A Comparative Study of Environmental Impact of Mine Drainage from Enugu Coal Mines, Southeastern Nigeria

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Abstract

This research carried out a comparative study of the impact of acid mine drainage from the abandoned Enugu coal mines, southeastern Nigeria. Surface water samples were collected from Onyeama, Ribadu, Okpara coal mines and Ekuu River draining the Onyeama coal mine. A total of twelve water samples were collected across the Enugu coal mines: four surface water samples from Onyeama coal mine, three samples from Ribadu coal mine, Ekuu River at confluence of Onyeama and Ribadu coal mines and three water samples from Okpara coal mine. The source of sample collection was: springs, mine ponds, streams and river. Physicochemical parameters such as temperature, pH, turbidity, total dissolved solids (TDS), electrical conductivity (EC), total hardness, Biological oxygen demand (BOD), and dissolved oxygen (DO) were measured from the water samples. Anions such as SO\(_4^{2-}\), Cl\(^-\), NO\(_3^-\) and F included major cations such as Ca\(^{2+}\), Na\(^+\), K\(^+\) and Mg\(^{2+}\) were also analysed using standard laboratory procedures such as titration and chromatography. Heavy metals (Fe, Mn and Al) were analysed using atomic absorption spectrometer (AAS). Comparison of the results with World Standards such as WHO, USEPA and SON for potable water shows that levels of pH, turbidity, F, Mg\(^{2+}\) and K\(^+\) exceeded these standards in some locations. The heavy metals concentrations were above the limits for potable water in all the mines. The average abundance of major anions was in this order: SO\(_4^{2-}\) > Cl\(^-\) > NO\(_3^-\) > F. The major cationic average abundance was in the order: Mg\(^{2+}\) > K\(^+\) > Na\(^+\) > Ca\(^{2+}\). Classification of water samples based on pH hardness shows that the water ranged from soft to moderately soft. Based on BOD values, some of the water samples contain high amounts of organic wastes. There was low dissolution of oxygen in the water samples leading to moderately acidic waters. Levels of Fe, Al and Mn were above permissible limits for potable water and show a trend of Fe > Al > Mn. Results also showed that Onyeama and Okpara coal mines have more acid mine drainage conditions than other mine locations. Acid mine drainage causes high acidity, which corrodes metals in borehole installation, dewetgation, and decrease in biodiversity among aquatic biota. Bioreactors can be used in remediying acid mine drainage sites.

1. Introduction

The mining industry is noted to be a major producer of acidic sulphur rich waste waters which drain active and most especially abandoned mines. These mine wastes are often acidic or very acidic in some instances. Such mine wastes constitute an additional environmental hazard due to their elevated levels of heavy metals such as Fe, Al and Mn [1]. The major contaminant in Enugu coal mine sites is shales of the Mamu formation, which contain pyrite (FeS\(_2\)), associated with toxic heavy metals such as As, Cr, Gd, Ni, Pb, Cu, Zn, Al, and Mn [2]. When the pyrites are oxidized, the water forms a soluble hydrous iron sulphate, which shows whitish, reddish or yellowish salt crust on weathered rock surfaces. These heavy metals found in pyrite, because of their toxicity are harmful to human health. These acidic waters which are sulphur rich are generated in both active and abandoned mines on a continuous basis and discharged into existing water sources used for human consumption such as drinking, irrigation and other domestic uses. Acid mine drainage caused by mining activities is the major problem facing the inhabitants in terms of potable water supply in Enugu and environs [3].

Enugu has an estimated population of 915,025. The discovery of coal in Enugu in 1909 led to the development of coal mines in Enugu such as Ogbeke, Onyeama, Okpara, Ribadu, Iva and Asata which constitute what is today referred to as Enugu coal field [4]. Enugu is faced with water supply shortages as a major problem with the inhabitants. Enugu area is also faced with acidic mine drainage pollution from underground coal mines and mine spoil scattered around the mines [5, 6]. Research has proven that acid mine drainage production results from both mining processes and after the land has been reclaimed. Acidity is stored in acid salts if it is not flushed as generated. Salts produced during mining operation can dissolve and contribute to acid mine drainage long after mining has ceased and the area has been reclaimed. When a percentages of 0.11%, 10% and 100% of the total pyrite is oxidized, it takes 1.2 years, 108 years and 1,080 respectively, to flushed out acid generated by pyrite oxidation [7]. Equations showing pyrite oxidation can be found in [3, 6]. Previous works on the hydrochemistry and mine drainage in some of the Enugu coal mines can be found in [2-4, 6, 8]. In most cases, these mine drainage assessment was on individual mine basis, rather than a composite evaluation of mine drainage from the Enugu coal mines for comparison. Since the research on Enugu coals by Ezeigbo and Ezeanyim [8], subsequent works have not brought their investigations in the different mines to determine the level of mine drainage in these mines currently.

The ultimate goal of this paper is to examine the level of mine drainage in the abandoned Enugu coal fields on a comparative basis and determine the environmental impact. To achieve this end the paper will assess the level of acidity caused by relative degree of pyritization in the Enugu coal mines. Evaluate the water quality in the different coal mines with the view of identifying the most contaminated and suggest remedial measures. Also, to compare the extent of mine drainage by previous workers and determine the extent of water degradation in the coal mines. The water that refill these mines dissolves any acidic salts that built up on the pore spaces of exposed walls and ceilings of underground chambers. This initial drainage water is potentially polluting [1].

2. Experimental Methods

2.1 Study Area Description

The study area which consist of Onyeama, Ribadu, Ekuu River and Okpara mines is delimited by latitudes 6° 28’ 18.72’’ to 6° 24’ 94” N and...
longitudes 7° 26’ 47.52” to 7° 27’ 11’’ E (Fig. 1). Onyeama mine is located about 6.5 km northwest of Enugu metropolis. The coal field is surrounded on the northeastern side by Ribadu mine. The mine is a catchment of Ekulu River. Okpara mine is situated in the present day Enugu West. It is located about 6.5 km from Enugu Port Harcourt express way. In terms of relief the highest elevation of about 350 m above sea level was measured at a location in Ekulu River, while the lowest of 202 m was obtained at Onyeama mine site location. The valleys in the study area are characterized by gorges and ravines.

The study area is characterized by a hot and humid climate with an annual temperature in the range 25 °C to 30 °C. The climate is of the typical rainforest type with an average annual precipitation of 1895 mm and total evaporation of around 724 mm/year. The rains are usually heavy during the rainy season, which last from March to October. The heavy rains precipitate flows which affects the mine and the rivers or stream surrounding them. The mines are often flooded during the rainy season [4].

The drainage in the study area is dendritic. Major rivers draining the Enugu coal mines are: Ekulu, Ogbette, Asata and Nyaba Rivers (Fig. 1). Ephemeral streams rise from about 300 m asl as springs and flow through deep V-shaped valleys incised in soil materials and the Ajali sandstone, but more perennial streams rise from the middle level of the escarpment near the base of the Ajali sandstone. The area is properly drained. The streams or rivers some of which are fractured controlled in the flow path give rise to dendritic drainage pattern [8].

![Fig 1 Drainage map of study area showing sample locations](image1)

### 2.2 Geology and Hydrogeology

Enugu coal mines is underlain by the Enugu shale formation which consist of soft to dark gray shale mudstone and intercalation of sandstone and sandy shale. It is underlain by the Mamu formation and Ajali Sandstones. The Ajali Sandstone overlies the Mamu formation which in turn overlies the Enugu shale (Fig. 2). The Mamu formation is the most important geologic formation with respect to coal formation, occurrence and mining [3]. The Mamu formation underlies the Ajali sandstone, but are generally affected by late cretaceous tectonic events leading to faulting, folding and fracturing of the rock materials. The faults and fractures encountered in the mine tunnels continue through the Ajali sandstone to the surface providing a conduit for perennial flow at almost all of the sides of the hill cuts. Enugu shale was deposited under shallow marine environment. It consist of light to dark grey shale, mudstone intercalations of sand stone and sandy shale. The Mamu formation was deposited under paralic conditions [9].

The Mamu formation exhibit cyclic sequence with heterogeneous lithology ranging from bottom to top by shale, or sandy shale/ shaly coal at the top, carbonaceous shale passing down into shale sandstone / shale or sandy shale. Among the five coal seams in Enugu, with the Mamu formation the third seam with a workable thickness of more than 1.5 m is exploited. The Mamu formation is highly fractured about 395 m thick [8].

![Fig 2 Geological map of the study area](image2)

### 2.3 Sample Collection

A total of twelve water samples were collected across Enugu coal field: Onyeama, Ribadu, Okpara mines and Ekulu River. Details of the locations of the water samples are given in Fig. 1. The water samples were collected from the mines at sedimentation ponds, stream channels, stream confluence, springs, tributaries and shallow hand dug well. Field equipment such as global positioning system (GPS), for measuring coordinates and elevations, plastic water sample bottles, conductivity meter, pH meter, spectrophotometers for TDS and turbidity measurement, clinical thermometer and concentrated HNO₃ were used for the field work. The water sample bottles were rinsed with water to be sampled three times before sample collection. Two drops of concentrated HNO₃ were added to each water sample bottle to avoid loss of ions before analysis. Parameters such as temperature, pH, conductivity, turbidity and TDS were measured in situ using standard field kits listed above. The water samples were labeled using marker tape and stored in a cool refrigerator and then transported to the laboratory for analysis the following day.

### 2.4 Analytical Procedures

Measurable physical properties notably, pH, temperature, electrical conductivity, turbidity and total dissolved solids (TDS) were evaluated in the field using standard field kits digital Mv Redox pH meter to measure pH, mercury-in-glass thermometer was used to measure temperature. Electrical conductivity and turbidity were measured with conductivity meter WA 3000 and spectrophotometer Dr 3000 respectively. Chemical parameters such as major ions were analyzed using ion chromatography and titration. Major cations and heavy metals (Fe, Mn and Al) were analyzed using atomic absorption spectrophotometer (AAS) all the chemical analysis were performed at the cross river water board limited, world bank assisted at Calabar Southeastern Nigeria.
### 3. Results and Discussion

#### 3.1 Water Quality

The physicochemical characteristics and concentrations of inorganic ions/salts in the shallow groundwater samples from abandoned Enugu coal mines are presented in Table 1 together with the maximum permissible limits contained in Nigerian standards for drinking water quality [15], world health organization [16] and United States environmental agency [17].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temp. (°C)</td>
<td>23.2</td>
</tr>
<tr>
<td>pH</td>
<td>7.0</td>
</tr>
<tr>
<td>EC (µS/cm)</td>
<td>1.24</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>0.5</td>
</tr>
<tr>
<td>TDS (mg/L)</td>
<td>1100</td>
</tr>
</tbody>
</table>

As presented in Table 1 and 2, almost all the water sampled in Enugu coal mines exhibited a constant temperature range of 24.2 °C except in Okpara mine located at Okpara and Okpara River where the temperature range from 29.8 °C to 28.8 °C. This temperature reflects air temperature since the water surface is directly contacted with solar rays. This air temperature always differ from groundwater temperature because, ground water temperature is due to the heat retained by the aquiferous layers. The water temperature in the study area ranged from 23.4 °C to 28.8 °C with a mean of 25.2 °C and a standard deviation of 1.90 with a median value of 24.3 °C. Temperature affects organisms as well as physical and chemical characteristics of water [18]. The temperature seems to have no substantial impact on water quality. Generally, rules of reactions to form acid mine drainage increases with increasing temperature. Acid mine drainage is formed faster at warmer pyritic material [19].

The pH values of the water samples ranged from 4 to 5.89, which is acidic condition. The mean pH level is 4.98 with a standard deviation of 0.68 and a median of 5.04. Low pH value is evidence of acid mine drainage due to the oxidation of pyrite (FeS) in abundance. This shows that the degree of acidity is more at Ribadu mine next to Okpara mine followed by Onyeama mine. Onyeama and Okpara have established pH values of 2.8 and 2.3 in Onyeama and Okpara mines respectively. Utom, et al., [5] discovered a pH range from 2.6 to 6.7 and a mean of 4.17 in Okpara mine. Temperature and pH are some of the various factors responsible for acid mine generation at reaction sites [19]. Temperature and pH also affects the solubility of heavy metals and hence their mobility and dispersion [20]. Acid mine drainage is a key factor in fish loss. A study of fish distribution in Pennsylvania streams affected by acid mine drainage revealed severe impact on fish at pH 4.5 to 5. Ten species showed tolerance to acid conditions at pH 5.5 and below and 38 species were found living in waters with pH values ranging from 5.6 to 6.4 while 68 species were only at pH levels higher than 6.4. There was complete loss of fish in 90% of the stream in waters with pH of 4.5 and total acidity of 15 mg/L [21]. Jamel, et al., [22] found in Churchill coal mine that pH values ranged from 2.62 to 3.80 and temperature range from 29.8°C to 34°C. Temperature of acid mine waters should not be 50 °C above the receiving water temperature to attain tolerance level by aquatic biota [22].

Electrical conductivity is indicative of heavy metal level of dissolution and desorption [23]. The conductivity of most fresh waters ranged from 10 to 1000 µS/cm, but may exceed 1000 µS/cm in polluted waters or those receiving large quantities of land run off or acid mine drainage runoff [24]. Ezegbo and Ezemayim, [8] reported values of 700 µS/cm and 1550 µS/cm at Onyeama and Okpara mine respectively. Utom, et al., [5] found EC range of 28 to 1043 µS/cm in Okpara mine. In this study, all the locations have EC values below 1000 µS/cm prescribed by Standard Organization of Nigeria [25]. The total dissolved solid (TDS) which gives a good indication of salinity ranged from 150.54 to 283 mg/L. The Standard Organization of Nigeria [25], stipulated an acceptable level of TDS for potable water at 50 mg/L. The mean level of TDS of this study ranged from 68 to 283 mg/L, falls above acceptable limits for potable water. Total dissolved solids in Okpara mine were found to be above acceptable levels for potable water, while TDS levels in Onyeama, Ribadu and Eku River were below acceptable limit of 500 mg/L for potable water. The low TDS values recorded in Onyeama, Ribadu mines and Eku River could be due to low dissolution of sulphides, presence of buffers such as calcium carbonates and bicarbonates or dilution [26]. Ezegbo and Ezemayim, [8] reported TDS values of 330 mg/L and 785 mg/L for Okpara and Okpara mines respectively.

### 3.2 Total Hardness

Hardness is a measure of the occurrence and abundance of divalent cations (Ca²⁺, Mg²⁺). When hardness values are very high in potential acid drainage waters indicates that the water differs from the more common type of hard waters, to the extent that sulphate with no bicarbonate is the dominant anion [5]. The total hardness values recorded in this study are below permissible limits of 500 mg/L for potable water (Table 3). The mean hardness value is 61.29 mg/L and ranged from 25.02 to 96 mg/L. All the coal mines reported in this study recorded low total hardness values. Such waters are highly desirable for direct potable use. Total hardness of water samples of this study ranged from 8 to 621 mg/L in Okpara mine. Ezegbo and Ezemayim, [8] reported average values of 100 mg/L and 100mg/L for Okpara and Okpara coal mines respectively.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hardness (mg/L)</td>
<td>31.48</td>
</tr>
<tr>
<td>Water classification</td>
<td>soft</td>
</tr>
</tbody>
</table>

### 3.3 Biological Oxygen Demand (BOD) and Dissolved Oxygen (DO)

Atmospheric oxygen is prerequisite for direct oxidation of pyrite and for regeneration of Fe²⁺. Thus, if air and oxygenated or Fe²⁺-rich waters can be excluded from pyritic material, pyrite oxidation can be inhibited and little or no acid will be generated [19]. Dissolved oxygen had a mean of 2.28±0.60 and ranged from 1.6 to 3.6 mg/L and a median of 2.1 mg/L. Eku River and the confluence of Ribadu and Onyeama mine had a high dissolved oxygen of 22 mg/L and Onyeama mine had lower dissolved oxygen. Oxygen is a key reactant in forming acid mine drainage and has low solubility in water. 10 mg/L of dissolved oxygen will produce acid with a pH of 3.2 [19]. There is low dissolved oxygen in this study, hence there is moderate acidity due to insufficient pyritization. The permissible level of DO by WHO [16] is 4.6 ppm for potable water. None of the DO values recorded in this study area locations is up to this standard.

The BOD permissible limit is 6.0 ppm, from the samples, most locations recorded levels of BOD above 6 ppm, so we can deduce that most samples contain high amounts of organic wastes, as such are highly polluted with organic matter. Low dissolved oxygen and high BOD are some of the biological parameters that characterized the degradation of water quality [28].
which is the permissible standard by WHO [16] for potable water (Table 1). Concentrations of sulphate in Ribadu mine and Ekulu River were below this permissible level. High concentration of sulphate is mainly due to the presence of iron sulphide in coal and rocks and its reaction with water and oxygen [28]. Sulphate levels of 310 mg/L and 240 mg/L were established by Ezeigbo and Ezeanyim, [8] in Onyeama and Okpara coal mines respectively. Waters samples from acid mine drainage discharges generally has high concentration of acidity, Fe, Mn and Al [29]. Ezemokwe et al., [30] established sulphate range from <20-700 mg/L, and a mean of 226.03 mg/L in Onyeama mine.

Chloride concentrations in the sampled waters from Enugu coal mines ranged from 5 to 150 mg/L with a mean and standard deviation of 31.48±46.52 with a median of 7.03 mg/L. The levels of chloride in all the mines studied were below the permissible level of 250 mg/L of potable water recommended by USEPA. Ezeigbo and Ezeanyim, [8] reported chloride values from 10.42-1.99 mg/L in Onyeama and Okpara mines respectively. Ezemokwe, et al., [31] discovered chloride values ranging from 27.90-160.30 mg/L with a mean of 83.44 mg/L in Onyeama mine. Also, Utom, et al., [5] established chloride mean values of 5.6 and range from 6.65-26.03 mg/L in Okpara mine. All these preceding finding fell within permissible standard for potable water. In this study, Okpara mine recorded the highest chloride values than other mines. Onwuka et al., [32], reported chloride mean value of 230 mg/L in groundwater in Enugu town. High chloride causes salty taste in water.

Nitrate levels in this study recorded a mean and standard deviation of 93.9±24.65 mg/L with a range of 49-92 mg/L and a median of 1.56 mg/L. Okpara coal mine recorded the highest level of nitrate among the mines investigated. Nitrate levels in this mine exceeded permissible level of 50 mg/L for potable water prescribed by SON [25]. High nitrate levels renders the water prone to potential risk, most especially expectant mothers and infants [19, 33, 35]. Nitrate also causes interferences in water supply and public health propagates in infants [35]. The source of nitrate is mostly from nitrate fertilizers [20].

Fluoride concentration in Enugu coal mine recorded a mean of 1.05±1.77 and ranged from 0.04 to 6.59 mg/L and a median of 0.64 mg/L. Fluoride levels were below permissible level of 1.5 mg/L set for potable water by SON [25], except in Onyeama mine where the value of 6.59 mg/L exceeded the level prescribed for potable water. Excess fluoride causes teeth mottling and at concentration above 4 ppm reduces the prevalence of osteoporosis and collapse vertebra [36]. Deficiency of fluoride in human body causes pains and tenderness of bones [36]. Anion levels in water of this study show a trend of SO<3 Cl->NO>3 F-

3.5 Major Cations

With respect to the major dissolved cations, the general trend of the mean values in water samples shows Mg<sup>2+</sup> K<sup>+</sup> Na<sup>+</sup>Ca<sup>2+</sup>. Magnesium content which is the most abundant vary between 10.61-908 mg/L with a mean and standard deviation of 285.2±734.6. The highest magnesium value of 908 mg/L was recorded in a location at Onyeama mine. The least magnesium value of 10.61 mg/L was obtained at Okpara coal mine. Ezeigbo and Ezeanyim, [8] reported Mg values of 185.08 mg/L and 85.12 mg/L in Onyeama and Okpara mines respectively. Utom, et al., [5] reported Mg range of 1.2-102 mg/L and a mean value of 19.5 mg/L in Okpara coal mine. Magnesium values ranging from 1.45-63.8 mg/L were obtained in surface water samples in a coal mine drainage from in Brazil by Campaner, et al., [37]. The source of Mg may be from magnesium carbonates. Next on the line is potassium, a biophilous element with low geochemical mobility [23]. Potassium levels in this study from 0.62-12.7 mg/L with a mean value of 11.02±3.64. Potassium values obtained from Okpara coal mine were above a permissible limit of 12 mg/L recommended by WHO [16] for potable water. Potassium values at Onyeama, Ribadu coal mines and Ekulu river were below this standard. Sodium and potassium are mostly from dissolution of felspar from adjoining basement areas. Calcium and Magnesium are responsible for temporary hardness in water. Sodium levels ranged from 6.95-121.2 mg/L with a mean and standard deviation of 8.9±17.3. Calcium recorded a mean of 6.72±5.09 and ranged from 2.09-20.02 mg/L. Both calcium and sodium levels were low compared to WHO [16] and SON [25] values of 12 mg/L and 75 mg/L recommended for potable water. Both calcium and sodium levels exhibit low level of metal pollution according to Ezemokwe, et al., [31].

3.6 Heavy Metal Contamination

Rock and earth layers above the coal removed during mining commonly contain traces of iron, manganese and aluminum and also contain other heavy metals. Such metals can be toxic to fish and other aquatic organisms when present in highly dissolved concentrations [28]. Iron concentrations in the study ranged from 0.41-11.95 mg/L with a mean and standard deviation of 5.53±3.45. This mean value is above the permissible level of 0.2 mg/L for potable water by WHO [16] standard. The highest level of Fe was obtained at the Onyeama coal mine. Iron concentrations in all the locations were above allowable limit for potable water. Ezeigbo and Ezeanyim, [8] obtained Fe levels of 8.40 mg/L and 25.76 mg/L in Onyeama and Okpara coal mines respectively, which exceed the values obtained in this study. Utom, et al., [5] recorded a mean value of 1.6 mg/L for Fe in Okpara coal mine. Ezemokwe, et al., [30] recorded iron range of 0.35-0.96 mg/L in water samples at Okpara mine. The mean levels of Al (1.08 mg/L) and Mn (1.30±1.00) obtained in this study also exceeded the standards stipulated by World Health Organization (WHO) and USEPA. This is mainly attributed to the mobility of these metals in acid mine waters. This study considered BO, Mg, Na, Ca, K, Al, Mn, Fe, SO, Cl, NO, and F as potentially metal mobilization. Warm temperatures are amenable for acid mine drainage conditions. Temperature and pH are essential for heavy metal mobilization. Warm temperatures are amenable for acid mine generation. Temperatures at Okpara mine are warmer than, Onyeama, Ribadu coal mines and Ekulu River. The degree of acidity is moderate in all the examined mines, but fell outside the range prescribed by WHO for potable water in all the coal mines. The water samples can be described as soft and springs are strong based on the total hardness values obtained in this study. High sulphate levels above world standards for potable water was obtained at Onyeama coal mine. Higher nitrates and chloride concentrations were recorded at Okpara coal mine than other mines suggesting probable sources from nitrogenous fertilizers and coal vitrains respectively. Fluoride levels were below world standards for potable waters except in one location at Onyeama mine that is above this standard. The average abundance of major cations is in the trend: S0<Cl<Na>Ca>Fe>

4. Conclusion

Acid mine drainage can be generated from both active and abandoned coal mines. Acid mine drainage is caused by the oxidation of pyrite. Low sulphate content at mine sites is evidence of insufficient pyritization. Acid mine drainage is characterized by acidic to moderately acid condition of acid mine waters. This study considered BOD and mobilization of heavy metals such as Fe, Mn, Al and other heavy metals that characterized acid mine drainage conditions. Temperature and pH are essential for heavy metal mobilization. Warm temperatures are amenable for acid mine generation. Temperatures at Okpara mine are warmer than, Onyeama, Ribadu coal mines and Ekulu River. The degree of acidity is moderate in all the examined mines, but fell outside the range prescribed by WHO for potable water in all the coal mines. The water samples can be described as soft and springs are strong based on the total hardness values obtained in this study. High sulphate levels above world standards for potable water was obtained at Onyeama coal mine. Higher nitrates and chloride concentrations were recorded at Okpara coal mine than other mines suggesting probable sources from nitrogenous fertilizers and coal vitrains respectively. Fluoride levels were below world standards for potable waters except in one location at Onyeama mine that is above this standard. The average abundance of major cations is in the trend: S0<Cl<Na>Ca>Fe>

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References

