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Geochemical Stability of a Sediment-Water System in a Reclaimed Post-Gold Mining Watershed: A PHREEQC-Based Scenario Assessment From North Sulawesi, Indonesia

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Abstract: Post-gold-mining reclamation areas commonly undergo prolonged geochemical adjustment, yet the processes governing long-term metal stability within sediment-water systems remain insufficiently understood. This study investigates the geochemical processes controlling dissolved metal behavior in a reclaimed post-gold-mining watershed in North Sulawesi, Indonesia. Long-term surface-water monitoring data collected between 2017 and 2023 were integrated with a pH-REdox-EQuilibrium C (PHREEQC)-based geochemical scenario assessment to evaluate sediment-water geochemical stability through changes in pH and dissolved metal mobility. River pH remained within a slightly alkaline to near-neutral range throughout the monitoring period. Dissolved concentrations of Cr, Fe, Mn, Pb, and Zn showed only minor temporal variation, whereas the representative PHREEQC scenarios indicated greater geochemical sensitivity for aluminum and arsenic. In particular, the representative PHREEQC scenarios suggested greater arsenic mobility under changing pH conditions, indicating that arsenic may be more responsive than the other evaluated elements to sediment-water geochemical adjustment. The PHREEQC-based geochemical scenario assessment supported the interpretation that pH buffering plays an important role in regulating sediment-water interactions. Collectively, the findings suggest that pH-related geochemical processes contribute to the long-term geochemical stability of reclaimed sediment-water systems, while arsenic may serve as a sensitive indicator of early geochemical change. The integrated approach provides a process-based framework for environmental monitoring and adaptive post-mining management, extending reclamation assessment beyond conventional compliance-based evaluation in tropical mining watersheds.

Keywords: Post-mining reclamation; Sediment-water system; Geochemical stability; PHREEQC-based geochemical scenario assessment; Arsenic mobility; pH dynamics; Environmental monitoring

1 Introduction

Gold mining activities often result in long-term geochemical disturbances that may persist well beyond mine closure. These disturbances include acid mine drainage (AMD), the release of metals and metalloids, and changes in sediment-water interactions. In post-mining river systems, sediments function not only as repositories of contaminants but also as potential secondary sources of metals that can be remobilized by changes in pH and redox conditions [1–4].

pH-REdox-EQuilibrium C (PHREEQC) has been widely applied to support geochemical interpretation of sediment-water systems, including assessments of chemical evolution, mixing processes, and metal behavior in mining-affected environments. Previous studies have shown that secondary minerals, particularly iron and manganese oxides, play an important role in buffering pH and regulating metal mobility through adsorption–desorption and precipitation processes [5, 6]. However, these findings cannot be directly extrapolated to reclaimed tropical post-gold

mining environments, where high rainfall and intense weathering are likely to accelerate geochemical adjustment and influence sediment-water interactions.

Although substantial research has focused on AMD, tailings geochemistry, and metal mobility in mining-affected environments, studies that integrate long-term monitoring records with process-based geochemical assessment remain scarce, particularly in reclaimed tropical post-gold mining watersheds. This limitation is especially important in tropical environments, where high rainfall, intense weathering, and heterogeneous reclamation stages can accelerate geochemical adjustment processes and influence sediment-water interactions differently from those observed in temperate mining systems. Consequently, the long-term trajectory of geochemical recovery in these environments remains poorly constrained. Furthermore, it remains uncertain whether compliance with environmental quality standards reflects long-term geochemical stability or merely a temporary condition within an ongoing adjustment process. Limited evidence also exists regarding which geochemical indicators are most sensitive for detecting early shifts in sediment-water equilibrium, particularly metalloids such as arsenic, whose mobility is strongly influenced by pH-dependent adsorption-desorption reactions on iron and manganese oxide surfaces. These limitations restrict the ability of environmental managers to distinguish between regulatory compliance and long-term geochemical resilience, as well as to identify early indicators of emerging environmental change [7].

To address these gaps, this study integrates a seven-year environmental monitoring dataset (2017–2023) with a PHREEQC-based geochemical scenario assessment to evaluate long-term sediment-water geochemical stability and the associated geochemical adjustment processes within a reclaimed post-gold-mining watershed in North Sulawesi, Indonesia. Rather than relying solely on compliance-oriented assessments, this research focuses on the trajectory of geochemical stability and the processes controlling sediment-water interactions under post-reclamation conditions. It further evaluates the relative sensitivity of dissolved metals and metalloids to changing pH conditions and examines whether arsenic may serve as an early indicator of geochemical imbalance. By combining long-term monitoring with process-based geochemical assessment in a tropical post-mining setting, this study contributes to a more comprehensive framework for evaluating environmental recovery and supporting adaptive post-mining management.

In Indonesia, post-mining reclamation success is commonly evaluated using compliance-based indicators. The findings of this study support the inclusion of process-based geochemical monitoring within long-term environmental management programs. Monitoring changes in pH together with geochemically sensitive elements such as arsenic and aluminum may provide earlier indications of sediment-water geochemical adjustment than reliance on regulatory threshold exceedances alone. Integrating routine environmental monitoring with process-based geochemical assessment may therefore strengthen adaptive post-mining management by supporting the early identification of geochemical change and improving the long-term sustainability of reclaimed mining watersheds.

In this study, we hypothesized that pH-mediated geochemical processes play a major role in controlling sediment-water interactions in the reclaimed watershed and that arsenic may respond more sensitively than the other monitored metals and metalloids to changes in geochemical conditions. This study evaluated long-term surface-water and river sediment quality monitoring data collected from the Batupangah River over seven years (2017–2023), comprising 14 monitoring events, together with a PHREEQC-based geochemical scenario assessment. Soil chemical properties and carbon stock assessments were included as supporting indicators of reclamation development and ecosystem recovery. These variables were not used as primary indicators of sediment-water geochemical stability but to provide complementary information on reclamation progress. Accordingly, the objectives of this study were to evaluate long-term sediment-water geochemical stability, examine the response of dissolved metals and metalloids to changing pH conditions, and assess the implications of these geochemical processes for adaptive post-mining environmental management.

2 Methodology

2.1 Study Area and Sampling Design

This study was conducted in the post-gold mining reclamation area of the Toka Tindung Project, North Sulawesi, Indonesia. The Batupangah River serves as the primary sediment-water system receiving runoff from the reclaimed area (Figure 1). To evaluate long-term sediment-water geochemical stability, the study integrated several complementary datasets, including soil chemistry, long-term surface-water monitoring, river sediment quality, vegetation-based carbon stock assessment, and PHREEQC-based geochemical scenario assessment. Multiple datasets with different temporal coverage and analytical objectives were combined to provide complementary information on post-mining environmental recovery. Surface-water and river sediment quality monitoring was conducted concurrently at the Batupangah River during 14 monitoring events between 2017 and 2023, with approximately two sampling campaigns per year. Both datasets therefore cover the same seven-year monitoring period. The monitoring dates and sampling frequencies have been clarified in Table 1.

Surface-water chemistry data were compiled from a long-term environmental monitoring program conducted at a representative monitoring location within the Batupangah River catchment. The dataset spans from February 2017 to November 2023 and consists of fourteen monitoring campaigns with approximately two sampling campaigns per

year. The measured parameters included physicochemical variables, major anions, nutrients, and dissolved metals, which were used for descriptive evaluation, trend analysis, and PHREEQC-based geochemical scenario assessment.

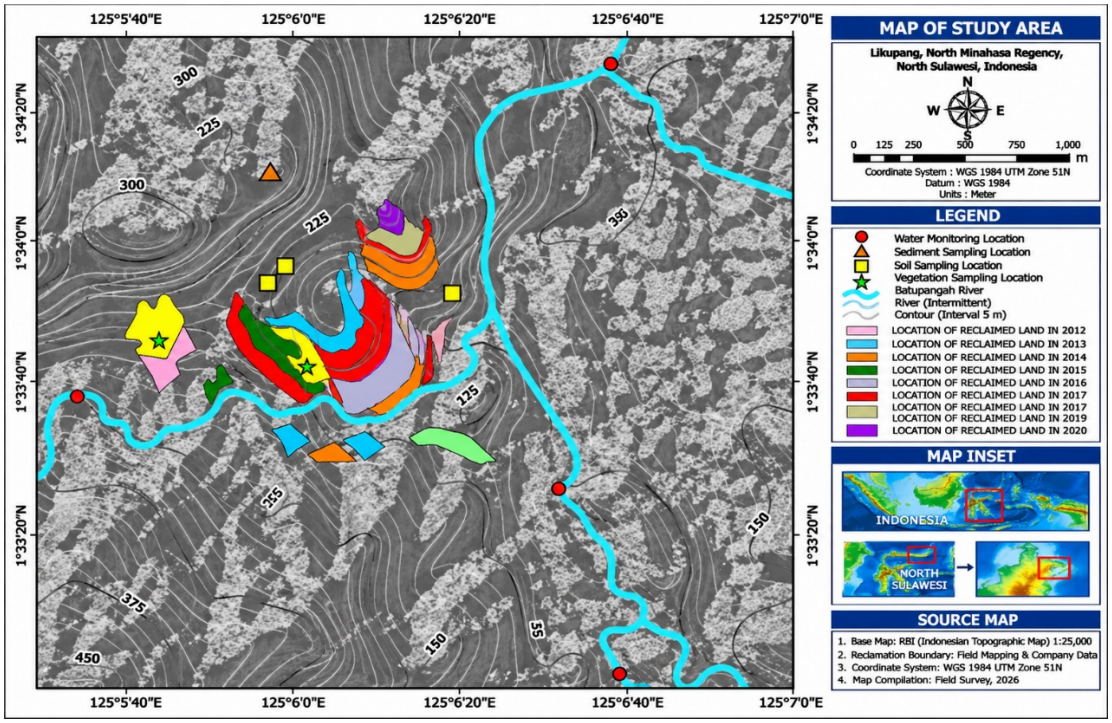

Figure 1. Map of the Toka Tindung Project reclamation area (research location)

Table 1. Summary of datasets used in this study

Dataset	Sampling Unit	Monitoring Period	Number of Observations	Parameters
Surface-water monitoring	Representative monitoring location within the Batupangah River catchment	February 2017–November 2023	14 monitoring events	Temperature, pH, alkalinity, Cl^-, SO_4^{2-}, NH_3, NO_3^-, PO_4^{3-}, Cr, As, Al, B, Ca, Fe, Mg, Mn, Pb, Se, Ag, Zn
Soil chemistry	Reclamation sites (2011–2019 planting years) and reference forest	2021 soil assessment; documented monitoring on 21 April and 21 September 2021	20 soil samples (10 locations × 2 depths)	pH (H_2O), pH (KCl)
River sediment quality	Composite sediment samples collected from the Batupangah River system	February 2017–November 2023	14 monitoring events	Pb, Cd, Cu, Zn, Cr, Hg
Vegetation inventory and carbon stock	Permanent sampling plots within the reclamation area	2021 field inventory	6 plots (20 m × 20 m each; 0.24 ha sampled area); reclamation area used for carbon stock extrapolation: 20.45 ha	Biomass and carbon stock

Source: Compiled from environmental monitoring records, laboratory analyses, and field surveys conducted within the Batupangah River reclamation area.

Soil sampling was conducted across nine reclamation sites representing different revegetation ages (2011–2019) and one reference forest site. Samples were collected at two depth intervals (0–30 cm and 30–60 cm) and analyzed for pH in H_2O and KCl. River sediment quality was evaluated using composite sediment samples collected from the Batupangah River system and analyzed for selected heavy metals. Vegetation inventory data from six permanent 20 m × 20 m plots (total sampled area 0.24 ha) were used to estimate aboveground biomass and carbon stock as supporting indicators of reclamation performance. The estimated carbon stock per hectare was extrapolated to the 20.45 ha reclamation area.

2.2 Laboratory Analyses and Carbon Assessment

Throughout the 2017–2023 monitoring period, surface-water sampling followed the company's standardized environmental monitoring protocol. Sample collection, preservation, laboratory analysis, and quality assurance/ quality control (QA/QC) procedures were applied consistently across all monitoring campaigns to ensure data comparability.

Soil samples were taken at depths of 0–30 cm and 30–60 cm. Soil pH was measured using distilled water (pH H_2O) and 1 M KCl (pH KCl) to separate active and potential acidity, following established soil chemical methods. Surface-water samples were filtered through 0.45 µm membranes, acidified to a pH less than 2, and analyzed for dissolved aluminum, arsenic, chromium, iron, manganese, lead, and zinc using Atomic Absorption Spectroscopy (AAS). In situ pH, temperature, electrical conductivity, and dissolved oxygen were measured using calibrated multiparameter instruments. Composite sediment samples were collected (0–10 cm), dried and homogenized, and analyzed for total metal concentrations (lead, cadmium, copper, zinc, chromium, and mercury) using AAS [5, 7].

Soil organic carbon was determined with UV–Vis spectrophotometry [8]. Aboveground carbon stocks were estimated from vegetation inventories conducted in 20 m × 20 m plots, covering a total sampled area of 0.24 ha. Tree biomass was calculated using established allometric equations and converted to carbon stock values (t/ha).

2.3 Geochemical Modeling and Data Analysis

A PHREEQC-based geochemical scenario assessment was conducted to evaluate how changes in water chemistry may influence sediment-water interactions within the reclaimed watershed. The analysis was intended to support the interpretation of long-term monitoring records and explore potential geochemical adjustment pathways under changing environmental conditions. Accordingly, the approach should be regarded as a process-based geochemical scenario assessment rather than a reactive-transport simulation or predictive forecasting model.

Model inputs were derived from surface-water chemistry data obtained during the long-term environmental monitoring program. Two representative monitoring conditions, corresponding to 2017 and 2023, were selected as end-member chemical states because they represent the earliest and most recent observations available within the monitoring record (Figure 2). Model inputs included pH, alkalinity, major anions, nutrients, and dissolved metal concentrations obtained from the monitoring dataset [6, 9]. The representative 2017 and 2023 end-member solutions were extracted directly from the long-term monitoring dataset, which provides the complete physicochemical composition used as model input. Geochemical calculations were performed using PHREEQC Version 3 (U.S. Geological Survey) with the minteq.v4.dat thermodynamic database. Laboratory-derived water chemistry data were entered using mg/L as the concentration unit. The simulations employed the SOLUTION, MIX, and SELECTED_OUTPUT modules.

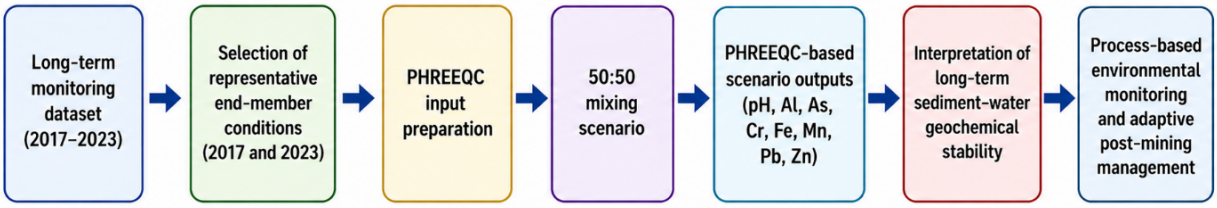

Figure 2. Workflow of the PHREEQC-based geochemical scenario assessment used to integrate long-term monitoring data with representative end-member conditions and a 50:50 mixing scenario

A 50:50 mixing scenario was implemented to represent an intermediate chemical condition between the two end-member states. The purpose of this scenario was not to simulate a physical mixing process within the river system but to evaluate the direction and relative sensitivity of geochemical responses during the transition from earlier to more recent environmental conditions. Model outputs focused on changes in pH and dissolved concentrations of selected metals and metalloids (Al, As, Cr, Fe, Mn, Pb, and Zn). The simulated outputs were interpreted alongside

the monitoring observations to support the interpretation of long-term geochemical evolution within the sediment-water system. The EQUILIBRIUM_PHASES, SURFACE, EXCHANGE, and KINETICS modules were not applied because the objective of this study was to evaluate changes in aqueous geochemistry using long-term monitoring data rather than to simulate mineral equilibrium, surface reactions, ion exchange, or reaction-path evolution. Therefore, the PHREEQC calculations should be regarded as a mixing-based geochemical scenario assessment rather than as a comprehensive reactive geochemical model.

Physicochemical and geochemical data were first summarized using descriptive statistics. To complement the descriptive evaluation, a non-parametric Mann–Kendall trend test was applied to selected water-quality parameters with sufficient uncensored observations to assess monotonic temporal trends during the 2017–2023 monitoring period. Trend analysis was performed to complement the process-based interpretation of the monitoring record rather than to establish causal relationships. For the Mann–Kendall trend analysis, values reported below the analytical detection limit, where present, were replaced with one-half of the corresponding detection limit ($\frac{1}{2}$DL) for parameters with a limited proportion of censored observations. Parameters dominated by censored measurements or affected by changes in analytical reporting limits were excluded from formal trend analysis because reliable statistical inference could not be achieved.

Surface-water quality was evaluated against the Class II river water quality standards specified in Annex VI of Government Regulation No. 22 of 2021 [10]. These standards were applied retrospectively as a common benchmark for the 2017–2023 monitoring dataset and do not imply that the regulation was in force throughout the entire monitoring period. The PHREEQC input files and supporting datasets generated and analysed during this study are available from the corresponding author upon reasonable request. No ethics approval was required because this study did not involve human or animal subjects.

3 Results

3.1 Soil Chemical Characteristics of the Reclamation Area

Soil chemical analyses showed relatively consistent pH values across the reclamation sites despite differences in revegetation age. Soil pH measured with H_2O ranged from 6.2 to 7.4, indicating conditions from slightly acidic to neutral. Conversely, KCl measurements were lower, ranging from 4.5 to 5.6, indicating potential acidity within the soil adsorption complex. The difference between pH H_2O and pH KCl indicates the existence of adsorbed acidity still retained within the soil system.

Table 2. Soil pH characteristics

No	Location/Reclamation Age	Soil Depth (cm)	pH (H_2O)	pH (KCl)	Soil Reaction Class*
1	TTN_2011 (10 years)	0–30	6.8	5.3	Neutral
		30–60	6.8	5.2	Neutral
2	TTN_2012 (9 years)	0–30	6.7	5.0	Neutral
		30–60	6.6	4.9	Neutral
3	TTN_2013 (8 years)	0–30	6.9	4.9	Neutral
		30–60	7.4	5.3	Neutral
4	TTN_2014 (7 years)	0–30	6.7	5.2	Neutral
		30–60	6.8	5.2	Neutral
5	TTN_2015 (6 years)	0–30	6.4	4.7	Slightly acidic
		30–60	6.6	4.6	Neutral
6	TTN_2016 (5 years)	0–30	6.5	5.0	Slightly acidic
		30–60	6.2	4.7	Slightly acidic
7	TTN_2017 (4 years)	0–30	6.8	5.0	Neutral
		30–60	6.7	4.5	Neutral
8	TTN_2018 (3 years)	0–30	7.3	5.6	Neutral
		30–60	6.9	5.0	Neutral
9	TTN_2019 (1–2 years)	0–30	6.7	5.0	Neutral
		30–60	6.6	5.3	Neutral
10	Primary forest (reference)	0–30	6.7	5.3	Neutral
		30–60	6.6	5.3	Neutral

Note: *Soil reaction classes are based on standard soil pH classification: slightly acidic (pH H_2O 5.6–6.5), neutral (pH H_2O 6.6–7.5). Site codes TTN_2011–TTN_2019 indicate the planting years of the revegetation areas. The reported reclamation ages were obtained from the original company-provided soil assessment records.

Similar pH values were observed at both sampling depths (0–30 cm and 30–60 cm), suggesting relatively uniform vertical soil conditions. The similarity in pH between the two sampling depths indicates relatively uniform soil chemical conditions throughout the sampled profile. Based on soil reaction classification, most locations fall

within the slightly acidic to neutral category. This is reflected in the tendency for slightly lower pH values in areas with younger revegetation ages. The natural forest area used as a reference also had pH values in the neutral range. Overall, the pH conditions are relatively stable, indicating no extreme acidity that could directly mobilize metals. Collectively, these observations indicate relatively stable soil chemical conditions within the reclaimed area (Table 2).

3.2 Surface-Water Physicochemical Characteristics

Surface-water physicochemical characteristics remained generally stable throughout the 2017–2023 monitoring period (Table 3). River water pH values ranged from slightly alkaline to near-neutral, with relatively lower values observed during the most recent monitoring campaigns. Nevertheless, all measured pH values remained within the limits established by Indonesian environmental quality standards.

Concentrations of major anions and nutrients were generally low throughout the monitoring period. However, the maximum total phosphate concentration (0.238 mg/L as P) slightly exceeded the Class II river-water reference criterion of 0.2 mg/L. Likewise, dissolved concentrations of chromium, iron, manganese, lead, and zinc remained relatively low throughout the monitoring period. In contrast, aluminum and arsenic exhibited greater temporal variability than the other dissolved metals and were therefore selected for further evaluation using the PHREEQC-based geochemical scenario assessment.

Table 3. Physicochemical characteristics of surface water and dissolved metals in the Batupangah River (2017–2023)

Parameter	Unit	Minimum	Maximum	Mean	Standard* (mg/L)
pH	–	7.06	8.34	7.85	6.0–9.0
Temperature	°C	23	26.5	25.2	±3 (natural)
Alkalinity	mg/L	54	85	71	–
Sulphate (SO_4^{2-})	mg/L	<5	15	9	300
Chloride (Cl^-)	mg/L	<0.5	3.2	1.7	300
Nitrate (as N)	mg/L	0.018	0.216	0.085	10
Total Phosphate (as P)	mg/L	0.032	0.238	0.098	0.2
Aluminium (Al)	mg/L	0.007	0.302	0.095	n/a
Hexavalent chromium, Cr(VI)	mg/L	<0.005	0.005	<0.005	0.05
Arsenic (As)	mg/L	<0.0005	0.005	0.002	0.05
Iron (Fe)	mg/L	<0.02	0.17	0.06	n/a
Manganese (Mn)	mg/L	<0.001	0.032	0.006	n/a
Lead (Pb)	mg/L	<0.001	0.001	<0.001	0.03
Zinc (Zn)	mg/L	<0.005	0.005	<0.005	0.05

Note: *Class II river-water reference criteria are based on Government Regulation of the Republic of Indonesia No. 22/2021, Annex VI, Section I. Nitrate is reported as N, total phosphate as P, and chromium as Cr(VI), consistent with the laboratory analytical records. A dash (–) indicates that no corresponding Class II river-water criterion is specified for the parameter. The criteria are used as reference benchmarks for comparison with the 2017–2023 monitoring data. n/a = not applicable.

Table 4. Summary of Mann–Kendall trend analysis for selected surface-water parameters during the 2017–2023 monitoring period

Parameter	Statistical Analysis*	Result	Interpretation
pH	Mann–Kendall	$p > 0.05$	No significant monotonic trend
Aluminum (Al)	Mann–Kendall	$p > 0.05$	No significant monotonic trend
Iron (Fe)	Mann–Kendall	$p > 0.05$	No significant monotonic trend
Arsenic (As)	Not analysed	–	Predominantly censored observations and changing reporting limits
Chromium (Cr)	Not analysed	–	Predominantly censored observations
Manganese (Mn)	Not analysed	–	Predominantly censored observations
Lead (Pb)	Not analysed	–	All observations below detection limit
Zinc (Zn)	Not analysed	–	All observations below detection limit

Note: *Formal trend analysis was performed only for parameters with sufficient quantitative observations. Parameters dominated by measurements below analytical detection limits or affected by changing reporting limits were excluded because reliable statistical inference could not be achieved. "–" = not analysed.

To further support the descriptive interpretation, the monitoring data were evaluated using a non-parametric Mann-Kendall trend analysis (Table 4). No statistically significant monotonic trends were detected for pH, aluminum, or iron during the monitoring period ($p > 0.05$). These findings suggest that temporal variability within the monitoring record was greater than any consistent monotonic temporal trend. Formal trend analysis was not applied to arsenic, chromium, manganese, lead, or zinc because their datasets were dominated by measurements below analytical detection limits or were affected by changes in analytical reporting limits. Consequently, interpretations for these elements were based on descriptive monitoring records together with the PHREEQC-based geochemical scenario assessment rather than formal statistical trend analysis.

3.3 Dissolved Heavy Metals and Implications for Sediment-Water Geochemical Stability

The assessment of dissolved heavy metals in the Batupangah River was based on long-term monitoring of physicochemical parameters and dissolved metal concentrations (Table 3). These observations provide the basis for evaluating geochemical stability within the sediment-water system because dissolved metals represent the mobile fraction that responds directly to changes in environmental conditions. During the 2017–2023 monitoring period, dissolved concentrations of Cr(VI), arsenic, lead, and zinc remained within their respective Class II river-water reference criteria (Figure 3). Aluminium, iron, and manganese were evaluated descriptively because the referenced Class II river-water standards do not specify criteria for these metals. The monitoring results generally indicate low dissolved metal concentrations, although the absence of Class II criteria for Al, Fe, and Mn precludes a regulatory compliance assessment for these parameters.

Aluminum and arsenic exhibited greater temporal variability than the other metals, although their concentrations generally remained within the applicable environmental quality standards. Overall, the monitoring results indicate stable dissolved metal conditions during the observation period. Because of their greater temporal variability, aluminum and arsenic were selected for a representative PHREEQC-based geochemical scenario assessment to evaluate their sensitivity to pH changes.

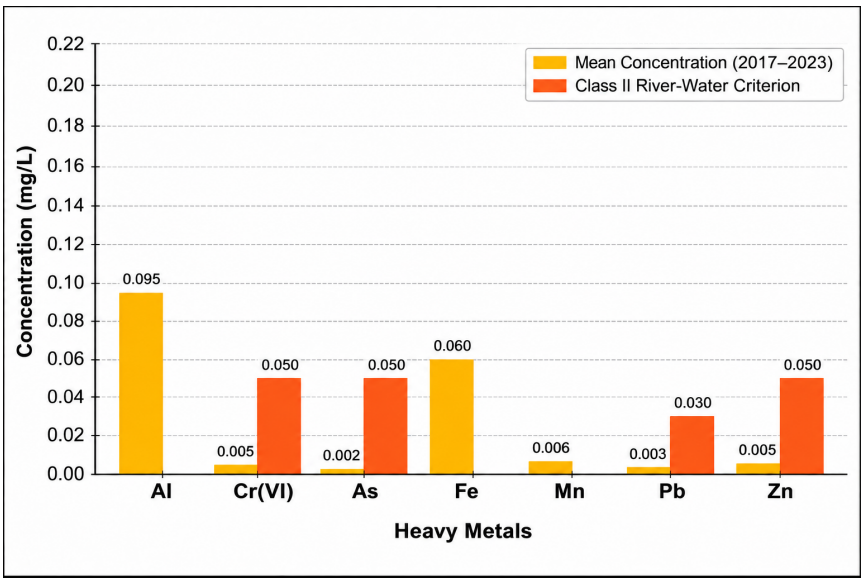

Figure 3. Mean dissolved metal concentrations and Class II river-water reference criteria during 2017–2023

Note: Values reported below analytical detection limits are shown for visualization purposes only and should not be interpreted as quantified mean concentrations.

3.4 Carbon Stock Study in Surface Organic Material

Carbon stock varied among the reclamation plots, ranging from 20.53 to 93.12 Mg C/ha (Table 5), with an arithmetic mean of 47.10 Mg C/ha and a sample standard deviation of 27.15 Mg C/ha (n = 6). Because all sampling plots were of equal size (20 m $\times$ 20 m), an unweighted arithmetic mean was used. Based on the unrounded mean calculated from the reported plot-level values (47.095 Mg C/ha), the indicative carbon stock extrapolated to the 20.45 ha reclamation area was 963.09 Mg C. Tree biomass contributed the largest proportion of the total carbon stock, whereas necromass and litter contributed only a small proportion. The observed variability reflects differences in vegetation development among the reclamation plots. The site-wide extrapolation should be interpreted cautiously because spatial variability and the representativeness of the six plots introduce uncertainty into the estimate. Although

carbon stock was not incorporated into the PHREEQC-based geochemical scenario assessment, it provides supporting information on the ecological recovery of the reclaimed watershed.

Table 5. Surface carbon stock components in the Toka Tindung reclamation areas

No. Plot	Stake, Pole, and Tree (Mg C/ha)	Seedlings and Undergrowth (Mg C/ha)	Subsurface (Mg C/ha)	Necromass (Mg C/ha)	Litter (Mg C/ha)	Soil Organic Carbon (Mg C/ha)	Amount (Mg C/ha)
1	22.82	0.34	6.16	0.00	0.85	9.34	39.5
2	8.97	0.18	2.42	0.00	0.70	8.73	21.00
3	40.73	0.40	11.00	0.00	0.51	4.90	57.55
4	33.91	0.22	9.16	0.00	0.00	7.58	50.87
5	67.58	0.29	18.25	0.20	0.62	6.18	93.12
6	9.57	0.22	2.58	0.00	0.00	8.16	20.53
Sum of plot-level value	183.58	1.65	49.57	0.20	2.68	44.89	282.57
						Average	47.10 Mg C/ha

Note: Values are expressed in Mg C/ha. The mean and sample standard deviation were calculated from six equally sized plots (20 × 20 m). Minor differences between component sums and plot totals may reflect rounding. The indicative carbon stock for the 20.45 ha reclamation area was estimated using the unrounded mean of the reported plot totals (47.095 × 20.45 = 963.09 Mg C).

3.5 PHREEQC-Based Geochemical Scenario Assessment

The PHREEQC-based geochemical scenario assessment used measured pH values of 8.18 (2017) and 7.61 (2023) as inputs representing two monitoring conditions. The lower pH observed in 2023 relative to 2017 represents a difference between the selected observations, rather than evidence of a sustained decline over the seven-year monitoring period. Consistent with this distinction, the Mann–Kendall test showed no statistically significant monotonic trend in pH during 2017–2023 ($p > 0.05$; Table 4). The PHREEQC-calculated pH for the hypothetical 50:50 mixing scenario was 7.97 (Table 6; Figure 4). This value represents a modeled intermediate chemical condition and should not be interpreted as an observed temporal stage or a forecast of future river-water pH.

Table 6. Measured end-member inputs and PHREEQC-calculated outputs for the 50:50 mixing scenario in the Batupangah River system

Parameter	Unit	2017-Measured Input	2023-Measured Input	50:50 Mixture-PHREEQC Calculated Output
pH	–	8.18	7.61	7.97
Aluminium (Al)	mg/L	0.0490	0.0160	0.0320
Arsenic (As)	mg/L	0.0005	0.0050	0.0028
Chromium (Cr)	mg/L	0.0050	0.0050	0.0050
Iron (Fe)	mg/L	0.0200	0.0200	0.0200
Manganese (Mn)	mg/L	0.0050	0.0050	0.0050
Lead (Pb)	mg/L	0.0010	0.0010	0.0010
Zinc (Zn)	mg/L	0.0050	0.0050	0.0050

Note: The 2017 and 2023 columns represent measured water-chemistry inputs, whereas the 50:50 mixing column represents PHREEQC-calculated outputs. The mixing scenario is hypothetical and does not represent a temporal trend or forecast. "–" = dimensionless.

The combined monitoring observations and representative PHREEQC scenarios indicate that pH is the principal variable influencing sediment-water geochemical evolution (Table 7). The PHREEQC-based geochemical scenario assessment further indicates that pH-related geochemical processes play an important role in controlling dissolved metal behavior within the Batupangah River sediment-water system. Accordingly, aluminum and arsenic were identified as the most sensitive indicators for evaluating long-term geochemical adjustment in the reclaimed post-gold mining watershed.

The measured dissolved aluminum concentration was lower in the selected 2023 monitoring condition (0.016 mg/L) than in the 2017 condition (0.049 mg/L). Conversely, the measured dissolved arsenic concentration was higher in 2023 (0.005 mg/L) than in 2017 (0.0005 mg/L). The PHREEQC-calculated 50:50 mixture yielded intermediate

concentrations for aluminum (0.032 mg/L) and arsenic (0.0028 mg/L). These differences describe the selected measured end-member conditions and the hypothetical mixing scenario; they do not establish a temporal trend or demonstrate specific sediment-water reaction mechanisms. Chromium, iron, manganese, lead, and zinc showed little variation among the reported conditions (Table 6; Figure 5).

Table 7. Summary of monitoring observations and pH-controlled geochemical responses from the PHREEQC-based geochemical scenario assessment

Parameter	Monitoring Observations	PHREEQC-Based Geochemical Scenario Assessment	Overall Interpretation
pH	Remained within slightly alkaline to near-neutral conditions throughout the monitoring period	Principal variable controlling dissolved metal behavior under representative scenarios	Primary control on sediment-water geochemical evolution
Aluminum (Al)	Exhibited temporal variability; no corresponding Class II river-water criterion is specified	Greater sensitivity to pH changes under representative scenarios	Sensitive indicator of geochemical adjustment
Arsenic (As)	Exhibited greater temporal variability than the other monitored elements, while remaining below environmental quality standards	Greater sensitivity to pH changes under representative scenarios	Sensitive indicator of geochemical adjustment
Chromium (Cr)	Relatively stable throughout the monitoring period	Little response across representative scenarios	Limited pH sensitivity
Iron (Fe)	Relatively stable throughout the monitoring period	Little response across representative scenarios	Limited pH sensitivity
Manganese (Mn)	Relatively stable throughout the monitoring period	Little response across representative scenarios	Limited pH sensitivity
Lead (Pb)	Relatively stable throughout the monitoring period	Little response across representative scenarios	Limited pH sensitivity
Zinc (Zn)	Relatively stable throughout the monitoring period	Little response across representative scenarios	Limited pH sensitivity

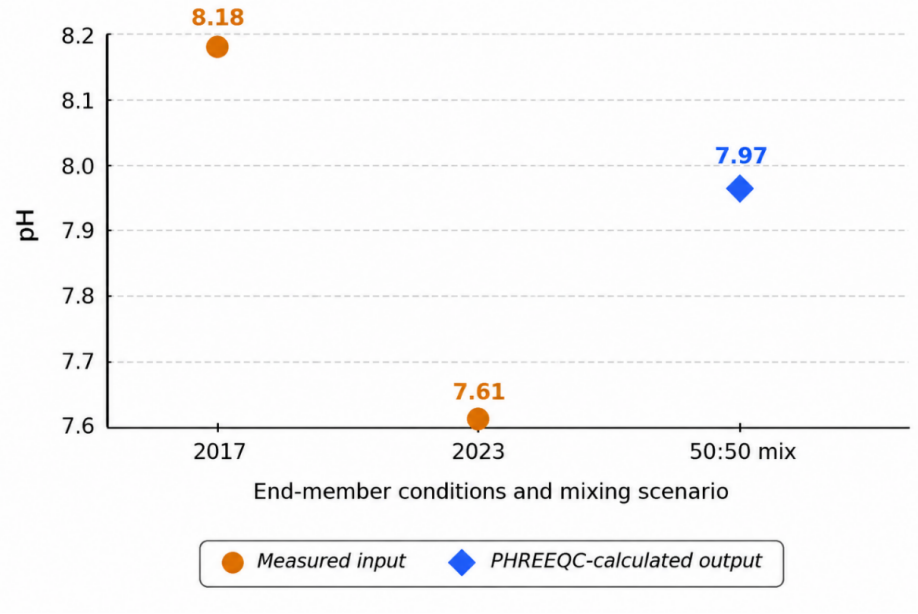

Figure 4. Measured pH end-members and PHREEQC-calculated pH for the 50:50 mixing scenario

Note: Measured pH values are 8.18 (2017) and 7.61 (2023); the modeled pH for the 50:50 mixing scenario is 7.97. These points are discrete end-member and modeled results and do not represent a continuous temporal trend.

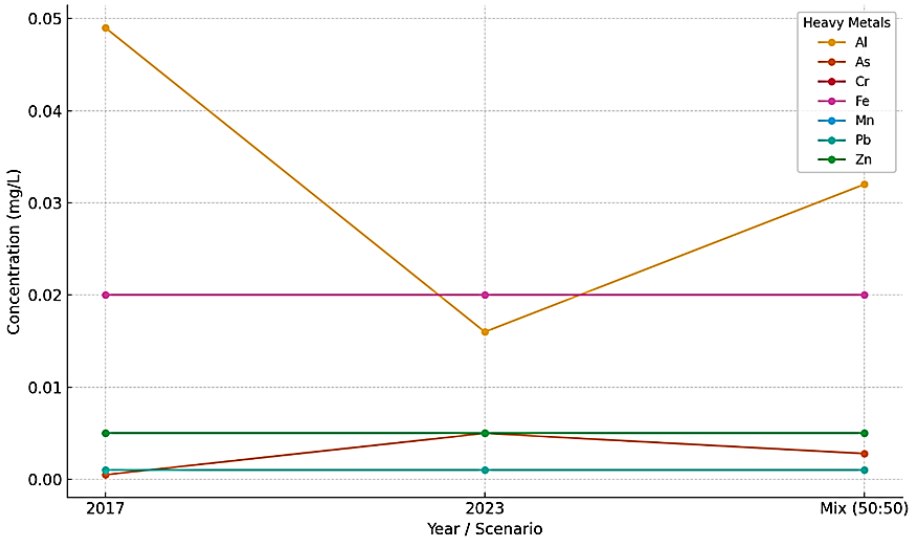

Figure 5. Changes in heavy metal concentrations (Al, As, Cr, Fe, Mn, Pb, Zn) based on conditions in 2017, 2023, and a 50:50 mixing scenario in the Batupangah River sediment-water system

4 Discussion

4.1 Geochemical Control of Sediment-Water Systems and Response of Sensitive Elements

The findings suggest that pH is a major control on the geochemical dynamics of the sediment-water system. In general, river sediments act as both sinks and potential sources of dissolved metals, with metal remobilization occurring when environmental conditions change, particularly pH, redox potential, and organic matter availability [11, 12]. In this context, even relatively small variations in pH may influence adsorption–desorption processes at mineral surfaces and thereby modify the behavior of dissolved metals and metalloids within the aquatic environment.

The PHREEQC-based geochemical scenario assessment showed that most evaluated metals (Cr, Fe, Mn, Pb, and Zn) remained relatively stable across the representative geochemical scenarios (Table 6). Adsorption, precipitation, and interactions with sediment mineral phases are potential processes influencing metal mobility in aquatic environments, although these mechanisms were not directly evaluated in the present modeling framework. Previous studies have highlighted the importance of sediment properties and mineral-surface interactions in controlling arsenic adsorption and desorption [13]. In contrast, the representative PHREEQC scenarios revealed greater sensitivity for aluminum and arsenic (Table 6; Figure 5), indicating that these elements respond more readily to changes in sediment-water geochemical conditions.

The lower dissolved aluminum concentration in the selected 2023 monitoring condition may be consistent with changes in aqueous geochemical conditions. However, mineral precipitation and adsorption onto sediment surfaces were not explicitly simulated; therefore, the mechanisms responsible for this difference cannot be established from the present PHREEQC mixing scenarios. Conversely, the selected 2023 monitoring condition showed a higher dissolved arsenic concentration than the 2017 condition, indicating a difference between the two measured end-member states. Shifts in pH and redox conditions may influence arsenic interactions with mineral surfaces, particularly iron and manganese oxides. Although the underlying mechanism cannot be directly verified using the available dataset, this behavior is consistent with processes that may enhance arsenic mobility under changing sediment-water geochemical conditions [13, 14]. In addition to pH-mediated processes, seasonal hydrological variability, fluctuations in redox conditions, organic-matter dynamics, and upstream sediment disturbance may also contribute to arsenic mobility in reclaimed mining watersheds. These factors were beyond the scope of the present study and warrant further investigation. Collectively, these findings are consistent with previous studies indicating that mining-affected sediments may pose greater ecological risks associated with sensitive metalloids such as arsenic [15].

The geochemical trends observed in the Batupangah River are broadly consistent with findings reported from other reclaimed and mining-affected watersheds in tropical environments. Previous studies in Indonesia, Southeast Asia, and other gold-mining regions have shown that the development of relatively stable near-neutral pH conditions following reclamation is commonly accompanied by low dissolved concentrations of major metals, whereas metalloids such as arsenic may remain more responsive to subtle changes in sediment-water geochemical conditions [4, 7, 13, 16]. Similar patterns have been observed in mining-impacted sediments where arsenic mobility is influenced by sediment-water exchange processes despite overall compliance with environmental quality standards [16]. The relatively stable concentrations of Cr, Fe, Mn, Pb, and Zn observed in this study are therefore consistent with broader post-mining recovery reported in tropical reclaimed watersheds. In contrast, the PHREEQC-

based geochemical scenario assessment suggests that arsenic may remain more responsive to subtle geochemical changes at the sediment-water interface than other dissolved metals. Differences among mining watersheds may arise from variations in rainfall intensity, lithology, reclamation age, and hydrological connectivity, all of which can influence sediment transport, weathering processes, and metal mobility. Consequently, the findings from the Batupangah River should be interpreted within the context of a tropical reclaimed watershed where long-term geochemical recovery is influenced by both reclamation practices and watershed-scale environmental controls.

From a methodological perspective, the PHREEQC-based geochemical scenario assessment provides a process-based framework for evaluating pH dynamics and metal responses in the sediment-water system. The 50:50 mixing scenario was used to represent an intermediate chemical condition between the selected end-member monitoring states and to examine the direction and relative sensitivity of geochemical responses under changing environmental conditions. This approach facilitates the interpretation of observed chemical changes during long-term environmental monitoring. Complementary river-system modeling studies have highlighted the importance of sediment characteristics, metal partitioning, and transport processes in understanding trace-metal behavior in aquatic environments [17]. The statistical trend analysis complements the descriptive interpretation by indicating that no significant monotonic trends were detected for the evaluated parameters during the monitoring period. Consequently, the interpretation of long-term geochemical adjustment is based primarily on representative monitoring conditions together with the PHREEQC-based geochemical scenario assessment rather than on statistically significant temporal trends analysis alone.

Although the geochemical responses observed in this study are influenced by local watershed characteristics, the integrated use of long-term environmental monitoring and PHREEQC-based geochemical scenario assessment offers a practical approach for evaluating sediment-water geochemical stability in reclaimed mining environments. This integrated approach may assist environmental managers in identifying geochemically sensitive elements and in supporting adaptive monitoring strategies during long-term post-mining ecosystem recovery. By complementing routine water-quality monitoring with process-based geochemical assessment, environmental managers may be better equipped to detect subtle changes in sediment-water interactions and to prioritize future monitoring efforts based on geochemical sensitivity rather than solely on regulatory compliance.

4.2 Implications for Post-Mining Management and Environmental Recovery Strategies

From a management perspective, the results of this study support a shift from compliance-based evaluation focused solely on regulatory threshold values toward process-based monitoring. Because pH was identified as the principal variable associated with the observed geochemical trends, it is appropriate to adopt pH as a core operational indicator in monitoring programs. Accordingly, pH may serve as a practical operational indicator for preventive mitigation decision-making and for assessing system stability [18, 19].

The contrasting responses of aluminum and arsenic suggest that these elements, together with pH, may warrant additional attention in future monitoring programs. A targeted monitoring approach could be explored as a complement to routine water-quality monitoring, rather than as a replacement for the existing monitoring of other elements. However, its effectiveness and potential efficiency would require validation through comparisons of monitoring costs, detection performance, and the risk of overlooking relevant changes in water quality. In particular, predominantly censored observations do not provide sufficient evidence to justify reducing the monitoring frequency of other metals [16].

From a risk-management perspective, the greater geochemical sensitivity of arsenic indicated by the representative PHREEQC scenarios should not be interpreted as evidence of current contamination because all measured concentrations remained well below the applicable environmental quality standard (0.05 mg/L). Representative monitoring conditions together with the PHREEQC scenarios indicated dissolved arsenic concentrations ranging from approximately 0.0005 mg/L to 0.005 mg/L. Although the absolute values remain low, this pattern suggests that arsenic is more responsive to changing geochemical conditions than the other monitored elements. Consequently, arsenic may serve as an early-warning indicator of ongoing geochemical adjustment within the sediment-water system. In practical terms, the greater geochemical sensitivity of arsenic warrants closer evaluation of pH trends, sediment conditions, runoff pathways, and reclamation performance, particularly when similar patterns are observed over multiple monitoring cycles. While the present data do not indicate an immediate ecological risk, continued monitoring of arsenic together with pH can support proactive environmental management and help identify emerging geochemical changes before regulatory thresholds are approached.

Variability in revegetation, as reflected by differences in carbon stocks, indicates that biological recovery is spatially uneven. This spatial variability may influence sediment stability and micro-hydrological conditions through erosion and infiltration processes [20]. Although carbon stocks were not directly incorporated into the geochemical modeling, revegetation heterogeneity has the potential to modify sediment-water interface conditions that are relevant to the mobility of sensitive elements [21]. Therefore, post-mining management strategies may integrate revegetation enhancement for erosion control, routine pH monitoring, and additional attention to arsenic and aluminum as

potentially sensitive geochemical indicators, while maintaining the broader monitoring program.

The PHREEQC-based geochemical scenario assessment provides a practical early-warning framework in post-mining environmental management. When changes in pH are observed, or when dissolved arsenic concentrations warrant closer evaluation, even while remaining below regulatory limits, such conditions may serve as early signals of system change. Such information enables managers to prioritize field inspections, evaluate sediment sources and runoff pathways sooner, and enhance erosion and drainage control measures. This approach emphasizes the importance of integrating field monitoring and modeling to assess metal-related risks in mining-affected areas [22, 23].

This study is based on long-term environmental monitoring data and a PHREEQC-based geochemical scenario assessment. Because no mineralogical characterization, sequential extraction, or adsorption-desorption experiments were conducted, the proposed geochemical mechanisms should be regarded as conceptual interpretations rather than directly verified mechanisms. Future studies integrating mineralogical and geochemical characterization would provide a stronger basis for evaluating sediment-water interaction mechanisms in reclaimed tropical mining environments. In addition, the monitoring dataset was evaluated using descriptive statistics and the Mann–Kendall test for selected parameters (pH, Al, and Fe). However, the limited number of monitoring observations (n = 14) constrained the statistical power to detect subtle monotonic trends, while predominantly censored measurements and changes in analytical reporting limits precluded reliable trend analysis for several other metals. Therefore, the absence of statistically significant trends should not be interpreted as evidence of no environmental change. Future studies should expand the monitoring dataset and consider statistical methods appropriate for censored observations, together with complementary trend estimators where applicable.

5 Conclusions

This study indicates that the sediment-water system within the reclaimed post-gold mining watershed has undergone progressive geochemical adjustment during the post-reclamation period. Long-term environmental monitoring conducted between 2017 and 2023 showed that river pH remained within a slightly alkaline to near-neutral range, with lower values observed during the most recent monitoring campaigns. Dissolved concentrations of Cr, Fe, Mn, Pb, and Zn remained relatively stable throughout the monitoring period, whereas the representative PHREEQC scenarios indicated greater geochemical sensitivity for aluminum and arsenic.

By integrating long-term environmental monitoring with a PHREEQC-based geochemical scenario assessment, this study provides a process-based framework for evaluating sediment-water geochemical stability beyond conventional compliance-based assessment. The findings suggest that pH-related geochemical processes play an important role in the long-term evolution of reclaimed mining watersheds and that arsenic may serve as a sensitive indicator of ongoing geochemical adjustment. The integrated approach presented here may support adaptive post-mining environmental management by complementing routine water-quality monitoring and assisting in the identification of geochemically sensitive elements for long-term monitoring in tropical reclaimed watersheds.

Author Contributions

Conceptualization, A.M., I.N., and U.R.I.; methodology, A.T.M. and U.R.I.; software, A.T.M.; validation, A.M., I.N., and U.R.I.; formal analysis, A.M.; investigation, A.T.M.; resources, A.T.M.; data curation, I.N.; writing, original draft preparation, A.T.M. and I.N.; writing, review and editing, A.M. and U.R.I.; visualization, A.T.M.; supervision, A.M. and I.N.; project administration, U.R.I. All authors have read and agreed to the published version of the manuscript.

Data Availability

The data used to support the research findings are available from the corresponding author upon request.

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Conflicts of Interest

The authors declare no conflicts of interest.

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