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The Interaction between Hg and N on Ex-gold Mining Soil

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ABSTRACT

Mercury (Hg) contamination poses a major hazard to ecosystems because it alters plant physiology, biochemistry, and metabolism. The impact is a competition for plant absorption space between Hg and nutrients (such as nitrogen) in the soil and plant system. The goal of this research was to quantitatively examine the interplay between mercury and nitrogen in ex-gold mining soil in Dharmasraya, West Sumatra. This study used a survey method to assess the diversity of ex-gold mining areas owned by each region (area and mining spots) at depths ranging from 0−20 cm to 20−40 cm, with three (Tebing Tinggi at spot 1 and Gunung Medan) to five (Tebing Tinggi at spot 2, Sikabau, and Koto Padang) replicates and a total of 54 samples. The ex-gold mining soil at Dharmasraya, has low fertility levels, including pH (4.03), CEC [7.15 cmol(+) kg^{-1}], OC (0.04% C), and total N (0.09% N), as well as a very high Hg content of 4.18 mg kg^{-1}. The interaction between mercury and nitrogen was non-significant at the 0.01 (2-tailed) level, with r = 0.167 and a linear equation $y = 3.2164x + 3.8849$; R^2 = 0.0276, indicating that mercury does not compete with N nutrients in ex-gold mining soil. However, the release of N by vegetation decomposition (OM-N) through the process of mineralization of C, as evidenced by the positive correlation between N and organic C (r = 0.645 with linear equation $y = 0.5445x - 0.0115$; R^2 = 0.4153), and also the release of Hg, which is absorbed from OM-N, as evidenced by the positive correlation between Hg and OC (r = 0.417** with linear equation $y = 0.0182x - 0.0379$; R^2 = 0.1744).**

Keywords: Dharmasraya, ex-gold mining soil, mercury, nitrogen

INTRODUCTION

Mercury (Hg) toxicity has become a major environmental concern, particularly in illegal gold mining in Dharmasraya, West Sumatra, Indonesia. Hg sources can infiltrate agroecosystems through both natural and anthropogenic mechanisms, with gold mining activities being the primary source of Hg pollution via the amalgamation process (Ayangbenro & Babalola 2017). The amalgamation process is responsible for most of the increase in Hg concentration at previous gold mining sites. Hg residues can contaminate soil, water, air, and plants in the form of Hg^0, Hg^+, and, most notably, Hg^{2+} in soil.

Because of its similarities to nutrients required by plants such as nitrogen (N), accumulated Hg in the soil can be absorbed by the cortical root tissue and transmitted to other regions of the plant via the xylem arteries (Ali *et al*. 2013). This demonstrates that Hg in the soil might compete for nutrient absorption space in plants. The N is one of the most important macronutrients. The total N absorbed by plants in the form of nitrate (NO^{3-}) and ammonium (NH_4^+), both of which are naturally available in the soil, is controlled by the activity of enzymes involved in the conversion of inorganic N to organic N. Direct Hg contamination can interfere with physiological, biochemical, and molecular processes in soil and plant systems, slowing growth and changing cell function, ultimately leading to cell death in plants (Fashola *et al*. 2016).

The Hg can produce drought stress by lowering stomatal conductance, transpiration, and water content in plants due to decreased xylem channels and incorrect cell elongation (Ashraf *et al*. 2017; Riaz *et al*. 2018). Hg-induced toxicity can damage cell membranes and degrade macromolecules (such as proteins and lipids) and cellular organelles in plants. This heavy metal changes the structure of biomolecules and proteins, affecting a variety of biological processes and can cause plants to grow more slowly by inhibiting photosynthetic activity and chlorophyll production. The Hg can also harm plants by suppressing, degrading, and disrupting the stability of enzyme macromolecules and antioxidants (Choppala *et al*. 2014). High Hg toxicity harms ecosystems and can delay N metabolism in the soil. Inhibiting the N cycle in the soil can diminish plant absorption of NO^{3-} and NH_4^+. Thus, it is interesting to investigate the interplay between Hg and total N in historical gold mining soils as a starting point for demonstrating their potential as productive land. The goal of this research

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is to investigate and evaluate the relationship between Hg and total N in ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia.

METHODS

Study Area

This research was carried out at Dharmasraya and was followed by soil analysis at the Soil Science Laboratory, Faculty of Agriculture, Andalas University, from May to July 2022. The research area was 100−131 m above sea level, with an average annual rainfall of ±2,469 mm and a temperature of ±27.5°C (BMKG 2023). This investigation was undertaken at seven ex-gold mining locations (Figure 1A). The sites were chosen based on the assessment of satellite imaging data and land use history obtained through interviews with community leaders and local miners. Each mining site was part of a minor river system that connected to the Batanghari River, with the possibility for water flow and Hg-containing soil deposits to enter the river. Field investigations suggest that ex-gold mining soil is generally sandy, with pebbles on the surface covered in shrubs and grass (Figure 1B).

Soil Samples

Soil sampling in the field was done using a Belgian-type mineral soil drill at each depth, following the purposive random sampling technique. This strategy was based on the variety of gold mining locations in each region. Soil sampling was conducted at depths ranging from 0 to 20 cm and 20 to 40 cm, with three (Tebing Tinggi at point 1 and Gunung Medan) to five (Tebing Tinggi at point 2, Sikabau, and Koto Padang) replicates, for a total of 54 samples.

Soil Analysis

Soil samples taken in the field were then processed in the lab for analysis. Soil analysis included pH measurement using a glass electrode pH meter, organic C using the Walkley and Black method, total N using the Kjeldahl method, CEC using the 1N NH_4OAc extraction method at pH 7, and Hg extraction and quantification with CV-AAS (Eviati & Sulaeman 2012). The data were statistically evaluated using Excel 2016 and SPSS software, specifically descriptive and correlation tests, and displayed as graphs and tables. Pearson's correlation coefficient and 2-tailed significance tests were utilized for bivariate correlations, along with the R^2 coefficient and *p*-value. [** = Significant correlation at the 0.01 and 0.05 (2-tailed) levels]. The maps of the ex-gold mining areas were processed with ArcGIS 10.8.

RESULTS AND DISCUSSION

The ex-gold mining land is composed of Oxisols with low fertility (Prima *et al*. 2023). Table 1 demonstrates that ex-gold mining land has a highly acidic soil response (pH 4.03), extremely low organic C (0.04% C), very low total N (0.09% N), low CEC [6.97 cmol(+)kg^{-1}], and Hg 4.18 mg kg^{-1}, all of which are

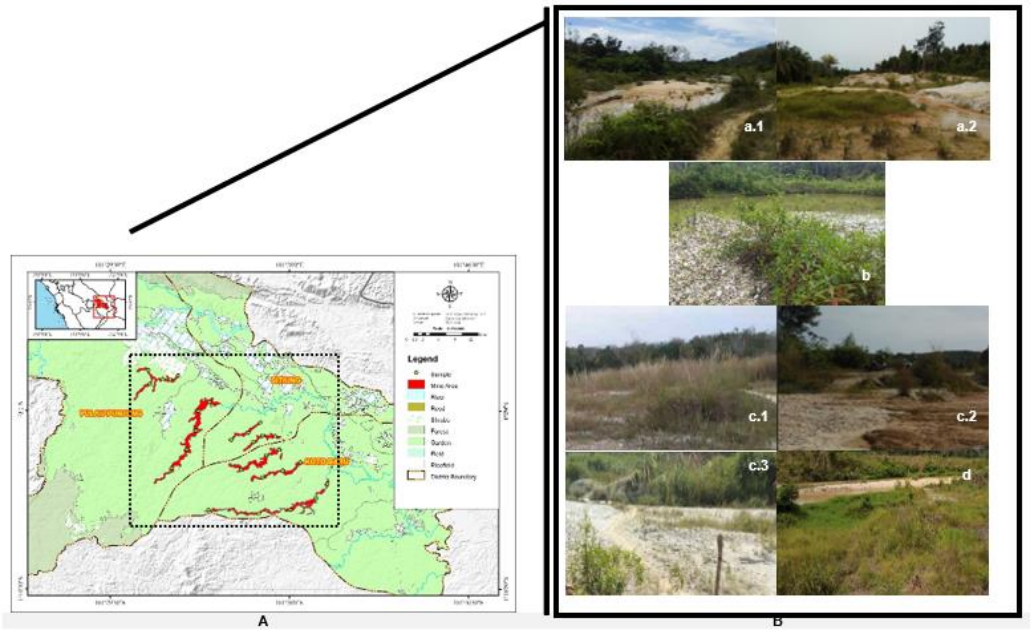

Figure 1 (A) Map of soil sampling locations and (B) view of ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia: Pulau Punjung, Tebing Tinggi at locations 1 (a.1) and 2 (a.2); and Sikabu (b); Sitiung, Gunung Medan at locations 1 (c.1), 2 (c.2), and 3 (c.3); and Koto Baru, Koto Padang (d).

beyond the safe limit. The rise in soil acidity in ex-gold mining soil owing to Hg pollution is caused by the reaction of Hg oxide with H^+ ions in solution (Al-Sulaiti *et al*. 2022). This causes the pH of ex-gold mining soil to become extremely acidic. Grass and shrubs dominate the low organic vegetation (Figure 1B). The carbon input from vegetation was rather minimal. Meanwhile, the mix of organic matter in ex-gold mining soil has a significant impact on total N. The lack of flora on ex-gold mining ground can prevent organic matter accumulation and disrupt the N cycle. The low CEC is caused by soil organic matter loss, Hg pollution of ex-gold mining land, and changes in soil structure dominated by sand with low clay content because of the tailings process. The ex-gold mining soil has degraded due to Hg contamination, indicating that Hg contamination has above the standard quality limit for Hg in soil. The standard quality limit for mercury in soil is 0.3 mg kg^{-1} (Alloway 2012). This figure exceeds the safe limit (0.05 mg kg^{-1}) in agricultural soil based on WHO standards (Chiroma *et al*. 2014), 0.08 mg kg^{-1} based on EU (Horvart *et al*. 2019), and 1 mg kg^{-1} based on the US EPA.

Observations and interpretation of satellite imagery data reveal that most ex-gold mining regions are in plantation sectors, which may pose a risk of Hg contamination to plantation land. Ex-gold mining locations are divided into inactive (Sikabau) and active (Tebing Tinggi, Gunung Medan, and Koto Padang). Ex-gold mining processes involve removing topsoil to a depth of ±50 cm and washing it to separate the ore from the soil or rock, causing changes in the soil's chemical characteristics. The use of mercury in the gold amalgamation process causes Hg accumulation in the soil, as well as poisoning of the environment and ecosystems surrounding gold mines.

The chemical properties of ex-gold mining soil in Dharmasraya differ at each gold mining site (Table 2). The highest Hg content was discovered at a depth of 0–20 cm at location 1, an operating gold mine in Gunung Medan, at 5.32 mg kg^{-1}, and at a depth of 20–40 cm at location 2, an inactive gold mining soil in Tebing Tinggi, at 5.17 mg kg^{-1}. This shows that Hg concentrations in the Dharmasraya mining area surpass national and international limits. If mining area is used effectively for reforestation, Hg waste can build in the soil and seep into plant tissues. The amount and type of Hg chemical compounds, proportion of sand and clay, soil pH, redox potential, and soil organic

Table 1 Descriptive statistics on the chemical properties of ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia

Analysis	Min	Max	Mean	SE	SD	*Criteria
Total N (%)	0.02	0.24	**0.09**	0.008	0.06	very low
pH H_2O (unit)	3.20	4.70	**4.03**	0.05	0.36	very acidic
OC (%)	0.0001	0.19	**0.04**	0.007	0.05	very low
CEC [cmol(+) kg^{-1}]	2.46	22.22	**7.15**	0.74	5.41	low
Hg (mg kg^{-1})	2.61	7.42	**4.18**	0.16	1.15	**above safe limits

Remarks: OC = organic C; CEC = cation exchange capacity; *Based on average values; **The standard limit for Hg in soil is 0.3 mg kg^{-1} (Alloway 2012); These values are above the safe limit (0.05 mg kg^{-1}) in soil for agriculture based on WHO criteria (Chiroma *et al*. 2014) and 0.08 mg kg^{-1} based on the European Union (Horvart *et al*. 2019) and 1 mg kg^{-1} based on the US EPA, SE = standard error; SD = standard deviation, *n* = 54 samples.

Table 2 Chemical characteristics of ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia

Subdistrict	Nagari	Location	Depth	pH H_2O	OC	Total N	CEC	Hg
			cm		%		cmol kg^{-1}	mg kg^{-1}
Pulau Punjung	Tebing Tinggi	1	0–20	4.00	0.026	0.10	4.13	4.90
			20–40	3.90	0.076	0.10	3.92	5.17
		2	0–20	4.14	0.005	0.05	4.98	3.46
			20–40	4.00	0.074	0.11	10.88	3.87
	Sikabau	1	0–20	4.18	0.049	0.14	10.16	3.76
			20–40	4.20	0.058	0.16	14.96	3.69
Sitiung	Gunung Medan	1	0–20	3.80	0.012	0.06	3.40	5.32
			20–40	3.30	0.004	0.03	3.94	4.73
		2	0–20	3.93	0.015	0.08	5.60	3.67
			20–40	3.87	0.029	0.06	5.91	4.36
		3	0–20	3.90	0.051	0.08	6.40	3.79
			20–40	3.90	0.025	0.07	5.93	4.68
Koto Baru	Koto Padang	1	0–20	4.34	0.043	0.08	7.36	4.37
			20–40	4.34	0.042	0.09	5.36	4.01

Remarks: OC = organic C; CEC = cation exchange capacity, and *n* = 54 samples.

matter all have an impact on Hg adsorption in soil (Velásquez Ramírez *et al*. 2021).

At location 1, an active gold mining area in Koto Baru with a pH of 4.34, the ex-gold mining soil pH was highest at depths of 0−20 and 20−40 cm. The maximum soil CEC in the mining area of Koto Padang was 4.34 cmol(+)kg^{-1} (Table 2). However, the CEC of the ex-gold mining soil is relatively low. At the active gold mining site in Tebing Tinggi, the highest CEC of ex-gold mining soil at 20−40 cm depth was 0.076% C (location 1) and 0.074% C (location 2). Meanwhile, in the dormant mining area of Sikabau, the depth of 20−40 cm was 0.058% C. This demonstrates that excavation from mining activities causes soil organic matter to collect in the lower strata. The inactive mining area at position 1 in Sikabau had the greatest total N in ex-gold mining soil, with 0.14% N and 0.16% N at depths of 0−20 cm and 20−40 cm, respectively.

The pH of soil in ex-gold illegal mining sites was 4.25, organic C is 0.08%, total N was 0.02%, and CEC was 2.54 cmol(+) kg^{-1} (Nyaing *et al*. 2021). The chemical parameters of soil following ex-gold illegal mining soil in Kuantan Singingi were pH 3.99, CEC [0.92 cmol(+) kg^{-1}], total N (0.03% N), organic C (0.57% C), and C/N ratio (19.77) (Aryanti & Hera 2019). Organic matter content ranges from very low to very low, with fluctuating data at three locations on ex-gold mining land, but one location, Inuman, has a relatively high organic matter content, with a content of 2.05% C at a depth of 20−40 cm when compared to the other locations. The average soil organic matter (SOM) on ex-gold mining soil ranges from 1.04% C at 0−20 cm to 1.42% C at 20−40 cm (Henrianto *et al*. 2019). The low fertility quality of the soil in forests degraded by ex-gold mining in Bojong Pari, where the nutrient content of N, C, and P was very low, as well as K and CEC, when compared to unmined forest land. Furthermore, nutrients gathered as a kind of wealth from varied flora in the forest have dropped substantially because of land use change-induced deterioration. This demonstrates that traditional gold mining has altered the nutritional status in the Bojong Pari region (Juhaeti & Naiola 1997).

The chemical composition of ex-gold mining soil in Dharmasraya (Table 3) shows that Hg is significantly positively correlated with organic C (r = 0.682** with the linear equation y = 0.0182x − 0.0379; R^2 = 0.1744) (Figure 2A). Meanwhile, Hg has no significant correlation with pH (r = 0.016), total N (r = 0.167), or CEC (r = 0.140). However, organic C showed a substantial positive correlation with total N (0.645**) and CEC (r = 0.525**) (Figure 2B). This suggests that the transit and retention of Hg in ex-gold mining soils is frequently linked to organic matter and nitrogen (N-OM). The stoichiometric relationship between Hg, organic C, and total N offers a foundation for determining the causes and ultimate destiny of Hg absorbed in the ecosystem (Wang *et al.* 2016). The relationship between Hg and organic C, and then between organic C and total N, is regulated by C and N fractionation during the mineralization process, as well as N-SOM transformation. This mechanism is also strongly linked to Hg biogeochemical processes, influencing the quantity and dynamics of Hg fluxes and emissions in soil. The interaction of Hg and organic C, as well as its transition in soil via the decomposition of SOM, releases absorbed Hg, which then undergoes Hg reduction, resulting in mass-dependence (MDF) and mass-independent (MIF) isotope fractionation for odd kinetics (Odd-MIF) (Yuan *et al.* 2020). Conversely, there is a considerable link between Hg and N concentrations (Blackwell & Driscoll 2015), which accounts for 80% of Hg fluctuation (Obrist *et al*. 2018). This event suggests a favorable link between N and the Hg biogeochemical cycle in soil. Litter from vegetation found on ex-gold mining soil decomposes and provides nutrients and energy to plants, animals, and bacteria. This will have an impact on soil nitrogen availability due to the process of N mineralization in the soil. Hg interaction in N-SOM has been connected to a reduction in organic N/O and S functional groups (Manceau *et al*. 2018). Hg accumulation in soil is thought to be related to the formation of N/O functional groups during N mineralization. However, the relationship between Hg and total N is not significant (r = 0.167 with the linear equation y = 3.2164x + 3.8849; R^2 = 0.0276) in ex-gold mining soils (Table 2 and Fig. 2A). However, as illustrated in Figure 3, Hg pollution is also likely to have an impact on the N cycle in ex-gold mining soils (Stein & Klotz 2016).

High and low N concentrations can alter the release and immobilization indices of N in the soil because N

Table 3 Pearson correlation matrix of soil chemical properties in ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia

Pearson correlation	Total N	pH H_2O	OC	CEC	Hg
Total N (%)	1	0.175	**0.645****	**0.682****	0.167
pH H_2O (unit)		1	0.124	0.174	0.016
OC (%)			1	**0.525****	**0.417****
CEC [cmol(+) kg^{-1}]				1	0.140
Hg (mg kg^{-1})					1

Remarks: OC = organic C; CEC = cation exchange capacity; ** = Significant correlation at the 0.01 level (2-tailed), and n = 54 samples.

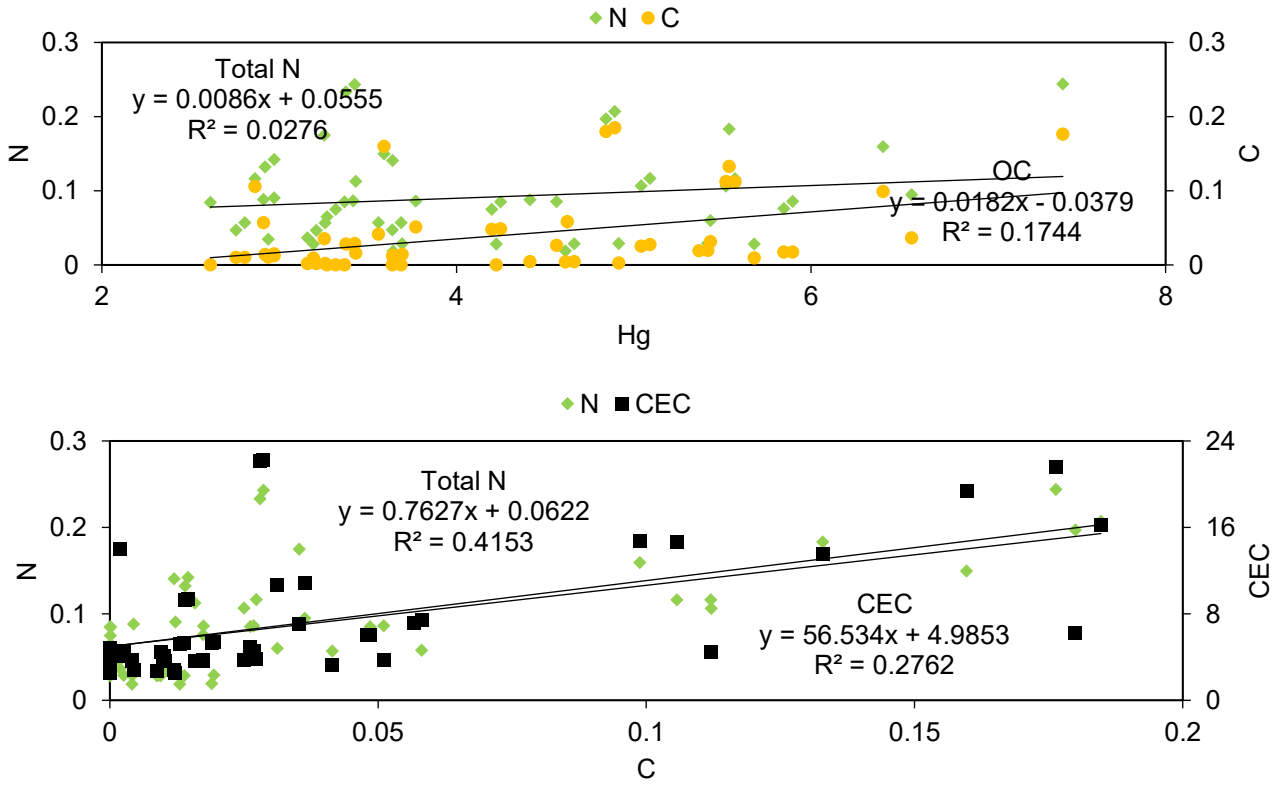

Figure 2 Correlation between Hg with total N and organic C (A) and organic C and total N and KTK (B) in ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia.

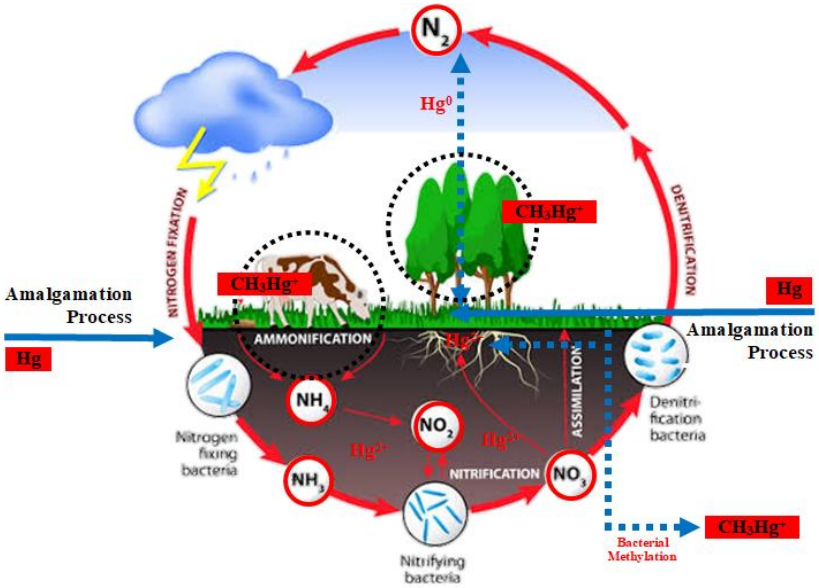

Figure 3 Modification of the N cycle in Hg-contaminated ex-gold mining land (Stein & Klotz 2016).

release occurs when there is enough N supply for breakdown and immobilization of N from the environment, which often occurs when N levels are low (Lin *et al*. 2023). The low total N of ex-gold mining soil in Dharmasraya (Tables 1 and 2) suggests that the relationship between N and Hg is minimal. The release of nitrogen from vegetation litter on ex-gold mining land, as well as nitrogen present in soil organic matter (N-SOM), is linked to C mineralization via functional groups such as N/O. A decrease in S and organic N content in soil organic matter will alter the release of Hg absorbed from N-SOM. The significant correlation

between total N and organic C (r = 0.645**) suggests that C loss during decomposition in ex-gold mine soils will indirectly release N and Hg (Hg^{+} and Hg^{2+}). The significant association between Hg and organic C in ex-gold mining soil will influence the collinearity between organic C and total N. Climate change-induced changes in nitrogen deposition can have an impact on the Hg cycle in ecosystems in ex-gold mining sites. Global changes impact the Hg cycle by influencing physical, biogeochemical, and ecological aspects (Sonke *et al*. 2023). Global changes, such as biome shifts and deforestation, will influence Hg cycles and exposure. Industrialization has resulted in considerable increases in anthropogenic nitrogen compound emissions and deposition in ecosystems. Furthermore, increasing nitrogen deposition in ex-gold mining sites reduces soil abundance and composition, resulting in reduced respiration and increased carbon sequestration in the soil (Maaroufi *et al*. 2015; Zhang *et al*. 2018). Increased carbon sequestration due to greater N deposition is anticipated to alter Hg uptake in soil (Lu *et al*. 2021).

CONCLUSION

Ex-gold mining soil in Dharmasraya, West Sumatra, Indonesia has low fertility levels, including pH (4.03), CEC (7.15 cmol(+) kg^{-1}), organic C (0.04% C), and total N (0.09% N), as well as a very high Hg of 4.18 mg kg^{-1}. The interaction between Hg and total N is not significant at the 0.01 level (2-tailed), with r = 0.167 and a linear equation of $y = 3.2164x + 3.8849$; $R^2 = 0.0276$, indicating that Hg does not compete with nutrient N in ex-gold mining land. Organic matter decomposition (N-SOM) through C mineralization releases N (r = 0.645** with the linear equation $y = 0.5445x - 0.0115$; $R^2 = 0.4153$), and Hg (r = 0.417** with the linear equation $y = 0.0182x - 0.0379$; $R^2 = 0.1744$).

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