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Article **Geochemical Classification of Global Mine Water Drainage**

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Abstract: This paper evaluates the geochemical distribution and classification of global Acid Mine Drainage (AMD) sources. The geochemical compositions of AMD from 72 mine water sites in 18 countries across 6 continents were referenced from literature. The secondary data were analysed for statistical distribution and mine water classification against the Hill (1968) framework. The research found that the global mine water displayed geochemical concentrations within 2%, 11%, 5%, 9% and 8% of the aluminium, sulphate, acidity, total iron and zinc distribution ranges, respectively, at the 75th percentile. The study also found that 46%, 11.1% and 2.7% of mine water sites met the criteria for Class I, Class II and Class III of the Hill (1968) framework, respectively, while the remaining 40% of sites were omitted by the framework's geochemical specifications. The results were used to optimise the Hill (1968) framework. The revised framework was proposed for effective AMD geochemical classification, regulation and remediation.

Keywords: Acid Mine Drainage (AMD); geochemical classification; mine water geochemistry; mine water characteristics

1. Introduction

The formation of Acid Mine Drainage (AMD) water streams is a naturally occurring phenomena and occurs in suitable environments where oxygenated water comes into contact with sulphide minerals in the presence of aerobic microorganisms [\[1–](#page-11-0)[3\]](#page-11-1). Large-scale commercial mining operations enabled by technological advancements and the growing economic need for mineral resources has resulted in significant increases in the volumes and toxicity of AMD generated globally [\[4](#page-11-2)[,5\]](#page-11-3). This is a result of the fragmentation of rocks during mining operations leading to increased surface area of rock faces with an abundance of sulphide minerals [\[4](#page-11-2)[,6\]](#page-11-4). AMD streams are typically characterised by low pH and high concentrations of heavy metals and sulphate [\[7](#page-11-5)[,8\]](#page-11-6). The geochemical processes leading to the formation of AMD can be summarised into four interdependent reactions as defined in Equations (1) – (4) [\[9,](#page-11-7)[10\]](#page-11-8).

$$
2FeS2(s) + 2H2O + 7O2(aq) = 2Fe2+ + 4SO42- + 4H+
$$
 (1)

$$
4Fe^{2+} + 4H^{+} + O_2 = 4Fe^{3+} + 2H_2O
$$
 (2)

$$
Fe^{3+} + 3H_20 = Fe(OH)_3(s) + 3H^+
$$
\n(3)

$$
FeS_2(s) + 14Fe^{3+} + 8H_2O = 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \tag{4}
$$

The decant of AMD from mine tailings, of abandoned mines in particular, poses serious environmental hazards including the contamination of rivers, destroyal of aquatic life, bioaccumulation

of toxic metals by organisms and plants, impairment to biodiversity and damaging natural habitats [\[2,](#page-11-9)[11](#page-11-10)[–13\]](#page-11-11). The release of AMD and the associated pollution poses long-term environmental hazard due to sulphide mineral rich mine sites being able to continue generating AMD centuries after commercial mining operations have ceased $[6,14]$ $[6,14]$. This long-term effect coupled with the severity of pollution has made AMD one of the greatest environmental challenges associated with the mining industry globally [\[5,](#page-11-3)[15,](#page-11-13)[16\]](#page-11-14). Prevention of AMD formation is the most ideal solution and may be achieved using techniques such as coating of the exposed mine rock surface and creating oxygen barriers to prevent the dissolution and oxidation of sulphide minerals respectively [\[17,](#page-11-15)[18\]](#page-11-16). However, the majority of AMD is associated with abandoned or closed mining sites where the ground water table has risen inside of mine shafts and pits allowing for exposure to sulphide minerals [\[19–](#page-11-17)[21\]](#page-11-18). water table has risen inside of mine shafts and pits allowing for exposure to sulphide minerals [19–21].
Once AMD is formed in these abandoned and closed mining sites the process is difficult to control and remediation is the most immediate solution [\[17\]](#page-11-15). Figure 1 shows the effects of abandoned mine [A](#page-2-0)MD on the surrounding environment.

Figure 1. Environmental effects of Acid Mine Drainage (AMD) from an abandoned coal mine in **Figure 1.** Environmental effects of Acid Mine Drainage (AMD) from an abandoned coal mine in KwaZulu-Natal, South Africa**.** KwaZulu-Natal, South Africa.

In 2010, a total of 6152 abandoned mines were identified in South Africa [\[22,](#page-11-19)[23\]](#page-11-20). In the United States, it has been estimated that there are more than 500,000 abandoned or closed mines affecting 25,000 km of water streams [\[24](#page-12-0)[,25\]](#page-12-1). Australia has recorded more than 50,000 abandoned mines [\[26\]](#page-12-2) and in the United Kingdom, the number of abandoned mines is estimated to be over 2000 [\[18,](#page-11-16)[27\]](#page-12-3). Many of these and other abandoned mines across the globe continue to generate AMD leading to numerous scholarly works on the remediation of the pollutant to limit environmental degradation $[6,17,28-34]$ $[6,17,28-34]$ $[6,17,28-34]$ $[6,17,28-34]$. Conventional pH correction has been the most widely implemented AMD remediation technique globally due to its operational simplicity, low capital investment costs, and scalability $[6,35]$ $[6,35]$. However, this remediation method, like any other, has its limitations. The selection of suitable remediation methods for mine water sites remains a critical task for local governments and organisations managing AMD streams. The suitability of an AMD remediation technique is influenced by many site-specific factors including the AMD geochemical composition, AMD flow rates, topography and site location amongst others [\[17,](#page-11-15)[34,](#page-12-5)[35\]](#page-12-6).

Over the past three decades, mine water management has become a key regulatory and policy requirement in many mining nations to counter the environmental hazard posed by AMD [\[36–](#page-12-7)[39\]](#page-12-8). Legal frameworks governing mine water differ by jurisdiction and typically include discharge permits, tailing storage and post-mine closure obligations [\[40\]](#page-12-9). To avoid uncertainty amongst stakeholders and adverse effects on the environment, classification frameworks for AMD have been proposed to indicate the contaminant levels and geochemistry of AMD streams [\[38](#page-12-10)[,41\]](#page-12-11). These classification frameworks have proven useful for site rehabilitators and environmental regulators in decision making [\[42\]](#page-12-12). The most prominent classification method is the Global Acid Mine Drainage (GARD) guide [\[36\]](#page-12-7). The GARD

guide is a simple classification framework that uses pH and Total Dissolved Solids (TDS) as primary indicators for AMD categorisation [\[36,](#page-12-7)[43\]](#page-12-13). Other AMD categorisation methods include the Water Accountability Framework (WAF), which uses pH, TDS and coliforms [\[44](#page-12-14)[,45\]](#page-12-15), the Gray Acid Mine Drainage Index (AMDI), which uses pH, SO_4 , Fe, Zn, Al, Cu and Cd [\[46,](#page-13-0)[47\]](#page-13-1) and the framework by Hill (1968), which classifies AMD using acidity, SO_4 , pH, Al and Fe concentrations [\[36](#page-12-7)[,48\]](#page-13-2).

The use of existing mine water classification frameworks has been limited to specific regions due to practical limitations and the high geochemical variability of global mine water [\[45](#page-12-15)[,49](#page-13-3)[,50\]](#page-13-4). Some practical limitations of existing frameworks include the criteria used for classification and the frameworks' applicability to mine waters generated from vastly different rock geology and environmental conditions [\[47](#page-13-1)[,51\]](#page-13-5). The high geochemical variability of global mine water sources requires additional research to improve mine water classification frameworks for global adoption. An ideal global AMD classification framework would enable mine water regulation and site rehabilitation while being simply understood, comprehensively specified and applicable to any mine water system. The framework by Hill (1968) remains one of the most comprehensive yet simple AMD geochemical classification methods available today and was therefore evaluated and optimised for AMD categorisation in this study. Table [1](#page-3-0) shows the Hill (1968) framework.

Table 1. Acid mine drainage classification [\[48\]](#page-13-2) adopted from [\[36\]](#page-12-7).

The Hill (1968) framework was developed based on distance of AMD streams from the original AMD sources [\[36,](#page-12-7)[48\]](#page-13-2). Naturally occurring neutralisation and oxidation processes take place over the distance travelled by a stream, which affects the AMD stream's quality until a completely oxidised and neutralised stream is achieved. In this framework, the greater the class number the better the water quality with the AMD stream at the mine source being presented as Class I and the fully oxidised and neutralised AMD stream presented as Class IV [\[36,](#page-12-7)[48\]](#page-13-2). The Hill framework indicates the pollution potential of an AMD stream and the level of oxidation and/or neutralisation required to achieve Class IV. This framework accounts for metals $Fe²⁺$ and Al, which tend to fully precipitate within the neutral pH range resulting in the neutralised and oxidised Class IV being specified with dissolved Fe²⁺, Al and acidity concentrations of zero. However, the framework does not include an indicator species of the cytotoxic metals present in AMD, which can cause serious ecological damage and require alkaline conditions to effectively precipitate as metal hydroxides. These cytotoxic metals, which include zinc (Zn), nickel (Ni), lead (Pb), arsenic (As) and cadmium (Cd), all tend to precipitate at pH greater than 8.5 [\[52\]](#page-13-6). The presence of these cytotoxic metals in AMD influences the selection of AMD treatment technology and the ecological pollution potential of AMD, therefore understanding their concentration distribution in global AMD sources is essential for AMD categorisation [\[33\]](#page-12-16).

This paper evaluates the geochemical distribution and classification of global AMD sources using secondary data. The Hill (1961) framework was evaluated as a categorisation baseline. Cation Zn^{2+} was added to the analyses as an indicator species for the cytotoxic metals present in AMD. The geochemical compositions of AMD from 72 mine sites from 18 countries across 6 continents were referenced from literature. The dataset was analysed for statistical distribution with the results used to propose improvements to the Hill framework for effective global AMD classification. The resulting

improvements to the framework were proposed to enable effective mine water remediation decision making and legislative governance.

2. Research Methodology

Chemical composition data of AMD from mine sites across the world were gathered from literature to form an indicative global AMD sample dataset. Secondary data were gathered for mine water sites in Africa, Asia, Europe, North America, Oceania, and South America. In each continent, with the exception of Oceania, three major mining countries were selected, and in each country, four AMD sites were randomly selected. In Oceania, due to the limitation of mining nations, two countries (Australia and New Zealand) were added to the dataset with four AMD sites selected in each country. Additionally, four mining sites from major mining nation Russia were added to complete the dataset. The referenced dataset comprised of coal, cupper (Cu), diamond, gold (Au), iron (Fe), lead (Pb), nickel (Ni), pyrite, rare earth minerals (REMs), silver (Ag), tin (Sn), uranium (U) and zinc (Zn) ore mines. The data were comprised of the highest concentrations of acidity, Al, pH, SO_4 , total Fe and Zn recorded in the referenced literature for each site. Table [2](#page-6-0) shows the referenced sites, countries located, mineral ores mined and the literature references. The global AMD dataset's geochemical distribution range was analysed using quartile interval scales and distribution plots. The global AMD dataset was also analysed for classification in the Hill (1968) framework. The analysis results gathered were used to propose an optimised Hill (1968) framework for effective mine water geochemistry classification.

Table 2. Global AMD sites.

Table 2. *Cont.*

3. Results and Discussions

3.1. Geochemical Distribution of the Global AMD Dataset T_{tot} secondment suggests that the sites can be classified with a narrow $\frac{1}{2}$

Figure 2 shows box and whisker plots of the pH, Al, SO₄, acidity and total Fe distribution for the global AMD referenced dataset. The dataset distribution ranges were 0.5 to 7.6 for pH, 350 mg/L to 56,240 mg/L for SO $_4$, 0.6 to 12,240 mg/L for total Fe, 4 to 38,342 mg/L for acidity and 0.01 to 17,689 mg/L for Al. The large distribution ranges indicate the high degree of AMD chemistry variation across the sites and illustrate the complexity of developing classification frameworks for global mine water. It was found that the distribution of Al, SO₄, acidity and total Fe data was heavily skewed towards the bottom end of the total range with the fourth quartile (upper 25th percentile) accounting for more than 80% of the total range. The 75th percentile data distribution for Al, SO_4 , acidity and total Fe were determined to be within the initial 2%, 11%, 5% and 9% of the total range, respectively. This finding suggests that the majority of global mine water sites can be classified within a narrow geochemical range.

Figure 2. Box plots of the geochemical distribution of global AMD sources. **Figure 2.** Box plots of the geochemical distribution of global AMD sources.

The pH distribution was more symmetrical with a median pH of 3.0 and maximum pH of 7.55. of Neutral Mine Drainage (NMD) sites in the referenced global AMD dataset. NMD has been distinguished as an independent mine water effluent due to its unique characteristics [\[36\]](#page-12-7). Mine water from five sites in the referenced dataset may be characterised as NMD. These sites displayed high From the stres in the referenced dataset may be enarteded as failed free stres disputy of rights represented the magnetic stress may be under the stre and between 0 to 800 mg/L for Al. The 75th percentile data distribution for pH was determined to be The observed circum-neutral pH range of between 6 and 7.55 may be attributed to the presence within the initial 49% of the distribution range. Table [3](#page-7-0) summarises the quartile interval scales of the global AMD dataset's geochemical composition.

Distribution	pΗ	Acidity	Aluminium	Sulphate	Total Iron
25th percentile $(Q1)$	$0 - 2.6$	$0 - 215$ mg/L	$0 - 11 \text{ mg/L}$	$0 - 1217$ mg/L	$0\text{--}40 \text{ mg/L}$
50th percentile (Q2)	$2.6 - 3.1$	215–712 mg/L	11–56 mg/L	1217–2444 mg/L	40–209 mg/L
75th percentile (Q3)	$3.1 - 4.0$	712–1788 mg/L	56–343 mg/L	$2444 - 6081$ mg/L	209–988 mg/L

Table 3. pH, Acidity, Al, total Fe and SO₄ global distribution summary.

Figure [3](#page-7-1) shows a box and whisker plot of the concentration distribution of Zn from the referenced dataset. Figure [4](#page-7-2) shows a distribution plot of pH vs. zinc for the referenced dataset. The concentration of Zn at the first quartile exceeded the agricultural irrigation and safe permissible discharge limit for industrial effluents of 1 mg/L, respectively [\[41,](#page-12-11)[125\]](#page-16-8). In total, 95% of the mine water data exceeded the safe environmental discharge limits of Zn. The total data distribution range for Zn was between 0 to 1912 mg/L. The dataset distribution of Zn was also found to be heavily skewed towards the bottom end of the range. The 75th percentile data distribution for Zn was determined to be within the initial 8% of the total range. Table [4](#page-8-0) summarises the distribution range of the Zn dataset by quartiles. The sample frequency was greatest between the pH range of 2.0 and 4.0, which accounted for 78% of total site data. The highest concentrations of Zn were found between pH 3.0 and 4.0.

Figure 3. Box plot of Zn concentration distribution.

Figure 4. Data distribution of Zn vs. pH. **Figure 4.** Data distribution of Zn vs. pH.

Distribution	Zinc		
25th percentile (Q1)	$0-5$ mg/L		
50th percentile (Q2)	$5-25$ mg/L		
75th percentile (Q3)	25-152 mg/L		

Table 4. Zn global distribution summary.

3.2. Classification of the Global AMD Dataset Using the Hill (1968) Framework

Figure [5](#page-8-1) shows the distribution of Al, SO₄, acidity and total Fe concentrations vs. pH with the data displayed against the Hill (1968) framework's classifications. Approximately 75% of the referenced global mine water sites had pH values between 2.0 and 4.5. This pH range is categorised as Class I AMD in the Hill (1968) framework. When evaluating the dataset distribution against the entirety of the Hill (1968) framework it was found that only 46% of the mine sites met the criteria for Class I AMD. The other 29% of sites within the Class I pH range exceeded the specification limits for either Al, SO_4 , acidity or Fe.

Figure 5. Dataset distribution against Hill (1968) framework.

A total of 11.1% of the mine sites met the criteria for Class II and 2.7% of the sites met the criteria for Class III. The referenced sites in Class II and Class III were comprised of the five mine water sites considered to be NMD. The remaining 40% of the referenced mine water sites were outside of the Hill (1968) framework. Of the referenced sites outside of the Hill (1968) framework 10.7% exceed the 20,000 mg/L SO⁴ upper limit for Class I AMD, 23.6% were below the 500 mg/L Fe limit and/or below the 1000 mg/L SO⁴ lower limit for Class I AMD and 4.2% were below the pH 2 limit for Class I AMD. The classification percentage of the global AMD dataset against each geochemical parameter of the Hill (1968) framework is summarised in Table [5.](#page-9-0)

Classification	AMD Geochemistry Distribution				
	Acidity vs. pH	Total Fe vs. pH	Aluminium vs. pH	Sulphate vs. pH	
Class I	53.3%	46%	76.2%	69.5%	
Class II	20%	12%	11.1%	11.1%	
Class III	10%	4%	2.7%	4.2%	
Class IV	$\overline{}$	-	1.6%	$\overline{}$	
Outliers	16.7%	38%	22.2%	15.2%	

Table 5. Global AMD distribution on Hill (1968) framework.

3.3. Evaluation of Results and Framework Optimisation

As mentioned in the introduction, an ideal AMD classification framework would enable mine water regulation and site rehabilitation while being simply understood, comprehensively specified and applicable to any mine water system. The Hill (1968) framework provides a good geochemical categorisation baseline for mine water sources; however, the framework's classification omits some mine water sites as shown in this study. In addition, the cytotoxic metal ions Zn, Pb, As, Co and Ni are not allowed for in the framework, which can limit the ability to conceptualise remediation solutions and AMD ecological hazards when making use of the framework. The framework required optimisation to increase applicability to global mine water sources and improve data comprehension for decision making.

Class I specifications of the Hill (1968) framework limit the upper geochemical concentrations of AMD. These specifications were all well exceeding by some sites in the global AMD dataset, which were highly acidic and highly contaminated. The highly contaminated and highly acidic sites accounted for 11.1% of the total mine water dataset in this research. The mine waters from these sites are highly toxic when considering geochemical composition and they require extensive remediation processes to neutralise, oxidise, precipitate solids, and manage sludge. The researchers propose the inclusion of a Class 0 into the framework, which can categorise these highly acidic and highly contaminated AMD mine waters.

For Class I of the framework, a total of seven sites had acidity concentrations below the 1000 mg/L lower limit and were therefore unclassifiable. The highest omissions on Class I, totalling 23.6%, were as a result of the lower limit of 500 mg/L on Fe^{2+} and the lower limit of 1000 mg/L for SO₄. The researchers propose a reduction of the lower limits for Fe^{2+} , SO_4 and acidity to cater for the vast geochemical variation amongst Class I AMD sites.

The results for Zn showed that the highest concentrations were between pH 3.0 and 4.0 while 75% of the Zn samples had a total concentration below 153 mg/L. The Zn concentration distribution was found to be highly variable across all pH and acidity ranges of the mine water sites. The researchers proposed adding a simple category for Zn^{2+} at the bottom of the framework. The proposed categorisation includes a category for concentrations up to the discharge limit of 1 mg/L represented as low (L) , concentrations up to the dataset median of 25 mg/L represented as medium (M), and concentrations exceeding the dataset median represented as high (H).

Class II and Class III of the Hill (1968) framework were considered effective at categorising the global AMD referenced dataset with all but one site unclassified. Table [6](#page-10-0) shows the proposed optimised Hill framework based on the discussed findings and proposal.

Class	Class Description	Thresholds		
Class 0 ^{**}	Highly concentrated and acidic mine drainage **	$pH = 0.5 - 3$ ** Acidity = $5-45$ g/L **	Total Fe = $1000-12,000$ mg/L **	$SO_4 = 10 - 60$ g/L ** $Al = 1000-18,000$ mg/L **
Class I	Acid mine drainage	$pH = 2.0 - 4.5$ Acidity = $0 - 15$ g/L **	$\text{Fe}^{2+} = 0 - 10,000 \text{ mg/L}$ ** $Fe^{3+} = 0$ mg/L	$SO_4 = 0-20$ g/L ** $Al = 0-2000$ mg/L
Class II	Partially oxidised and/or neutralised	$pH = 3.5 - 6.6$ Acidity = $0-1$ g/L	$Fe^{2+} = 0 - 500$ mg/L $Fe^{3+} = 0 - 1.000$ mg/L	$SO_4 = 500 - 10,000$ mg/L $Al = 0 - 20$ mg/L
Class III	Neutral and not oxidised	$pH = 6.5 - 8.5$ Acidity = 0 mg/L	$\text{Fe}^{2+} = 0 - 500 \text{ mg/L}$ $Fe^{3+} = 0$ mg/L	$SO_4 = 500 - 10,000$ mg/L $Al = 0 - 2000$ mg/L
Class IV	Oxidised and neutralised/alkaline	$pH = 6.5 - 8.5$ Acidity = 0 mg/L	$\text{Fe}^{2+} = 0 \text{ mg/L}$ $Fe^{3+} = 0$ mg/L	$SO_4 = 500 - 10,000$ mg/L $Al = 0 mg/L$
	Category ^{**}	$L = Zinc \leq 1 mg/L^{**}$	$M = Zinc \leq 25$ mg/L **	$H = Zinc > 25$ mg/L **

Table 6. Proposed Optimised Hill Framework.

** indicate the revisions proposed to improve the framework.

4. Conclusions

This paper investigated the geochemical distribution and classification of 72 global mine water sites from 18 countries across 6 continents using quartile interval scales and distribution plots. The Hill (1968) framework was tested for global mine water classification and results were used to propose improvements to the framework. The research found that the global mine water displayed geochemical concentrations within 2%, 11%, 5%, 9% and 8% of the Al, SO_4 , acidity, Fe and Zn total distribution ranges, respectively, at the 75th percentile. The Hill (1968) framework was found to be inefficient at global mine water categorisation with 40% of the referenced mine water sites being omitted from the classification's specifications. To contribute towards effective global mine water classification for regulators and mine water remediators, the following revisions were proposed by the researchers to optimise the Hill (1968) framework:

- 1. The addition of Class 0 to the framework for highly acidic and high concentration baring AMD. The research results found that 11% of the referenced mine water sites exceeded Class I specifications. Class 0 is proposed as an addition to aid policy makers identify these sites as uniquely contaminated mine waters and aid remediators to identify suitable remediation techniques.
- 2. Revisions to Class I to enable the classification of all the geochemical variations of non-neutralised and unoxidised mine water sources. The research results showed that 38.7% of non-neutralised and unoxidised referenced mine waters did not meet the specification of the original Class I of the Hill (1968) framework. The proposed revisions comprised of changes to the lower limits of $Fe²⁺$, acidity and SO₄ concentrations to enable the classification of all mine water sources.
- 3. The addition of an indicator species for cytotoxic cation AMD contaminants Zn, Ni, Pb, As and Cd. Zinc was selected as the indicator species for these contaminants with a categorisation of low, median and high proposed for classification. The proposed addition of Zn will enable regulators and mine water remediators to greater understand the environmental impacts of the AMD source and the mine water remediation requirements.

Author Contributions: D.V.V.K. conceived of the manuscript concept. S.K.T. conducted the research and wrote the manuscript. D.V.V.K. and P.B. supervised the work and contributed to the discussions and the results formulated in this work. All authors have read and agreed to the published version of the manuscript.

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