

Research Article

The Combined Application of Surface Floating Wetlands and Bottom Anaerobic to Remediate AMD-Contaminated Lakes

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Acid mine drainage (AMD) causes environmental pollution that affects many countries with historic or current mining industries. The eco-remediation system (RW) which combined surface floating wetlands and bottom anaerobic sediments (SFW-BAS) was selected for AMD-contaminated lakes (AMDW). Meanwhile, AMDW and nature aquatic ecosystems (NW) were set as the control groups, respectively. The parameters, including pH, Eh, Fe, Mn, SO_4^{2-} , and the degradation rate of the native dominant plant litter were investigated to assess the effects of remediation. The results showed that the average of pH, Eh, and EC, was 2.73, 484.08 mv, and 2395.33 μ s·cm⁻¹, respectively. The average content of SO_4^{2-} , Fe, Mn, Cu, Zn, and Pb was 2190 mg·L⁻¹, 40.2 mg·L⁻¹, 4.6 mg·L⁻¹, 249.2 μ g·L⁻¹, 1563 μ g·L⁻¹, and 112.9 μ g·L⁻¹, respectively. The degradation rate of plant litters in AMDW ranged from 14.5% to 22.6%. However, RW ultimately improved the water quality and the degradation of litters. RW has a good effect on buffering the acidity, ranging from 3.96 to 7.41. The pH of RW (6.14) is close to that of NW (7.41). The average content of SO_4^{2-} , Fe, Mn, Cu, Zn, and Pb was 2071 mg·L⁻¹, 3.4 mg·L⁻¹, 2.4 mg·L⁻¹, 85.3 μ g·L⁻¹, 607.4 μ g·L⁻¹, and 47.8 μ g·L⁻¹, respectively, which showed good pollutant removal performance. The degradation rate of plant litters in RW ranged from 27.8% to 32.6%. Therefore, RW can be used to remediate AMDW.

1. Introduction

Upon exposure to water and oxygen and as a result of the activities of indigenous microbial populations, iron pyrite (FeS₂) and other sulphide minerals can be oxidized to form acidic and sulphate-rich drainage, which is a consequence of the mining and processing of metal ores and coals [1–3]. The oxidation of pyritic mining waste is a self-perpetuating corrosive process that has generated acid mine drainage (AMD) effluent for centuries or longer [4]. AMD is characterized by a low pH and high levels of sulphate and toxic metallic ions [2]. It is an important environmental issue since it can degrade the waterways in and around mine districts [5]. Both the terrestrial and aquatic ecosystems and processes may be disturbed [6, 7]. In aquatic ecosystems, it can be observed as a severe disturbance or even destruction of the water quality and the functions, habitats, biological

communities, and species composition of these ecosystems. Studies have shown that AMD could significantly decrease the water quality [8, 9] and the activity of the invertebrates, aquatic plants, or zoobenthos [10]. Additionally, it may also affect the degradation of plant litter in the streams [11].

Therefore, there have been numerous studies devoted to reducing AMD formation or to treating these waste waters [6, 12]. However, few studies are designed to evaluate the techniques for ecological restoration of AMD-contaminated land or water. The optimized strategies are aimed to suppress the formation of AMD by treating the acid-producing rock directly and stopping/retarding the production of acidity at the source. These techniques include the use of alkaline amendments (e.g., limestone, hydrated lime, fly ash, and waste phosphate rock) [13, 14], covering the tailings with a layer of sediment or organic material (e.g., sewage sludge, manure mixture, and olive pomace) [15], coating technologies (e.g., solid-phase phosphates, phospholipids, and lipids) [16], the use of bactericides (e.g., sodium dodecyl sulphate, benzoic acid, and sodium benzoate), or the use of natural organic acids (e.g., fulvic acids, tannic acids, oxalic acid, and humic acids) [17]. Since large quantities of AMD are produced in the wider environment, the traditional methods could not meet the requirements. The current abiotic and bioremediative strategies for the remediation of AMD have been considered "active" and "passive" processes. The active treatment process involves abiotic remediation strategies (the addition of a chemical-neutralizing agent, e.g., limestone, hydrated lime, pebble quicklime, soda ash, or caustic soda) [14] and biological systems, such as sulphidogenic bioreactors [18]. In the passive treatment process, the abiotic technologies are mainly anoxic limestone drains (ALD) [19], but the biological remediation strategies have wider choices, such as wetlands [20] or compost bioreactors [18], the reducing and alkalinity-producing system (RAPS) [21], successive alkalinity-producing systems (SAPS), permeable reactive barriers (PRBs) [22], and iron-oxidizing bioreactors. Nevertheless, these strategies are purposed to suppress the formation of the AMD or to treat the production before it enters the environment. It will not work on already polluted water but on the wider environment, where the natural ecological system has been destroyed by the AMD. Surface floating wetlands are easy and cheap to construct and therefore used to treat mine tailings water and polluted rivers [23]. Surface floating wetlands are based on the self-cleaning capability of plants to augment aquatic projects and rapidly remove pollutants from water bodies [23]. Furthermore, there was the potential of activated sludge for the remediation of sulphur-rich wastewaters, owing to its simplicity and low cost [24]. Activated sludge could form suitable microbial community for AMD treatment [24]. If SFW-BAS could be constructed, the removal effects of contaminants from AMDW will increase with easy and low cost.

In this study, there are characterized by an inherently fragile ecosystem and sensitive processes of biogeochemical cycling in the karst regions in southwest China. RW was constructed to remediate AMDW, and RW included floating aerobic wetlands on the surface and anaerobic sediments in the bottom. AMDW and NW were set as the control groups, respectively. The water quality (pH, Eh, EC, Fe, Mn, and SO_4^{2-}) and litter decomposition rate of NW, AMDW, and RW were evaluated to compare to the remediation effect for the contaminated waters. These findings would provide the basic data of AMDW treatment.

2. Materials and Methods

2.1. Study Regions. The study was carried out at the Guizhou province, located in the central karst region of Southwest China, which has a well-developed karst landform [25]. It has a moderate, subtropical, humid monsoon climate, which is characterized by hot and wet summers and mild and humid winters. Karst ecosystems are rich in water and mineral resources and can provide unique habitats for numerous fish and wildlife. Unfortunately, they are

inherently fragile, at risk of degradation and vulnerable to the environmental change (e.g., human habitation, mining, and draining the sewage) [26]. Coal resources are extremely abundant in the Guizhou Province, but the sulphur contents in the coal are relatively high, with an average of 2.34% [27]. Because of the long time and large-scale mining activities of the coal and metal mines, large volumes of sulphide waste rock or tailings were discarded in this region. The main iron sulphide minerals are pyrite and marcasite (FeS₂), but others, such as chalcopyrite (CuFeS₂), covellite (CuS), galena (PbS), and sphalerite (ZnS), also exist [3, 28].

2.2. Simulated Experiments. The simulation of the microcosm aquatic ecosystems was carried out with six cylindrical tanks, each with a height of 900 mm and an internal diameter of 950 mm. A 15-20 cm layer of sediment from the Huaxi Reservoir and a 60 cm layer of lake water were placed in these tanks. The Huaxi Reservoir is a typical karst lake. The submerged vegetation, including Vallisneria natans Hara, Hydrilla verticillata, Ceratophyllum demersum L, pondweed, and Ottelia acuminate, was planted in these tanks. These microcosms were incubated in the open air, receiving natural lighting and temperature throughout the year. The cultivation lasted approximately three months, until these microcosms could be self-sustaining, and they were set as the simulation of the natural aquatic ecosystem (NW). Then, four microcosms were treated with the AMD on a regular basis, which were set as the simulation of the AMD-contaminated aquatic ecosystem (AMDW). Approximately two months later, two of the AMDW microcosms were equipped with an SFW-BAS treatment system, which was set as the eco-remediation aquatic ecosystem (RW). The surface floating wetlands consist of plastic foam or an empty bottle made of lightweight material that floats, has basins with holes in the bottom to be beneficial to the roots and for the water to penetrate, has soil or another suitable medium to support the vegetation, and contains vegetation that is a mix of cattail, stick tight, Lolium perenne, Alternanthera philoxeroides, and another native species. The bottom anaerobic sediments were formed by the original addition of the activated sludge and by constantly obtaining organic substances from the surface floating wetlands. The layout of the system is shown in Figure 1.

RW was successfully built for the remediation of AMDW (Figure 2). RW was in normal operation for 210 d. Plants grew well in the floating aerobic wetlands. There were fluffy anaerobic sediments at the bottom. RW could promote vertical interactions between the aerobic vegetation and anaerobic microorganisms to remediate AMDW.

2.3. Sampling and Analytical Methods. The litter of the native dominant woody species at the local shore, such as Salix babylonica and Broussonetia papyrifera, and submerged plants, such as Vallisneria natans Hara, Hydrilla verticillata, and Ceratophyllum demersum L, was collected in this study. The above selected litters were thoroughly cleaned by water and air-dried prior to the reuse or storage. Five grams of air-dried leaves were packed into 15 * 30 cm nylon



FIGURE 1: Microcosm experimental setup. NW: simulating the natural aquatic ecosystem; RW: simulating the eco-remediation AMD-contaminated aquatic ecosystem; AMDW: simulating the AMD-contaminated aquatic ecosystem. In the microcosm RW, an eco-remediation system that combines floating aerobic wetlands on the surface with constructed anaerobic sediments in the bottom (SFW-BAS) was employed.



FIGURE 2: A pilot passive system for the remediation of an AMD-polluted ecosystem.

mesh bags with a mesh size of 2 * 2 mm. The bags were placed at the bottom of the microcosm and on the surface of the sediments in quintuplicate at the three habitats representing NW, AMDW, and RW. The bags were retrieved 0, 15, 30, 60, 120, and 210 d after they were mounted. The decomposition rate was determined by direct measurements of the weight losses from the litter bags [29]. The mass losses were determined in oven-dried values (48 h, 60°C or until constant weight) after a strict cleaning.

Synchronously, water samples were collected at a depth of 0.2 m. The pH, electrical conductivity (EC), redox potential (Eh), and dissolved oxygen (DO) were determined in the water of the microcosm systems. The pH and EC were detected by a portable pH/EC/TDS instrument (HI 98129, HANNA, Germany). The Eh and DO were recorded by a potentiometer and DO6 probes, respectively. The water samples for the metal analysis were filtered through 0.45 μ m membrane filters, acidified with nitric acid to pH <1, and

analysed by flame atomic absorption spectroscopy (WFX100 Beijing Ruili, China). The SO_4^{2-} concentrations were determined by the conventional ignition method in gravimetry.

2.4. Data Analysis. All data were analysed with SPSS 13.0. The differences in each evaluation index at different times were compared with a single-factor analysis of variance (ANOVA), and those in different microcosms were compared with two-factor ANOVA. Moreover, Duncan's multiple range was used to treat the average significant difference among the multiple comparisons.

3. Results

3.1. Effects of Remediation on pH, EC, Eh, and DO. The water in the microcosm NW was crystal clear, the submerged



FIGURE 3: The temporal variation of pH, EC, Eh, and DO in NW, RW, and AMDW.

plants (*Vallisneria natans* Hara, *Ceratophyllum demersum* L, *Hydrilla verticillata, Potamogeton distinctus,* and *Ottelia acuminata*) were growing well, and fish and shrimps were shuttling back and forth in the microcosms. The pH ranged from 6.45 to 8.06 (with a mean of 7.46), and the DO varied from 5.4 to 8.1 mg·L⁻¹ (with a mean of 7.26 mg·L⁻¹) (Figures 3(a) and 3(d)). The ECs were relatively low, with a range of 310 to 810 μ s·cm⁻¹ (Figure 3(b)), and the Eh varied from 150 to -100 mV during the experiment time (Figure 3(c)). These indexes agreed well with those of the natural waters. The Fe, Mn, Cu, Zn, and SO₄²⁻ concentrations remained at a very low level during the period of observation (Figure 4), which can meet class III of the environmental quality standards for surface water (GB3838-2002, China).

In the microcosms AMDW, the water colour changed to dark red, and the bottoms and walls became coated with a

layer of ochre particles. All the submerged vegetation, filamentous algae, fish, and shrimps disappeared. The average of the pH was 2.73, with the lowest value being 1.38 (Figure 3(a)), and the water presented an obvious acidification with a high salinity (1200–3750 μ s·cm⁻¹; Figure 3(b)) and Eh (420–690 mv; Figure 3(c)). Simultaneously, the concentrations of the contaminated ions and sulphate in the AMDW were continuously increasing, with values dozens or hundreds of times higher than those in NW (Figure 4), which presented a typical AMD contamination [3].

Compared to the microcosm AMDW, RW had good performance in raising the pH to neutral (Figure 3(a)), buffering the strong oxidizing nature (Figure 3(c)) and reducing the metal levels to environmentally permissible limits (Table 1; Figure 4; GB3838-2002, China). The average of pH in RW was 6.14, ranging from 3.96 to 7.41. The pH of RW (6.14) is close to that of NW (7.41), indicating that RW had



FIGURE 4: Concentration variation of Fe, Mn, Cu, Zn, Pb, and $\mathrm{SO_4}^{2-}$ in NW, RW, and AMDW.

TABLE 1: The Duncan multiple comparison of the typical pollution ions among NW, RW, and AMDW.

	Fe (mg·L ^{-1})	Mn (mg· L^{-1})	Cu ($\mu g \cdot L^{-1}$)	Zn ($\mu g \cdot L^{-1}$)	Pb $(\mu g \cdot L^{-1})$	SO_4^{2-} (mg·L ⁻¹)	
NW	$1.1 \pm 0.8 \text{ A}^1 \text{a}^2$	$0.0 \pm 0.0 * Aa$	79.7 ± 55.8Aa	$279.4 \pm 40.4 \text{Aa}$	$0.0 \pm 0.0 * Aa$	324 ± 144 Aa	
RW	$3.4 \pm 3.2 \text{Aa}$	2.4 ± 1.2 Bb	85.3 ± 62.3Aa	607.4 ± 389Aa	$47.8 \pm 56.7 Aa$	$2071 \pm 352Bb$	
AMDW	40.2 ± 14.0 Bb	$4.6 \pm 0.8 Cc$	$249.2 \pm 76Bb$	1563 ± 315Bb	112.9 ± 96.7Aa	2190 ± 390 Bb	
F	48.478	41.108	17.009	31.6	2.651	70.95	
р	0.00001	0.00001	0.0003	0.00001	0.1111	0.00001	

 A^1 is p value <1%; a^2 is p value <5%. *The value is less than IDLs (instrument detection limit).

good effects on buffering the acidity of AMDW. The microcosm RW also ensured regular growth and activity for the microorganisms and benthonic animals (porous texture, well-agglomerated microbes could be seen in the bottom of the system, and there was a large amount of *chironormus larva* in the water). However, these microorganisms and benthonic animals consumed the DO intensively resulted in anaerobic conditions and maintained a higher salinity than in NW (Figures 3(d) and 3(b)).

3.2. Effects of Remediation on Fe, Mn, Cu, Zn, Pb, and SO_4^{2-} . Compared to the microcosm AMDW, the ecological remediation significantly reduced the concentrations of harmful ions such as Mn, Cu, Zn, and Pb (Table 1 and Figure 4), which have a trend of decreasing with time (Figure 4). Except for Mn, the other metals meet class III of the environmental quality standards for surface water. For SO_4^{2-} , remarkable reduction occurred from 120 to 210 d (at the surface of the constructed sediments, Eh: from -90 mV to -255 mV) [8]. Compared to the control microcosms (AMDW), the sulphate concentrations decreased dramatically from 2500 to $1700 \text{ mg} \cdot \text{L}^{-1}$ (Figure 4(f); Table 1) due to the high rates of sulphur removal [30]. However, the effect was limited. Despite its sharp decline, it remains at a high level with a value of over 1600 mg L^{-1} after 210 d, which was significantly higher than those in the blank microcosms (NW) ($<500 \text{ mg} \cdot \text{L}^{-1}$).

3.3. Effects of Remediation of Degradation of the Litters. The AMD significantly decreased the degradation rates of the plants growing on the bank or submerged plants compared with NW (Figure 5) and affected the normal ecosystem function of the waters. Under NW conditions, the highest values of the degradation rates of the litters were observed (25-35%), and the rates continuously increased and then became constant with time. After 210 d, the degradation rates of Salix babylonica and Broussonetia papyrifera were 58.1% (Figure 5(a)) and 52.6% (Figure 5(b)), respectively, and those of Ceratophyllum demersum L, Vallisneria natans Hara, and Hydrilla verticillata were 54.8% (Figure 5(c)), 44.6% (Figure 5(d)), and 54.1% (Figure 5(e)), respectively. The degradation rates in AMDW were notably lower than those in NW at the same time (Figure 5), and there are significant differences (Table 2). Additionally, the degradation rates of the above ordered litters after 210 d were only 36.58%, 35.09%, 20.7%, 30.5%, and 25.1%, respectively.

The degradation trend in RW was consistent with that of NW. RW remediation significantly improved the degradation rate of the litters and became more similar to that of NW over time, which was preferable for the ecosystem function (Figure 5). The rates after 210 d were 44.7%, 46.6%, 49.4%, 43.4%, and 54.3%, respectively, and there were no significant differences in *Salix babylonica Ceratophyllum demersum* L, *Vallisneria natans* Hara, and *Hydrilla verticillata* compared with NW (Table 2). However, the rates still had large variations within NW (Figure 5), which indicated that it will be hard to recover the ecosystem once it is destroyed.

4. Discussion

In this study, we developed a pilot passive system for the remediation of an AMD-polluted ecosystem. This system combined both the floating aerobic wetlands on the surface and constructed anaerobic sediments at the bottom, which can promote vertical interactions between the aerobic vegetation and anaerobic microorganisms (Figure 6). On the lake's surface, the floating aerobic wetlands are partially installed, and the associated oxidation and hydrolysis reactions can eventually result in the precipitation of dissolved metals [31]. In addition, to some extent, the filtering, uptake, adsorption and exchange by plants, soil, and other biological materials could remove the metals and other ions [3, 32]. At the lake's bottom, the constructed anaerobic sediments can promote the anaerobic bacterial activity, which ultimately results in a series of reduction reactions and the subsequent precipitation of metal sulphides and generation of alkalinity [12, 33]. The low-cost natural products and wastes such as straw, wood chips and saw dust, spent mushroom compost, mixed manure, and potatoes [34, 35] were used as organic substrates in the AMD treatment systems to create reducing conditions. In this study, we used residual sludge from the municipal wastewater treatment plant as the first organic substances. The sewage sludge contains abundant and easily degradable organic matter, nitrogen, and phosphorus, which can quickly create anaerobic environments for the microbial consortia within the sediments in the pilot experiments. When RW is stable, the organic substrates could continuously be obtained from the surface wetland. Overall, this remediation system involves two parts of the surface and the bottom, providing a combination of aerobic and anaerobic environments and their interactions. The surface floating wetlands provide an ongoing supply of organic substances to the bottom (like acid reduction using microbiology (ARUM) by Kalin [36], which continuously provides nutrients for the reducing bacteria to establish and maintain reductive conditions in the sediment and ameliorate the lake's pH. Within





FIGURE 5: The degradation trend of the five plant litters in NW, AMDW, or RW. Salix babylonica and Broussonetia papyrifera are native dominant woody species at the local shore; Vallisneria natans Hara, Hydrilla verticillata, and Ceratophyllum demersum L are native dominant submerged plants in the Huaxi Reservoir.

TABLE 2: The Duncan multiple comparison of the litter decomposition rate among NW, RW, and AMDW (%).

	Salix babylonica	Broussonetia papyrifera	Ceratophyllum demersum L	Vallisneria natans Hara	Hydrilla verticillata
NW	$33.2 \pm 18.4 \text{ A}^{1}a^{2}$	37.4 ± 19.6 Aa	34.9 ± 19.3 Aa	33.0 ± 17.2 Aa	34.8 ± 20.1 Aa
RW	30.4 ± 16.2 ABa	29.9 ± 16.9 ABb	30.9 ± 18.0 Aa	27.8 ± 16.0 ABa	32.6±19.4 Aa
AMDW	24.2 ± 12.7 Bb	22.6 ± 11.7 Bc	17.1 ± 10.5 Bb	17.3 ± 10.4 Bb	14.5 ± 8.7 Bb
F	9.762	17.067	12.274	9.985	13.331
р	0.0045	0.0006	0.002	0.0041	0.0015

 $A^1 b^2$: A^1 is p value <1%; b^2 is p value <5%.



FIGURE 6: The possible remediated strategy of RW for AMD-contaminated lakes.

the bottom constructed sediments, the decomposition of the additive sewage sludge or organic substances from the surface wetlands is always occurring. These processes can form inorganic C, N, and P, which will diffuse to the surface waters for the surface wetland plants to grow and remove metals by the adsorption or exchange from the water column.

Effectively buffering the acidity in these systems is a crucial issue in this study. The generation of alkalinity to buffer the acidity in acidic aquatic ecosystems is a remediation process that occurs spontaneously in nature [37]. The primary production of the photosynthetic organisms (e.g., phytoplankton, periphytic algae, or moss) is associated with natural alkalinity-generating processes in acidic systems [12, 33]. However, consuming acidity by biological reduction in the sediments or under anaerobic conditions plays an important role [33, 38]. In most aquatic ecosystems, the bottom sediments are under anoxic conditions. Under these anaerobic conditions, the nitrate reduction (denitrification), manganese reduction, iron reduction, and sulphate reduction would occur in an orderly fashion [33]. These reduction reactions are mediated by the indigenous microorganisms in anoxic environments where the appropriate electron acceptors (NO₃⁻, Mn⁴⁺, Fe³⁺, and SO_4^{2-}) and electron donors (usually organic substances) are present. These processes consume hydrogen ions (H⁺), which leads to an increase in the pH [12, 33].

For those alkalinity-generating processes, RW effectively increased the pH of the waters in the lakes (Figure 3(a)), which promoted the precipitation of metals. Therefore, most metal ions with low K_{sp}, such as Fe, Mn, Cu, and Zn, could form soluble hydroxides or carbonates [39, 40]. The main metal ions, such as Fe and Al, formed hydroxide colloids, which can be adsorbed into organic compounds and several kinds of ions, and then coprecipitated [41, 42]. When these series of reactions progresses, the precipitation of oxides, hydroxides, and other organic particulates can result in the movement of these metals from the water column into the sediments [43]. The bottom constructed sediments will create anaerobic environments. Under these conditions, the anaerobic and sulphate-reducing bacteria can use the sulphate to oxidize the organic matter and release bicarbonate and hydrogen sulphide [44]. Then, the produced hydrogen sulphide readily reacts with the dissolved metals to form insoluble metal sulphides that subsequently precipitate. Ultimately, the elimination of most of the above ions (Figure 4) results in a gradual decline in EC (Figure 3(b)).

The plants on the banks and those that are submerged are more important to the biological component in aquatic ecosystem [45]. The decomposition of these litters plays a vital role in the normal circulation of materials, energy flow, and the health and stability of the aquatic ecosystem [29]. It is of great significance to employ litter degradation as an indicator for evaluating the effects of contamination and eco-remediation. The organic matter will be converted into inorganic matter by a large number of microbes and benthonic animal communities in NW for the continuous degradation of the litter [46], in which the microbes are active, breed normally, and have a better degradation effect (Figure 4) due to the neutral pH and the absence of harmful substances. However, in AMDW, not only are the activity of the aquatic organisms and the richness and quantity of the microbes and benthonic animals markedly inhibited due to the low pH (2.73); high salinity; high concentrations of Fe, Mn, Cu, Zn, Pb and other toxic elements; and the rust-like suspended particles [33], but the degradation rates of the litters are also decreased. AMD pollution could cause long-term impairment to the waterways and biodiversity, which has serious human health and ecological implications [47].

5. Conclusions

The ecological remediation conducted in this study had good effects on buffering the acidity and removing metals and sulphate, which ultimately improved the water quality and the degradation of litters. However, there are large differences between the NW and RW in the anaerobic situation, primarily in terms of increasing the organic matter. The ecotechnological remediation technology may be used as a pretreatment stage for the lake's water column. More studies are needed to develop a real and a sustainable remediation system in AMDW.

Data Availability

The main table and figure data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Kaiju Chen, Yan Zeng, Zhongzheng Wen, Zhengyan Ran, and Li He worked on conceptualization, investigation, and data curation. Tianling Fu and Hu Wang worked on writing the original draft, review and editing, and visualization. Tianling Fu supervised, reviewed, and edited the manuscript and took part in project administration. Yonggui Wu worked on supervision.

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