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Charles D. Spellman Jr.

Travis L. Tasker

Joseph E. Goodwill

William H.J. Strosnider

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Potential Implications of Acid Mine Drainage and Wastewater Co-treatment on Solids Handling: A Review

Charles D. Spellman Jr[1], Travis L. Tasker Ph.D.[2], Joseph E. Goodwill Ph.D. P.E. [3], William H.J. Strosnider Ph.D.[4]

[1] *Dept. of Civil and Environmental Engineering, University of Rhode Island, Kingston, RI, 02881. Email: cspellman@uri.edu*
[2] *Dept. of Environmental Engineering, Saint Francis University, Loretto, PA 15940. Email: ttasker@francis.edu*
[3] *Dept. of Civil and Environmental Engineering, University of Rhode Island, Kingston, RI, 02881 (Corresponding author). Email: goodwill@uri.edu*
[4] *Belle W. Baruch Marine Field Laboratory, University of South Carolina, Georgetown, SC 29440. Email: bill@baruch.sc.edu*

Abstract

Acid mine drainage (AMD) is a persistent and extensive source of water pollution and ecological degradation. Co-treating municipal wastewater (MWW) with AMD using existing infrastructure at conventional wastewater treatment plants (WWTPs) may serve as a potential option for AMD abatement. However, commonly elevated iron and aluminum concentrations and low pH of AMD could negatively impact various processes at a WWTP. The focus of this mini-review was to determine how co-treating MWW with AMD could impact the solids handling processes at a WWTP. While no studies have explored the solids that could be generated during co-treatment in a WWTP, there are numerous articles that separately discuss the solids generated during AMD or MWW treatment. Reviewing this literature revealed that iron and aluminum, common metals in AMD, are already present in MWW sludge and typically benefit most solids handling processes. The addition of AMD would elevate iron and aluminum concentration but would likely result in improved sludge dewatering, removal of odor-causing compounds during processing, and a decreased bioavailability of trace metals and water-soluble P in land applications. This review concludes that co-treating MWW with moderate-to-low volumes (< 50%) of AMD within WWTPs will have minimal impact, and likely improve, solids handling processes.

Keywords: Acid Mine Drainage; Wastewater treatment; Co-treatment; Iron; Waste management; Sewage Sludge.

Introduction

Global industrialization has brought about a plethora of legacy pollution issues, including acid mine drainage (AMD). AMD is created when sulfide-containing minerals, such as pyrite (FeS_2), are exposed to oxygen and water after mining or other types of land disturbance (Younger et al. 2002). The resulting discharges often have elevated acidity (some coal drainages may be net neutral), elevated sulfate and iron (Fe) concentrations from the oxidation of sulfide rock, and a variety of trace metals [e.g., aluminum (Al), manganese (Mn), copper (Cu), zinc (Zn), arsenic (As), and lead (Pb)] from low pH driven dissolution of surrounding rocks (Evangelou and Zhang 1995; Jacobs et al. 2014; Strosnider et al. 2011; Younger et al. 2002).

AMD abatement can be obtained by both passive (e.g. limestone dissolution, engineered wetlands) and active (e.g., chemical addition) treatment approaches (Hedin et al. 1994; Johnson and Hallberg 2005; Watzlaf et al. 2004). However, additional approaches can include co-treating AMD with other wastes such as organic solid waste substrates, agricultural slurry, fracking flowback water, or municipal wastewater (MWW) (Chang et al. 2000; He et al. 2016; Hughes and Gray 2013a; McDevitt et al. 2020). Benefits of co-treating AMD with other wastewaters includes providing low cost AMD abatement, improving effluent quality from treatment systems, and mitigating AMD impacts on receiving bodies.

Co-treating MWW with AMD could enhance conventional waste water treatment plant (WWTP) processes, including improved colloid destabilization (i.e. coagulation) during metal hydrolysis (Metcalf & Eddy et al. 2013), precipitative removal of biochemical oxygen demand (i.e. “enhanced coagulation”, Edzwald and Tobiason 1999), increased nutrient removal by phosphate adsorption onto metal hydroxides (Ruihua et al. 2011), and enhanced inactivation of fecal coliforms (Winfrey et al. 2010). Co-treatment may also offer opportunities for bioelectricity generation (Vélez-Pérez et al. 2020). Although successful co-treatment with AMD and MWW has been noted primarily in passive systems (e.g. Johnson and Younger 2006; McCullough et al. 2008; Strosnider and Nairn 2010), effective co-treatment has also been demonstrated in more conventional MWW treatment scenarios (Deng and Lin 2013; Ruihua et al. 2011; Wei et al. 2008). In a comprehensive bench scale examination of AMD and MWW co-treatment, Hughes and Gray (2012, 2013a; b) demonstrated improved phosphate adsorption, metal (e.g., Fe and Al) removal, decreased effluent chemical oxygen demand (COD) concentrations, and concluded that co-treatment should not degrade activated sludge system performance. Although the literature suggests that co-treating AMD and MWW within the existing infrastructure of WWTPs is feasible, there are substantial research gaps prohibiting full-scale adaptation. One overlooked factor is the impacts of AMD addition on a MWW facility's waste solids handling and subsequent disposal processes.

Nearly all WWTP processes generate physical byproducts classified as "solids" that require separate treatment and disposal. Solids, generalized as "sludge", encompasses pre-treatment grit, sludge from primary sedimentation, wasted activated sludge, and filtration backwash solids (Carnes and Eller 1972). Larger objects removed by screening (e.g. "rags") which are typically directly landfilled will not be classified as solids in the scope of this review. Solids treatment and disposal (i.e. "solids handling") is equally as intricate and important as liquid-phase treatment. Solids may require pre-treatment such as chemical-conditioning, thickening, and/or digestion which is traditionally followed by mechanical dewatering (filter pressing, centrifugation, etc.) (Carnes and Eller 1972). Treated solids can either be landfilled, incinerated, or conditioned for beneficial reuse. Conditioned solids for the purpose of land application are defined as "biosolids."

AMD strength (acidity, pH, metal concentrations, etc.) can vary greatly with mine type (coal versus hard rock drainage) and geographic location, meaning no two drainages are alike. However, the authors suggest that co-treating MWW with AMD, in general, could result in elevated concentrations of Fe and Al (found in most mine drainages) in solids generated during MWW treatment. These elevated metal concentrations may impact facilities solids handling processes. The primary objective of this review is to identify how certain AMD metals (i.e., elevated Fe and Al loads) from co-treatment with MWW may influence traditional WWTP solids handling processes.

Review Methodology

This review identified relevant peer-reviewed research that highlight the impact of Al and Fe on MWW solids handling processes. Relevant literature was identified through Google Scholar searches and was extracted from bibliography sections in relevant textbooks. Keywords that were used alone and in various combinations to find literature included “activated sludge”, “trace metals”, “acid mine drainage”, “iron”, “aluminum”, “metal hydroxides”, and “sludge handling.” There was no bias towards certain publications and all works were reviewed equally. The authors acknowledge the limited number of articles published within the last ten years, with the majority being published prior to 2010. However, all cited studies were screened via the Elsevier Scopus citation database (www.scopus.com) to ensure the cited information was the most recent and relevant.

Review Results

It is not uncommon for metals, especially Fe, to appear in MWW solids in substantial amounts. Typical concentrations of Fe range from 1 to 300 g per dry kilogram of MWW solids, with little information on Al or Mn (Environmental Protection Agency 2009). These metals are of little concern for WWTPs as they are relatively unregulated as sludge constituents. Neither Fe nor Al in solids is currently regulated as a pollutant for land application or landfilling (per U.S. Code of Federal Regulations Title 40, Part 503); Fe and Al are not mentioned in any of these regulations (as of March. 27, 2020) nor regulations for other countries (per European Union Directive 86/278/EEC). Generally, increasing Fe and Al concentrations in a facility's secondary treatment processes may have overall benefits for the WWTP. Elevated Fe and Al concentrations in sludge have been correlated with lower COD concentrations in plant final effluents, likely by coagulation mechanisms (Park et al. 2006). Improved effluent water quality is the primary aim for a WWTP, but Al and Fe addition by co-treatment will likely benefit other MWW treatment processes, such as solids handling. The benefits that could be provided by MWW co-treatment with AMD are summarized in Table 1 and discussed in depth in the following text.

Conditioning and Dewatering

Introduction of increased Fe and Al concentrations from AMD could improve sludge dewatering during co-treatment of MWW. Al and Fe salts that undergo hydrolysis are often used for sludge conditioning and to improve coagulation of suspended particles (Davis and Edwards 2014; Novak 2006). The addition of these metal salts decreases raw sludge specific resistance to filtration (SRF) and lowers the percent of “bound water” within the sludge, thus reducing the time needed for dewatering (Katsiris and Kouzeli-Katsiri 1987). Yu *et al.* (2016) demonstrated a negative curvilinear correlation between Fe(III) and sludge water (Fig. 1).

In comparison between Fe and Al coagulants, ferric Fe [Fe(III)] based coagulants remove approximately double the bound water compared to those treated with Al [82% vs only 48%

removal, respectively] (Katsiris and Kouzeli-Katsiri 1987). The decrease in bound water leads to more efficient and cost-effective sludge dewatering. Therefore, increasing Fe(III) concentrations by co-treating MWW and AMD may improve sludge settling and dewatering. It is also not uncommon for drinking water utilities that use metal coagulants to send their Fe/Al-rich sludge to a WWTP for disposal as an alternative to landfilling, as many drinking water facilities do not operate an on-site sludge handling system. WWTPs accepting these sludges have generally experienced no negative impacts on their treatment processes (Asada et al. 2010; Marguti et al. 2018).

The presence of Al in secondary MWW waste sludge has benefits that are similar to those provided by Fe. $Al(OH)_3$ concentration was demonstrated to be inversely proportional to the SRF, implying Al also improves sludge dewaterability (Hsu and Pipes 1973). Furthermore, anaerobic digestion of Al-rich sludge before dewatering further improved dewaterability by nearly two orders of magnitude. The Al particles act as “skeleton builders”, significantly strengthening the solids bulk structure and improving water movement out of the sludge (Lai and Liu 2004). However, certain Al species or complexes can lead to variability in dewatering performance. For example, polymerized forms (mixed with polymer) of hydrolyzed Al improve sludge dewatering by allowing higher resistance to compression (Cao et al. 2016). This, however, only holds implications for co-treatment scenarios where polymers are also added during dewatering.

Co-treating MWW with AMD may also serve as a low-cost alternative to implementation of an advanced oxidation process (AOP). AOP is a technique that can be used at WWTPs to generate numerous radicals that improve oxidation processes and sludge conditioning (Glaze et al. 1987; Neyens and Baeyens 2003). AOP can occur by mixing ferrous Fe [Fe(II)] with hydrogen peroxide, facilitating a Fenton reaction that generates hydroxyl radicals and Fe(III). Co-treatment with Fe-rich AMD could replace a Fenton AOP and retain comparable dewatering efficiency. Yu *et al.* (2016) directly compared sludge dewatering characteristics with addition of Fe in the form of either Fe(II), Fe(III), or a variation of the Fenton AOP process. The experiments mixed sludge and Fe (always 48 mg Fe/g sludge) in a conditioning tank followed by pumping the mixture to a pressure-controlled feed tank, and then dewatering the mixture via a laboratory diaphragm filter press. The addition of Fe improved dewatering to some degree in all cases when compared to control raw sludge (Fig. 2).

Although Yu *et al.* noted that Fenton reactions achieved the best performance, Fe(III)-alone without an AOP still significantly improved sludge water content. Increased Fe(III) content decreases the sludge cake water content by up to 15% compared to raw sludge (RS), suggesting that adding AMD through co-treatment could improve sludge processing. Conversely, Fe(II) yielded little to no improvement over the RS. An AMD discharge with an increased Fe(II) fraction would require significant oxidation for enhanced sludge processing. Although the Fe(II) results are noteworthy, it is of minimal concern for co-treatment adaptability as it will be rapidly oxidized to Fe(III) in a WWTPs aeration basin. However, this could be of concern for WWTPs co-treating MWW with AMD that store sludge in an anaerobic system with long detention times where Fe reduction would likely occur (Rasmussen et al. 1994). The stability of Al in the +3 oxidation state could be more suitable during anaerobic storage and processing (Park et al. 2006).

Co-treating MWW with AMD will also impact the pH of sludge and further influence the performance of the solids handling system. Sludge pH is inversely proportional to its dewaterability, with an optimum dewatering pH of 2.5 for both centrifugal or filtration dewatering (Chen et al. 2001). However, operating at extremely acidic pH values is likely not feasible due to elevated corrosion risks and slower oxidation rates of odor-causing compounds (Nielsen et al. 2006). Yet sludge flocs have been shown to still maintain structural stability over a pH range of 4.5 to 9.5 (Liao et al. 2002) while also maintaining SRF between pH 3 and 7, with less desirable SRF at higher pH values (Raynaud et al. 2012). Over a pH range of 3.2 to 9.1, lower pH values also correlated with a lower sludge shear sensitivity (i.e. stronger flocs) (Mikkelsen et al. 1996). The improved sludge dewaterability over a lower pH range is attributed to positively charged ions from acid compounds (e.g. H^+, Fe^{+3}, etc.) neutralizing the sludge particles surface charges. The neutralization leads to improved aggregation and a particle size distribution more conducive to dewatering (Karr and Keinath 1978). At a higher pH, the size distribution shifts to high concentrations of smaller particles that fill voids, trap water, and clog filtration pores reducing overall bound water movement (Raynaud et al. 2012). Although lower pH may improve dewaterability, dewatered sludge will require amendments (i.e., lime, etc.) to obtain the pH necessary for post-dewatering processes [e.g., minimum pH for biosolids land application is 12 (Doyle 1967)].

Odor Control and Anaerobic Processes

Managing odor is a common nuisance and cost burden at many WWTPs (Dague 1972). Fe from co-treatment may help mitigate odor at WWTPs and could present positive economic benefits for the immediate community by increasing surrounding property values by up to 15% (Lebrero et al. 2011). Divalent metal species in AMD, including unoxidized Fe(II), can scavenge and react with the primary odor-causing compound H_2S to form insoluble metal sulfide complexes which are non-odorous (Johnson and Hallberg 2005). This suggests that the addition of Fe(II) in mostly anaerobic settings (e.g. AMD added after aeration) would assist in decreasing odor causing compounds during solids processing. Oxidized Fe in aerobic co-treatment systems will also enhance odor reduction. The addition of zero-valent Fe (Fe0) nanoparticles at various doses to MWW sludge demonstrated improved oxidation of H_2S to form Fe sulfides and increased the final biosolids nutrient bioaccessibility (Li et al. 2007). The resultant Fe-sulfides further reacted with H_2S to form Fe polysulfides without the need for additional Fe input. Although the aforementioned study utilized Fe0, only the core of the nanoparticles contained Fe0 while the shell was oxidized and consisted of hydroxides/oxyhydroxides. These hydrolyzed Fe compounds are similar to those that would form after oxidation of AMD Fe.

Al addition also improves the overall anaerobic sludge digestion processes. Biogas often contains volatile sulfur compounds (e.g. H_2S, CH_3SH, CS_2) that cause nuisance odors and corrosion issues. Dosing Al can remove high percentages of these dangerous sulfur compounds from biogas while maintaining consistent digester performance (Akgul et al. 2017). Additionally, the total volume of biogas generated would be expected to decrease (Hsu and Pipes 1973) likely resulting from significant removal of volatile compounds. Furthermore, the same study showed a noticeable decrease in digester coliform counts as well as improved dewaterability after digestion. All of the aforementioned improvements could equate to significant cost savings for a WWTP, in addition to benefits from reduced odors. These results suggest that co-treatment with Al-rich AMD (which is relatively rare) would be most advantageous at a WWTP operating an anaerobic digestion system, due to the valance-stability of Al.

Biosolids Composition and Land Application

Although MWW solids may contain 1 to 300 g of Fe per dry kilogram of solids (Environmental Protection Agency 2009), only a handful of studies have examined the relationship between Fe and Al content in water and resulting biosolids Fe and Al content. As previously discussed, there are minimal regulatory standards for common AMD metals in biosolids. However, trace metals and metalloids (e.g. Pb, Hg, and As) in biosolids can have environmental and human health implications if they bioaccumulate or leach after land application (Arulrajah et al. 2011). Both As and Hg have frequently been investigated for their role in biosolids toxicity during land use. AMD from the eastern part of the United States rarely has As and Hg concentrations above drinking water standards (Herlihy et al. 1990), but elevated concentrations of various metals and metalloids of concern (e.g., As, Cd, Pb) can be found in other geographic locations which would have negative implications for co-treatment feasibility (Cheng et al. 2009; Rytuba 2000; Strosnider et al. 2011). Decreasing the bioavailability of trace metals and metalloids in soil is important when considering if biosolids can be applied to land. Fig. 3 demonstrates the difference in bioavailability of Pb during a field study when 99% Fe-powder was added to biosolids compost (109 g Fe/kg) and mixed with soil (Brown et al. 2012). Experimental analysis showed that 75% of the Fe in the amended biosolids was Fe(III), similar to what might be expected of co-treatment biosolids.

Although Fe amended biosolids decreased the bioavailability of Pb (Fig. 3) there was significantly less impact on As bioavailability (Brown et al., 2012). The increased retention of toxic compounds by elevated-Fe biosolids during soil application renders amended biosolids marketable not just as compost, but also as remediation substrate for sequestering trace metals (e.g. Pb) in soils (Farfel et al. 2005). It is important to note that the substantial concentration of Fe added (>80 g/kg) in the Brown *et al.* experiment would only be expected under co-treatment with a high ratio of Fe-rich AMD. This Fe concentration is likely orders of magnitude higher than what a typical AMD discharge [AMD Fe varies 1 µg/L to > 600 mg/L (Johnson 2003;

Strosnider et al. 2011; Younger et al. 2002) and 0.2-70 mg/L Fe for coal mine drainage (Strosnider et al. 2020)] might contribute in a co-treatment system. In these situations, solids trace metal bioavailability would likely not be improved as demonstrated in the Brown *et al*. low Fe (5 g Fe/kg) experiments. These results imply that decreased toxic compound concentrations could only be expected during co-treatment on a case by case basis as a function of AMD and MWW influent Fe concentrations and system Fe removal capabilities.

Both Fe and Al may benefit agricultural land application of biosolids. AMD metals have demonstrated potential related to improving soil phosphorus (P) availability. Adler and Sibrell (2003) showed that additions of neutralized AMD "flocs" to high-P soil (20 g floc / kg soil) could sequester roughly 70% of water-extractable P. A similar result was noted in a larger scale study, where application of manure mixed with AMD treatment residuals to a large parcel of farmland decreased the water-soluble P content (Sibrell et al. 2015). Similarly, mixing biosolids with Al-rich water treatment alum sludge improved agricultural crop yields in traditional potting soil by retaining higher concentrations of P at both laboratory (60 days) and greenhouse (105 days) scales (Mahdy et al. 2009). Furthermore, the application of Al-hydroxides (1 to 4% w/w) in the aforementioned study also decreased the total nutrient loading in greenhouse runoff. The reduction of soluble P in biosolids amended with Al-rich water treatment sludge is caused by the formation and precipitation of Al/Fe-P complexes or P adsorption unto hydroxides (Huang et al. 2007). Commercially available Fe(III)-rich biosolids have varying results on agricultural use, demonstrating improved growth size of oranges but no impact on pear growth (Pérez-Sanz et al. 2002). However, most studies examining agricultural Fe-rich biosolids applications demonstrated non-negative yet neutral impacts on fruit growth. There is a strong potential for AMD co-treatment biosolids to support localized agriculture. Co-treatment could reduce demands for artificial fertilizers and potentially decrease nutrient loading on waterways without negatively impacting agriculture processes.

Incineration Considerations

Co-treatment has the potential to impact sludge incineration operations. The aforementioned inverse relationship between Al and Fe content and sludge bound water would also improve the combustibility of the dewatered sludge, reducing stress on incinerator processes. Furthermore, the resulting ash would have increased amounts of extractable P (Farfel et al. 2005). Ash generated by incinerating sludge from co-treated MWW with AMD could improve nutrient recovery and be viewed as a beneficial reuse product. Due to increasing global stress on P demand, WWTP processes have long been a point of focus as a source of potential P recovery and recycling (Farfel et al. 2005; Ottosen et al. 2013). Ash product produced from a co-treating incineration facility with a high extractable P could alleviate local P demand. Incinerated sludge ash can contain up to 10% P by mass (Donatello and Cheeseman 2013) and the amount of P that is recoverable is directly proportional to ash value. Sludge ash can successfully be applied to land as a fertilizer (Bierman and Rosen 1994). Therefore, this beneficial use ash also carries economic incentives, as it is now a product to boost revenue rather than a waste. Furthermore, the extractable P-rich ash is significantly less dense than a dried and stabilized sludge making it more economically viable to transport.

There are also disadvantages to be considered for incineration facilities. Depending on the water chemistry of the AMD, the ash could contain higher weight-percentages of toxic trace metals (e.g. As and Pb). Ash containing > 100 mg/kg of Pb would be considered a hazardous waste and could not be disposed of in a traditional municipal landfill. Pb concentrations in municipal landfills can be indirectly associated with a variety of health issues for neighboring communities (Kim and Williams 2017), and remains a liability for the generator. Furthermore, As is a primary contaminant in landfill leachates (Pinel-Raffaitin et al. 2006), and a landfill would likely not accept As-containing wastes due to the potential costs required for As treatment after leaching.

Conclusions

From a solids handling perspective, co-treating MWW with AMD could provide numerous benefits for a WWTP. The metals common to AMD (e.g., Fe, Al) are already present in conventional MWW sludges, and additional loads from co-treatment would not result in concentrations above those seen in some facilities. Current regulations indicate that sludges with high concentrations of these metals can be easily disposed in landfills or land applied. When Fe and Al concentrations are elevated, they may provide additional benefits that could make co-treating MWW with AMD more economically viable. For example, elevated Fe and Al can improve sludge dewatering, potentially lowering operating costs. The Al in these sludges can also decrease concentrations of odor causing compounds that are often challenging to control at WWTPs. Other opportunities might exist to use the biosolids or incinerated sludges (i.e., ash) from co-treating MWW with AMD for soil remediation or agricultural amendments (e.g. immobilizing trace metals in contaminated soils). While the findings of this review suggest that there are potential benefits from co-treating MWW with AMD, many questions remain to be answered before full-scale implementation. Further research into potential impacts from other common AMD metals (Cu, Mn, Zn, etc.) is needed. Future work should also include laboratory scale studies to investigate the outcomes of this review in various co-treatment scenarios (e.g. dewatering, digestion systems, incineration, and land application).

Data Availability Statement

No data, models, or code were generated or used during the study

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Table 1. Summary of key improvements on solids handling from co-treatment.

Potential benefits of co-treating MWW with AMD	Relevant Citation
AMD addition would elevate iron concentrations above a facility's current levels, but resulting concentrations would likely not exceed those already commonly observed in typical MWW sludges (1 to 300 g Fe per dry kg)	EPA, 2009
Iron and aluminum concentrations are correlated with a decrease in COD concentrations	Park et al., 2006
Increases in iron concentration generally reduces sludge water content	Katsiris and Kouzeli-Katsiri, 1987; Yu et al. (2016)
Aluminum decreases sludge specific resistance to filtration	Hsu and Pipes, 1973
WWTPs co-treating MWW with AMD containing high iron concentrations have ability to easily adapt to a Fenton AOP	Yu et al. (2016)
Decrease in pH improves dewaterability	Raynaud et al 2012; Karr and Keinath 1978
AMD metals may precipitate nuisance odor-causing compounds	Johnson and Hallberg, 2005
Aluminum addition is advantageous for WWTPs with anaerobic digestion	Akgul et al., 2017; Hsu & Pipes, 1973
Iron can decrease bioavailability and mobility of trace metals in land application	Farfel et al., 2005; Brown et al. 2012
Iron-rich biosolids decrease water-soluble phosphorus content when added to fertilizer	Adler and Sibrell 2003; Farfel et al., 2005; Sibrell et al., 2015
Incinerated sludge may be rich in phosphorus and used for land application	Farfel et al., 2005; Donatello & Cheeseman, 2013