Environmental Research 245 (2024) 118073

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Spatio-temporal trends of mercury levels in alluvial gold mining spoils areas monitored between rainy and dry seasons in the Peruvian Amazon

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ARTICLE INFO

Keywords:
Rainforest
ASGM
Kriging
Pollution
Degradation
Madre de Dios

ABSTRACT

Artisanal and small-scale gold mining (ASGM) in the Amazon has degraded tropical forests and escalated mercury (Hg) pollution, affecting biodiversity, ecological processes and rural livelihoods. In the Peruvian Amazon, ASGM annually releases some 181 tons of Hg into the environment. Despite some recent advances in understanding the spatial distribution of Hg within gold mine spoils and the surrounding landscape, temporal dynamics in Hg movement are not well understood. We aimed to reveal spatio-temporal trends of soil Hg in areas degraded by ASGM.,. We analyzed soil and sediment samples during the dry and rainy seasons across 14 ha of potentially contaminated sites and natural forests, in the vicinities of the Native community of San Jacinto in Madre de Dios, Peru. Soil Hg levels of areas impacted by ASGM (0.02 ± 0.02 mg kg^{-1}) were generally below soil environmental quality standards (6.60 mg kg^{-1}). However, they showed high variability, mainly explained by the type of natural cover vegetation, soil organic matter (SOM), clay and sand particles. Temporal trends in Hg levels in soils between seasons differed between landscape units distinguished in the mine spoils. During the rainy season, Hg levels decreased up to 45.5% in uncovered soils, while in artificial pond sediments Hg increased by up to 961%. During the dry season, uncovered degraded soils were more prone to lose Hg than sites covered by vegetation, mainly due to higher soil temperatures and concomitantly increasing volatilization. Soils from natural forests and degraded soil covered by regenerating vegetation showed a high capacity to retain Hg mainly due to the higher plant biomass, higher SOM, and increasing concentrations of clay particles. Disturbingly, our

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https://doi.org/10.1016/j.envres.2023.118073
Received 28 September 2023; Received in revised form 23 December 2023; Accepted 27 December 2023
Available online 28 December 2023

findings suggest high Hg mobility from gold mine spoil to close by sedimentary materials, mainly in artificial ponds through alluvial deposition and pluvial lixiviation. Thus, further research is needed on monitoring, and remediation of sediments in artificial to design sustainable land use strategies.

1. Introduction

Mercury (Hg) is one of the most dangerous and prioritized environmental contaminants in the world (WHO, 2019). It is characterized by high toxicity, mobility and long-term persistence in the environment and living organisms (Kumar et al., 2023; Meyer et al., 2023). The United Nations Minamata Convention on Mercury was created in 2013 and ratified in 2017, with the purpose of protecting human health, and the environment, and reducing emissions and anthropogenic releases of mercury and its compounds (Joy and Qureshi, 2023).

Artisanal and small-scale gold mining (ASGM) is the principal source of atmospheric Hg pollution (UN Environment, 2019) and a main driver of deforestation, especially in the Peruvian Amazon (Hänggli et al., 2023; Larrea-Gallegos et al., 2023). It is estimated that ASGM releases between 675 and 1000 t of Hg globally into the environment per year, which represents 37.7% of the total Hg (2220 t) emitted annually by anthropogenic sources (UN Environment, 2019). In the department of Madre de Dios located in the Southern Peruvian Amazon, the annual Hg releases has been estimated to amount to 181 t (Arana and Montoya, 2017). The main sources of Hg release to the environment are the discharge of Hg tailings to land and water, and the burning of the gold-Hg amalgam (Esdaile and Chalker, 2018). ASGM is also a major driver of large-scale deforestation (Dethier et al., 2023). In Madre de Dios, one of the most biodiverse tropical regions in the world (Ocañas and Thomsen, 2023). About 40,178 ha of forest has been lost over a period of 19 years (Alarcón et al., 2021), at rates of about 7432 ha per year (Caballero et al., 2018). Sadly, deforestation rates due to mining in Madre de Dios have been on the rise in recent years, at 11,500 ha year^{-1} during 2019–2020 (Engstrand, 2021), and 9210 ha year^{-1} during 2021–2022 (MAAP, 2023).

A wide range of Hg pollution levels associated with ASGM has been registered in the Peruvian Amazon. Riverine sediments near ASGM have been found to contain around 0.95 mg Hg kg^{-1} in the Madre de Dios region (Diringer et al., 2019) while river outlets downriver of ASGM areas showed elevated Hg levels (15−50 ng Hg l$^{-1)}$, potentially leading to bioaccumulation in fish and exposure of communities downstream (Diringer et al., 2019). Some of the fish analyzed had concentrations >0.5 mg Hg kg^{-1}, exceeding the World Health Organization (WHO) standard (Barocas et al., 2023). On the other hand, Hg levels in mine spoils (Roman-Dañobeytia et al., 2021; Soto-Benavente et al., 2020; Velásquez-Ramírez et al., 2020, 2021) have consistently been found below 6.6 mg kg^{-1}, the Environmental Quality Standards for Soil of Peru (ECA) (MINAM, 2017).

Considering these previous studies, understanding the dynamics of Hg in soils and sediments is essential to know the temporal and spatial patterns that control Hg fluxes in ecosystems degraded by gold mining in the Peruvian Amazon and beyond. Here we aimed to unravel spatio-temporal trends in Hg levels in soils and sediments of artificial ponds between rainy and dry over two years of evaluation in ASGM-degraded areas in the Peruvian Amazon.

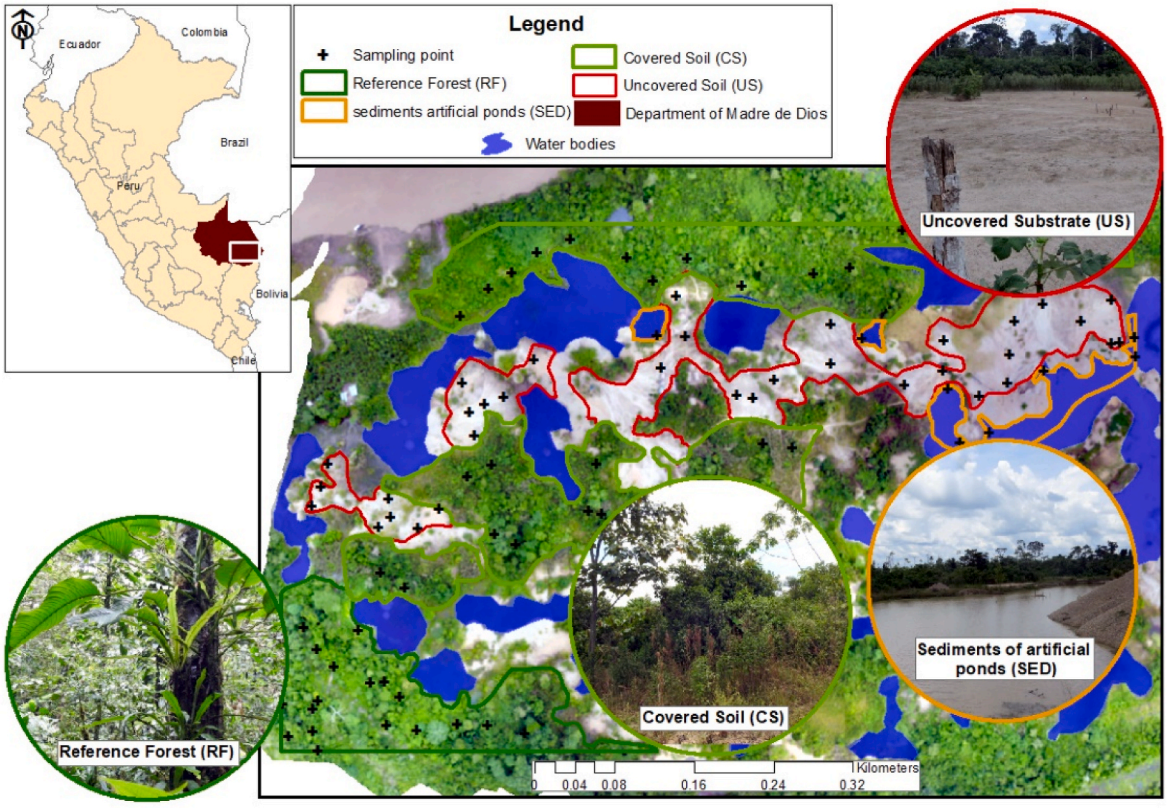

Fig. 1. Study area located in San Jacinto Native Community, in the Peruvian Amazon, Madre de Dios. Sample sites are marked by crosses.

2. Materials and method

2.1. Study area

We studied areas degraded by ASGM in the vicinities of San Jacinto Native Community and in mine spoils of a formal mining concession (453155E, 8606510N; 454017E, 8606515N; 454019E, 8605982N; 453157E, 8605982N) in the Peruvian department Madre de Dios (Fig. 1), considered "the Peruvian capital of biodiversity" (Congreso Constituyente Democrático, 1994). The study region is characterized by a mega thermal climate (tropical climate), with little or no water deficiency, and a saturated environment according to the Thornthwaite Climate Classification (SENAMHI, 2021). It is located on a low terrace at about 220 m. a.s.l. (meters above sea level), in recent alluvial deposits (Quaternary formations). Climax vegetation is composed of humid tropical forests according to the Holdridge Life Zones System classification (Holdridge, 1967). The climate in the study area is seasonal, with a mean annual rainfall of 2156 mm (SENAMHI, 2021) and an average temperature of 26 °C and a maximum of up to 36 °C (SENAMHI, 2021). Soil moisture is high throughout most of the year, classified as a Udic soil moisture regime. The soil temperature regime is classified as hyperthermic with annual mean of soil temperatures above 22 °C (Soil Science Division Staff, 2017). Soils are characterized as Entisols (Velásquez--Ramírez et al., 2021).

2.2. Soil sampling

In 2018 and 2019, a comprehensive sampling strategy was implemented in the ASGM-degraded areas covering both the dry and rainy seasons, and including the main landscape units described by Velásquez-Ramírez et al. (2021) (Table S1). More concretely, we collected representative samples in landscape units characterized by Uncovered Substrate (US, 34 samples) and Covered Soil (CS, 27 samples), as well as in Reference Forest (RF, 18 samples) composed of undisturbed vegetation. Additionally, we sampled sediments from artificial ponds in downstream areas of mine spoils (SED, 8 samples) (Table S1) as well as three sediments sampled in 2020 (dry season) and 2021 (rainy season).

Samples were taken through stratified random sampling across a total area of 14 ha. Each sample consisted in 0.5 kg of surface soil (0–20 cm depth). And we repeated sampling at the same locations during rainy and dry seasons over a two year period. Sampling was carried out in accordance with the Guide for Soil Sampling of Peru (MINAM, 2014), Environmental Quality Standards for Soil of Peru (ECA) (MINAM, 2017) and the Canadian Environmental Quality Guidelines (CEQGs) (CCM, 2007).

2.3. Soil and Hg analysis

Samples were mixed, stored, air dried, and passed through a 2.0 mm sieve, after which we determined particle size distribution (Bouyucos method), soil acidity in water extract 1:1 (weight of water/weight of dry soil) was determined using a multiparametric tabletop meter (C1020, Consort, Turnhout, Belgium), soil organic soil matter (SOM) (Walkley Black Method), and cation exchange capacity (CEC) (Ammonium acetate pH = 7) (ISRIC, 2002). These analyses were carried out on two replicates per sample at the Soil Fertility Analysis Laboratory (Laboratorio de análisis de fertilidad de suelos - LAFES) of the IIAP Research Center in Puerto Maldonado, Madre de Dios.

Hg levels in soils and sediments samples were analyzed according to the Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry method USEPA 7473 (U.S. EPA, 2007), by a Direct Mercury Analyzer DMA-80 (Milestone, Sorisole, BG, Italy). Calibration of the DMA-80 was performed using the 1000 ± 10 μg/mL liquid standard in nitric acid (Inorganic Ventures, Christiansburg, Virginia, USA), recording 11 points on the analytical curve of the equipment. The quality of the analyses was verified both at the beginning and the end of each daily reading using a batch of approximately 20 samples plus their respective repetitions with certified values of reference materials; TILL-1 Geochemical Soil and Till Reference Materials (92 ± 0.00 μg Hg kg^{-1}) and BCR–320 R Channel Sediment (0.85 ± 0.09 mg Hg kg^{-1}). The detection limit was 0.005 ng. Each sample yielded duplicate results, exhibiting less than a 10% error margin. All analyses were conducted at the Mercury and Environmental Chemistry Laboratory (Laboratorio de Mercurio y Quimica Ambiental -LAMQA), and Plant and Soil Fertility Analysis Laboratory (Laboratorio de Análisis de Plantas y Fertilidad de Suelos - LAFES) of the IIAP Research Center in Peru, in Puerto Maldonado, Madre de Dios.

2.4. Interpolation method and data analysis

As data were not homoscedastic and residuals did not present normal distributions, we used non-parametric statistical tests for comparing Hg concentrations across landscape units, such as Kruskal-Wallis ($p < 0.05$) and Wilcoxon as post-hoc test for comparisons of medians and multiple comparisons, respectively. Additionally, Principal Components Analysis (PCA) was used to understand the variability of Hg levels in the soil and sediments (Yotova et al., 2023). We applied a generalized linear model (GLM) with Gamma distribution and an inverse link to explain the most influential variables explaining variation in Hg levels (Faraway, 2016). We constructed several multivariate models and considered that the model with the lowest Akaike Information Criterion (AIC) value was the most appropriate model solution. To determine the quality and significance of the models, we calculated pseudo R^2. Geostatistical methods were applied to understand and analyze the spatial pattern of total Hg (Jianshu, 2019), by applying Empirical Bayesian Kriging (EBK) (Wang et al., 2023). Calculating various semivariogram models during EBK interpolation (Agyeman et al., 2022) has been done in ArcGIS 10.1 (ESRI Inc., USA). Experimental variogram models were generated in the GeoR (v1.7–1) package for R statistical program v 4.3.2 (R Development Core Team, 2023).

3. Results

3.1. Mercury levels in degraded and non-degraded areas

ASGM-degraded areas showed high variability (CV > 25%) of soil Hg levels. Soil Hg levels in RF (Median: 0.08 mg Hg kg^{-1} DM) were significantly higher (Kruskal-Wallis $p < 0.001$) than CS (median: 0.06 mg Hg kg^{-1} DM) and US (median: 0.01 mg Hg kg^{-1} DM) and sediments of artificial ponds (SED) (median: 0.02 mg Hg kg^{-1} DM) in ASGM spoils (Table S2).

Sediments from artificial (SED) ponds and ASGM soils (US) had sandy texture (90.50 ± 7.74% and 89.50 ± 7.66%, respectively), combined with low clay content (6.17 ± 4.65% and 5.70 ± 4.33%, respectively), low SOM (0.42 ± 0.27% and 0.37 ± 0.27%, respectively) and low CEC (5.64 ± 1.08 Cmol (+) kg^{-1} and 5.47 ± 1.12 Cmol (+) kg^{-1}). In contrast, reference forest samples were characterized by clayey soils (26.7 ± 8.72%), with low sand particle content (20.50 ± 16.1%), high SOM (3.14 ± 1.88%), and high CEC (18.4 ± 5.24 Cmol (+) kg^{-1}). Soil pH showed significant differences (Kruskal-Wallis $p < 0.001$) across US (5.71 ± 0.51), CS (5.22 ± 0.71) and pond sediment (5.45 ± 0.41) from ASGM spoils and RF (5.54 ± 0.78) (Table S2).

3.2. Soil Hg levels in rainy and dry season

Soil Hg levels between rainy and dry seasons were not significantly different for pooled CS and US samples (Wilcoxon, $p = 0.53$) nor sediments from artificial ponds (Wilcoxon, $p = 0.93$) in ASGM spoils (Fig. 2). However, Hg levels differed significantly between landscape units in both the rainy (Kruskal-Wallis $p < 0.001$) and dry seasons (Kruskal-Wallis $p < 0.001$) over the two years of evaluation. Hg levels in vegetated soils (CS and RF) were significantly higher than in US and SED

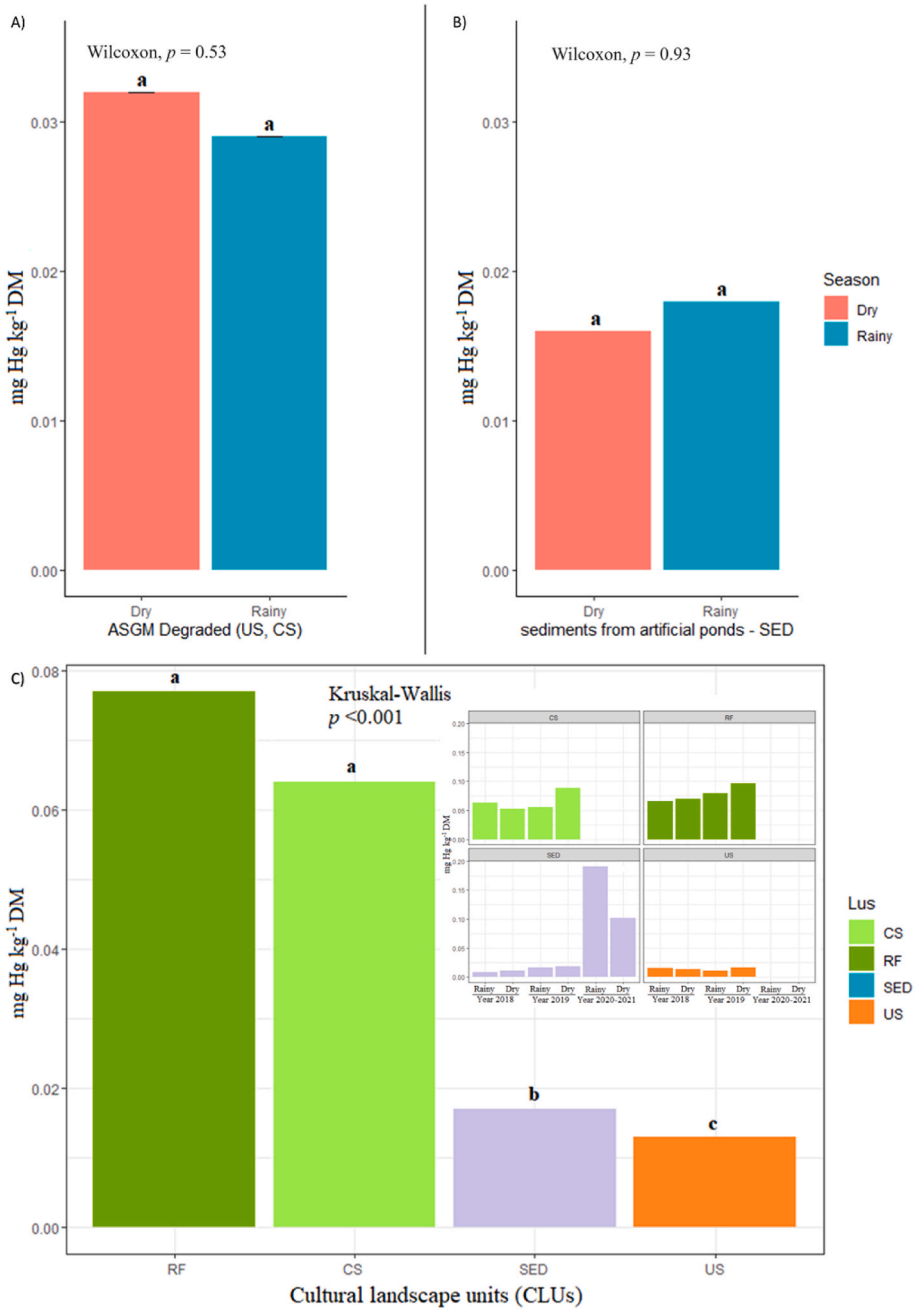

Fig. 2. Bar charts of Hg levels in the rainy and dry seasons (a, b) and Hg levels in degraded areas per each year of evaluation and CLUs (CS, US and RF) including SED, Degraded area (CS and US) and Sediment of artificial ponds (SED).

both during the rainy (Kruskal-Wallis $p < 0.001$) and dry season (Kruskal-Wallis $p < 0.001$) over the two years of evaluation. In addition, there was a tendency of increasing Hg levels over time in CS and RF, while US samples presented only minimal variation in Hg levels in the rainy and dry seasons during the two years of study. On the other hand, Hg levels in the SED show a slight increase in the first two years and presented a dramatic increase in Hg levels in the rainy and dry seasons of the third and fourth sampling years (Fig. 2).

In the degraded areas, CS yielded higher levels of Hg both in the rainy (median: 0.062 mg Hg kg^{-1} DM) and dry seasons (median: 0.073 mg Hg kg^{-1} DM) compared to US soils (rainy season: 0.012 mg Hg kg^{-1} DM; dry season: 0.014 mg Hg kg^{-1} DM). Hg concentrations of CS were quite similar to those recorded for RF soils in both seasons (rainy: 0.073 mg Hg kg^{-1} DM; dry: 0.082 mg Hg kg^{-1} DM). Average Hg levels in SED during the rainy season (median: 0.018 mg Hg kg-1 DM) were slightly higher than during the dry season (median: 0.016 Hg mg kg-1 DM), though the trends were quite variable from year to year (Table 1).

3.3. Correlation analysis

Spearman rank correlation coefficients revealed high ($p < 0.05$) correlation of soil Hg with soil particles (clay, rho = 0.63, $p < 0.05$, and silt, rho = 0.66, $p < 0.05$), SOM (rho = 0.62, $p < 0.05$), CEC (rho = 0.65, $p < 0.05$), and lower with the pH (rho = −0.18, $p < 0.05$) (Fig. S1). The first two axes of a PCA plot (explaining 79.1% of variation) confirm these results, showing a direct relation with CEC and SOM and major negative relation with sand particles and pH. In addition, the highest levels of Hg were found in the RF, CS, and in the SED (Fig. 3).

To explain the variation in Hg levels, we selected the GLM model with the lowest AIC and the highest value of R^2 (0.82) (Table S3), in

Table 1
Mann-Whitney post-hoc comparisons of Hg levels, taken in dry and rainy season. Samples correspond to Uncovered Substrate (US), Covered Soil (CS), Sediment (SED) and Reference Forest (RF).

Parameters		RF		CS		US		SED	
Hg (mg kg^{-1} wd)		Median	Min Max	Median	Min Max	Median	Min Max	Median	Min Max
Year 2018	Rainy season	**a** 0.066	0.039 0.098	**a** 0.063	0.011 0.194	**b** 0.015	0.005 0.077	**b** 0.008	0.005 0.27
	Dry season	**a** 0.07	0.033 0.108	**a** 0.053	0.017 0.195	**b** 0.014	0.005 0.088	**b** 0.011	0.005 11.13
Year 2019	Rainy season	**a** 0.08	0.045 0.126	**a** 0.055	0.01 0.241	**b** 0.011	0.005 0.1	**b** 0.016	0.004 0.23
	Dry season	**a** 0.097	0.05 0.152	**a** 0.089	0.009 0.5	**b** 0.016	0.006 0.11	**b** 0.018	0.005 2.046
Year 2020	Rainy season	–	–	–	–	–	–	**a** 0.191	0.154 0.216
Year 2021	Dry season	–	–	–	–	–	–	**b** 0.102	0.08 0.135
Rainy season		**a** 0.073	– –	**a** 0.062	– –	**b** 0.012	– –	**b** 0.018	– –
Dry season		**a** 0.082	–	**a** 0.073	–	**b** 0.014	–	**b** 0.016	–

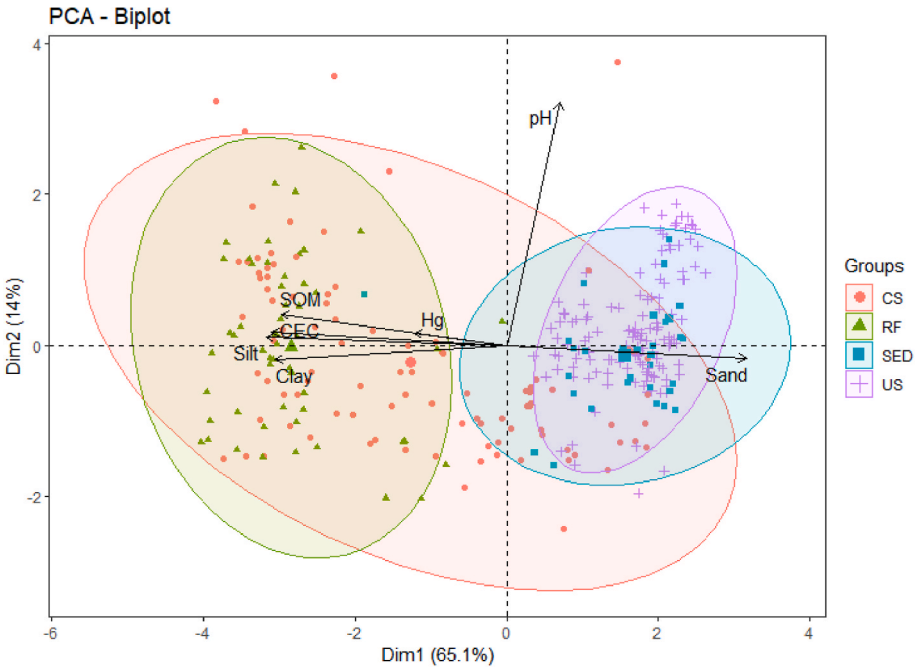

Fig. 3. PCA loading plot showing the relations between Hg levels and the selected soil characteristics. The first two axes explain 79.1% of the variance in data.

which sand content (GLM, $p = 0.006$), SOM ($p = 0.004$), and landscape units ($p = 0.001$) represented the most important variables explaining soil Hg levels (Table 2).

3.4. Maps and geostatistical results

We mapped the soil Hg levels between seasons in two years of evaluation, using the Empirical Bayesian Kriging method to determine the focal points of contamination in the areas degraded by ASGM (Dmytruk and Cherlinka, 2023). The highest levels of Hg (>0.2 mg Hg kg^{-1}) were found in a SED point near the mining camp during the dry season in the two years of evaluation (Fig. 4 A, B). During the rainy season the Hg was more dispersed near water bodies (Fig. 4 C, D).

4. Discussion

4.1. Soil and sediment Hg levels in ASGM degraded areas in Madre de Dios

ASGM degraded areas are the product of extreme soil remodeling whereby old sediments from deeper layers are transported to the surface (Caballero et al., 2018). This explains why ASGM soil in our case are characterized by high sand content (69.40–96.6 %), low SOM (0.037–1.35%), variable CEC (2.93–8.8 Cmol (+) kg^{-1}) and variable pH (4.33–6.9 pH), which in turn partly explains the Hg variability we found in soils (up to 25% CV) (Table S2). Our findings show Hg variability depended mainly on CEC (r = 0.65) and SOM (r = 0.62) (Fig. S1), which is lower than documented previously by Velasquez-Ramírez et al. (2021) (r = 0.84 In both cases), which may due to the broader geographical

Table 2
Influence of the explanatory variables on Hg levels in the GLM with lower AIC.

Predictors	Hg		
	Estimates	CI	*p*
(Intercept)	21992.63	0.00–148600799434.11	0.241
pH	0.43	0.05–3.93	0.444
Sand	1.20	1.06–1.36	**0.006**
Clay	1.07	0.86–1.36	0.591
SOM	0.15	0.04–0.54	**0.004**
CEC	1.44	0.96–2.21	0.092
Season [Rainy]	266.64	0.10–2026004.78	0.189
Vegetation [CS]	0.01	0.00–2.17	0.104
Sediment [SED]	0.00	0.00–0.00	**<0.001**
No vegetation [US]	47054458.11	373.78–11720470054750.97	**0.004**
Season [Rainy]: CLUs [CS]	0.19	0.00–1251.78	0.722
Season [Rainy]: CLUs [SED]	892371.80	1.39–5008043865151.13	0.059
Season [Rainy]: CLUs [US]	0.07	0.00–1256807.79	0.751
Observations	315		
R2 Nagelkerke	0.822		

range of sampling points in the latter study. On a positive note, our soil Hg levels were below Peruvian and Canadian environmental soil quality standards (6.60 mg kg-1DM) (Table 1, Table S2).

In the Peruvian department of Madre de Dios, Hg levels have generally been found to be below 0.024 mg Hg kg^{-1} DM ((Roman-Dañobeytia et al., 2021; Soto-Benavente et al., 2020; Velásquez-Ramírez et al., 2020, 2021) (Table S4). However, other Latin-American findings reported higher Hg levels such as Quinchía (Colombia) with 7.1 mg kg^{-1} (Camargo et al., 2015); Bolívar (Venezuela) with 0.11 mg kg^{-1} (Santos-Francés et al., 2011), Combat Creek watershed (French Guiana) with 0.09–9.22 mg kg^{-1} (Guedron et al., 2009); and Serra da Santa Cruz (Brasil) with 10 mg kg^{-1} (de Andrade Lima, Bernardez, and Barbosa 2008).

ASGM soils covered by regenerating vegetation (CS) displayed higher Hg levels, up to 233% higher than in uncovered soil (US), which thus reflects the lower Hg retention capacity of the latter (Table S2). This might be explained by the fact that plants absorb Hg through their stomas (Gačnik and Sexauer Gustin, 2023; Gustin et al., 2022; Mahida et al., 2023), and gets incorporated into the soil through leaf litter (Casagrande et al., 2023). It was estimated that such Hg incorporation can represent up to 40% of the total Hg deposition in the Amazon rainforest (Fostier et al., 2015). As leaf litter is gradually incorporated in soil it leads to increasing SOM which further enhances sorption of Hg in soil. For example, in CS we found more SOM (2.09 ± 1.55 %) than US (0.37 ± 0.27 %) (Table S2), showing a direct significant correlation with Hg (r = 0.62) (Fig. S1). Most of the Hg is adsorbed as oxided forms (Hg^{+2}) to organic functional group, such as thiols (-SH) (Ajsuvakova et al., 2020).

Unsurprisingly, we found a high correlation of Hg with soil particles, especially clay (r = 0.63), which is an absorber of metals such as Hg (Najamuddin et al., 2023; Qin et al., 2023), mainly explained by high specific surface and secondary minerals such as oxides and hydroxides of Fe and Al (Akinyemi et al., 2022; Jelecevic et al., 2021; Ugwu and Anthony Igbokwe, 2019). In contrast, sand content was inversely correlated with Hg (r = −0.67) (Figueiredo et al., 2018; Méndez-López et al., 2023) due to the fact that sand is a weak sorbent for Hg (II) (Ballabio et al., 2021). In our case, US (>30% Sand), presents low SOM and clay contents, causing a decrease in Hg (II) sorbents, and therefore, low Hg retention capacity (Ballabio et al., 2021).

The Hg levels in the sediments of the artificial ponds in ASGM spoils ranged between 0.004 mg Hg kg^{-1} and 11.13 mg Hg kg^{-1} (median: 0.017 mg Hg kg^{-1}) (Table S2; Table 1), similar to other gold mining ponds. Yet most of the values we obtained are below the Interim

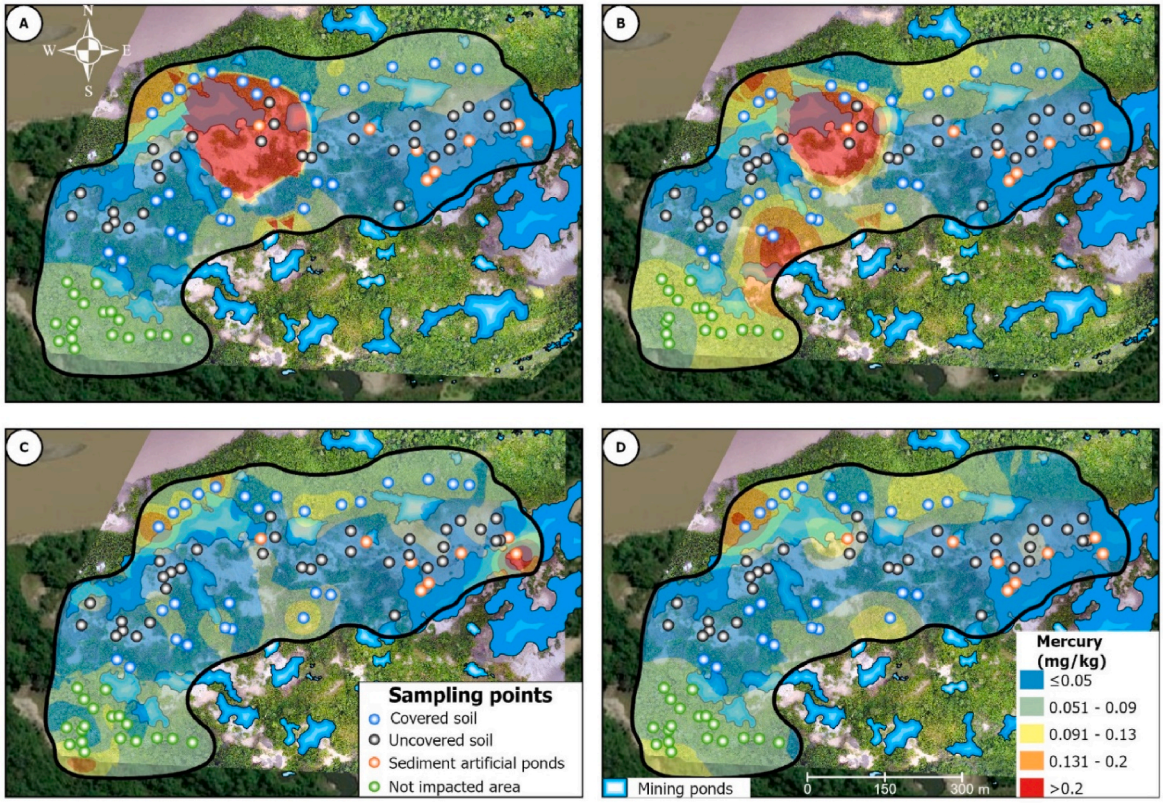

Fig. 4. Interpolation mapping of Hg levels per each season and year of evaluation, in the San Jacinto Native Community. (A) First year: dry season, (B) second year: dry season, (C) first year: rainy season, and (D) second year: rainy season.

Sediment Quality Guidelines (ISQG) for freshwater (0.17 mg Hg kg^{-1} DM; CCME), suggesting less severe contamination than in other comparable contexts. For example, in the Colombian department of Chocó levels around 0.210 mg Hg kg^{-1} were reported (Gutiérrez-Mosquera et al., 2020) and 0.52 ± 0.25 mg Hg kg^{-1} in Mojana (Pinedo-Hernández et al., 2015). Similarly, value of 19.24 mg Hg kg^{-1} were recorded in Venezuela (Santos-Francés et al., 2011), 0.22 ± 0.19 mg Hg kg^{-1} in Guyana (Howard et al., 2011), and 0.08–0.24 mg Hg kg^{-1} in Brazil (Cesar et al., 2011). Our highest levels of 11.10 mg Hg kg^{-1} and 2.54 mg Hg kg^{-1} (Table 1), were recorded in ponds near mining camps, where Hg was presumably used during gold recovery. Generating Hg as metallic droplets or amalgamated with gold (Moreno-Brush et al., 2020) can be a potential source of mercury methylation (Malone et al., 2023) Indeed, generally these kind of mining ponds (lentic systems) can produce up to 7 times more methylmercury than rivers (lotic systems) (Gerson et al., 2020), increasing local health risks for humans and the environment.

4.2. Mercury in the dry and rainy season

The spatial distribution of Hg levels in the area degraded by ASGM differed across the landscape units, seasons and years of evaluation (Fig. 4). In hot regions, Hg^0 emissions are high, decreasing Hg levels on the soil surface (Ballabio et al., 2021).

In the dry season, solar radiation increases the temperature, thus causing Hg volatilization (Feng et al., 2023; Liu et al., 2023; MacSween et al., 2020; Yang et al., 2019), In our study sites the maximum ambient temperature is around 36 °C (SENAMHI, 2021), while at US sites the surface temperature is above 36 °C causing the release of Hg from soil or vegetation through activation energy reduction (MacSween et al., 2020; Yang et al., 2019) and kinetically induced photoreduction of Hg^{2+} to Hg^0 (Fan et al., 2018).

Pluvial precipitation influences the variability of Hg in soil (Gębka et al., 2020) and conduces movement of Hg throughout aquatic ecosystems by erosion, runoff and percolation (Li et al., 2024). Water erosion has been estimated to displace at least 0.1% of the surface soil (Panagos et al., 2021) and the Hg bound to soil particles is transferred to the liquid phase at percentages between 17% and 20% (Zheng et al., 2016). Accordingly, agricultural areas have been calculated to lose 8.4 Mg Hg Yr^{-1} and forests 3 Mg Hg Yr^{-1} (Panagos et al., 2021). The estimated sediment flow in areas impacted by ASGM without vegetation is more than 1000 t km^{-2} Yr^{-1} (Diringer et al., 2019), and an average of 4.9 and 41 g Hg Km^{-2} Yr^{-1} is released in watersheds (Diringer et al., 2019). This would explain the trend of increasing Hg concentrations during the rainy season, which in our case results in a 961 % increase of Hg SED (median: 0.191 mg Hg kg^{-1}) in the rainy season compared to the dry season (median: 0.018 mg Hg kg^{-1}). Meanwhile, in the US sites Hg decreased by 45.5% (median: 0.011 mg Hg kg^{-1}) in the rainy season (Table 1), likely due to run off process associated with intense rainfall events, the sloping topography of degraded areas and displacement of the tailings towards aquatic ecosystems (Benoit, 2018; Chen et al., 2016; Gerson et al., 2020). Hg pollution in rivers and ASGM artificial ponds is particularly worrying as it is an important source of Hg biomagnification within the food chain, mainly through accumulation in fish muscle tissue (Diringer et al., 2015).

Hg levels in river sediments are linked to particle settling and re entrainment (Diringer et al., 2015). We found significantly higher Hg content in sediment during the rainy season (median: 0.018 mg Hg kg^{-1}) than in the dry season (median: 0.016 mg Hg kg^{-1}, Kruskal-Wallis, $p = 0.72$) (Table 1; Fig. 2), This may suggest that Hg settling and re entrainment during the rainy season is increased and as a consequence produces Hg mobilization of Hg-bound soil particles from upland terrain, adsorption to suspended particles from dissolved forms, and resuspension o in the river channel (Diringer et al., 2019). This Hg remobilization can be dramatically higher near US, due to higher Hg release from sandy soils than clayey soils (Couic et al., 2021; O'Connor et al., 2019) and its lower Hg retention capacity.

5. Conclusion

Mercury levels in soil and sediment from artificial ponds in ASGM degraded areas showed significant variability between seasons (rainy and dry) mainly influenced by vegetation, soil organic matter (SOM), clay particles, and cation exchange capacity (CEC). Most Hg levels were below the upper limits of the Peruvian and Canadian environmental quality standards. However, in the sediment of the artificial pool near the mining camp, high levels of Hg were present, representing a potential source of contamination. During the wet season, rainfall decreased Hg levels in degraded soils by up to 45.5%, while it increased significantly in sediments by up to 961%. This mobility suggests a high potential risk of Hg contamination downstream, mainly in artificial water bodies during rainfall. In contrast to the uncovered degraded areas, natural forest soils alongside the covered degraded soils showed a high capacity to retain Hg, mainly influenced by higher levels of CEC and SOM. Considering the risks associated with Hg accumulation in sediments its monitoring and remediation must be prioritized to support ecological restoration practices in the Peruvian Amazon.

Funding

This study was funded by PROCIENCIA and the National Council for Science, Technology and Technological Innovation (CONCYTEC) through the Project "Recovery of soils in areas degraded by alluvial gold mining through the succession of cover crops and plants of potential agroforestry use in the Madre de Dios region" (Contract number 141–2017 FONDECYT).

CRediT authorship contribution statement

Edwin Becerra-Lira: Conceptualization, Data curation, Methodology, Validation, Visualization, Writing - original draft, Writing - review & editing. **Liset Rodriguez-Achata:** Conceptualization, Methodology. **Adenka Muñoz Ushñahua:** Investigation, Validation. **Ronald Corvera Gomringer:** Resources, Writing - review & editing. **Evert Thomas:** Visualization, Writing - review & editing. **Jorge Garate-Quispe:** Data curation, Visualization, Writing - review & editing. **Litcely Hilares Vargas:** Investigation, Validation. **Pedro Romel Nascimento Herbay:** Investigation, Validation, Writing - review & editing. **Luis Alfredo Gamarra Miranda:** Data curation. **Eleuterio Umpiérrez:** Methodology, Writing - review & editing. **Juan Antonio Guerrero Barrantes:** Validation, Visualization, Writing - review & editing. **Martin Pillaca:** Methodology, Validation. **Edgar Cusi Auca:** Investigation, Writing - review & editing. **Joel Peña Valdeiglesias:** Writing - review & editing. **Ricardo Russo:** Writing - review & editing. **Dennis del Castillo Torres:** Conceptualization, Resources, Writing - review & editing. **Manuel Gabriel Velasquez Ramírez:** Conceptualization, Methodology, Project administration, Resources, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

We are grateful for the support of specialists from the Research Institute of the Peruvian Amazon (IIAP), Bioversity International, Universidad LaSalle (ULASALLE), the National Agrarian University La Molina (UNALM) and Centro de Innovación Científica Amazónica

(CINCIA). Special thanks are due to PROCIENCIA and Consejo Nacional de Ciencia, Tecnología e Innovación Tecnológica (CONCYTEC) from Peru.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2023.118073.

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