorption coefficient Kd was small.

5 PRODUCTION OF BIOCHAR

Biochar can be made from biomass waste, which is an environmentally friendly and economical option. Biomass waste materials suitable for biochar production include crop residues (field residues and processing residues, such as nut shells, fruit pits, bagasse, etc.), as well as yard, food and forestry wastes, and animal manures. Currently, a large amount of agricultural, municipal and forestry biomass is burned or decomposed, and then releases CO_2 and CH_4 into the atmosphere. They may also pollute local groundwater and surface water, which is a big problem with livestock waste. Hence, the use of these materials to make biochar can not only remove it from the pollution cycle but also obtain biochar as a by-product of energy production from the biomass. Raw materials must not contain unacceptable levels of toxins, such as heavy metals, which can be found in sewage sludge, industrial waste or landfills (International Biochar Initiative [IBI]).

The choice of raw materials is usually affected by biomass resources and availability in nearby areas. Owing to collection, transportation and storage costs, the use of local raw materials (if they are also an environmentally sustainable option) often makes the most economic sense.

Biochar has different physical and chemical properties ascribe to the thermochemical operating parameters and inherent property of biomass. For the purpose of higher output and target product quality, several units and reactors have been developed to produce biomass. The usage differs in oxygen, heating rate and final temperature may cause changes in the quality and distribution of the final product, though the principle of the reactors is similar (Wang et al. 2020).

Biochar production systems are usually divided into pyrolysis or gasification systems. The pyrolysis system uses kilns, retort tanks and other special equipment to contain the roasted biomass while excluding oxygen. Vent the reaction vessel to allow the pyrolysis gas to escape. The pyrolysis gas is often referred to as "syngas". As the generated syngas burns and releases heat, the process becomes self-sustaining.

The basic process for making biochar is called pyrolysis. Pyrolysis is the breaking down (lysis) of material by heat (pyro). As the material is broken down, it releases gas. This is the first step in the combustion or gasification of biomass (International Biochar Initiative [IBI]).

There are two types of pyrolysis systems currently in use: fast pyrolysis and slow pyrolysis. Fast pyrolysis tends to produce more oil and liquid, while slow pyrolysis produces more syngas. Figure 3 shows a variety of thermochemical conversion technologies for biochar production including slow pyrolysis, fast pyrolysis, gasification and torrefaction. Depending on the reaction conditions, especially the amount of available oxygen, a large difference in the yield and quality of biochar is produced in these processes. For example, by extending the residence time of biochar at a pyrolysis temperature of about 400°C to several hours or even days, high yield and high-quality biochar can be produced, which usually belongs to slow pyrolysis of biomass (Wang et al. 2020).

The gasification system produces a smaller amount of biochar in the directly heated reaction vessel where the air is introduced. The more oxygen a production unit can release, the more biochar it produces. The production of biochar is optimized in the absence of oxygen (International Biochar Initiative [IBI]).

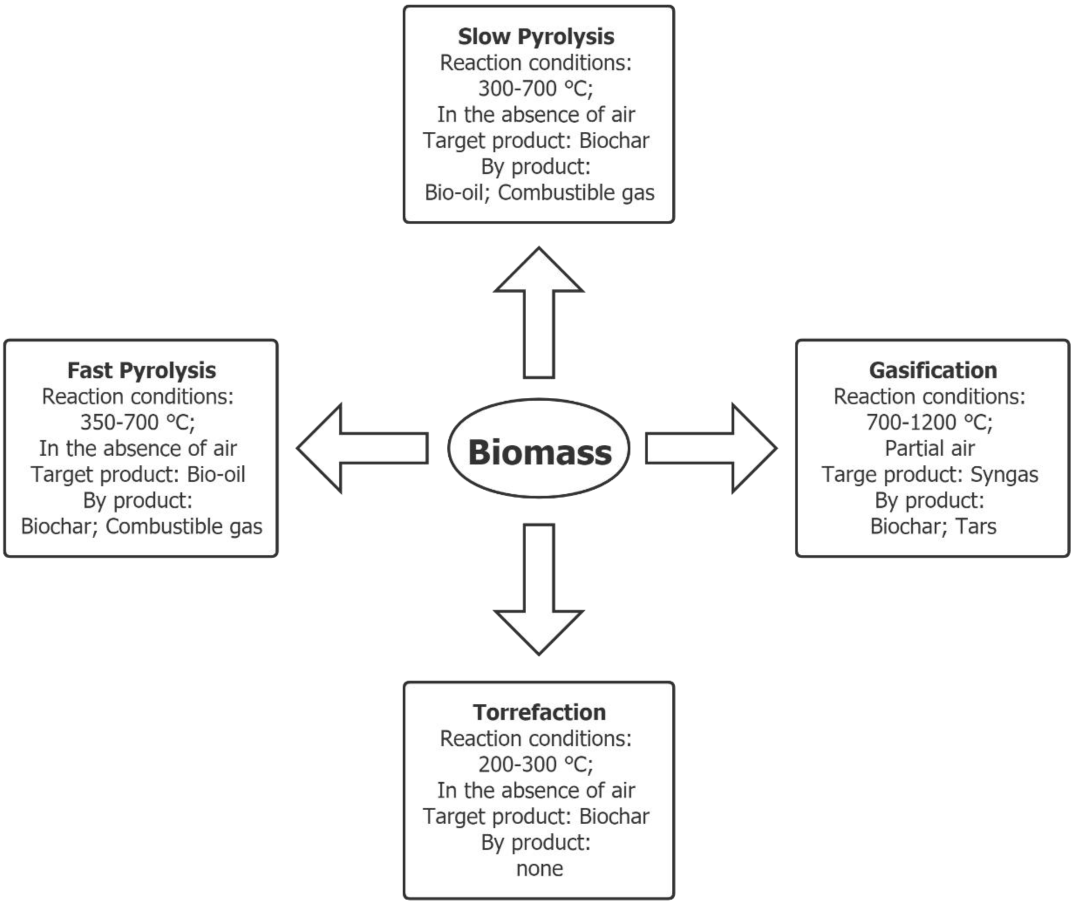

FIGURE 3. Biomass thermo-chemical processes for biochar production (Wang et al. 2020.)

5.1 Production of biochar in Finland

According to Converting Waste Agricultural Biomass into a Resource, Compendium of Technologies by the United Nations Environmental Programme (2009), 140 billion metric tons of biomass is generated annually from agriculture all around the world, which includes agricultural wastes, such as corn stalks, straw, sugarcane leavings, bagasse, nutshells, and manure from cattle, poultry, and hogs; forestry residues, such as wood chips, bark, sawdust, timber slash, and mill scrap; municipal waste, such as waste paper and yard clippings. The great volume of biomass can be converted to energy and raw materials.

In the European Union, renewable raw materials resources available are mainly forestry and agricultural residues. Forestry residues is the main source of the residues, while field residues and post-consumer wood or wood industry residues rank second and third, respectively (Marmiroli et al. 2018). It was estimated that from the amount of dry matter from forestry and agriculture separately about 9 and 85 Mt had been used for other purposes (Searle and Malins 2016), which could be available for transformation into biochar applicable to agriculture (Marmiroli et al. 2018). On the ways to dealing with the waste in the European Union, according to the EU Waste Framework Directive, there are three basic principles that should be obeyed in order of importance: (i) preventing waste, (ii) recycling and reusing, and (iii) improving final disposal and monitoring (Marmiroli et al. 2018). In view of this, production of biochar through upgrading waste material from forestry and other plant maintenance residues is an active step in activities aimed at improving the environmental and economic superiority.

Finland is currently involved with biochar related projects and biochar market has just recently started to emerge. According to Salo (2018-10-26), 2017 was the first year when considerable quantities of biochar were sold and utilised in Finland. During 2018 Noireco companies had made significant investments to biochar production and the annual production capacity was estimated to increase by at least 10000 thousand kilograms. Further investments to increase production capacity was planned as well. It is estimated that the market of biochar in Finland will increase tenfold by 2022 (Noireco Oy. 2018).

Biochar quality is greatly affected by its raw material. According to studies conducted by VTT Technical Research Centre of Finland and Natural Resources Institute Finland (LUKE), willow is the best raw material for biochar production, as it has an excellent pore structure: 200 m^2 / g, target 700-800 m^2 / g. The more pores on the bichar means its functional area is larger. The advantageous pore distribution, on the other hand, enables various properties. In addition, it binds water and dissolved nutrients (1 day 0.6 m^3 water / 300 kg or about 1 m^3 and 3 days 0.9 m^3 water / 300 kg or about 1 m^3) (Carbons Finland Oy 2020).

Willow is the fastest growing tree species in Finland, which is another reason why it is well suited for biochar production. Willow-based biochar has proven to be the most functional raw material in a variety of tests. The properties of the biochar produced from willow can be modified by manufacturing methods or chemically or, for example, by inoculating suitable microbes. (Carbons Finland Oy 2020.)

Figure 4 shows the porous structure of biochar microscopically.

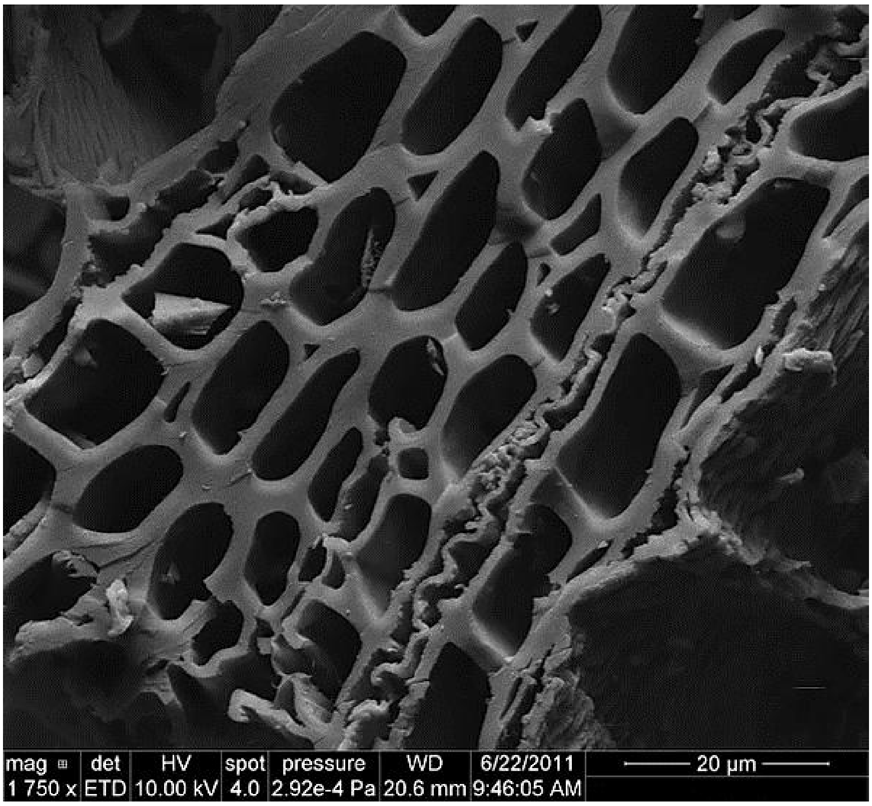

FIGURE 4. Microscopic biochar (Carbofex Oy 2019.)

5.2 Production of biochar in China

In the period of traditional agriculture, crop waste incineration is also a general field of processing, it is a commonly used method of soil covering the lit achieve smokeless combustion of biomass under anaerobic conditions, biochar after burning remain in the soil, which can improve soil and improve soil fertility. With the development of technology, nowadays, the preparation of biochar is mostly carried out in kilns, which improves efficiency, but the basic principles are the same as traditional agricultural methods (Lü et al. 2015).

At present, the commonly used method for preparing biochar is the thermal cracking, that is, oxygen-limited calefactive carbonization method. According to different reaction conditions, thermal cracking method can be divided into two types: one is rapid cracking method, the reaction temperature of which is generally above 700 °C, biofuels are mainly prepared by this method; the other is conventional cracking method, the temperature of which is generally below 700 °C, and it is usually used for the preparation of biochar. Studies have shown that the properties (spatial structure and characters) of biochar can be affected by the types of biomass raw materials. biochar from different biomass materials not only has different stability, but also has different effects on the adsorption capacity of pollutants, under the same pyrolysis conditions (Lü et al. 2015). Ameloot et al. (2013) reported the influence of different biomass sources on the performance of biochar. The higher the lignin content in the raw material, the higher the aromatic C content and the C:N ratio in the prepared biochar material, and the lower the salinity of biochar. In addition to the types of biomass raw materials, the pyrolysis temperature is also a very critical factor in the process of biochar preparation. It can not only affect the yield of biochar, but also control the surface structure and adsorption properties of biochar. Chun et al. (2004) reported the difference between biochar prepared at different temperatures (300-700 °C) and found that as the temperature increased (300-600 °C), the specific surface area of biochar increased (116-438 $m^2 \cdot g^{-1}$), and while the pyrolysis temperature was increased to 700 °C, the specific surface area of biochar decreased (363 $m^2 \cdot g^{-1}$), which indicates that some fine pore structures on the surface of biochar are destroyed at 700 °C, indicating that temperature is an important factor determining the surface structure of biochar.

The physical properties of biochar include specific surface area, pore structure, bulk density, etc. The chemical properties include pH, carbon content, hydrogen to carbon ratio, oxygen to carbon ratio, ash content, volatile matter, functional group types and cation exchange capacity, etc. The physical and chemical properties of biochar are directly affect its adsorption performance. The physical and chemical properties of biochar are affected by the preparation conditions and the types of raw materials. Table 2 shows the comparison of the physical and chemical properties of different raw materials. The temperature in the preparation conditions has the greatest influence on its physical and chemical properties and performance. Generally, the specific surface area and carbon content of biochar prepared under high temperature conditions are relatively high (Wang et al. 2020). Wang et al. (2020) found that pyrolysis temperature is a key parameter that affects the performance of biochar, and pH, carbon content, ash, conductivity, oxidation stability and specific surface area of biochar are all positively related to it.

TABLE 2. Comparison of physical and chemical properties of biochar from different raw materials

Feedstock	Production Temperature (°C)	pH	Organic Carbon Content (%)	Surface Area ($m^2 \cdot g^{-1}$)	Reference
Wheat straw	450	8.34	42.90	0.93	Wang et al. 2019
Sludge	500	6.76	16.82	34.348	Xu et al. 2020
Bamboo	675	9.4	58.43	13.9	He et al. 2019
Cow manure	700	6.04	47.66	3.153	Zhang et al. 2020
Walnut	700	9.42	47.67	4.631	Zhang et al. 2020
Sawdust	550	6.2	41.00	9.6	Ghanim et al. 2020
Hickory	600	8.4	83.50	277.4	Li et al. 2020
Hickory wood	600	7.1	69.10	221.5	Xiang et al. 2020

According to Roberts et al. (2010) and Shackley et al. (2011), it is showed by the existing biochar system analysis report that in order to have a better economic and environmental preference, waste biomass, instead of wood or other original biomass, can be used as biochar raw materials. Jiang et al. (2012) found that China had shown great potential in that regard, producing 800 million tons of agricultural straw residues each year. While enough straw is retained to maintain soil quality, it is estimated that 505 million tons of residues can be produced (Clare et al. 2015). Therefore, let those straw residues be translated into bioenergy products or biochar is conducive to the reduction of GHG emission in China.

Pinewood, microalgae, crop residues and other raw materials have been used in the pyrolysis production of biochar. Compared with lignocellulosic biochar, the stability of algae and crop residual biochar is poor, and the pH and ash content are also high. In different lignocellulosic biomass, the ratio of hemicellulose, cellulose and lignin, as well as their connectivity and crystallinity are different. The changes in thermal degradation behavior are attributed to changes in the structure and chemical properties of specific components of raw materials. Due to the different decomposition temperatures of cellulose (315-400°C), hemicellulose (220-315°C) and lignin (160-900°C), and the different proportions of these three components in the biomass, biological The pathways and mechanisms of char formation are different (Wang et al. 2017; Yang et al. 2007).

6 SEPARATION OF USED BIOCHAR

Magnetic biochar can be easily separated and recovered from the medium under a magnetic field environment. After the reaction, the depleted magnetic biochar can usually be reused after regeneration to remove contaminants. Meanwhile, the reuse time of magnetic biochar is also important when judging the performance of magnetic biochar. Nowadays, due to the different nature of pollutants, commonly used regeneration reagents include hydrochloric acid, sodium hydroxide, chelating agents, organic solvents and so on (Yi et al. 2019).

According to an article about magnetic separation for Cd^{2+} (Huang et al. 2021), iron ions modified magnetic biochar, compared with its original one, has a higher separation efficiency. While the turbidity of its original biochar slowly decreases with time and maintains a relatively high level after 30 minutes. Besides, when the initial pH is lower than 3.0, the turbidity of the separated original biochar is lower than that of its respective magnetic biochar. This difference is because the release of iron ions or a huge amount of hydrogen ions into the solution is enhanced by the strong acidic conditions. With the increase in the initial pH value, and decreased turbidity of stabilized at the initial pH about> 5.0. These turbidity results indicate that although magnetic biochar is easily separated from the solution, pH value of the solution is a key factor in magnetic separation (Huang et al. 2021). After separation, the magnetic biochar is recycled through an appropriate desorption process to avoid secondary pollution (Huang et al. 2021).

Moreover, some physical means are usually used to help the regeneration of magnetic biochar. For example, ultrasonication and oven-drying are used to assist the regeneration of magnetic biochar (Heo et al. 2019). Generally, the adsorption capacity of the regenerated magnetic biochar is severely reduced, which is mainly because of the destruction of the structure of magnetic biochar after adsorption. Therefore, how to ensure the reactivity of regenerated magnetic biochar is very important for the application of magnetic biochar (Yi et al. 2019).

7 LABORATORY STUDY: REMOVAL OF NITROGEN FROM WATER USING BIOCHAR

The goal of the experiment was to compare two different types of biochar for nitrogen and other nutrients removal from industrial water and landfill water. The results were obtained by measuring the content of nitrogen and assessing the performance of biochar.

7.1 Materials and equipment

The two types of biochar used in the experiment were named as H5 and UEF. The landfill water tested in the experiment was from Jätekukko, Kuopio, and the industrial water was mine water from Lapland, Finland.

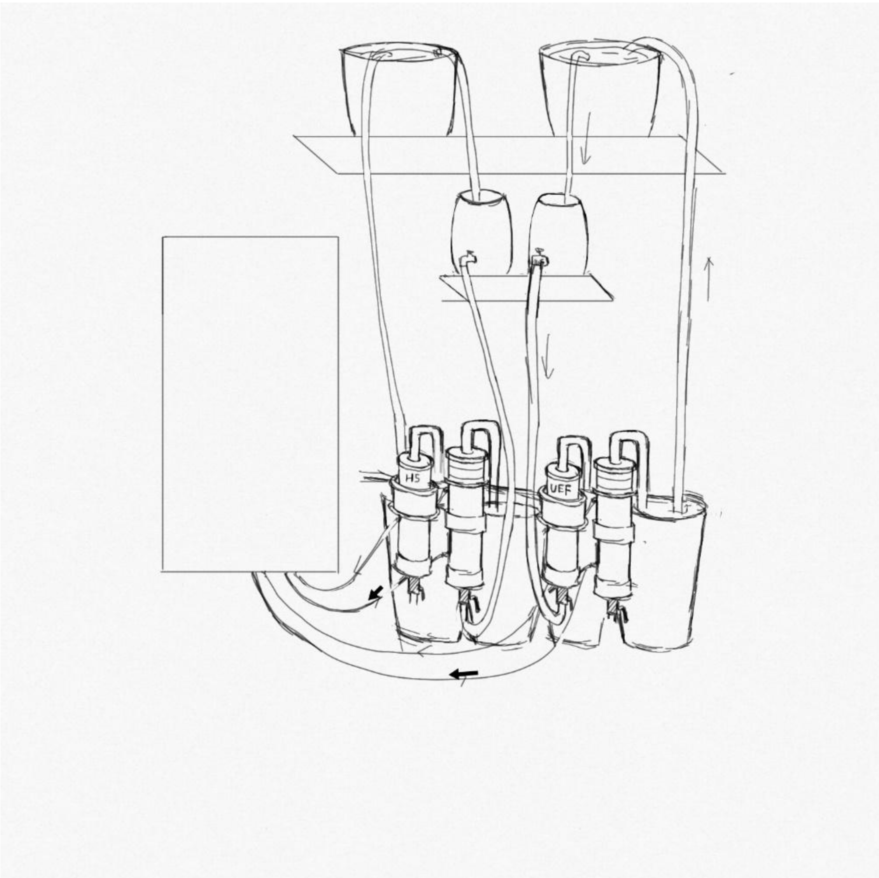

FIGURE 5. Diagram of the experiment device

FIGURE 6. The equipment used in the experiment

Figure 5 and 6 showed the diagram and the photograph of the experiment equipment.

There were two top tanks above which were used to contain the unprocessed water. Water was directed from above to the middle white tanks and then the filtration columns. After being filtrated, it flew to the two big tanks on the ground and was pumped into the middle tanks. The middle white tanks were used to adjust excessive pressure difference due to height.

The top and bottom of the biochar column are covered with two metal mesh to hold the biochar in place. The piezometers on the left were for monitoring the water pressure and measuring the pressure difference caused by filtration.

Biochar was washed by water and dried before use. After that, biochar was put into the filtration columns tightly to have a better filtering effect. Column I was filled with Carbons H5, and Column II was filled with UEF biochar. Deionized water was put in and circulated through the equipment before using the raw water.

7.2 Analytical methods

The adsorption effects of biochar were tested by LCK tests. The samples were disposed of by high-speed digestion. After heating and cooling down, COD, total-N and total-P can be tested. Water from two top tanks and four columns after filtrating by biochar were taken as samples. Figure 7 shows samples transferred into tubes for testing.

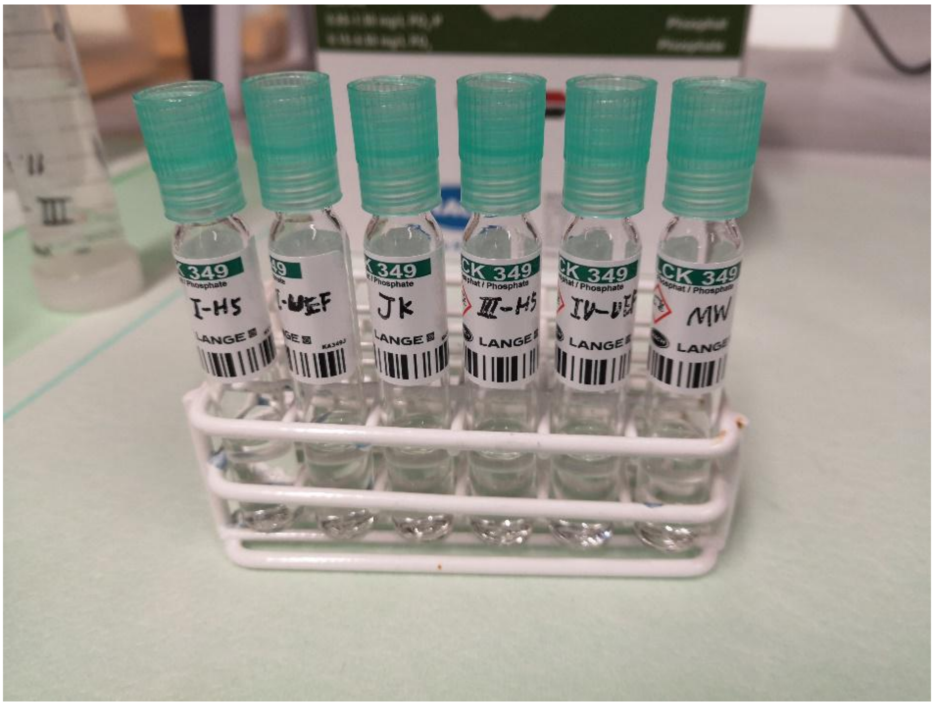

FIGURE 7. Samples from six different points

Figure 8 shows the samples under high-speed digestion.

FIGURE 8. Samples in the HT 200S high-speed digester

8 RESULTS

Water was from the same container for all of the tests. The flow rate was constant.

The particles size of the biochar was first tested by sieves to know what kind of biochar was used and know whether it will stick the column affecting the results. In this test, the most suitable size of the particles was supposed to be between 2-4 mm because that won’t clog the column and the flow was still easy to adjust.

The particle size distribution of H5 and UEF biochar was shown in Figure 9 and 10. From those charts, it could be known that about 39.29 % of H5 biochar and 41.87 % of UEF biochar for the experiment was under this range.

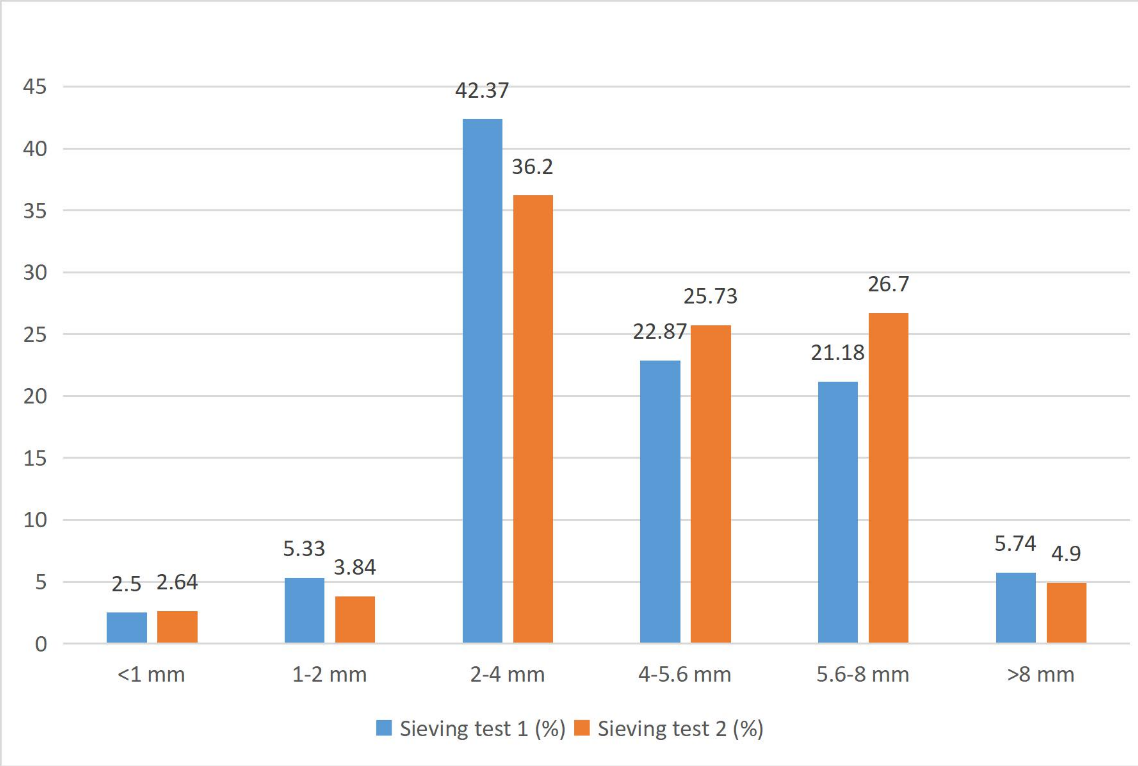

FIGURE 9. The particle size distribution of H5 biochar

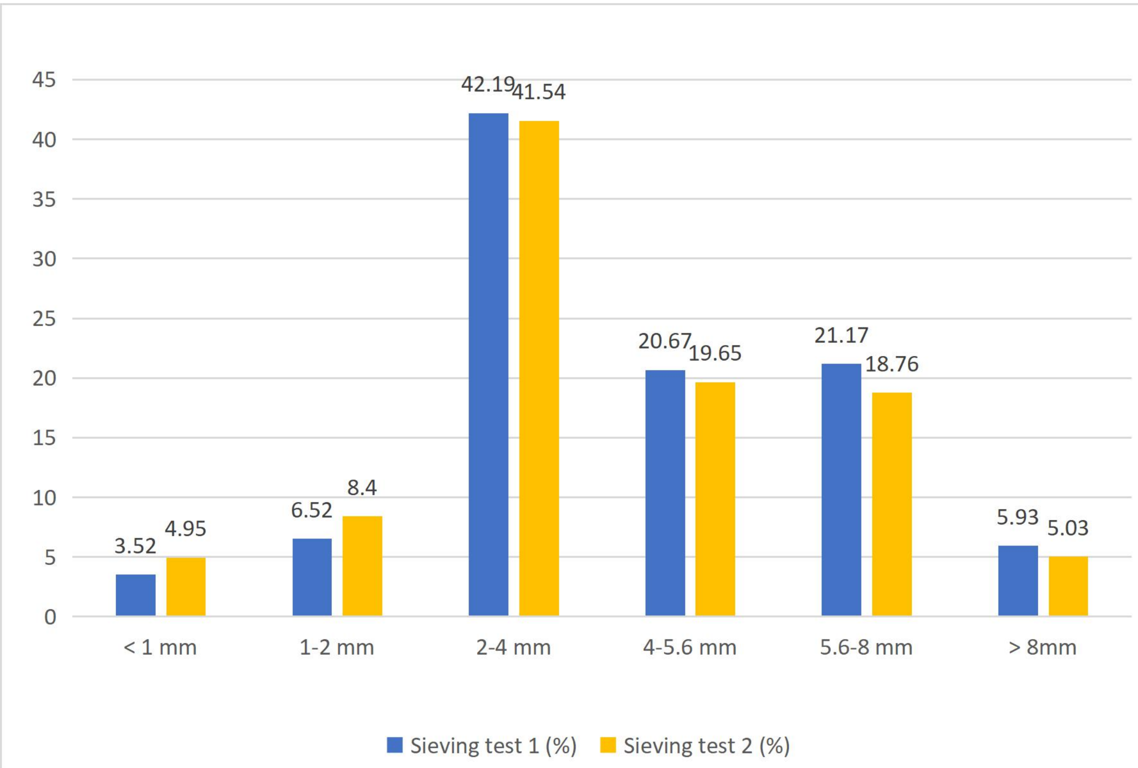

FIGURE 10. The particle size distribution of UEF biochar

After testing and recording the nutrients content in water from different tanks, the adsorption rate had been calculated and part of the data had been shown in Table 3.

TABLE 3. N adsorption efficiency of biochar H5 and UEF for mine water

Test Date	Type of Biochar	Nitrate (mg/, A)	Nitrite (mg/l, A)	Ammonium (mg/l, K)
11.03.2020	H5	3.3%	15.2%	10.1%
	UEF	2.6%	9.7%	6.9%
18.03.2020	H5	2.3%	1.7%	12.5%
	UEF	3.9%	4.6%	12.0%
25.03.2020	H5	4.6%	5.9%	12.0%
	UEF	6.2%	11.5%	28.9%

The two bar charts (Figure 11 and 12) show the adsorption efficiency of H5 and UEF biochar to nitrogen in mine water respectively. In the two bar charts, the adsorption efficiency is expressed as a plus or minus percentage.

A good filtering effect was indicated by a positive value indicates, which means the adsorption of the measured elements decreases after filtering, while a negative value means that the amount increases instead.

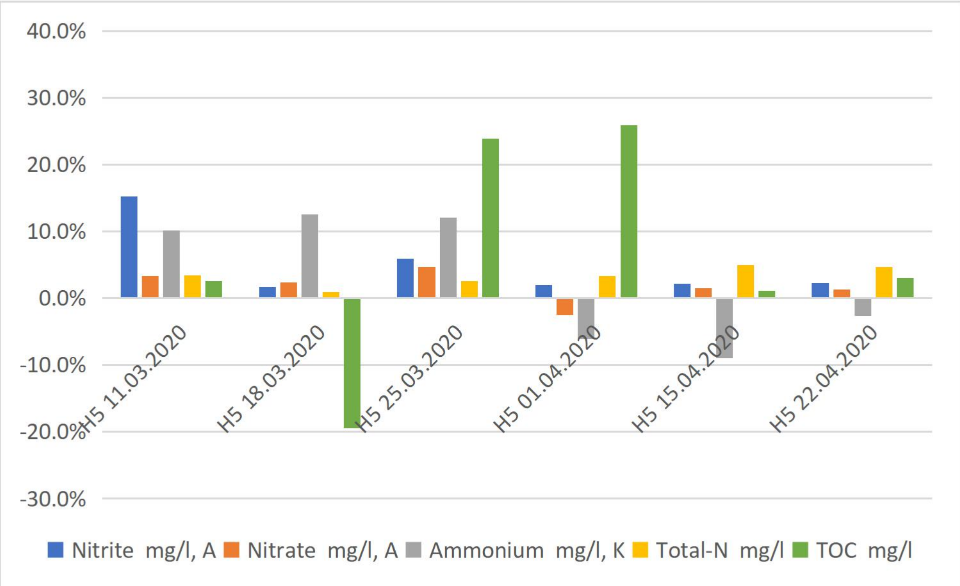

FIGURE 11. The reduction percentage of nutrients in mine water after filtration by H5

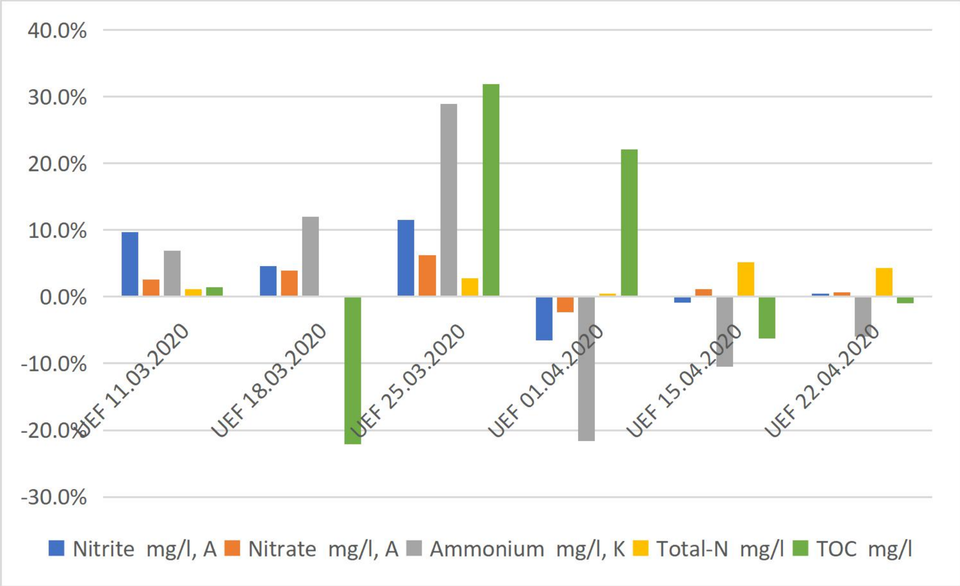

FIGURE 12. The reduction percentage of nutrients in mine water after filtration by UEF

Also, Figure 13 and 14 show the reduction percentage of Jätekukko landfill water after filtrated by two kinds of biochar, H5 and UEF, respectively.

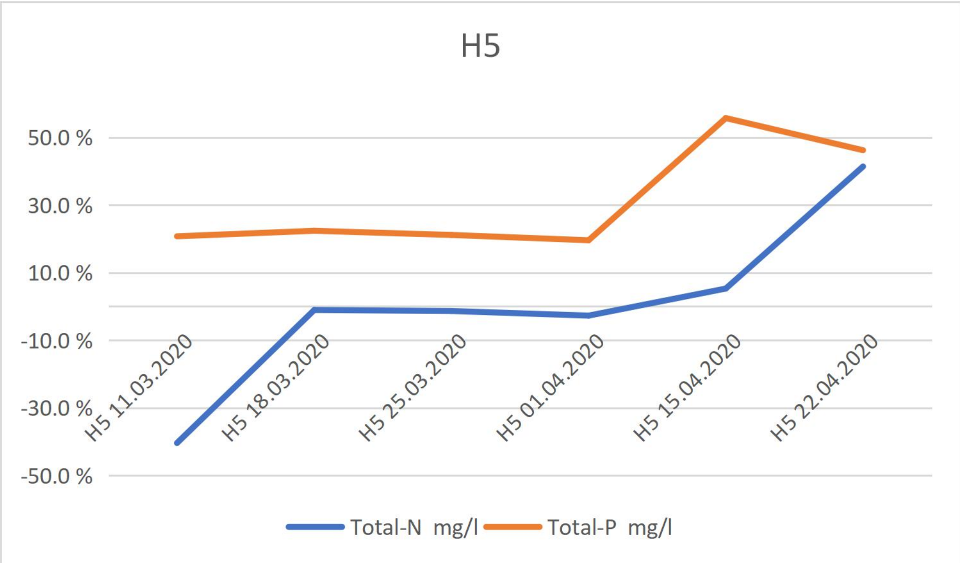

FIGURE 13. The reduction percentage of Jätekukko landfill water after filtration by H5

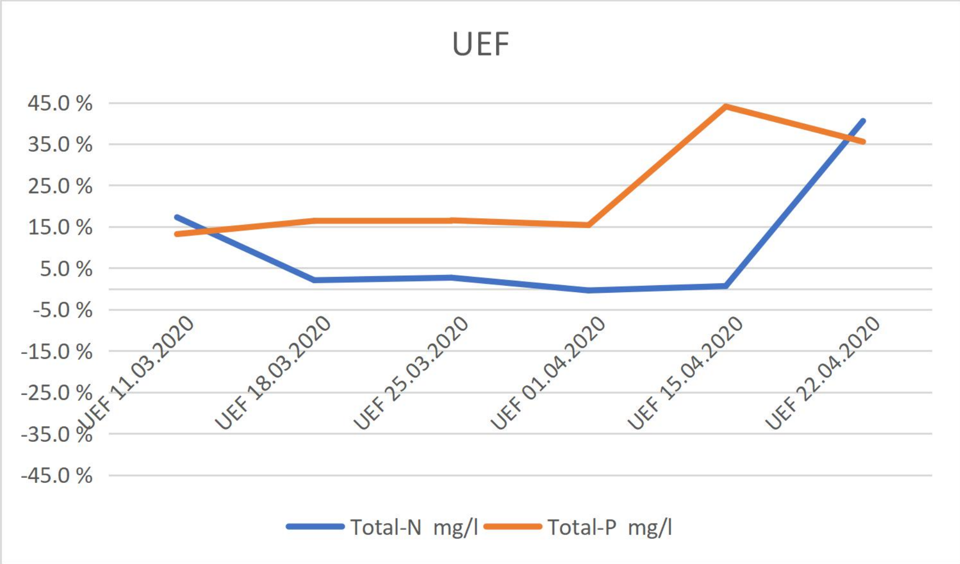

FIGURE 14. The reduction percentage of Jätekukko landfill water after filtration by UEF

9 DISCUSSION

From Figure 11 and 12, we can see that nitrite, nitrate and total nitrogen in mine water had been removed partly during the experiment and had a positive reduction rate especially in the first half of the experiment. However, there was no significant correlation between the reduction of total organic carbon (TOC) and the adsorption of biochar. Typical amount of TOC in the results was 3-4 mg/L but the result from March 18 was 1.97 mg/L which made the percentage result looked different. The amount of ammonium was shown to be decreased in the first half of the experiment while to be increased later. It looks like the biochar cannot adsorb ammonium anymore after few weeks.

There was no phosphorous in mine water so the absorption of total phosphorous cannot be seen in this experiment.

The unprocessed water is the same but the TOC result of top tank mine water on April 22 was 28.84 mg/L which was much higher than others.

From Figure 13 and 14 we cannot draw any conclusion from the irregular data.

10 CONCLUSIONS

From the experimental results, we can conclude that biochar H5 performs better than biochar UEF in the adsorption capacity of total nitrogen. Both of them have a certain adsorption effect on bromide, sulfate, lithium, sodium, potassium, calcium as well.

However, due to the long term of the experiment, the water may be lack of mixing which may cause some problems on the data.

Besides, the pH of the water sample had not been tested and there were a lot of salt materials in it. As time went on, some of these became more which may affect the pH level of the water. Bacteria in the water working for adsorption needs nutrients especially phosphorous and nitrogen to support their anaerobic digestion. The bacteria flushed into the biochar columns supposed to contribute to the adsorption process. Whereas the consumption of nitrogen and the lack of phosphorous in mine water may also affect the adsorption effect of bacteria. All these possible reasons eventually lead to an increase in nitrogen.

The exact material of those two kinds of biochar was unsure and the temperature of the experiment has not been tested as well.

In summary, many factors have been affected the adsorption of nitrogen by biochar, but there are still some factors that have not been explored or sufficiently studied (such as bacteria, salinity and co-existing ions), and this is expected to be improved in future experiments.

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