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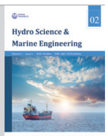

ARTICLE

Stream Monitoring and Preliminary Co-Treatment of Acid Mine Drainage and Municipal Wastewater along Dunkard Creek Area

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ARTICLE INFO

Article history
Received: 10 October 2020
Accepted: 26 October 2020
Published Online: 30 October 2020

Keywords:
Combined treatment
Mixing experiments
Anaerobic batch reactor treatment

ABSTRACT

This study investigated coal-mine drainage (AMD) and municipal wastewater (MWW) contaminant concentrations and conducted the combined treatment in phases I and II: phase I, evaluating the effects of mixing the two based on the extent of acid neutralization and metals removal; phase II: conducting anaerobic batch reactor treatment of AMD and MWW under varying COD/sulfate ratios (0.04-5.0). In phase I, acid mine drainage water quality conditions are as follows: pH 4.5, acidity 467.5 mg/L as $CaCO_3$, alkalinity 96.0 mg/L as $CaCO_3$, Cl^- 11.8 mg/L, SO_4^{2-} 1722 mg/L, TDS 2757.5 mg/L, TSS 9.8 mg/L, BOD 14.7 mg/L, Fe 138.1 mg/L, Mg 110.8 mg/L. Mn 7.5 mg/L, Al 8.1 mg/L, Na 114.2 mg/L, and Ca 233.5 mg/L. Results of the mixing experiments indicated significant removal of selected metals (Fe 85~98%, Mg 0~65%, Mn 63~89%, Al 98~99%, Na 0~30%), acidity (77~95%) from the mine water and pH was raised to above 6.3. The Phase II results suggested under the wide range of COD/sulfate ratios, COD and sulfate removal varied from 37.4%-100% and 0%-93.5% respectively. During biological treatment, alkalinity was generated which leads to pH increase to around 7.6-8.5. The results suggested feasibility of the proposed technology for co-treatment of AMD and MWW. A conceptual design of co-treatment system which is expected to remove a matrix of pollutants has been provided to utilize all the locally available water resources to achieve the optimum treatment efficiency. The technology also offers an opportunity to significantly reduce capital and operating costs compared to the existing treatment methodologies used.

Featured Application: In this study, we have measured the concentrations of contaminants in acid mine drainage (AMD) and municipal wastewater (MWW) and conducted the combined treatment in phases I and II. This is significant because in previous years there was a massive fish kill that is linked to the high total dissolved solids (TDS) and salinity content in the water. With the current proposed combined treatment technology, it shows high potential in reducing TDS and salinity content in the combined wastewater which will prevent similar accident (Dunkard creek fish kill in 2019) happening again.

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DOI: https://doi.org/10.30564/hsme.v2i2.2448

1. Introduction

Acid mine drainage (AMD) originates from oxidation of mine rocks exposed to air and water during mineral extraction which leads to the generation of sulfate, dissolved metals (mainly iron) and low pH [1,2]. It degrades water quality in rivers near mining regions. It is estimated that over 3,000 miles of streams are impaired in the Appalachia due to acid mine drainage [3]. As such, the remediation of these bodies is of primary environmental importance in watersheds with historical mining practices. The key focus on treating AMD is pH neutralization, metals precipitation, and sulfate reduction. In order to remediate AMD, a variety of treatment and prevention methods have been utilized throughout different stages of AMD formation process [4]. Generally, these treatments can be grouped into passive or active systems. Passive treatment generally includes biological activities in systems such as bioreactors, lagoons and wetlands [5]. Based on the sulfate concentration in AMD, one of the widely utilized passive treatment methods is sulfidogenic process (utilizing sulfate-reducing bacteria (SRB)) to reduce sulfate to sulfide, and to promote alkalinity production and metal sulfides formation [6,7]. For biotic sulfate reduction, it generally requires electron donors from organic sources [8-13]. A previous study from applied various organic sources (e.g., wood chips, sawdust, leaf mulch and sheep manure) for sulfate reduction and suggested higher sulfate reduction rates with addition of sewage sludge compared to no sludge addition [7]. Additional studies also showed that diversified sources of electron donors from municipal wastewater (MWW) were more preferable for sulfate reducing bacteria growth [12,14]. Active systems are often required for higher flow rates or more complex treatment requirements. This approach offers enhanced treatment flexibility to adapt to fluctuations in contaminant concentration, flow rate, and other environmental conditions. After treatment, any number of separation methods can be employed, with common applications including sedimentation, filtration through granular media, and membrane filtration [15]. Although active chemical treatment can effectively treat AMD, it is generally associated with high operational costs and massive amounts of sludge being produced [16].

Municipal wastewater (MWW) generally features high pH, TSS, alkalinity, COD and the typical range is: pH of 7-8, alkalinity from 200-250 mg/L as $CaCO_3$ [17,18]. Generally, MWW treatment needs to reduce the suspended solids, phosphorus, nitrogen and biochemical oxygen demand concentrations to certain limits in order to maintain receiving water bodies from water quality degradation and negative human health impact. Traditional MWW treatment consumes significant amount of economic, material and energy resources. Sludge scraping, mechanical aeration, sludge and effluent pumping, ultraviolet disinfection, clarifier skimming, and other conventional MWW treatment methods consume substantial amount of energy [18-20]. Among the various treatment processes, aeration of activated sludge is the most energy consuming process, typically accounting for 45% of total energy consumption [21], and energy usage is around 0.28-0.71 kWh/m^3 [22].

In MWW, suspended solids are generally removed by either filtration, flocculation, biodegradation, or settling [18]. Phosphorus are often removed by ferric iron or aluminum salt dosing [23]. However, these chemical dosing can be considerably expensive and consumption of these flocculants has increased over recent years [24,25]. This indicated that AMD which contain high amount of ferric iron salts can be utilized for phosphorus adsorption [26]. In anaerobic treatment of AMD which contains sulfate reducing bacteria, nutrients (nitrogen, phosphorus) can be supplied from MWW for cost-effective operation [5,27]. Organics (Biochemical oxygen demand) in MWW can also be lowered by bacterial respiration through sulfidogenic reaction. So MWW and AMD serve as perfect combination to remediate the overall water quality.

Co-treating AMD and MWW could be cost-effective and mitigate infrastructure challenges of building two separate treatment systems in places where these two wastewaters are existing but financial resources are limited. The study of co-treatment of AMD and municipal wastewater (MWW) has long been explored. A paper first discussed mixing of AMD and MWW has great potentials in reducing pathogens by low pHs and high metal concentrations in AMD, but has not studied the combined treatment potentials [28]. Another paper discovered that Escherichia coli population was significantly decreased when adding AMD to MWW which indicated combined treatment has promoted disinfection effect [29]. Another study investigated varying pH impact on sewage related microbes and found a rapid decreasing trend in bacteria concentration with lowering pH values [30].

The AMD can be added directly to the activated sludge tank, either as a raw stream or after a pre-treatment such as mixing with digested sludge or screened MWW. The effect is a mutual benefit to both wastes while maintaining the integrity of the parent wastewater treatment facility. A combination of dilution by and alkalinity of the MWW and anaerobic digester sludge neutralized the acidic discharge, which in turn causes metal ions to precipitate from the solution. The AMD, meanwhile, aids in removing MWW constituents such as phosphorus, which adsorb and

DOI: https://doi.org/10.30564/hsme.v2i2.2448

co-precipitate with the metal (oxy) hydroxides in a loose sludge as with the previous processes.

The AMD and MWW sampling sites (supplementary Figure S1) are located around a Dunkard river network affected by AMD associated with the intensive mining of metal sulfides. Releases of AMD have low pH, high concentrations of iron, calcium, aluminum, sodium and manganese, high sulfate and chloride, high specific conductivity, high total dissolved solids, and low concentrations of toxic heavy metals. Currently the lower Dunkard Creek water quality is found to be affected by three sources: AMD discharges from abandoned mines: high metal loadings; wastewater from approximately 1,000 residents which is treated to various degrees; alkaline mine drainage discharge via Steele Shaft treatment plant: high TDS. This study is to evaluate an innovative process for addressing all of these problems in a single, comprehensive treatment system. The research objective is to be achieved by conducting the following using a phased approach: Phase I: evaluate pollutant concentrations and trends, and preliminary treatment of combined AMD, municipal wastewater and steel shaft discharge; phase II: conduct the treatment process utilizing anaerobic batch reactor analysis and study microbial sulfate reduction reaction kinetics to develop a conceptual design of the combined treatment process.

2. Materials and Methods

2.1 Site Description

The location of sampling points has been provided in supplementary Figure S1 and Table 1 shows values for pH, acidity, alkalinity, chloride, sulfate, TDS, TSS, BOD, and metals content (iron, Magnesium, Manganese, aluminum, sodium, calcium) in the sampled sites. Site 2A drainage comes from abandoned mines. Site 2B is very close to 2A, and the distance of the two is no more than 10 meters. The water is also from abandoned mines and has similar characteristics for parameters with 2A. Site 7A sample had a nearly neutral pH. However, it has higher acidity than 2A and 2B which can be explained by the Fe content (228 mg/L) compared to 2A and 2B (33 mg/L, 28 mg/L). Site 7B is further upstream and not far from 7A. It had significantly different characteristics from 7A. Site 8A has high acidity which is attributed to its Fe concentration. Site 8B is close to 8A. These two had similar chemical characteristics. Water coming out from Steele Shaft facility has been treated with alkaline materials to raise the pH of acid mine drainage. The discharges were high in sulfate, and sodium concentration. The primary influent, MWW(P), was collected at a location after the bar screen and the secondary influent, MWW(S), was at a location after the clarifier.

2.2 Field Sampling

Phase I: water samples were collected from ten (10) sites including mine drainages (2A, 2B, 7A, 7B, 8A, 8B), Steel Shaft discharges (two separate pipes for replicates analysis – SSD-A, SSD-B), and primary and secondary influents from the Bobtown wastewater treatment facility (MWW(P), MWW(S)). Steel shaft discharges refer to a mine drainage treatment plant by adding alkaline chemi-

Table 1. Acid mine drainage, municipal wastewater and steel shaft discharge characteristics summary in the Dunkard Creek Sampling Sites

Parameters	**Unit**	**2A**	**2B**	**7A**	**7B**	**8A**	**8B**	**SSD-A**	**SSD-B**	**MWW(P)**	**MWW(S)**
pH		2.7	2.7	5.9	6.9	5	4	7.8	8	7.3	7
Acidity	mg/L as $CaCO_3$	351	284	616	48	736	770	8.6	3.2	47.5	20.2
Alkalinity	mg/L as $CaCO_3$	0	0	156	360	48	11.7	153	187	237.2	84.5
Cl^-	mg/L	9.1	8.6	13.3	9.8	16.6	13.6	71	79	101.3	98
SO_4^{2-}	mg/L	1603	1335	1991	512	2463	2428	7057	5680	92.5	82.4
TDS	mg/L	2478	2032	3200	1144	3863	3828	10395	8390	340	290
TSS	mg/L	4.6	11.3	12.4	9.6	13.3	7.8	6.4	19.3	86.6	11.6
BOD	mg/L	6.7	7.9	25.7	10.9	12.2	24.6	7.8	8.9	>136.2	>26.1
Fe	mg/L	33	28	228	1.5	274	264	0.2	1.3	0.4	0.2
Mg	mg/L	121	111	117	50	131	135	217	156.5	7.3	7.5
Mn	mg/L	3.8	3.8	10	1.3	13	13	0.4	0.1	0	0
Al	mg/L	24	23.5	0.2	0.1	0.3	0.5	0.1	0.1	0.2	0
Na	mg/L	33	35	165	148	152	152	2508	3765	65.9	67.4
Ca	mg/L	238	226	257	131	270	279	565	355	163	159

cals (limestone, sodium hydroxide, etc.) to neutralize the water turn it into alkaline mine drainage. Field measurements of pH, temperature, turbidity and electrical conductivity were taken. SSD, AMD and MWW samples were collected and transported under refrigeration to laboratories and were stored at 4˚C before analysis. Sample holding time for acidity, alkalinity, TSS, TDS, and nutrients was less than 24 hrs. Concentrations of Cl^-, SO_4^{2-}, COD, and metals were analyzed within 1 week of the sampling. AMD samples collected from the above-mentioned six (6) locations were mixed together in equal volumes to make a combined AMD solution for experiments with MWW sample and SSD sample.

In phase I (1) the AMD samples were mixed with MWW at different ratios to investigate the feasibility of co-treatment of the two waste streams. In phase I (2), mixing experiments were conducted using AMD, alkaline SSD, and MWW to evaluate the effects of combined mixing on removing metals, acidity, and other constituents in the mine drainage samples, since it is more closely simulate what actually will happen in real environment based on the location of the three streams.

Phase II: water samples were collected from AMD site 8A along Dunkard Creek, Pennsylvania, and wastewater samples (MWW (P)) from the Bobtown, Pennsylvania and Star City, West Virginia wastewater treatment plant. Field measurements of pH, temperature, turbidity and electrical conductivity were taken. SSD, AMD and MWW samples were collected and transported under refrigeration to laboratories and were stored at 4˚C before analysis. Sample holding time for acidity, alkalinity, TSS, TDS, and nutrients was less than 24 hrs. Concentrations of SO_4^{2-} and COD were analyzed within 1 week of the sampling.

2.3 Experimental Procedure

2.3.1 The Phase I Mainly Involves the Mixing of AMD, MWW, SSD to Promote Chemical Precipitation

(1) Mixing experiment 1

The first set of mixing experiments was performed using mixture of all the mine drainages from the six locations collected (1:1:1:1:1:l volume ratios) and mixture of the primary and secondary influents (1:1 volume ratio). Two different mixing ratios between the mine drainage mixture and wastewater mixture were used: 200mL (AMD mixture) + 400 mL (sewage MWW mixture) (abbreviation:200A+400S), 300 mL (AMD mixture) + 300mL (sewage MWW mixture) (abbreviation:300A+300S). The results are summarized in Figure 1.

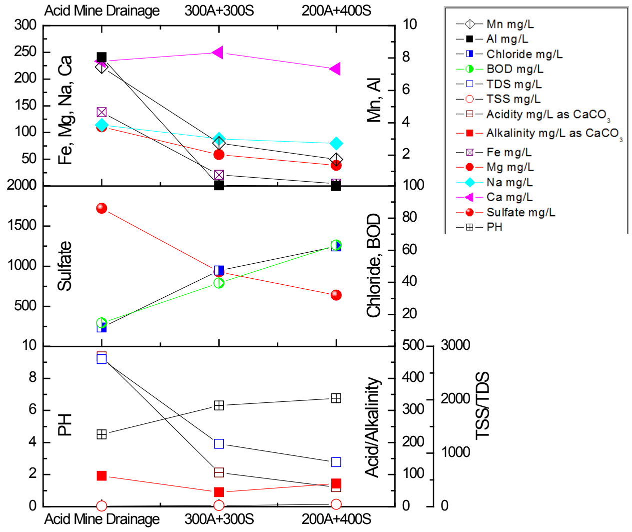

Figure 1. Results of mixing experiments 1

(2) Mixing experiment 2

The second set of mixing experiments was conducted using mixture of all the AMD collected (1:1:1:1:1:l volume ratios), mixture of the primary and secondary influents (1:1 volume ratio), and the alkaline SSD. Two different ratios for the mixing of the mine drainage mixture, wastewater mixture and steel shaft mine drainage were used (the ratio is determined based on the in-situ flowrate of AMD and alkaline SSD: 200mL (AMD mixture) + 400 mL (MWW mixture) + 400 mL (alkaline SSD) (abbreviation:1+2+2), 100mL (AMD mixture) + 400 mL (MWW mixture)+ 400 mL (alkaline SSD) (abbreviation:1+4+4). The results are summarized in Figure 2.

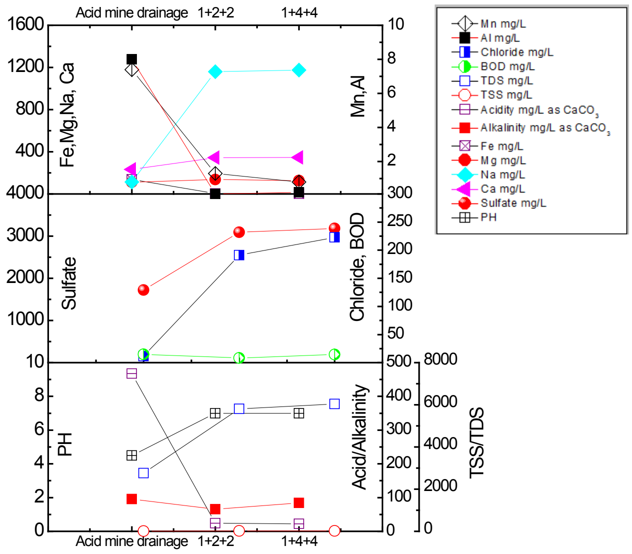

Figure 2. Results of mixing experiments 2

DOI: https://doi.org/10.30564/hsme.v2i2.2448

2.3.2 The Phase II Experiments Focus on Anaerobic Batch Reactor Treatment

The collected samples were mixed using different ratios and the mixture solutions (after settlement of particulate matters) were treated in anaerobic bioreactors for COD and sulfate removal in anaerobic conditions.

(1) Pre-setup mixing: a range of AMD: MWW mixing ratio was used to evaluate the chemical composition of the mixture solution.

(2) Reactor Series 1: Four different MWW: AMD mixing ratios (volume based): 1:1, 2.5:1, 2.5:1(with 20 g AMD soil addition to provide bacterial source for anaerobic degradation purposes, AMD soil refer to the soil sampled from AMD impacted sites), 5:1 were used. This resulted in a range of COD: sulfate ratios (0.2, 0.6, 0.8, and 5) and the mixture solutions were treated in four bioreactors respectively.

(3) Reactor Series 2: Another series of four bioreactors were set up to treat mixture solutions with lower COD: sulfate ratios (0.06, 0.04, 0.05, and 0.13) than those in Reactor Series 1. Probably because of the heavy rain season, organics content in municipal wastewater (Bobtown, PA) has been relatively low.

(4) Reactor Series 3: In order to achieve COD to sulfate ratios similar to Series 1, Star City Wastewater Treatment Plant was sampled instead of Bobtown Water Treatment Plant to obtain higher content of COD and acid mine drainage was sampled from the same place around Dunkard creek area. COD/sulfate ratios of 1.6, 2, 2.4, and 3.1 (This is COD to Sulfate ratio in terms of concentration) were obtained.

2.4 Chemical Analysis Procedures

All samples were analyzed after filtration with 0.45 μm filter paper. pH, TSS, TDS, and BOD were measured according to Standard Methods [31]. pH and conductivity were tested using pH/conductivity meter (YSI 63). COD and sulfate concentrations were analyzed using a UV-Vis spectrophotometer (Hach DR2800). Alkalinity and acidity were analyzed using autotitrators (Thermo Scientific Orion 950 and MettlerToledo DL50). Samples for metals analysis were digested by concentrated nitric acid (~70%, trace metal grade), and measured using an atomic adsorption spectroscopy (Perkin Elmer 3100). Duplicate sample has been analyzed to ensure accuracy of experiments.

3. Results and Discussion

3.1. Phase I Study

Mixing Experiments

From Figure 1 and 2, the two series of mixing experiments in phase I exhibit similar results in almost all the parameters, each AMD + MWW and AMD + MWW + SSD treatment produced substantial alkalinity and raised pH to around 6-7, which is neutral, so it does indicate an ideal environmental for microbial growth. Besides the added alkalinity and dilution of acidity that resulted from mixing MWW and AMD, significantly increased pH was observed. These results indicate that the full advantages of mixing AMD with MWW has been realized and further acidity can be buffered during anaerobic incubation phase. It is unknown if the primary mechanism for acid neutralization is abiotic (i.e., sorption to organic ligands) or biotic (i.e., biological sulfate reduction) because no control study has been performed in phase I. Additionally, based on the paper [5] substantial biological sulfate reduction is unlikely to happen due to the relatively short period of mixing time (24 hours) and the present metal concentrations (Fe) are much greater than those reported to be severely inhibitive to sulfate reducing bacteria [32]. Therefore, it is assumed that the majority of the acid neutralization and alkalinity generation observed when mixing AMD + MWW and AMD+MWW+SSD were via abiotic process. In the mixing experiment 2, the acid neutralization capacity provided by SSD (mainly composed of limestone chemicals) was sufficient to produce net-alkaline effluent. And this is highly promising for treatment purposes because it serves as alkaline addition to co-treat AMD and MWW, which could result in significant cost savings. In addition, further alkalinity may be generated by sulfate reducing bacteria in phase II using the substantial amount of biodegradable organic material in MWW.

As expected, dissolved concentrations of major metal elements (in the mg/L range) of interest in the mixed AMD decreased with MWW exposure (Figure 1). The AMD + MWW mixtures resulted in decreased dissolved Al, Fe, Mn, Mg and Na. The AMD + MWW + SSD exposures resulted in less removal of some metals (Mg, Na and Ca) due to the high concentration of these metals in SSD (Table 1). Overall, results indicate that passive mixing and incubation of AMD with WW can remove significant quantities of heavy metals (Mn, Al and Fe) from solution. The removal mechanisms are likely due to the increase in pH which promoted the formation of the insoluble $Al(OH)_3$. Besides, Al and Fe can combine with phosphate to form a stable solid, and they can also complex with organic materials to form ligands [23]. Additional studies are needed to evaluate more variation of ratio of AMD to MWW and mixing duration to optimize Al and Fe removal.

In general, Mixing experiment 1 and 2 both resulted in significant decreases in acidity, iron, manganese, alumi-

DOI: https://doi.org/10.30564/hsme.v2i2.2448

num, and the rest of the parameters relatively remaining on the similar level with the mine mixture (mixing experiment 1) or those even higher than the mine mixture (mixing experiment 2), this is probably because of the municipal wastewater and Steel Shaft water (as the Table 1 showing) exhibits fairly much higher concentration on those parameters, like sodium, calcium, chloride, sulfate and TDS, so this has influenced the mixing results in sulfate, chloride, sodium, calcium. However, the elevated metals and sulfate concentration can be removed in later stage, the anaerobic treatment process. And above all, the preliminary goal of the mixing is to decrease acidity and Fe which has been achieved, and the pH is raised to around 7. It is perfect for microorganisms, which suggests a promising future for microbial growth and further removal of metals and sulfate.

3.2. Phase II Study

Phase II study followed the concept of preliminary study of combined treatment in phase I and investigate further into biological treatment processes. In this phase, combined treatment has been conducted through two-step: mixing and biological treatment.

3.2.1 Pre-setup Mixing

Mixing ratios of 1:5, 1:10, and 1:15 (AMD: MWW, volume based) were performed to determine proper mixing ratio for subsequent anaerobic biological treatment. The mixing water characteristics are provided in Table 2. In order to provide sufficient organics for anaerobic biological degradation process, AMD: MWW mixing ratios were chosen based on COD/sulfate concentrations. So AMD: MWW mixing ratios of 1:1, 1:2.5, 1:2.5S (20g AMD soil added) and 1:5 have been chosen.

Table 2. Chemical Composition of the mixture solutions from the Pre-Setup Mixing

Parameters	**Unit**	**1:5**	**1:10**	**1:15**
pH		6.5	6.7	6.7
Acidity	mg/L as $CaCO_3$	98.4	88.3	93.8
Alkalinity	mg/L as $CaCO_3$	208.9	232.7	248.2
Cl^-	mg/L	95.9	95.9	101.4
SO_4^{2-}	mg/L	516.3	369.6	333.5
TDS	mg/L	510	694	1701
TSS	mg/L	307	340	351
BOD	mg/L	200.3	226.2	290.1
Fe	mg/L	31.5	14.1	12.6
Mg	mg/L	23.6	17.1	14.1
Mn	mg/L	1.4	0.8	0.5
Al	mg/L	0.7	0.4	0.3
Na	mg/L	123	113	107.5
Ca	mg/L	85.5	72	66.5
Cu	mg/L	0	0	0
NO_2^-	μg/L	1.7	10.5	10.6
NH_3	mg/L	37.1	34.8	37.5
NO_3^-	mg/L	0.2	0.4	0
PO_4^{3-}	mg/L	0.1	0.3	0.8

3.2.2 Anaerobic Bioreactor Treatment

(1) Reactor Series 1

A series of four bioreactors (Series 1) were set up using Boston Round bottles. The bioreactors were used to treat mixture solutions of different AMD: MWW volume ratios – 1:1, 1:2.5, 1:2.5 and 1:5 with 20 g of AMD soil. The volume mixing ratios resulted in initial COD: sulfate ratios of 0.2, 0.8, 0.6, and 5, respectively. Each reactor contained solid surfaces for biofilm development (i.e., attached growth of microorganisms). The results show that all mixing ratios produced satisfactory sulfate and COD removal in the reactors (COD reduction of 67.1%-88.9% and sulfate reduction of 60.8%-93.6%) (Figure 3). COD was dramatically and significantly decreased in the bioreactors containing AMD+MWW mixtures (67.1%-88.9%). The activity of various heterotrophic microbes (i.e., sulfate reducing bacteria, nitrate reduction, fermenters, methanogenesis and iron reducing bacteria) likely served to decrease organics throughout the system and has reported in previous studies [33,34]. PCR and qPCR results indicated that sulfate-reducing bacteria were dominant in the microbial communities and the resulting dsr A gene concentrations ranged from 13.3-15.0 log gene copies/μL) which are significantly higher than the levels in the original of AMD (9.7 log gene copies/μL) and MWW (12.5 log gene copies/μL) [34]. Therefore, the authors concluded that the dsrA genes were enriched as a result of the biological treatment. It has been reported that there are 2 to 3.5 copies of dsrA gene per SRB cell [35], and the number of active sulfate reducing bacteria microorganisms can be reasonably estimated from the gene concentrations.

DOI: https://doi.org/10.30564/hsme.v2i2.2448

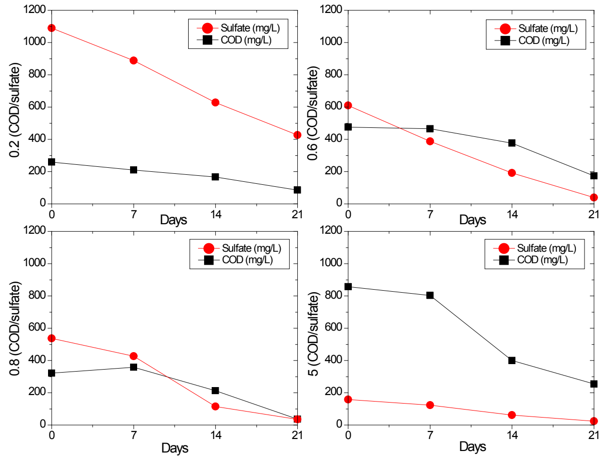

Figure 3. COD and sulfate concentrations in the reactor containing 0.2, 0.6, 0.8 and 5.0 (COD: sulfate) mixture solution ratio

(2) Reactor Series 2

Reactor series 2 refer to the second sampling trip taken to collect AMD and MWW water samples to test the treatability of the mixtures with the same volume mixing ratios of AMD and MWW but result in lower COD: sulfate concentration ratios (0.06, 0.04, 0.05 and 0.13). This could possibly be attributed to the rain season. The bioreactors (Figure 4) produced satisfactory reductions of both COD and sulfate as well (COD reduced 91.9%-100% and sulfate reduced 20.1%-73.2%). This indicates once the microbial community has been established, it can treat much lower COD/sulfate ratios and still achieve satisfying results. The limiting factor in the reactor series 2 is COD concentration since in all COD/sulfate ratios organics has been degraded to below detection level. And when the organics were below detection level, there would be no electron donors for sulfate reducing bacteria to continuously obtain electrons to reduce sulfate.

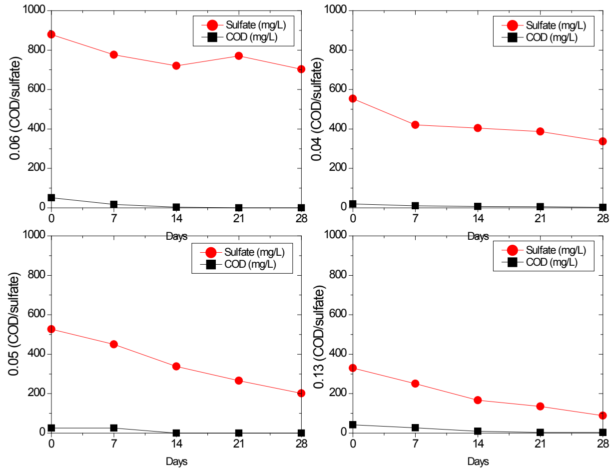

Figure 4. COD and sulfate concentrations in the reactor containing 0.06, 0.04, 0.05 and 0.13 (COD: sulfate) mixture solution ratio

(3) Reactor Series 3

In Reactor series 3, MWW was sampled from the Star City wastewater treatment plant in order to provide sufficient organics for biological sulfate reduction process which resulted in COD/sulfate concentration ratios of 1.6, 2, 2.4 and 3. COD reductions were observed in all four reactors. However, the bioreactors produced mixed results for sulfate reduction (Figure 5). There was a general trend of COD reduction in the bioreactors and for reactors containing COD/sulfate of 1.6, 2 and 3.1 the sulfate is also decreasing substantially. Possible reasons for the inconsistent sulfate concentration trends in reactor COD/sulfate of 2.4 include: a. Different source of wastewater: different composition of wastewater collected from Star City wastewater treatment plant may have caused different microbial responses due to carbon source, pH, and other potentially toxic chemicals. b. Increasing pH over the course of the treatment: pH 5-8 is favorable for microbial growth, and a pH above 8.0 is damaging to microbial community. Fromthe 2nd week, pHs in the bioreactors were higher than 8 which indicated unfavorable environment for the microbial community and resulted in unstable performance (Supplementary materials Figure S2).

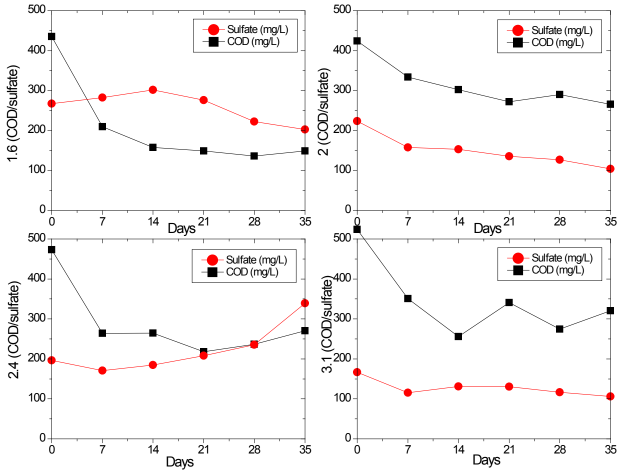

Figure 5. COD and sulfate concentrations in the reactor containing 1.6, 2, 2.4 and 3.1 (COD: sulfate) mixture solution ratio

Overall, the three series of reactors were conducted under a wide range of COD/sulfate ratios from 0.04-5.00. Almost all of the ratios (except COD/sulfate of 2.4) achieved promising results for COD/sulfate reduction, which indicate sulfate reduction related bacterial community has been established. The two step combined treatment process lead to pH increase, COD/sulfate reduction, metals reduced (Fe, Ca, Mg, Mn), and TDS/TSS removal (from Phase I and Phase II).

The full-scale AMD and MWW co-treatment systems

DOI: https://doi.org/10.30564/hsme.v2i2.2448

may result in energy, chemicals, and cost savings. Aside from low energy and low-cost set-up, these systems could be operated using gravity flow without ongoing purchased energy inputs. A field-scale co-treatment system could be applied in commonly engineered structures, such as ponds, aerobic wetlands, clarifiers, and vertical flow bioreactors, which could decrease engineering costs. Significant cost savings would result from eliminating the need to purchase and transport organic substrate, often a major cost of passive AMD treatment. Cost savings would also result by using AMD as a coagulant/flocculant and disinfectant for MWW treatment. In addition, the use of MWW as no-cost organic substrates consumes an item considered as "waste" to relatively-costly organic substrate, such as the compost or refined carbon sources (e.g., ethanol, methanol) often used in AMD treatment [36]. Raw MWW and high-strength AMD are often not independently addressed with passive methods because of the limitations of conventional passive treatment technologies and/or the lack of locally available suitable carbon substrate [37].

3.3 Conceptual Design and Recommendations

The bioreactors used in this study simply demonstrated feasibility of such treatment sequence (i.e., mixing followed by anaerobic biological treatment). So a conceptual design of the proposed treatment method is recommended for combined treatment of the mine drainages and sewage from the Bobtown, PA (Supplementary materials Figure S4 and Figure S5).

Conceptually, collection lines upstream of the proposed treatment pond (Steel Shaft lines and AMD discharge) could be run combined based on the mixing results given. In addition, a pumped line would convey MWW to the treatment system. The treatment pond could be configured to optimize residence time and reactivity under a collection system that combines Steele Shaft discharge and AML (Acid Mine Leachaye) discharges to form chemical precipitates and settle solids. Then the effluent is then mixed with MWW flow which provides organic matters for biological activities. A large pond would provide much of the biological reactions and settle remaining solids. Effluent could be discharged to Dunkard Creek at the downstream end of the treatment or piped to the Monongahela River.

Use the data obtained from reactor 1:1 of AMD/ SWW volume ratio, series 1 (supplementary material Table S1), the influent concentration of sulfate from the mixture water is 1090.2 mg/L, and the effluent is 158.3 mg/L. This ratio has been picked since locally AMD and SWW were pumped at similar flow rate.

The detention time,

$$\theta = -\frac{\ln\left(\frac{Ceff}{Cinf}\right)}{\kappa}, \qquad (1)\ [18]$$

Where

C_{inf}= pollutant concentration in influent (mg/L)

C_{eff}= pollutant concentration in effluent (mg/L)

k = apparent pollutant removal rate (day^{-1})

Since this equation is fit for first-order k, we calculate k value as follows (supplementary material Table S1 and S2, Figure S1): k=0.0247 day^{-1}

So $\theta = -\frac{\ln\left(\frac{427\ \text{mg}/1}{1090.2\ \text{mg}/\text{L}}\right)}{0.0247/d} = 37.9d$, assume void ratio □=0.7, flow rate Q=4.0 cfs = 9 786.3 m^3/d (medium value of flow rate from AML),

$$V = \frac{\theta Q}{\eta} = \frac{37.9d \times 9786.3m3/d}{0.7} = 5.31\times105m3\ , \qquad (2)\ [18]$$

So the volume of the treatment pond is 5.31×10^5 m^3, or 1.87×10^7 ft^3.

4. Conclusions

In conclusion, the first set of mixing was effective for removing Fe, magnesium, manganese, aluminum, acidity, sulfate, and increasing pH to around 6. The second has significant removal rate on acidity, Fe and pH is reaching neutral. And at the same time, since the second set is a mixture of the three different sources of water, it more closely depicts what will really happen in the natural environment. But above all, the two set of mixing both reduced significant amount of acidity, Fe, aluminum, manganese, which is the main goal for our mixing experiments, and the pH is favorable for microbial growth which lays a good foundation for the later stage of the project, cultivating sulfate-reducing bacteria

This purpose of this study was to obtain better understanding of water quality along mining affected Dunkard Creek area and propose an optimum treatment method to improve water quality. Along the Dunkard Creek streamline, steel shaft alkaline drainage, AMD and municipal wastewater in Bobtown can be combined to treat the water more efficiently. Through aerobic mixing, metals, TSS and TDS can be reduced and mixture pH has been raised, and the following anaerobic biological treatment promote further organics and sulfate reduction, which would co-precipitate more metals when sulfide is generated and precipitated as metal sulfides.

The conceptual design of co-treatment system has been proposed to achieve the purpose of the study. More details need to be considered since the water quality from AMD and municipal wastewater depend by the season and other variables, and more practical concerns need to be taken into account. For better understanding of the whole treatment performance, microbiological community analysis, fate of sulfate and metals needs to be further analyzed.

Supplementary Materials

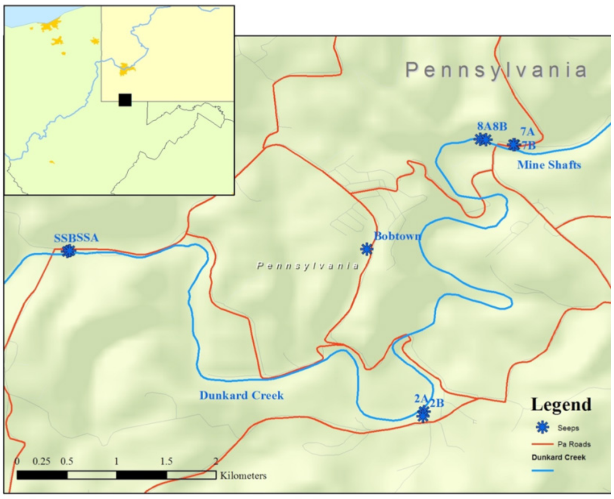

Figure S1. Sampling site locations of Acid Mine Drainage, Municipal Wastewater and Steel shaft discharge

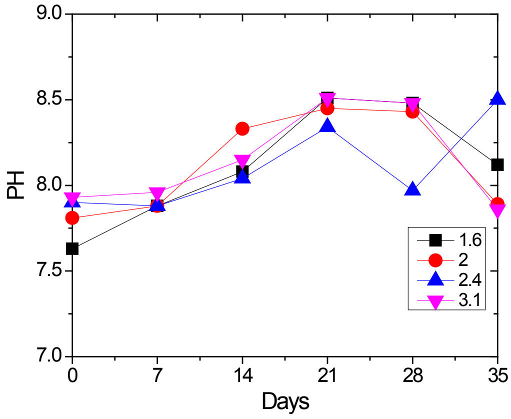

Figure S2. pH changes in the Reactor Series 3

Table S1. COD and Sulfate concentrations in 1:1 Reactors of Series 1

Time	Reactors	COD (mg/L)	Sulfate(mg/L)
1/24/2011	1:1	258.4	1090.2
2/4/2011	1:1	210.1	887.7
2/11/2011	1:1	166.6	627.8
2/18/2011	1:1	84.9	427

Table S2. $\ln C/C_0$ vs. Time in 1:1 Reactor of Series 1

$\ln(C/C_0)$	T(day)
1.00	0
0.81	11
0.58	18
0.39	25

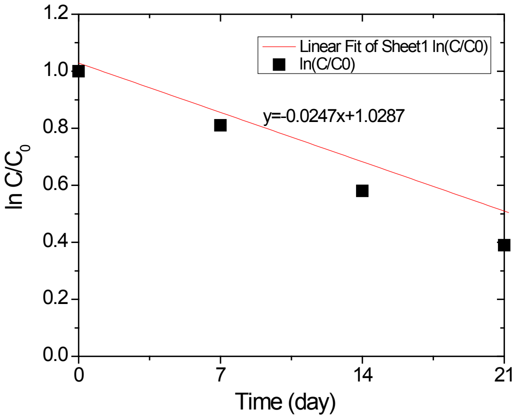

Figure S3. The linear relationship of $\ln C/C_0$ vs. time

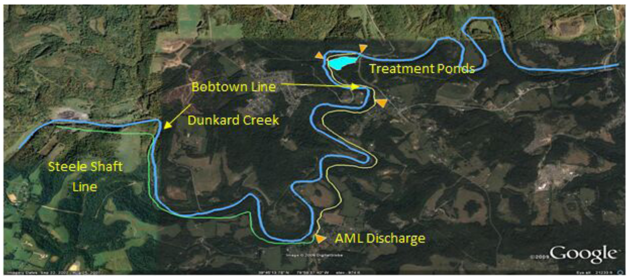

Figure S4. Combined mine water and wastewater flow to treatment system

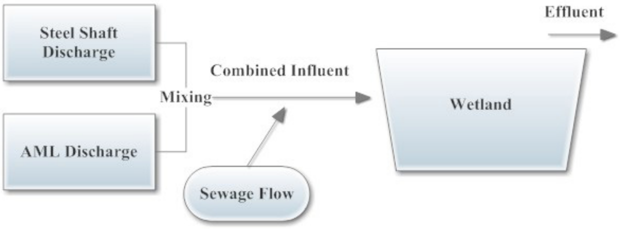

Figure S5. Conceptual Design of Treatment Process

Author Contributions

Conceptualization, D.D.Y., A.N.O-B. and L.-S.L..; methodology, D.D.Y. and L.-S.L..; validation, D.D.Y.. and L.-S.L..; formal analysis, D.D.Y.; investigation, D.D.Y. and L.-S.L..; resources, L.-S.L; writing—original draft preparation, D.D.Y. and L.-S.L.; writing—review and editing, D.D.Y., A.N.O-B. and L.-S.L..

DOI: https://doi.org/10.30564/hsme.v2i2.2448

Conflicts of Interest

The authors declare no conflict of interest.

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DOI: https://doi.org/10.30564/hsme.v2i2.2448