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Environmental contamination by metals, metalloids, and cyanides in the historic and active ASGM area of Kokumbo in Côte d'Ivoire

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Abstract

In Côte d'Ivoire, despite an intense development of artisanal and small-scale gold mining (ASGM) activities in the last two decades, the environmental impacts of this activity are poorly documented. This study aimed to document the concentrations of geogenic and exogenous contaminants potentially released by ASGM (metals and cyanides) in different sources (ore, mining wastes) and environmental compartments (soils, surface and ground waters, sediments) in the Kokumbo area, part of the Au-rich Birimian greenstone belt. Alluvial ore material is enriched in various metal(oid)s (As, Co, Cu, Cr, Fe, Mn, Ni, Sb, and V) compared to the average composition of the upper continental crust while other metals (Cd, Pb, Ti, Zn) show no geochemical anomalies. High Hg concentrations were found in cyanidation residues (up to 8.32 mg/kg) and sediments (up to 20.4 mg/kg) compared to unprocessed alluvial ores (0.06 ± 0.01 mg/kg) indicating that Hg used in amalgamation is the source of Hg contamination. Cyanidation residues contain up to 100 mg/kg of total cyanides but generally less than 3% are in the form of free cyanides, the most mobile and toxic form. Arsenic concentrations in water are low (< 2.5 µg/L) despite its relatively high content in sediments and soils (76 ± 54 mg/kg), showing a low mobility of As, likely due to its adsorption on iron oxy(hydr)oxides. Apart from Mn, metals have low dissolved concentrations in water except in a stream draining a cyanidation site suggesting that the cyanidation effluent discharge may contribute to metal dispersion in rivers.

Keywords Artisanal and small-scale gold mining · Contamination · Metal · Cyanide · Mercury · Arsenic · Ivory Coast

Introduction

Although often informal or illegal, artisanal and small-scale gold mining (ASGM) plays a very important socioeconomic role in many low- and middle-income countries mainly in Africa, Asia, and South America (Intergovernmental Forum on Mining 2017). At the global scale, about 20 million persons work in ASGM representing 90% of the workforce in gold mining. In Africa, it was estimated that in 2014, around 9 million people (100,000 in Côte d'Ivoire) worked in small artisanal mines (all minerals considered) and about 54 million people depended on the sector (600,000 in Côte d'Ivoire) (Intergovernmental Forum on Mining, 2017). ASGM helps to reduce poverty and supplements insufficient income from agriculture during the dry season. It provides employment opportunities for migrants, young people with limited education and women, while contributing to rural development, particularly in arid regions (Adranyi et al. 2023; Hilson and Hu 2022; Mwandiringana and Ye 2023; Soko 2019; Champy and Dessertine 2024). However, ASGM can have major adverse impacts on the environment, the society, and the health of miners and residents living near the mining sites (Champy and Dessertine 2024; Landrigan et al. 2022; Ondayo et al. 2023). The environmental impacts of ASGM include deforestation, land and river physical degradation (Ngom et al. 2022a), and contamination of soils, rivers, and groundwaters by chemicals that may have a geogenic source (metals and metalloids present in the ore) or that are used for ore processing (Timsina et al. 2022). Indeed, the various artisanal techniques for gold (Au) extraction use chemicals such as acids, mercury (Hg), and/or cyanide (CN). These compounds, once released into the environment, are harmful to living organisms and may affect human health (Malone et al. 2023; Timsina et al. 2022). ASGM is the largest anthropogenic source of Hg emissions. Estimations range from 640 to 1000 tons/year at the global scale, and from 54 to 567 tons/year (median value of 140) for Africa (Cheng et al. 2023; Streets et al. 2019). ASGM is also the larger source of Hg affecting inland aquatic ecosystems (Obrist et al. 2018). During Au-Hg amalgamation, large quantities of liquid elemental Hg(0) are released in surrounding soils and aquatic systems, lost in tailings and emitted as vapor possibly resulting in environmental contamination through atmospheric transport and subsequent Hg deposition (Moreno-Brush et al. 2020). Once inorganic Hg(0) enters aquatic systems, it is transported in association with organic matter and sediments and can undergo oxidation to Hg(II) and bacteria-mediated methylation processes (Moreno-Brush et al. 2020). Organic methylmercury ($\text{CH}_3\text{-Hg}^+ \text{X}^-$, where X^- design any anion, also noted Me(Hg)) is the most toxic form of

Hg. It is easily assimilated and accumulated in aquatic trophic chains through bioaccumulation and biomagnification processes (Eisler 2004). Few studies exist on human exposure to Hg in West Africa (Ondayo et al. 2023). Nevertheless, high Hg levels have been reported in ASGM workers and communities in several West African countries including Burkina Faso, Ghana, Nigeria, and Senegal. Human exposure to Hg is mainly associated with the consumption of fish contaminated with MeHg around mining areas and involvement in the amalgamation practice (Basu et al. 2015; Gibb and O'Leary 2014; Niane et al. 2015; Ondayo et al. 2023; Tomicic et al. 2011).

In addition to Hg, an increasing use of cyanides (CN) in ASGM is reported worldwide as it allows to recover Au more efficiently at a relatively low cost (Malone et al. 2023). This compound is highly toxic for organisms due to its ability to interfere with respiration at the cellular level. The health impact of a chronic exposure to non-lethal levels of CN is not well characterized, but its adverse health effects on ASGM workers, including loss of short-term memory, have been reported in Burkina Faso (Knoblauch et al. 2020). Cyanides exist in various forms in the environment, including free cyanide (F-CN), the most toxic form, and less toxic complexes with metals (e.g., Au, Cu, Fe, Hg, and Zn) (Johnson 2015). Cyanide-metal complexes are relatively stable but can dissociate and release F-CN in water under low pH and sunlight exposure (Johnson 2015). Microbial activity plays also an important role in the degradation of free and complexed cyanides in gold mining tailings (Akcil and Mudder 2003; Gupta et al. 2010; Welman-Purchase et al. 2024). However, even though F-CN can be abiotically and biotically degraded in the environment, persistent F-CN pollution and dissociation of cyanide-metal complexes can occur in ASGM-impacted soils and aquatic systems, inducing a long-term risk for the environment and human health (Guimaraes et al. 2011; Razanamahandry et al. 2016). For example, in Burkina Faso, F-CN were detected in soils and river water in a small watershed affected by ASGM despite the presence of CN degrading bacteria (Razanamahandry et al. 2016). Besides, the reprocessing of Hg-rich tailings with CN after Hg amalgamation represents an additional risk as CN can react with Hg(0) and promote its oxidation to Hg(II) and further form cyanide-mercury soluble complexes (Hg(CN)_n^{2-n}) increasing Hg mobility in ASGM-impacted aquatic systems (Malone et al. 2023). Complexation of Hg with CN may also favor Hg methylation (Malone et al. 2023). In Ghana, river sediments affected by ASGM activities using both Hg and CN had higher Me(Hg) concentrations and $\text{Me(Hg)/Hg}_{\text{total}}$ with respect to contexts using Hg alone (Tulasi et al. 2021).

Besides the chemicals used in ASGM, other toxic metals and metalloids such as arsenic (As) and lead (Pb) which are commonly associated with gold mineralization can

also be released to the environment during gold extraction. This occurs, for instance, during crushing, grinding, and lixiviation of ore rocks, or through the lixiviation and erosion of tailings (Ondayo et al. 2023; Plumlee et al. 2013; Tabelin et al. 2020). In particular, in West Africa, gold mineralization in volcano-sedimentary shales and volcanic rocks of the Birimian formations is often associated with As-rich sulfide minerals and their oxidation can lead to arsenic contamination of surface waters and groundwater (Ahoulé et al. 2015). For example, in Niger, high level of As and Mn were detected in groundwater in the ASGM region of Koma Bangou. The highest groundwater As concentrations in this region were found in old wells originally dug for gold extraction and later used for drinking water supply (Farouk et al. 2022). Farouk et al. (2022) hypothesized that As was mobilized as water interacted with the Au-As sulfide ore exposed during its exploitation. A lack of knowledge about possible co-contaminants in association with the gold mineralization can have dramatic consequences for the ASGM communities. In 2010, Pb poisoning due to the presence of high concentrations of Pb in Au ores led to the death of around 400 children under 5 years old and an acute poisoning requiring treatment of 2000 more children in Nigeria (Dooyema et al. 2011; Plumlee et al. 2013). Consequently, it is essential to carry out integrated studies looking at both geogenic and exogenous contaminants associated with potential ASGM-related sources (ores, tailings, effluents...) and looking at the dispersion of these contaminants in the environment. It is worth recalling that geogenic metals are defined as metals that are naturally present in the ore. Their release in the environment may occur during natural processes or may be enhanced by ore processing. Exogenous contaminants are not present in the ore and are brought and released in the mining sites during mining activities. Geogenic sources may also combine with exogenous sources. Contaminations of anthropic origin can include the enhanced release of geogenic metals as a result of ore processing and the release of exogenous contaminants (metals and CN).

To date, only a few studies exist on possible environmental contamination by metals and CN resulting from ASGM activities in Côte d'Ivoire (Kinimo et al. 2018a, b; Mason et al. 2019; N'Goran et al. 2022), which is very little in comparison with other regions of the world affected by AGSM, such as Ghana and South American countries. Mason et al. (2019) reported that ASGM has an influence on Hg levels in rivers and fish on a regional scale in Côte d'Ivoire. Kinimo et al. (2018a, b) showed that concentrations of several metal(loid)s (e.g., As, Pb, Cr, Ni, Mn) in wetland sediments from three different mining areas were affected by mining activity in an extent that depended on the type of activity (industrial vs ASGM) and the regional location. These results and the fact that both Hg and CN are

widely used in the country (Champy and Dessertine 2024) call for further studies about ASGM environmental impacts in Côte d'Ivoire.

The gold-mining area of Kokumbo, Central Côte d'Ivoire, was chosen to provide for the first time a comprehensive assessment of the environmental contamination by metals and CN from ASGM activities in Côte d'Ivoire. This zone is exploited for gold since the nineteenth century. ASGM activities expanded since the beginning of the 2000s and especially since 2013–2014 (Champy and Dessertine 2024; Soko 2019). In detail, the aim of this study was to document the concentrations and sources of contaminants in ASGM activities at Kokumbo including possible exogenous contaminants used in Au ore processing (e.g., Hg, Zn and CN) and geogenic contaminants (e.g., As, Hg, Pb, Mn and Zn) in different environmental compartments (groundwaters, surface water, sediments or soils) potentially affected by ASGM. For that, metal, metalloid, and cyanide concentrations were determined in representative samples of potential sources and impacted environments to determine their natural levels (i.e., local geochemical background) and to quantify the degree of anthropogenic contamination. In addition, petrological and mineralogical analyses were performed to determine major and minor phases potentially bearing metal co-contaminants (such as As-rich or Pb-rich sulfides). A statistical analysis was conducted to evaluate the contribution of ASGM to the observed metal concentrations and to identify the origin of the metals (geogenic vs exogenous).

Material and methods

Study site

The area of Kokumbo (sub-prefecture of the department of Toumodi, ~35,000 inhabitants) is located in central Côte d'Ivoire, about 30 km south of Yamoussoukro (Fig. 1). The studied area (~150 km²) comprises several villages (Akroukro, Assèkro, Niamkey-Konankro, Kplessou, Sérémé, Gbonty, Kimoukro) distributed around the Kokumbo Hills (including Mont Kokum, 510 m a.s.l) where artisanal gold mining operations are carried out. The humid equatorial climate with a dense semi-deciduous forest is characterized by four seasons: two rainy seasons (March–June and September–October) alternating with two dry seasons (July–August and November–February). The mean annual precipitation and temperature are 1100 mm and 27 °C, respectively (Ahoussi et al. 2020). The studied area is drained on its western side by the Bandama River (Fig. 1). The geology of the Kokumbo area is part of the Baoulé Mossi domain of the West African craton. Kokumbo lies within the Toumodi greenstone belt, a Birimian NE-SW metamorphized volcano-sedimentary sequence with greenschist facies surrounded by

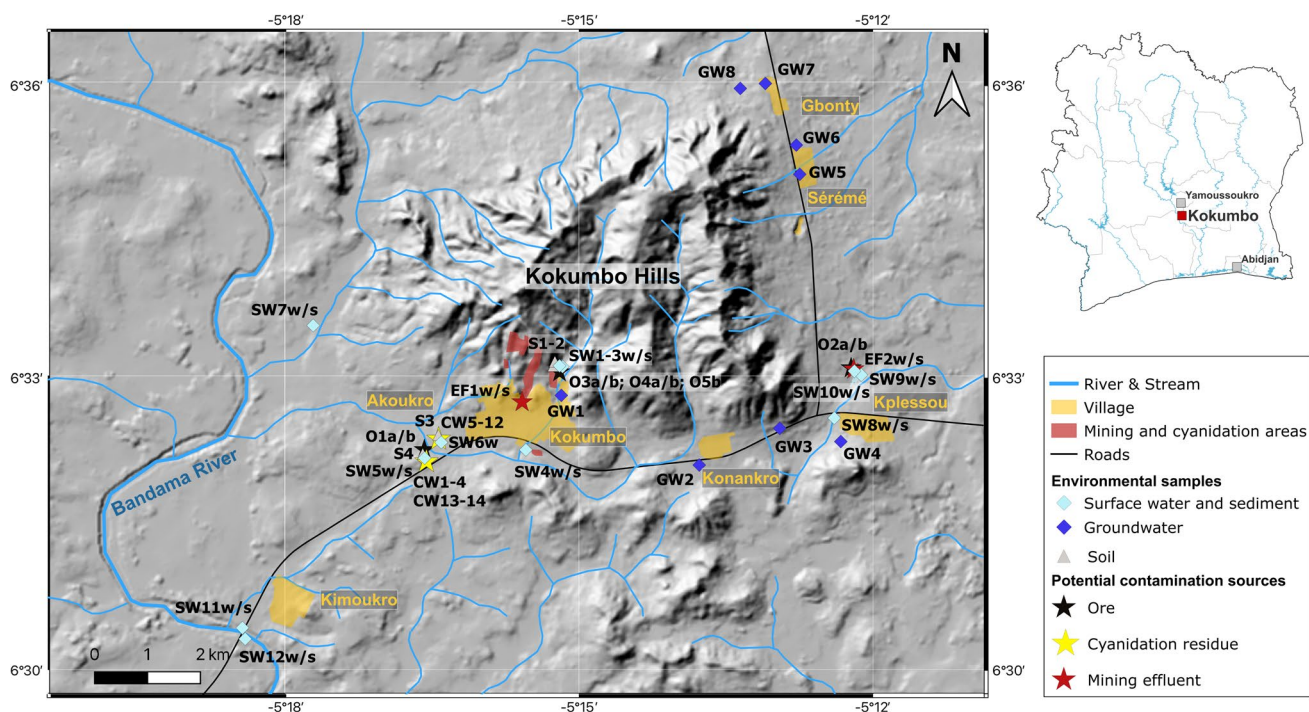


Fig. 1 Sample location of environmental samples (river water and sediments, soils, groundwater) and potential contamination sources (ores, mining effluent water and sediments and cyanidation residues)

in the area of Kokumbo. Background shaded relief has been derived from Forest and Buildings removed Copernicus DEM (FABDEM) (Hawker et al. 2022)

granite and gneiss rocks (Hayman et al. 2023). Gold was discovered in Kokumbo in 1850 and from then, alluvial, eluvial, and vein gold deposits were exploited by the local Baoulé population (Aguié and Kouamé 2019; Chauveau 1978). From the early to mid-twentieth century, during the French colonization period, gold was mainly exploited by European mining companies. The artisanal activity was then resumed by the local population from 1958 and it has been booming since 2002 with the beginning of the political-military crisis in Côte d'Ivoire and a sharp increase of gold price (1600 US \$ per ounce (31.1 g) in 2002, compared to 250 US\$ a few years before) (Soko 2019). The gold price was stable between 2014 and 2020, and then increased sharply again until present, approaching 3000 US\$ per ounce in first trimester of 2025.

ASGM is performed in Kokumbo similarly to what is done throughout West Africa; it mixes surface excavations of eluvium and alluvium material and vertical excavation of gold rich vein and hard rock with digging wells. Alluvial and eluvial gold is mined by mixing gold-rich sediment or soil material with river water using motor-pumps and passing it over an inclined ramp to gravimetrically pre-concentrate gold (Fig. 2(A–C)). The pre-concentrated ore is recovered from the ramp and gold is pre-concentrated using gold panning (Fig. 2(D)) and finally amalgamated with mercury. The ore material going through the ramp is generally further

treated with cyanide (Fig. 2(E–G)). In this latter process, several chemicals are used besides cyanides including hydrochloric and sulfuric acids and metal zinc sometimes replaced by activated charcoal. In addition, wells are dug (down to ~ 100 m) to extract gold-rich quartz veins or hard rocks (Fig. 2(H)). Wood is used to support shaft walls and ventilation is provided by plastic sheeting wound to bring air. These vein ores or hard rocks are crushed and milled (Fig. 2(I–K)). The flour obtained also first passes over an inclined ramp with water to gravimetrically pre-concentrate gold (Fig. 2(L)). The pre-concentrate is then processed using gold panning and Hg amalgamation (Fig. 2(D)), and the discarded ore material going through the ramp further treated with cyanide (Fig. 2(E–G)).

Sampling and sample pre-treatment

Two field campaigns were carried out in the Kokumbo area in October 2020 and September 2021 during the short rainy season. Sampling locations (Fig. 1) of streams, ponds, soils, and potential contamination sources have been selected to represent different contexts (alluvial ore extraction sites, ore processing sites, cyanidation sites, no mining) and by adapting to site accessibility. Groundwater sampling was made in wells from the different villages in the studied area (Fig. 1). Details about the samples are given in Table S1 in

Fig. 2 Main ASGM practices in Kokumbo. **A–B**: exploitation of gold-rich river sediments and soils; **C**: device for washing alluvial ore; **D**: gold panning, **E–G**: cyanidation site. **H**: vein ore extraction shaft; **I**: crushed vein ore c; **J–K**: vein ore milling; **L**: vein ore flour washing ramp



Appendix. Each location was sampled once either in 2020 or 2021, except the Balo pond which was sampled three times (SW1 - 3w/s). The potential sources of contamination consisted of raw alluvial and vein ores ($n = 4$, O1a to O4a), processed (crushed and washed) ores ($n = 5$, O1b to O5b), cyanidation residues ($n = 16$, CW1 to CW14c), and water (EF1w, EF2w) and sediment (EF1s, EF2s) from effluent discharges from an alluvial ore washing site (Fig. 1, Table S1 in Appendix). The approximate age of cyanidation residues was recorded based on the information provided by workers on cyanidation sites.

Eight samples of groundwaters (GW1 to GW8) used for human consumption were collected in Kokumbo and surrounding villages (Gbonty, Sérémé, Kplessou, Niamkey-Konankro, Fig. 1). Samples of surface waters

($n = 12$) and sediments ($n = 11$) were collected in streams or ponds supposedly unaffected by mining activity (SW8w/s; SW9w/s), in streams or ponds draining gold mining areas (SW1 - 7w/s and SW10w/s), and in the Bandama River (SW11w/s, SW12w/s). Physicochemical parameters of groundwaters and surface waters (temperature, pH, conductivity) were measured in the field using a calibrated multiparameter (WTW 3210 and 3310). In addition, four surface soil samples were collected in the vicinity of cyanidation areas (S1 to S4).

Solid samples (ores, cyanidation residues, surface soils, sediments) were collected using a spoon in plastic bags or vials and consisted of composite samples to be representative of the studied material (e.g., residue heap, 10 m² soil area). Groundwaters were sampled using a bucket

rinsed three times with the sampled water. Surface water and groundwater samples were filtrated using disposable polypropylene syringes and syringe filters (cellulose acetate, 0.22 µm). First, the syringe was rinsed three times with water from the site and the filter was rinsed by discarding ten drops of filtered water. Then, the filtered water was collected in an acid-cleaned HDPE bottle (30 mL) and acidified to 0.15 M with HNO₃ (Analpure, Analytika®). Water and solid samples were kept in a cool box for transport to the laboratory and then stored at 4 °C until their shipment to the laboratory HydroSciences Montpellier (France). Solid samples were freeze-dried and crushed in an agate mortar (ores and cyanidation residues) or using a mixer mill (MM 400, Retsch, soils and sediments) after sieving at 2 mm using a nylon sieve.

Six rock samples were collected at Kokumbo at ASGM sites located on the Kokumbo Hill: R1, representative of the oxidized facies, was collected at the surface near an artisanal extraction shaft intersecting a gold-rich quartz vein and R2, R3, R4, R5, and R6 were collected on the flanks of Kokumbo Hill and are representative of hard rocks exploited in extraction shafts.

Chemical and mineralogical analysis

Ore and sediment mineralogy

Polished thin sections of rock samples (R1 to R6) were made at the laboratory Geosciences Montpellier. Petrographic observations were done with a transmitted-reflected light-polarizing microscope.

The mineralogy of the solid phases (sediments, ore and cyanidation residues) was determined by X-ray diffraction using a PANalytical X'Pert Pro diffractometer equipped with a cobalt tube ($\lambda = 1.79 \text{ \AA}$) running at 40 kV and 40 mA at the CEREGE laboratory. Samples were ground in an agate mortar, deposited on low background silicon plates and analyzed from 5° (2 θ) to 70° (2 θ) with a step size of 0.033° and a total counting time of 5 h. Samples were also spun at 15 rpm to improve statistics. Phase identification was performed using the X'pert Highscore plus software (PANalytical) together with the PDF- 2 ICDD database (International Center for Diffraction data, Powder Diffraction Files 2).

Major and trace element determination

All consumable (e.g., vials, pipette tips, tubes) used for the preparation of water and solid samples for trace element analysis were acid-cleaned in a 1.5 M HNO₃ bath during 24 h and rinsed three-times with ultrapure water before use. Sample preparation was carried out in a class 10 000 cleanroom at HydroSciences Montpellier. High purity acids including 9.5 M HCl (Suprapur, Merck®), 15 M

HNO₃ (Analpure, Analytika®), and 22.6 M HF (Suprapur, Merck®) and ultrapure water (Milli-Q®, resistivity > 18.2 MΩ.cm, Q-POP Element system, Millipore®) were used for sample preparation and analysis.

Solid samples (ores, cyanidation residues, surface soils, sediments) were digested using a microwave-assisted digestion (ultraWAVE, Milestone®). Then, 50 mg of solid samples were weighed in a PTFE tube and a 5 mL mixture containing concentrated HNO₃:HCl:HF (3:1:1) was added. The samples were digested using the following microwave protocol: heating from ambient temperature to 200 °C in 25 min, then heating at 200 °C for 10 min. The samples were allowed to cool down to room temperature, transferred into 30 mL PFA vials, and evaporated to dryness at 75 °C. The dry residue was redissolved in 30 mL of 1.5 M HNO₃. The vials were placed in an ultrasound bath for 30 min and then heated with closed cap at 80 °C for one night to ensure a complete dissolution of the residue. A certified reference material (CRM) of stream sediment (NCS DC 70317) and a procedural blank was included in each set of samples to control the analytical quality of the procedure.

Water samples and digest solution of solid samples were analyzed using a routine procedure by ICP-MS (iCAP TQ, Thermo Scientific®, AETE-ISO platform, OSU OREME, Université de Montpellier) after an adequate dilution in 0.15 M HNO₃. Concentrations of major elements (Al, Ca, Fe, K, Mg, Na) and trace elements (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sb, Ti, V, Zn) were determined using external calibration and internal standard correction (Sc, Ge, Rh, and Ir) to adjust for sensitivity variations induced by matrix effect or instrumental drift. A collision cell with He gas and a KED mode (kinetic energy discrimination) were used to eliminate polyatomic interference created in the argon plasma. A CRM of water (SLRS-6) was analyzed. Accuracy was within 6% of certified values for all measured elements. The concentrations measured in the procedural blanks represented less than 1% of sample concentrations for all measured elements showing that no contamination occurred during the entire protocol. Finally, concentrations measured in the CRM of sediment (NCS DC 70317) were matching certified values within 12%, except Cr ($\pm 18\%$) and Ni and Sb ($- 23\%$ for 2020 sample digestion) (Table S2 in Appendix).

Total mercury concentration in solid samples was measured using a Direct Mercury Analyzer (DMA- 80, Milestone®) following the 7473 EPA standard method. About 0.02 g to 0.1 g of crushed freeze-dried sediments, soils, ores, or cyanidation residue were analyzed. CRM of sediments (NCS DC 70317 and PACS- 2 or PACS- 3) were analyzed at the beginning and the end of the analytical session. Accuracy was better than 10% compared to certified Hg concentration of NCS DC 70317 ($0.035 \pm 0.002 \text{ mg/kg}$, $n = 5$, $- 1.6\%$ of the certified value), PACS- 2 (2.77 mg/kg , $+ 8.7\%$ of the certified value), and PACS- 3 (2.90 mg/kg , $+ 3.2\%$ of the certified value).

Free and total cyanide determination

Total cyanide (T-CN) concentrations were determined in a selection of 20 cyanidation residues, soil, and sediment samples in an accredited laboratory (Wessling, France) using the certified method ISO 17380 for the photometric determination of total cyanide in soils using automated distillation/continuous-flow analysis. In addition, free cyanide (F-CN) concentrations in soils were estimated using an extraction of 2.5 g of solid samples in 50 mL of 1 M NaOH (adapted from EPA method 9013 A). Samples were wrapped in aluminum foil to protect the suspension from light and agitated for 24 h on a shaking table. Then, samples were centrifuged at 4000 rpm for 30 min and the supernatant was filtrated at 0.22 μ m to remove particles. Free cyanide (F-CN) concentrations were measured in the extract, after adjusting its pH to 7 with diluted HCl, using the pyridine pyrazalone method (HACH® method 8027) and a spectrophotometer (DR1900, HACH®) (Razanamahandry et al. 2016).

Data processing

Statistical tests were performed using R software. The student *t* test (when data followed a normal distribution) or the Mann–Whitney test (when normality was not verified) were applied to compare average values of the different groups of samples (e.g., raw ore vs processed ore, ore vs. cyanidation residues, ores vs. sediments). Correlations among elements were evaluated using the Spearman method. In an attempt to determine the origin of metal(loid)s, a principal component analysis (PCA) was performed with the “FactoMineR” and “factoextra” packages using standardized (centered-reduced) log-transformed concentrations of 15 metals (Al, Fe, Mn, Ti, As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Sb, V, and Zn) including all solid samples (ores, cyanidation residues, sediments, and soils).

Enrichment factors of metals and metalloids were calculated in soils and sediments by normalizing the concentration of the element (X) by the concentration of an immobile element (aluminum) in the sample $((X/Al)_{\text{sample}})$ and comparing with the same ratio in a reference $((X/Al)_{\text{ref}}$, Eq. 1). Two alternative references were used in this study. The average composition of the upper continental crust (UCC, Rudnick and Gao 2014) was used to investigate local enrichment(s) of the geochemical background. The average composition of local ore material was used to investigate the possible enrichments in exogenous metals brought by the Au recovery processes (amalgamation, cyanidation).

$$EF_{X/ref} = \frac{(X/Al)_{\text{sample}}}{(X/Al)_{\text{ref}}} \quad (1)$$

Comparison with other mining sites in West Africa

The results of this study shall be placed in a regional perspective. For this purpose, a review of the literature was carried out to produce a summary of published data on metal contamination levels in sediment and soils (Table 3) and water (Table 4) in ASGM impacted areas in West Africa. The criteria for including studies were as follows: (i) study area in a West African country part of the Birimian Greenstone Belt (mainly Côte d’Ivoire, Burkina Faso, Guinea, Ghana, Niger, Mali, and Senegal), (ii) study area potentially impacted by ASGM (industrial mining excluded at the exception of two sites in Côte d’Ivoire and Mali due to the scarcity of studies in these countries), (iii) data are available (raw data or statistical summary), (iv) appropriate information is provided about the analytical techniques and analytical controls to ensure metal data quality (e.g., detail about the analytical technique used for metal determination, results for certified reference materials, and analytical blanks). Data summaries of metal contamination in soils, sediments, and waters in mining regions have already been published for Africa with a broader geographical scope such as Gerson et al. (2018), Kohio et al. (2024), and Moreno-Brush et al. (2020) for mercury; Ahoulé et al. (2015) for arsenic; and Affum et al. (2016) and Ondayo et al. (2023) for polymetallic dataset.

Results

Characterization of potential sources of contamination

Mineralogy observations

Petrographic observations and mineralogical analysis were performed to identify the major and minor minerals in ores and to identify the possible presence of metal-bearing minerals in the gold mineralization such as metal sulfides. Petrographic observations were done on 6 rock samples representing mined ores (Fig. 3). Two kinds of ore are mined, an oxidized facies and less altered hard rocks. The rock R1 is a very oxidized breccia with a matrix mainly composed of fine-grained quartz. It represents the shallow oxidized facies that is mined at the surface. It can host quartz veins containing gold. Other minerals observed in R1 include iron oxides and titanium oxides (rutile). They represent less than 5% of the rock. The less altered hard rock is represented by samples R2–R6 and is mined deeper, in the flank and bottom of the Kokumbo Hill. Two distinctive rocks can be observed within this facies. The first rock type (R2 and R3) is composed of feldspar, chlorite, sericite, minor quartz, and

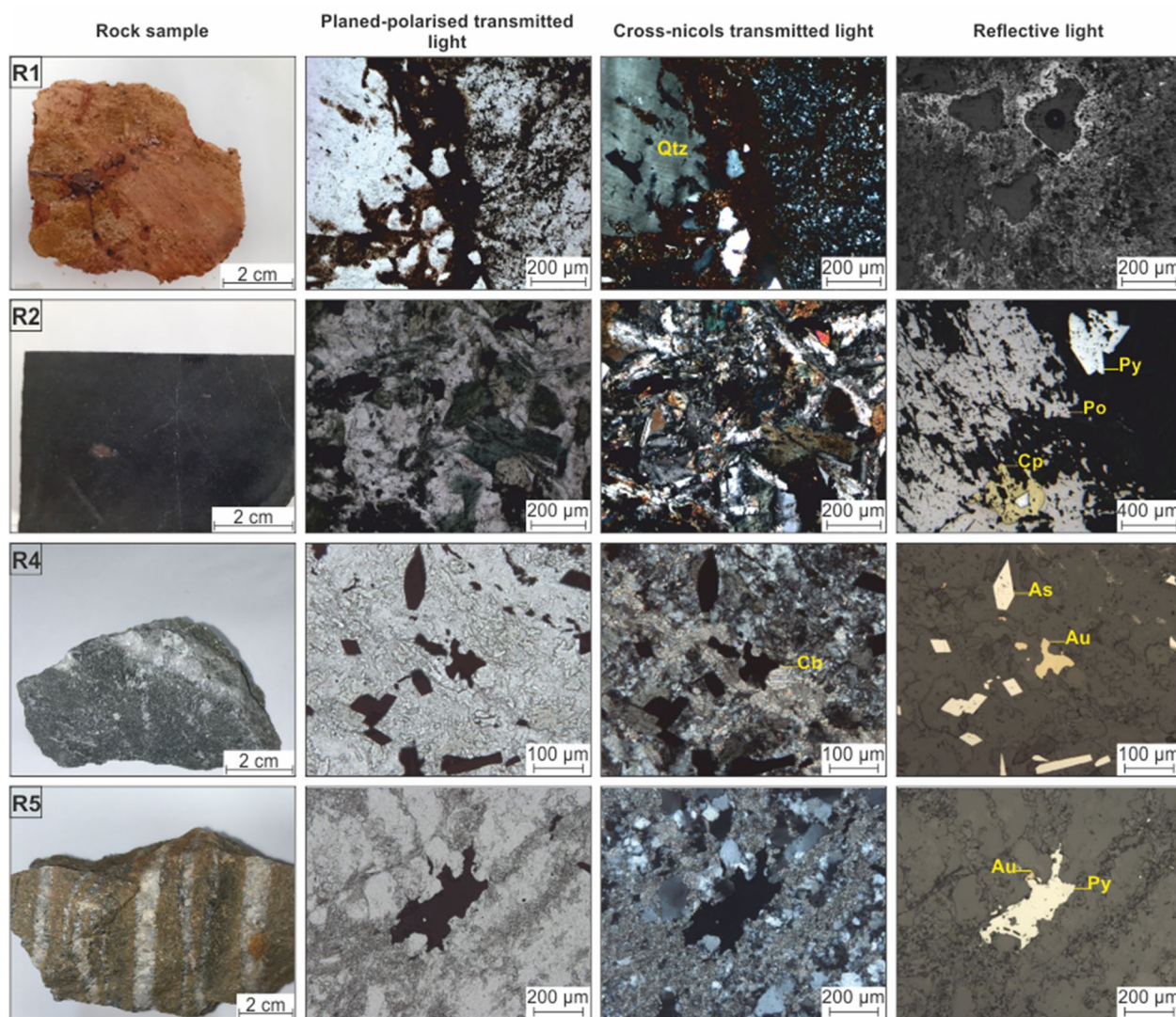


Fig. 3 Macroscopic and microscopic observation of R1, R2, R4, and R5 rock samples in planed-polarized transmitted light, Cross-nicols transmitted light and reflective light (As: arsenopyrite, Au: gold, Cb: Carbonate, Cp: chalcopyrite, Po: pyrrhotite, Py: pyrite, Qtz: quartz). R1: Highly oxidized breccia with quartz and very fine grain matrix. Rare iron oxides and Ti (rutile) minerals. R2: Metamorphic rock composed of K-feldspar ($\approx 50\%$), chlorite, and sericite ($\approx 40\%$), quartz ($\approx 5\%$) and rare epidote. Sulfides ($\approx 5\%$) are mainly pyrite, chalcopy-

rite and pyrrhotite. R4: Metamorphic rock with quartz veins mainly composed of deformed quartz, calcite, chlorite and rutile (rare). Sulfides are mainly pyrite and arsenopyrite. Gold is present as free grain and included in pyrite (cracks and border). R5: metamorphic rock with quartz veins mainly composed of deformed quartz, calcite, chlorite, muscovite, and rare epidote. Pyrite is the main sulfide. Gold is present as free grain and included in pyrite (cracks and border)

rare epidote. Locally sulfides are present up to 5% and correspond to pyrite, pyrrhotite, and chalcopyrite. Ilmenite is rare and no gold was found in those samples. The second type of rocks (R4, R5, R6) is composed of quartz, carbonates, chlorite, muscovite, and rare epidote. Sulfides (pyrite and arsenopyrite) are locally present up to 5%. Gold occurs as free gold, in inclusion within pyrite and at its edges.

According to XRD results (Fig. S1 and Table S3 in Appendix), alluvial ore material contained quartz, phyllosilicates (kaolinite, muscovite, paragonite), iron oxy(hydr)oxides (hematite, goethite), titanium oxides (anatase, rutile), and aluminum

oxy(hydr)oxides (gibbsite, boehmite). No significant mineralogical change occurred following the ore milling and washing step except in mineral phase relative abundances (e.g., less kaolinite and more quartz was found in crushed and washed ores O1b, O2b, and O3b compared to raw ores O1a, O2a, and O3a, respectively, Fig. S1). The mineral assemblage found in cyanidation residues (CW1 to CW4) was similar to that of ore materials (quartz, kaolinite, hematite, goethite and muscovite) except that titanium oxides were not detected. The sediments collected in the mine effluents also contained the same major mineral phases to those observed in alluvial ore material.

Major and trace element concentrations

The geochemical composition (concentrations of major and trace elements) was determined in raw and processed ores and in cyanidation residues to evaluate whether they represent potential contamination sources of metals and metalloids. Ore material was depleted in Al (31.0 ± 17.5 g/kg), Ca (0.69 ± 0.51 g/kg), K (3.27 ± 3.47 g/kg), Mg (0.49 ± 0.40 g/kg), Na (0.77 ± 1.04 g/kg) compared to the average composition of the upper continental crust (UCC, Table 1, Fig. 4). The depletion in alkali elements (Ca, K, Mg, and Na) is consistent with the observed mineralogical phases and with the high degree of weathering of source rocks. It is typical for soil and river sediments of the West African Craton (Asiedu et al. 2017; Roddaz et al. 2007). In contrast, metal and metalloid concentrations including As (96 ± 84 mg/kg), Co (65 ± 72 mg/kg), Cu (139 ± 80 mg/kg), Cr (353 ± 240 mg/kg), Fe (126 ± 91.9 g/kg), Mn (1.60 ± 1.32 g/kg), Ni (104 ± 58 mg/kg), Sb (6.1 ± 2.9 mg/kg), and V (411 ± 270 mg/kg) are 2 to 20 times higher in ores than UCC average composition while Cd (0.09 ± 0.07 mg/kg), Pb (12 ± 10 mg/kg), Ti (5.85 ± 3.69 g/kg), and Zn (84 ± 54 mg/kg) presented similar concentrations (Table 1, Fig. 4). There is no significant difference between these element concentrations in raw (O1a–O4a) and processed ores (i.e., milled and washed O1b–O5b) (p values > 0.05 , Fig. 4). Raw ore material (O1a–O4a) exhibits an average Hg concentration of 0.061 ± 0.017 mg/kg ($n = 4$) while processed ores (O1b, O2b, and O5b) and cyanidation residues (CW1–CW14c) have higher total mercury content, from 0.89 to 8.32 mg/kg (Table 1). Sediments (EF1 s and EF2 s) collected in mine effluents also have a high Hg concentration although slightly lower (0.24 and 0.77 mg/kg).

The average concentrations of the majority of the metals and metalloids (Al, As, Cd, Co, Cr, Fe, Mn, Sb, and Zn) in cyanidation residues are not significantly different from the ores (p values > 0.05) with the exception of Hg, Pb, Cu, Ni, Ti, and V. Cyanidation residues are associated with higher concentration for Hg (1.93 ± 1.77 mg/kg) and Pb (44 ± 24 mg/kg) and lower average concentrations for Cu, Ni, Ti, V (p values < 0.05 , Fig. 4). The concentrations of three groups of metal(loid)s show a positive correlation with each other in ore and cyanidation residues (Spearman correlation coefficients $r^2 > 0.5$, p value < 0.001) and were (1) Al, As, Cr, Cu, Fe, Ni, Ti, V; (2) Co and Mn; and (3) Cd and Zn (Table S4).

The dissolved concentration of metal(loid)s in mining effluents are relatively low and comparable to typical concentrations found in rivers (Table 2). For example, dissolved concentrations of metal(loid)s in mining effluents (EF1w and EF2w) are 0.3–0.4 $\mu\text{g/L}$ for As, 0.05–0.20 $\mu\text{g/L}$ for Cr, 0.03–0.08 $\mu\text{g/L}$ for Pb, and 0.02–0.20 $\mu\text{g/L}$ for Sb indicating a low mobility of these elements from the solid to the aqueous phase under the field environmental conditions. Only

Zn in EF1w and Mn in EF2w samples have a relatively high concentration (51 $\mu\text{g/L}$ and 352 $\mu\text{g/L}$, respectively) but they remain below the WHO guidelines for drinking water.

Cyanide content

Cyanidation residues contained 1.9 to 100 mg/kg of total cyanides T-CN (47 ± 27 mg/kg), no trend was observed when comparing fresh and recent cyanidation residues (Table 1). However, it is noteworthy that the older (> 1 year) residue pile naturally recolonized by vegetation (CW14a to CW14c) generally present lower T-CN content compared with more recent residues. Within this residue pile, the most recent layer at the surface has a lower CN content (1.9 mg/kg, CW14a), compared to the intermediate layer at 1 m (14 mg/kg, CW14b) and the deeper layer at 2.5 m (27 mg/kg, CW14c). Extractable free cyanides (F-CN) concentrations are comprised between 0.20 and 7.65 mg/kg and generally represent less than 3% of T-CN except for the fresh cyanidation residues (CW13) and the surface of the older residue pile (CW14a) for which F-CN represented 10% (7.65 mg/kg) and 27% (0.51 mg/kg) of T-CN, respectively.

Impacts on environmental compartments

Groundwaters

Metal(loid) concentrations were determined in groundwater collected from wells used by the population in different villages (Fig. 1). Physicochemical parameters and dissolved (< 0.22 μm) element concentrations measured in groundwater samples are presented in Table 2. Groundwaters are warm (> 27.7 $^{\circ}\text{C}$), have a slightly acidic to slightly alkaline pH (5.37–8.01) and a conductivity ranging from 48 to 713 $\mu\text{S/cm}$. Overall, dissolved concentrations of metal(loid)s are below the potability guidelines established by the WHO (Al, As, B, Cd, Cr, Cu, Mn, Pb, Sb, and Zn, Table 2) except for one sample (GW3) which has a high Zn concentration of 3000 $\mu\text{g/L}$. Toxic metal(loid)s enriched in ore, waste, and/or sediment samples relative to UCC values (Fig. 4) show low concentrations in groundwaters below or similar to the world river average values, such as for As (0.03 to 2.45 $\mu\text{g/L}$), Cr (0.01 to 0.56 $\mu\text{g/L}$), Pb (0.01 to 0.09 $\mu\text{g/L}$), and Sb (0.03 to 0.09 $\mu\text{g/L}$). However, the dissolved Mn concentrations are relatively high compared to the world river average (up to 203 $\mu\text{g/L}$) but remain below the WHO guidelines for drinking water (400 $\mu\text{g/L}$).

Surface waters

Water samples were collected from different streams draining or not AGSM sites (extraction, ore processing,

Table 1 Major, trace element, and cyanide concentration (total cyanide T-CN and extractable cyanide F-CN) in solid samples (raw and processed alluvial or vein ores (R-ore and P-ore, respectively), cyanide residues (CW), mining effluent sediments, river sediments, and soils) of the Kokumbo area and reference values

Sample name	Al g/kg	Ca g/kg	Fe g/kg	K g/kg	Mg g/kg	Mn g/kg	Na g/kg	Ti g/kg	As mg/kg	Cd mg/kg	Co mg/kg
Potential contamination sources											
O1a	43.2	0.5	124	1.05	0.311	2.17	0.11	9.95	141	0.092	109
O2a	34.7	0.171	115	3.75	0.124	0.612	2.84	11.1	10.2	0.088	11.1
O3a	39.2	0.403	166	6.03	0.402	0.872	0.273	6.97	138	0.053	12.4
O4a	3.94	0.302	13.4	0.731	0.144	0.388	0.208	0.229	7.08	0.032	5.22
O1b	29.6	1.23	276	0.974	0.728	4.08	0.312	6.42	155	0.146	223.9
O2b	21.7	0.764	60.9	11	0.34	1.46	2.34	4.91	90.3	0.219	65.7
O3b	37.6	0.733	243	3.92	0.744	3.06	0.341	5	253	0.146	90.3
O4b	8.44	0.397	13.3	1.38	0.243	0.093	0.193	0.636	8.13	0.012	3.26
O5b	60.3	1.75	119.6	0.558	1.38	1.68	0.308	7.45	57	0.056	63.1
Ore material aver- age $\pm$ sd	31 $\pm$ 17.5	0.69 $\pm$ 0.51	126 $\pm$ 91.9	3.27 $\pm$ 3.47	0.49 $\pm$ 0.40	1.60 $\pm$ 1.32	0.77 $\pm$ 1.04	5.85 $\pm$ 3.69	95.6 $\pm$ 84.1	0.094 $\pm$ 0.066	64.9 $\pm$ 71.5
CW1	17.8	1.36	62.4	3.39	0.727	1.68	1.16	2.66	73.6	0.177	89.9
CW2	24.5	1.66	169	1.48	0.539	1.68	0.792	6.09	103	0.083	61.2
CW3	12.7	1.41	43.4	3.35	0.532	1.22	0.956	2.16	152	0.126	52.4
CW4	15.4	1.32	59.4	3.27	0.587	0.842	1.61	2.44	40.7	0.354	31.2
CW5	16.5	1.39	37.2	1.55	0.925	0.723	0.926	1.42	30.2	0.07	55.6
CW6	12.5	1.21	25.7	1.25	0.772	0.534	0.982	1.08	30.2	0.072	23.5
CW7	24.4	1.84	38.7	1.97	0.986	0.606	0.735	2.09	33.2	0.292	18.1
CW8	20.7	1.37	47.3	1.53	0.861	3.47	0.555	1.94	41.7	0.094	178
CW9	25.6	2.21	47	2.6	1.45	1.09	1.06	2.29	73.9	0.127	35.1
CW10	18.6	1.88	35.5	1.93	1.37	0.591	0.798	1.82	47.5	0.122	18.6
CW11	12.9	1.02	23.7	1.3	0.689	0.445	0.503	1.11	18.6	0.051	11.2
CW12	20.2	2.05	48	1.98	1.22	0.725	1.09	1.94	42.6	0.099	25.7
CW13	23.5	1.43	78.6	2.13	0.559	2.24	0.741	2.33	65	0.134	77.7
CW14a	27.1	4	106	2.57	0.933	7.91	0.336	2.69	80.2	0.535	107
CW14b	19.9	0.601	62.2	2.11	0.33	2.02	0.41	1.9	57.5	0.101	67.1
CW14c	22.4	1.56	69.4	2.24	0.549	2.51	0.732	2.35	69.9	0.109	94.4

Table 1 (continued)

[illegible]

Table 1 (continued)

Sample name	Cr mg/kg	Cu mg/kg	Hg mg/kg	Ni mg/kg	Pb mg/kg	Sb mg/kg	V mg/kg	Zn mg/kg	T-CN mg/kg	F-CN mg/kg(% TCN)	Sample type
Potential contamination sources											
O1a	296	183	0.061	153	6.49	4.48	483	94.4	n.a	n.a	R-ore (all.)
O2a	338	161	0.044	140	2.06	8.21	540	81.1	n.a	n.a	R-ore (all.)
O3a	388	134	0.077	80	22.6	7.42	648	73.3	n.a	n.a	R-ore (all.)
O4a	64.7	31.9	0.063	28.4	8.93	4.21	18.2	12.6	n.a	n.a	R-ore (vein)
O1b	811	262	2.47	189	19.1	7.35	676	181	n.a	n.a	P-ore (all.)
O2b	254	149	1.1	130	6.52	6.33	243	118	n.a	n.a	P-ore (all.)
O3b	571	218	0.1	107	31.3	11.7	727	123	n.a	n.a	P-ore (all.)
O4b	32.4	19.8	0.09	9.68	2.69	2.28	30.7	11	n.a	n.a	P-ore (vein)
O5b	419	94.6	1.02	99.4	8.44	3.37	328	62	< 0.1	n.a	P-ore (all.)
Ore mate- rial aver- age $\pm$ sd	353 $\pm$ 240	139 $\pm$ 80.3	0.558 $\pm$ 0.835	104 $\pm$ 57.9	12.0 $\pm$ 10.0	6.15 $\pm$ 2.89	411 $\pm$ 270	84.1 $\pm$ 53.8			
CW1	263	94	1.68	55.2	76.6	20.8	221	183	46	n.a	CW (fresh)
CW2	811	120	1.1	83.5	25.2	7.13	581	134	56	n.a	CW (fresh)
CW3	155	87.5	1.57	42	65.9	4.97	125	236	2	n.a	CW (fresh)
CW4	221	72	1.06	43.4	101	6.37	186	194	61	n.a	CW (fresh)
CW5	104	51.4	1.74	29.7	17.6	5.54	93.7	75.1	74	0.20 (0.3%)	CW (< 3 m)
CW6	63.1	38.6	1.18	20	15.4	4.41	64.4	58.9	100	0.75 (0.8%)	CW (< 3 m)
CW7	110	49.7	0.893	30	19.3	5.87	110	91.2	47	1.22 (2.6%)	CW (< 3 m)
CW8	145	89.5	2.14	42.2	37.8	6.12	158	78.7	47	0.78 (1.7%)	CW (< 3 m)
CW9	124	67.1	8.32	37.6	48.6	7.49	128	95.2	35	0.87 (2.5%)	CW (> 6 m)
CW10	91.6	59.8	2.87	25.7	56.8	12	97.8	70.6	63	0.76 (1.2%)	CW (> 6 m)
CW11	65.6	28.2	1.19	15.9	14.8	3.43	62.4	55.1	53	1.00 (1.9%)	CW (< 3 m)
CW12	146	63.9	1.17	31.8	26.3	7.56	127	84.8	45	0.85 (1.9%)	CW (< 3 m)
CW13	207	82.1	1.39	50.7	57.1	8.28	183	107	80	7.65 (9.6%)	CW (fresh)
CW14a	287	98.1	1.24	62.2	50.5	5.83	248	143	1.9	0.51 (26.8%)	CW (> 1 y)
CW14b	137	69.8	1.76	44.9	41.2	6.56	137	98.4	14	0.77 (5.5%)	CW (> 1 y)
CW14c	187	87.5	1.51	57.3	43.9	8.48	177	113	27	0.50 (1.9%)	CW (> 1 y)
Cyanidation residue avg $\pm$ sd	195 $\pm$ 177	72.4 $\pm$ 23.8	1.93 $\pm$ 1.77	42 $\pm$ 17.3	43.6 $\pm$ 24.3	7.55 $\pm$ 4.04	169 $\pm$ 122	114 $\pm$ 51.9	47.0 $\pm$ 26.9	n.a	
EF1s	442	64.4	0.773	39.3	16.1	3.97	279	80.8	n.a	n.a	Mining effl
EF2s	286	172	0.237	149	5.08	7.37	325	98.7	n.a	n.a	Mining effl

Table 1 (continued)

Sample name	Cr mg/kg	Cu mg/kg	Hg mg/kg	Ni mg/kg	Pb mg/kg	Sb mg/kg	V mg/kg	Zn mg/kg	T-CN mg/kg	F-CN mg/kg(% TCN)	Sample type
Potential impacted environmental compartment											
S1	610	116	1.08	72.7	26	7.53	556	75.5	n.a	n.a	Soil
S2	164	24.5	0.473	23.6	14.1	1.59	133	16.9	n.a	n.a	Soil
S3	184	66.1	0.693	66.6	9.11	1.94	239	52.7	2.9	2.05 (70.7%)	Soil
S4	81	29.5	1.08	15	45.4	11.6	62.8	101	49	0.57 (1.2%)	Soil
SW3 s	573	90.5	20.4	60.3	20.4	5.2	465	77.9	0.23	n.a	River sed
SW1s	599	100	7.8	62.8	26.9	6.76	390	84.5	n.a	n.a	River sed
SW2s	792	121	9.89	72.6	23.2	5.42	649	66.3	n.a	n.a	River sed
SW4s	599	120	2.45	86.7	20.3	4.6	474	90.8	n.a	n.a	River sed
SW5s	1268	207	0.083	151	34.7	9.2	964	146	n.a	n.a	River sed
SW7s	647	68.6	0.474	44.6	13.8	3.31	563	29.8	n.a	n.a	River sed
SW8s	1086	72.9	0.557	38.2	15.2	6.26	601	62.6	n.a	n.a	River sed
SW9s	433	49.7	1.19	46.8	17.6	5.55	328	29.8	n.a	n.a	River sed
SW10s	631	168	0.957	136	13.8	13	721	83.8	n.a	n.a	River sed
SW11s	209	47.3	4.51	49.6	21.3	1.69	130	154	n.a	n.a	River sed
SW12s	520	159	0.587	113	30	7.17	547	115	n.a	n.a	River sed
Soil and sediment avg ± sd	560 ± 329	95.9 ± 52.9	3.48 ± 5.52	69.3 ± 38.8	22.1 ± 9.45	6.06 ± 3.39	455 ± 246	79.1 ± 39.7	n.a	n.a	River sed
Reference values											
UCC	92	28	0.05	47	17	0.4	97	67	n.a	n.a	
World average ¹	130	75.9	n.a	74.5	61.1	2.19	129	208	n.a	n.a	
SPM ²	130	53	n.a	78	46	n.a	116	130	n.a	n.a	
African SPM ²	90	197	0.5	–	91	–	–	315	–	–	
SQG ³											

n.a. data not available

¹Rudnick and Gao 2014: Average composition of the Upper Continental Crust (UCC)²Viers et al. 2009: Average composition of World and African river suspended particulate matter (SPM)³CCME, 1999: Sediment Quality Guidelines (SQG) for the protection of aquatic life, probable effect levels (PEL) data for freshwater sediments

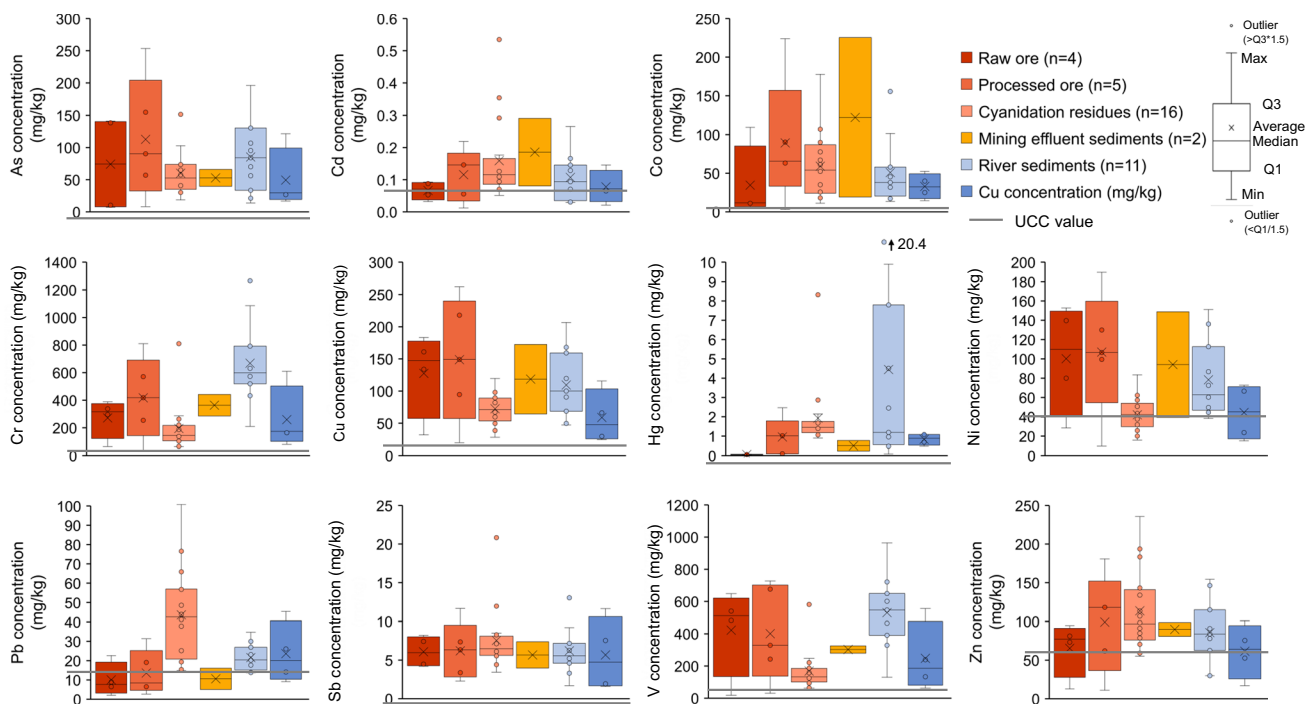


Fig. 4 Comparison of metal (Cd, Co, Cr, Cu, Hg, Ni, Pb, V, Zn) and metalloid (As, Sb) concentration distributions in raw and processed ores, cyanidation residues, mining effluent sediments, river sediments

and soils of the Kokumbo area. Q1 stands for first quartile, and Q3 for the third quartile

cyanidation) to investigate a possible mobilization of metals in a dissolved form from ASGM site toward the aquatic environment in Kokumbo. Surface waters are all warm (25.6–31.7 °C), with a near neutral pH (7.3 ± 0.4) and a conductivity (124–622 $\mu\text{S}/\text{cm}$) similar to those of groundwaters (Table 2). The dissolved metal(loid)s in surface waters show similar concentrations ranges compared to groundwaters for As (0.3–2.4 $\mu\text{g}/\text{L}$), Cd (0.001–0.08 $\mu\text{g}/\text{L}$), Pb (0.02–0.19, excluding SW5w) or Sb (0.02–0.14 $\mu\text{g}/\text{L}$) but higher concentrations of Al (3.4–3800 $\mu\text{g}/\text{L}$), Fe (25–3190 $\mu\text{g}/\text{L}$), Mn (15–1753 $\mu\text{g}/\text{L}$). SW5w sample, which corresponds to a stream downstream from a cyanide treatment site flowing through an abandoned cocoa plantation, shows higher metal concentrations including Al, Co, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn compared to other samples. The metal concentrations of surface water in this study are all below WHO guidelines for drinking waters except for Mn in half of surface water samples and Al in SW5w sample.

Sediments and soils

Sediments were collected in streams, along with water samples, to characterize the possible dispersion of metal-bearing particles in the Kokumbo area due to ASGM activities. The mineralogy obtained by XRD analysis of river sediments is similar to that of ores and cyanidation residues, including quartz, kaolinite,

hematite, goethite, with variable presence of muscovite, titanium oxides (anatase and rutile), and aluminum hydroxides (gibbsite) and other accessories minerals such as albite and microcline (Table S3).

The average concentrations of metals and metalloids are not statistically different between soil and sediment samples except for Cr and V which are slightly but significantly higher in sediments than in soils (p value < 0.05). Similarly, metal(loid) concentrations are not different on average in soils and sediments when compared to the ores except for Pb and Hg (p value < 0.02), which exhibit higher concentrations (22 ± 9 mg/kg for Pb and 3.5 ± 5.5 mg/kg for Hg) closer to the concentrations measured in cyanidation residues (Table 1, Fig. 4).

The enrichment factors of metals and metalloids in soils and sediments were first calculated using the UCC composition as reference to highlight a possible local geogenic enrichment, from natural release of metals present in the ore, and/or from an enhanced release of these metals as a result of ore processing (geogenic source, but anthropic origin) (Table S5). Enrichment factors showed a moderate to high enrichment in As ($EF_{\text{As/UCC}} = 5.5\text{--}110$), Cd ($EF_{\text{Cd/UCC}} = 0.6\text{--}12$), Co ($EF_{\text{Co/UCC}} = 1.7\text{--}23$), Cr ($EF_{\text{Cr/UCC}} = 2.8\text{--}37$), Cu ($EF_{\text{Cu/UCC}} = 3.3\text{--}22$), Hg ($EF_{\text{Hg/UCC}} = 4.5\text{--}1206$), Mn ($EF_{\text{Mn/UCC}} = 0.8\text{--}12$), Ni ($EF_{\text{Ni/UCC}} = 1.8\text{--}10$), Pb ($EF_{\text{Pb/UCC}} = 0.7\text{--}20$), Sb ($EF_{\text{Sb/UCC}} = 6.7\text{--}214$), Ti

Table 2 Physicochemical parameters, major cation and trace element concentrations in water samples (surface waters, groundwaters, and mining effluents) of the Kokumbo area and reference values (average composition of World and African rivers from (Gaillardet et al. 2003) and WHO guidelines for drinking water (World Health Organization 2017))

Sample name	pH	Cond µS/cm	Temp °C	Ca mg/L	K mg/L	Mg mg/L	Na mg/L	Si mg/L	Al µg/L	As µg/L	B µg/L	Cd µg/L
Surface waters												
SW1w	7.2	362	28.1	17.5	1.07	8.9	10.2	8.29	4.53	2.17	9.01	0.002
SW2w	7.7	377	29	17	1.32	8.37	12	8.36	4.64	2.36	8.22	0.003
SW3w	n.a	n.a	n.a	19.3	1	9.8	8.83	8.24	9.7	2.07	7.87	0.001
SW4w	7.37	286	27.6	13.7	8.43	4.7	6.44	4.59	45.7	1.14	16.8	0.004
SW5w	7.31	414	29.2	26	5.23	13.4	10.6	11.4	3800	0.745	14.2	0.083
SW6w	n.a	n.a	n.a	18.3	7.67	6.28	11.7	4.96	47.3	0.925	17.2	0.001
SW7w	6.94	348	25.6	16.9	7.41	4.49	14.2	10.3	65.4	0.641	20	0.002
SW8w	6.69	163	28.8	7.36	1.05	3.65	4.5	6.43	12.3	1	9.97	<0.001*
SW9w	7.28	277	31.7	15.5	3.46	5.96	5.58	8.22	3.36	0.921	15.1	<0.001*
SW10w	6.88	277	30.7	15.3	3.43	5.82	5.35	8.17	7.63	0.922	14.9	0.001
SW11w	7.78	181	28.2	9.12	4.38	2.99	6.52	8.15	71.9	0.354	10.4	0.001
SW12w	7.76	190	28.4	9.54	4.52	3.01	6.42	8.27	116	0.329	10.8	0.001
Avg ± sd	7.3 ± 0.4	287 ± 88	28.7 ± 1.7	15.5 ± 5.1	4.1 ± 2.7	6.4 ± 3.1	8.5 ± 3.2	7.9 ± 1.9	349 ± 1087	1.1 ± 0.7	12.9 ± 4	0.01 ± 0.03
Groundwaters												
GW1	5.37	578	28.2	13.1	17.6	4.99	22.5	5.69	35.8	0.195	3.69	0.09
GW2	5.48	48	28.3	2.25	0.16	0.6	1.37	4.69	16.5	0.026	5.37	0.005
GW3	5.38	346	28.7	4.73	4.55	1.95	21.56	4.95	6.92	0.05	4.1	0.019
GW4	5.95	104	29	3.53	3.13	1.35	6.17	4.53	1.23	0.113	12.4	0.004
GW5	8.01	310	29	24.5	2.24	1.36	6.37	5.96	65.3	0.733	12.4	<0.001*
GW6	7.35	713	29.6	41.5	0.49	14.4	24.8	24	0.432	2.45	10.7	0.037
GW7	6.42	501	27.7	27.3	0.34	12.5	10.9	9.9	2.09	0.025	13.7	0.002
GW8	6.89	622	28.1	37.6	0.31	16.3	10.1	12.7	0.333	0.451	8.54	0.002
Avg ± sd	6.4 ± 1	403 ± 243	28.6 ± 0.6	19.3 ± 15.6	3.6 ± 5.9	6.7 ± 6.6	13 ± 8.8	9.1 ± 6.7	16.1 ± 23.3	0.51 ± 0.82	8.9 ± 4	0.02 ± 0.03
Mining effluents												
EF1w	7.01	256	30.9	14.3	2.99	4.55	5.14	5.05	25.9	0.308	13.1	0.002
EF2w	5.38	124	31	1.89	3.36	0.97	5.31	7.39	14.4	0.352	26	0.016
Reference values												
World river average ¹	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	32	0.62	10.2	0.08
African rivers ¹ (min–max)	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	12–480	0.11–0.17	3.1–3.2	n.a
WHO guideline for drinking water ²									900	10	2400	3

Table 2 (continued)

Sample name	Co	Cr	Cu	Fe	Mn	Ni	Pb	Sb	Ti	V	Zn
	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L
Surface waters											
SW1w	1.55	0.25	0.581	415	744	0.826	0.035	0.084	0.07	0.462	0.614
SW2w	1.51	0.415	0.764	223	764	0.936	0.038	0.144	0.049	0.41	1.62
SW3w	1.3	0.187	0.366	838	686	0.831	0.064	0.058	0.182	0.479	0.888
SW4w	1.05	0.223	1.78	100	239	0.982	0.111	0.124	0.198	0.875	4.02
SW5w	65.3	9.79	31.8	3190	1753	34.9	5.29	0.032	0.356	4.93	45.5
SW6w	0.365	0.106	1.09	25	66.4	0.429	0.042	0.124	0.493	1.14	0.39
SW7w	0.344	0.374	1.31	279	22.7	1.02	0.083	0.045	0.89	1.97	1.51
SW8w	2.12	0.944	0.168	1481	241	0.759	0.016	0.023	0.258	0.288	0.358
SW9w	3.4	0.365	0.123	1076	670	1.17	0.029	0.043	0.082	0.186	0.57
SW10w	3.27	0.371	0.172	1077	627	1.25	0.019	0.044	0.149	0.265	1.69
SW11w	0.156	0.216	1.42	153	15.8	0.798	0.099	0.035	0.702	1.98	0.548
SW12w	0.157	0.28	1.41	337	15.1	0.871	0.191	0.026	1.09	2.17	0.967
Avg ± sd	6.7 ± 18.5	1.1 ± 2.7	3.4 ± 9	766 ± 894	487 ± 504	3.7 ± 9.8	0.5 ± 1.5	0.07 ± 0.04	0.4 ± 0.3	1.3 ± 1.4	4.9 ± 12.8
Groundwaters											
GW1	10.8	0.064	9.18	3.83	203	24.3	0.038	0.027	< 0.02*	0.168	30.6
GW2	1.21	0.563	0.379	22.3	181	0.99	0.015	0.007	0.13	0.126	6.66
GW3	5.87	0.01	1.17	20.9	86.1	2.85	0.039	0.012	< 0.02*	0.104	3024
GW4	1.01	0.131	0.485	35	35.1	0.947	0.023	0.015	< 0.02*	0.057	4.64
GW5	0.164	0.451	0.522	5.71	30.6	0.299	0.02	0.09	0.24	4.01	3.43
GW6	0.025	0.104	0.532	99	5.67	0.059	0.09	0.071	0.047	6.31	415
GW7	0.39	0.009	0.095	1.28	24	0.528	0.01	0.009	0.026	0.32	1.29
GW8	0.012	0.032	0.316	< 0.7*	0.824	0.311	0.017	0.003	0.027	1.1	16.4
Avg ± sd	2.4 ± 3.9	0.17 ± 0.21	1.6 ± 3.1	26.9 ± 34.1	70.8 ± 79.3	3.8 ± 8.3	0.03 ± 0.03	0.03 ± 0.03	0.09 ± 0.09	1.5 ± 2.4	438 ± 1054
Mining effluents											
EF1w	0.402	0.2	1.53	138	26.3	0.7	0.079	0.194	0.095	0.948	50.7
EF2w	2.15	0.046	0.589	5.08	352	1.33	0.025	0.023	< 0.02*	0.121	2.39
Reference values											
World river average ¹	0.148	0.7	1.48	66	34	0.8	0.079	0.07	0.489	0.71	0.6
African rivers ¹ (min–max)	0.04–0.43	0.38–0.53	0.63–2.03	31–614	0.44–29.7	0.29–5.04	0.04	n.a	0.23–5.81	0.59–0.65	0.04–0.59
WHO guideline for drinking water ²		50	2000		400	70	10	20			3000

¹ Gaillardet et al. 2003; ² World Health Organization 2017, * < detection limit; n.a. not available

($EF_{Ti/UCC} = 1.8\text{--}10$), V ($EF_{V/UCC} = 3.4\text{--}27$), and Zn ($EF_{Zn/UCC} = 0.9\text{--}11$). Then, enrichment factors were calculated using the average composition of local alluvial ore material to identify a possible enrichment with exogenous metals brought by the Au recovery processes (amalgamation, cyanidation) and distinguish between a geogenic vs exogenous origin of metals. When compared to the average value of alluvial ore material, only Hg ($EF_{Hg/ore} = 1.7\text{--}459$), Pb ($EF_{Pb/ore} = 0.4\text{--}11$) (Fig. S2), and in a lesser extent Cr ($EF_{Cr/ore}$ up to 4), Cd ($EF_{Cd/ore}$ up to 4.4), Sb ($EF_{Sb/ore}$ up to 5.7), and Zn ($EF_{Zn/ore}$ up to 3.3) show a significant enrichment in some samples of soils and sediments. The vegetated soil S4 located in a cyanidation area presented a moderate enrichment in Cd ($EF_{Cd/ore} = 4.4$), Sb ($EF_{Sb/ore} = 5.7$), Pb ($EF_{Pb/ore} = 11.3$), and Zn ($EF_{Zn/ore} = 3.3$). Finally, the sample SW5s corresponding to the water sample SW5w draining the cyanidation area of Akoukro shows a slight enrichment in As, Cd, Cr, Sb, V, and Zn ($EF_{metal/ore}$ from 1.6 to 3.6).

Total and extractable cyanides (T-CN and F-CN) were measured in the two soils sampled in cyanidation areas of Akoukro (samples S3 and S4, Table 1). The soil S4 has a high content in T-CN of 49 mg/kg of which only 1.2% are in an extractable form, it was covered by vegetation close to a cyanidation site. The soil S3 was sampled in a cultivated field next to the cyanidation site and has a lower T-CN concentration of 2.9 mg/kg but 70% of T-CN was present as extractable F-CN (2.05 mg/kg).

Discussion

Origin of metal(loid)s in the Kokumbo area

A principal component analysis (PCA) was performed with metal concentrations (Al, Fe, Mn, Ti, As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Sb, V, and Zn) in solid samples (ores, cyanidation residues, soils and sediments) and allowed to identify three main components (PC1, PC2, and PC3) accounting 80% of the variance of the dataset. PC1, PC2, and PC3 explained 53.5%, 18.2%, and 8.3% of the dataset variance, respectively.

PC1 is associated to a group of metals identified as likely being of geogenic origin: Al, As, Co, Cr, Cu, Fe, Mn, Ni, Ti, and V (Fig. 5(a)). Indeed, the concentration of these metals did not significantly differ in cyanidation residues compared to ore materials. Furthermore, enrichment factors for these elements with respect to the alluvial ore material ($EF_{metal/ore}$) do not show an enrichment in soils and sediments, except Cr in a few samples (Table S5). PC2 is associated, by order of importance, with Pb, Cd, Zn, Hg, and Sb (Fig. 5(a)) for which an exogenous source is suspected due to the higher concentrations found in

cyanidation residues than in ore materials, and to the enrichment observed in soils and sediments for Hg and Pb, and to a lesser extent for Cd, Sb and Zn (Fig. S2). PC3 is mainly associated with Hg.

Three endmembers are revealed in a PC1-PC2 biplot corresponding to (1) alluvial ores with a high score on PC1, (2) cyanidation residues with a high score on PC2, and (3) vein ores with low scores on PC1 and PC2 (Fig. 5(b)). This is consistent with the findings that most metals and metalloids have a predominantly geogenic origin, while Hg, Pb (and Cd, Sb, and Zn) have an exogenous source in direct or indirect relation with amalgamation and cyanidation processes.

In many ASGM-impacted context, Pb contamination in soil and sediments has been found and ascribed to the presence of Pb minerals such as galena in association with gold mineralization (Fonshiyinwa et al. 2024; Plumlee et al. 2013; Tabelin et al. 2020). Galena was not observed in Kokumbo ore samples and a slight enrichment in cyanidation residues and in soils and sediments was observed compared to ore material suggesting that an exogenous source is involved (Fig. S2). The source of Pb, Cd, and Sb might be related to the operation of engines on ore processing sites as they can be released by unleaded gasoline, diesel and engine oil combustion (Gupta 2020; Yang et al. 2019), but identifying the origin of these metal(loid)s which are not directly used in the Au recovery process requires further research. Interestingly, despite the fact that metal Zn is used in the Au recovery process after cyanidation, Zn was not present in high concentrations in cyanidation residues, nor was it enriched in most sediments and soils (except a moderate enrichment in a soil from the cyanidation area and in sediments of the Bandama River). This suggests that the release and dispersion of Zn through this mining practice is limited in the Kokumbo area. Nevertheless, the contribution of Zn in PC2 highlights an impact of cyanidation process on Zn concentration although moderate. This contrasts with other ASGM areas where important Zn contamination in cyanidation effluents, wastes, and soils have been found such as in Niger (Tankari Dan-Badjo et al. 2019).

PCA results for most sediments are positioned between the cyanidation residue and alluvial ore endmembers suggesting a combined impact of both types of ASGM sites (extraction areas and cyanidation areas) on streams in the Kokumbo area. The vein ores (O4a/b) constituted a third endmember with low scores on PC1 and PC2 (Fig. 5(b)) due to their lower metal concentration compared to alluvial ores, likely related to their different nature (quartz vein). The PCA result of the superficial soil S2 collected on the vein ore processing sites was detached from the other samples (Fig. 5(b)) and may reflect the influence of dust emitted by vein ore crushing (Fig. 2(I, J, K)). The PCA result of the soil S4, collected next to a cyanidation site, stood close to

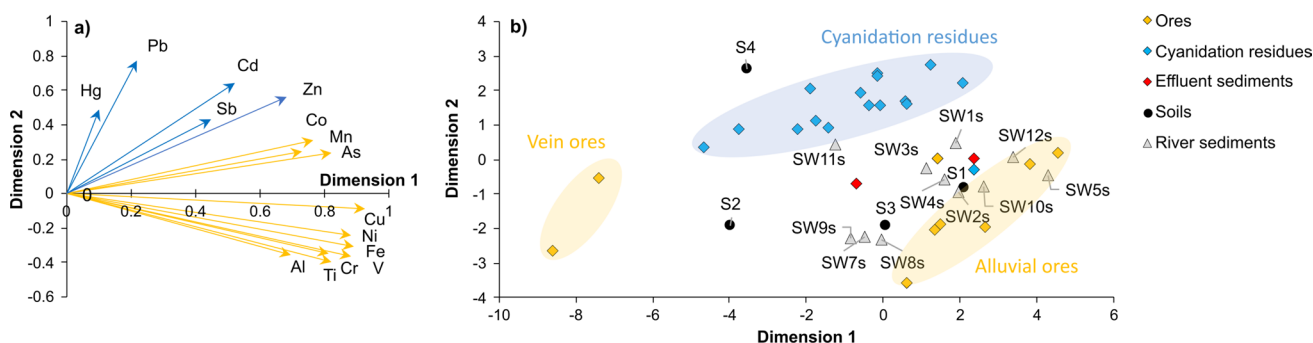


Fig. 5 PCA results representing (a) variables (i.e., metals and metalloids) and (b) observations (i.e., all solid samples) within the first two principal components PC1 (dimension 1) and PC2 (dimension 2)

the cyanidation residue end-member (Fig. 5(b)). This result, together with the high T-CN content, suggests that it corresponds to an old revegetated cyanidation residue, reflecting the capacity of the vegetation to recolonize these wastes.

The highest concentration of Hg (up to 20.4 mg/kg) and enrichment factor (up to 1206) are found in the Balo pond sediments (SW1s- 3s) suggesting that Hg amalgamation occurs in its vicinity despite a ban on gold-panning activities in the surrounding area or it could be a historic contamination inherited from former mining activities. In comparison, the raw ore material exhibits a low average Hg concentration (0.061 ± 0.017 mg/kg) typical of the sediment geochemical background encountered in Côte d'Ivoire (< 0.010 mg/kg according to Mason et al. 2019) and in West Africa (e.g., 0.030 ± 0.006 mg/kg in Senegal, Niane et al. 2019; 0.015 ± 0.007 mg/kg in Mali, Bokar et al. 2020). On the other hand, processed ore material and cyanidation residues show high levels of Hg (1.93 ± 1.77 mg/kg) confirming that large amounts of Hg are lost during the amalgamation process at the processing sites. ASGM activities are therefore the main source of Hg environmental contamination in Kokumbo.

Mercury contamination is not confined to the streams around Kokumbo but seems to reach the Bandama River as elevated Hg concentrations (0.587 and 4.51 mg/kg) and enrichment factors (11 and 150) were measured in its sediments (SW11s and SW12s, Fig. S2). Considering the recent expansion of artisanal mining activities along the Bandama River (Ngom et al. 2022b), the contamination of the river with Hg could represent a threat for local inhabitants due to a possible contamination of fishes. It is of note that the fish consumption in Côte d'Ivoire accounts for around 42% of the animal protein in the population diet (Mason et al. 2022). In other ASGM impacted rivers of Côte d'Ivoire presenting lower total Hg levels in sediments, Mason et al. (2019) evidenced MeHg (methylmercury) levels in fishes whose consumption is considered a risk to human health, with 14% of tested fish exceeding the United States Environmental Protection Agency reference value. In the Gambia River,

Niane et al. (2019) demonstrated that while the concentrations of total Hg in sediments decreased with increasing distance from the ASGM sites, those of MeHg increased due to more favorable conditions for the methylation process downstream. Considering the high level of total Hg measured in this study, an in-depth study of Hg transport and speciation in the Bandama River would be required to evaluate the risk of bioaccumulation and biomagnification of MeHg along the river trophic chain.

Metal concentrations in soils and sediments in the Kokumbo area compared to other ASGM regions in West Africa

As a general rule, metal and metalloid concentrations in soils and sediments affected by ASGM can be highly variable within and between areas due to many parameters at stake (e.g., proximity of the mining sites, intensity and age of the activity, nature of the mineralization exploited and of the bedrock, and mining practices) (e.g., El Farouk et al. 2022, Kinimo et al. 2018a, b, Niane et al. 2019, Ondo et al. 2023, Sako and Nimi 2018, Tankari Dan-Badjo et al. 2019). Nevertheless, the average concentrations of Al, Fe, Cd, Ni, and Zn measured in soils and sediments in Kokumbo are in the same range of values as those reported by other studies in the West African countries part of the Birimian Greenstone Belt (Table 3). It is notable that the average concentrations of Cr, Cu, and Pb in sediments at Kokumbo are the higher compared to values reported elsewhere in Côte d'Ivoire, Niger, Ghana, Burkina Faso, and Mali (Table 3). Manganese content in Kokumbo soils and sediments are some of the highest reported in Table 3 ($0.3\text{--}3.7$ g/kg) but similar to other ASGM areas in Burkina Faso (1.35 ± 1.37 g/kg at Kalsaka, Dabiré and Sako 2024) or in Ghana (1.17 ± 0.02 g/kg in the Birim River, Hadzi et al. 2024). The presence of Mn ore deposits in the volcano-sedimentary belts of the West African craton has been evidenced in Mali, Ghana, Burkina Faso and Côte d'Ivoire (Hein and Tshibubudze 2016)

and four Mn-deposits are currently being exploited in Côte d'Ivoire. Arsenic concentrations in sediments at Kokumbo (85.3 ± 54.7 mg/kg) were also among the highest reported in West Africa and were comparable to those of stream sediments impacted by industrial gold mines such as the Tongon mine in Côte d'Ivoire (125 ± 99 mg/kg, Sako et al. 2018) or the Morila mine in Mali (65.5 ± 65.6 mg/kg, Bokar et al. 2020) (Table 3). The high concentrations of As and Cu in Kokumbo is consistent with the petrographic observations of arsenopyrite and chalcopyrite in the ore rock samples (R2–R6) representative of the green schist facies.

At Kokumbo, total Hg concentrations are high in soils (0.83 ± 0.30 mg/kg) and especially in river and pond sediments (4.45 ± 6.22 mg/kg) compared to exploited alluvial ore material (0.061 ± 0.017 mg/kg). These concentrations are much higher than the values (0.035 ± 0.041 mg/kg) determined in rivers and ponds sediments collected near ASGM sites in four other regions of Côte d'Ivoire (Table 3, Mason et al. 2019) but comparable to those measured at an artisanal mining site along the Gambia River in Senegal (1.16 ± 0.8 mg/kg in river sediments and 3.27 ± 3.02 mg/kg in pond sediments (Niane et al. 2019) or in soils (median 0.68 mg/kg) and sediments (median of 0.82 mg/kg) close to ASGM-impacted villages in the South-West of Senegal (Gerson et al. 2018).

Even though most metals showed an enrichment relative to the average composition of the UCC, their levels remain moderate and on the lower end of the ranges reported by Ondayo et al. (2023) for As, Cd, Cr, Cu, Ni, and Pb in soils and sediments affected by ASGM across Africa. The levels of Cd, Cu, Pb, and Zn are also lower than the Canadian freshwater sediment quality guidelines (SQG in Table 1, CCME 1999, respectively 3.5, 197, 91, and 315 mg/kg). However, As, Cr, and Hg concentrations are significantly higher than the SQG (respectively 17, 90, and 0.5 mg/kg), suggesting a possible risk for the environment and human health, although the impact is yet to be assessed.

Cyanides persistence in mining residues

At Kokumbo, cyanidation residues contain relatively high concentrations of cyanides, from 1.9 to 100 mg/kg of total cyanides (T-CN) but generally less than 3% are in the form of free cyanides (F-CN), the most mobile and toxic form. The F-CN content in cyanidation residues (from 0.20 to 7.65 mg/kg), is in the range of F-CN detected in soils (0.15 to 13 mg/kg) around two cyanidation areas in Burkina Faso (Razanamahandry et al. 2018). The guidelines for cyanide in soils vary according to the land use, the cyanide form (T-CN, F-CN...), and the country legislations but they are generally between 50 and 500 mg/kg for T-CN and from 0.9 to 100 mg/kg for F-CN (Dzombak et al. 2005). Only seven and three out of the sixteen residues would exceed

the most stringent guideline value for T-CN and F-CN in soils, respectively. T-CN concentration seems to decrease with time as the older residue (> 1 year) shows the lowest concentration, indicating that natural abiotic and/or biotic processes of CN degradation are at work. However, T-CN showed a certain persistence, with no significant differences in T-CN content between fresh, < 3 months and > 6 months residues. The fresh residues present the highest concentration of F-CN (7.65 mg/kg, 9.6% of T-CN). This suggests that F-CN species are rapidly leached and/or degraded in the bare residue piles. The older residue naturally recolonized by vegetation presents a lower T-CN content compared with more recent residues but a higher proportion of F-CN (26.8%). The cultivated soil adjacent to the cyanidation site (S3) has a relatively high cyanide content (2.90 mg/kg) and shows the highest proportion of F-CN (70.7%). These higher proportions of F-CN in vegetated cyanidation residue and soil suggest that plant activity (e.g., root chelation) or cyanide-degrading microorganisms in the rhizosphere (Kumar et al. 2017) could favor CN bioavailability. These preliminary results call for a detailed study on the presence and speciation of CN in soils cultivated near the cyanidation sites and on the role of plant and microorganism in its degradation.

Potential metal mobility toward the surface waters and groundwaters

Dissolved metal (Al, B, Cd, Cr, Cu, Mn, Ni, Pb, and Zn) and metalloid (As and Sb) concentrations are below WHO guidelines in all groundwaters collected from wells used for drinking water supply (Table 2), except for Zn in one sample (GW3). This well is located in a village and had a slightly acid pH; this isolated high level of Zn (3024 μ g/L) might be related to the use of Zn material in the well construction. Arsenic has a high geochemical background in soils and sediments of Kokumbo but dissolved concentrations in groundwater are all below 2.5 μ g/L indicating that As is not mobile to the aqueous phase under the physicochemical conditions of the investigated wells. Studies of As spatial distribution in groundwaters within the Birimian formation in Burkina Faso have evidenced a high degree of spatial variability, even at the very local scale (< 100 m) with As contaminated wells located close to uncontaminated wells (Bretzler et al. 2017, 2019; Smedley et al. 2007) while surface water from 10 reservoirs had low As concentrations (0.51 ± 0.16 μ g/L) (Ouédraogo and Amyot 2013). The presence of high As levels in groundwater in Burkina Faso was related to the proximity of gold mineralization containing As-rich pyrite and arsenopyrite. Sulfide minerals, including arsenopyrite, have been observed in Kokumbo mineralization, so the presence of contaminated wells in the area cannot be ruled out and would require vigilance. The concentrations

Table 3 Comparative data of metal(loid)s concentrations in river sediments and soils from gold mining areas in West African countries forming part of the Birimian Greenstone Belt (average value $\pm$ standard deviation and/or (minimum–maximum) are reported depending on data availability)

Sample name	Context	Country	Al g/kg	Fe g/kg	Mn g/kg	As mg/kg	Cd mg/kg	Cr mg/kg	Cu mg/kg	Hg μ g/kg	Ni mg/kg	Pb mg/kg	Zn mg/kg	Reference
Soils ($n = 4$)	AGSM area (Kokumbo)	Côte d'Ivoire	32.8 $\pm$ 22.4 (11.1–59.0)	83.7 $\pm$ 79.8 (23.1–200)	1.4 $\pm$ 1.2 (0.4–3.2)	49.3 $\pm$ 48.5 (16.7–122)	0.07 $\pm$ 0.05 (0.02–0.15)	259 $\pm$ 237 (81–610)	58.9 $\pm$ 42.0 (24.5–116)	832 $\pm$ 301 (473–1085)	44.4 $\pm$ 29.3 (15.0–72.7)	23.6 $\pm$ 16.1 (9.1–45.4)	61.4 $\pm$ 35.6 (16.9–101)	This study
River sediments ($n = 11$)			30.8 $\pm$ 8.7	152 $\pm$ 64.8	1.3 $\pm$ 1.0	85.3 $\pm$ 54.7	0.10 $\pm$ 0.07	668 $\pm$ 292	109 $\pm$ 51.4	4446 $\pm$ 6224	78.3 $\pm$ 38.8	21.5 $\pm$ 6.7	85.5 $\pm$ 40.6	
Soils ($n = 8$)	Industrial mine area (Tongon)	Côte d'Ivoire	(18.6–42.5)	(64.8–320)	(0.3–3.7)	(13.8–196)	(0.03–0.27)	(210–1268)	(47.3–207)	(83–20410)	(38.2–151)	(13.8–34.7)	(29.8–154.4)	
			–	–	–	125 $\pm$ 99	–	–	44.8 $\pm$ 18.2	16 $\pm$ 9	23.6 $\pm$ 15.6	15.2 $\pm$ 4.8	21.0 $\pm$ 13.2	Sako et al. (2018)
			–	–	–	(11.3–279)	–	–	(23.9–77.3)	(< LD–30)	(9.8–53.3)	(8.7–22.7)	(8.0–45)	
River sediments ($n = 5$)	AGSM area (Afema)	Côte d'Ivoire	63.5 $\pm$ 8.5	33.1 $\pm$ 14	0.18 $\pm$ 0.16	5.1 $\pm$ 4.3	0.48 $\pm$ 0.27	151 $\pm$ 52	20.2 $\pm$ 4.6	–	40.1 $\pm$ 12.3	3.2 $\pm$ 1.5	55.4 $\pm$ 12.8	Kinimo et al. (2018a)
River sediments ($n = 5$)	AGSM area (Agbaou)		31.7 $\pm$ 0.4	40.4 $\pm$ 1.4	6.3 $\pm$ 0.1	< LD	0.48 $\pm$ 0.15	122 $\pm$ 3.9	34.1 $\pm$ 1.1	–	66.4 $\pm$ 2.2	3.4 $\pm$ 0.6	48.6 $\pm$ 1.3	
River sediments ($n = 5$)	AGSM area (Bonikro)		36.1 $\pm$ 18.5	29.3 $\pm$ 4.6	0.3 $\pm$ 0.1	< LD	0.54 $\pm$ 0.19	34.1 $\pm$ 15.9	21.6 $\pm$ 11	–	19.5 $\pm$ 6.7	7.7 $\pm$ 5.4	75.9 $\pm$ 32.3	
River sediments ($n = 30$)	Industrial and AGSM area (Tengrela and Korhogo)	Côte d'Ivoire	37.3 $\pm$ 26.3 (2.7–94.2)	41.1 $\pm$ 19.8 (8.3–82.2)	0.82 $\pm$ 0.85 (0.14–4.38)	–	–	–	29.1 $\pm$ 26.1 (2.0–127)	–	27.8 $\pm$ 21.9 (2.0–97)	–	–	N'goran et al. (2022)
River sediments ($n = 13$)	Four AGSM and industrial areas	Côte d'Ivoire	–	–	–	–	–	–	–	35 $\pm$ 41 (2.4–147)	–	–	–	Mason et al. (2019)
River sediments ($n = 11$)	AGSM area (Nigerien Liptako)	Niger	24.9 $\pm$ 24.0	0.38 $\pm$ 0.36	5.50 $\pm$ 5.96	0.034 $\pm$ 0.017	–	–	15.0 $\pm$ 11.2	–	18.2 $\pm$ 13.5	8.06 $\pm$ 4.18	26.7 $\pm$ 19.4	El Farouk et al. (2022)
Soils ($n = 117$)	AGSM area (Kalsaka)	Burkina Faso	–	–	–	(5.5–81.8)	(0.06–1.29)	177 $\pm$ 197	(3.56–37.1)	–	(3.52–48.1)	(2.17–15.6)	39.8 $\pm$ 31.3	Dabiré and Sako (2024)
			–	–	–	90.9 $\pm$ 167	–	(27–1690)	(3.66–227)	–	(5.6–571)	–	(6.1–161)	
Superficial soils ($n = 11$)	AGSM area (Bagassi)	Burkina Faso	12.1 $\pm$ 1.8 (8.7–14.7)	34.7 $\pm$ 5.4 (24.1–42.1)	0.95 $\pm$ 0.14 (0.82–1.27)	3.55 $\pm$ 1.23 (1.8–5.6)	0.05 $\pm$ 0.01 (0.03–0.06)	107 $\pm$ 22 (68.6–135)	43.2 $\pm$ 6.61 (27.7–49.3)	110 $\pm$ 20 (84–146)	36.8 $\pm$ 8.34 (23.9–52.7)	7.22 $\pm$ 2.15 (5.03–10.6)	47.4 $\pm$ 11.5 (29–69)	Sako and Nimi (2018)

Table 3 (continued)

Sample name	Context	Country	Al g/kg	Fe g/kg	Mn g/kg	As mg/kg	Cd mg/kg	Cr mg/kg	Cu mg/kg	Hg µg/kg	Ni mg/kg	Pb mg/kg	Zn mg/kg	Reference
River sediments (n = 6)	AGSM area (Bonsa river)	Ghana	10.8 ± 7.1 (1.1–19.2)	–	0.51 ± 0.03 (0.18–0.87)	0.74 ± 0.29 (0.35–1.09)	–	–	–	1080 ± 250 (680–1380)	–	–	–	Affum et al. (2016)
River sediments (n = 4)	AGSM areas (Nyam River)	Ghana	25.4 ± 0.11	18.2 ± 0.13	0.15 ± 0.01	397 ± 7.65	0.06 ± 0.02	59.5 ± 2.96	19.4 ± 1.99	150 ± 10	12.2 ± 0.26	7.93 ± 0.36	39.4 ± 3.63	Hadzi et al. (2024)
River sediments (n = 4)	AGSM areas (Subri River)		48.0 ± 0.03	36.1 ± 0.33	0.27 ± 0.03	9.63 ± 1.33	0.04 ± 0.02	120 ± 9.73	28.1 ± 1.87	30 ± 0	23.8 ± 0.81	10.2 ± 0.80	31.8 ± 4.57	
River sediments (n = 4)	AGSM areas (Birim River)		42.5 ± 0.42	56.4 ± 0.40	1.17 ± 0.02	35.0 ± 2.54	0.05 ± 0.02	134 ± 11.0	35.2 ± 2.18	60 ± 10	38.6 ± 2.43	10.3 ± 0.82	67.5 ± 9.05	
River sediments (n = 4)	AGSM areas (Bonsa River)		64.3 ± 0.63	42.1 ± 0.04	0.15 ± 0.01	4.84 ± 0.29	0.05 ± 0.03	134 ± 3.61	16.3 ± 0.11	–	27.3 ± 3.95	14.4 ± 1.50	27.8 ± 2.50	
River sediments (n = 16)	Industrial mining area (Morila)	Mali	9.8 ± 3.4 (5.9–17.4)	46 ± 33 (13–120)	0.22 ± 0.19 (0.04–0.79)	65.5 ± 65.6 (4–200)	0.01 ± 0 (0.01–0.02)	66.0 ± 49.7 (20.8–162)	40.1 ± 34.1 (8.37–115)	14 ± 6 (5–25)	6.46 ± 3.23 (3.7–15.7)	12.8 ± 3.32 (8.32–18.5)	11.8 ± 5.78 (6.1–31.3)	Bokar et al. (2020)
River sediments (n = 12)	AGSM area (Gambia River)	Senegal	–	–	–	–	–	–	–	1162 ± 799	–	–	–	Niane et al. (2019)
River sediments (n = 22)			–	–	–	–	–	–	–	350 ± 163	–	–	–	
Pond sediments (n = 22)			–	–	–	–	–	–	–	3273 ± 3017 (230–9260)	–	–	–	
River sediments (n = 22)	AGSM area (Kedougou region)	Senegal	–	–	–	–	–	–	–	820 (med.) (50–130000)	–	–	–	Gerson et al. (2018)
Soils (n = 12)			–	–	–	–	–	–	–	680 (med.) (130–19000)	–	–	–	

Table 4 Comparative data of dissolved metal(loid)s concentrations in surface waters and groundwaters from gold mining areas in West African countries forming part of the Birimian Greenstone Belt (average value $\pm$ standard deviation and/or (minimum–maximum) are reported depending on data availability)

Sample name	Context	Country	pH	Cond $\mu\text{S}/\text{cm}$	Al $\mu\text{g}/\text{L}$	Fe $\mu\text{g}/\text{L}$	Mn $\mu\text{g}/\text{L}$	As $\mu\text{g}/\text{L}$	Cd $\mu\text{g}/\text{L}$	Cr $\mu\text{g}/\text{L}$	Cu $\mu\text{g}/\text{L}$	Ni $\mu\text{g}/\text{L}$	Pb $\mu\text{g}/\text{L}$	Zn $\mu\text{g}/\text{L}$	Reference
Surface waters (n = 12)	AGSM area (Kokumbo)	Côte d'Ivoire	7.3 $\pm$ 0.4 (6.7–7.8)	287 $\pm$ 88 (163–414)	349 $\pm$ 1087 (3.36–3800)	766 $\pm$ 894 (25–3190)	487 $\pm$ 504 (15.1–1753)	1.1 $\pm$ 0.7 (0.33–2.36)	0.01 $\pm$ 0.03 (0.001–0.08)	1.1 $\pm$ 2.7 (0.11–9.8)	3.4 $\pm$ 9 (0.12–31.8)	3.7 $\pm$ 9.8 (0.43–34.9)	0.5 $\pm$ 1.5 (0.02–5.29)	4.9 $\pm$ 12.8 (0.36–45.5)	This study
Groundwaters (n = 8)			6.4 $\pm$ 1 (5.4–8)	403 $\pm$ 243 (48–713)	16.1 $\pm$ 23.3 (0.33–65.3)	26.9 $\pm$ 34.1 (1.28–99)	70.8 $\pm$ 79.3 (0.82–202.8)	0.51 $\pm$ 0.82 (0.03–2.45)	0.02 $\pm$ 0.03 (0.002–0.09)	0.17 $\pm$ 0.21 (0.01–0.56)	1.6 $\pm$ 3.1 (0.09–9.2)	3.8 $\pm$ 8.3 (0.06–24.3)	0.03 $\pm$ 0.03 (0.01–0.09)	438 $\pm$ 1054 (1.29–3023)	
Surface waters (n = 8)	Industrial mining area (Tongon)	Côte d'Ivoire	7.6 $\pm$ 0.5 (7.1–8.7)	1071 $\pm$ 1751 (60–4530)	–	2738 $\pm$ 4550 (300–12600)	–	4521 $\pm$ 8564 (2–19000)	0.13 $\pm$ 0.19 ($<$ LD–0.5)	4.6 $\pm$ 2.0 ($<$ LD–33)	2531 $\pm$ 4793 ($<$ LD–12000)	1966 $\pm$ 3658 ($<$ LD–8570)	1.5 $\pm$ 1.8 (0.7–5.8)	27 $\pm$ 8 (10–37)	Sako et al. (2018)
Groundwaters (n = 11)			6.9 $\pm$ 0.5 (6.3–8.0)	335 $\pm$ 309 (64–1110)	–	5609 $\pm$ 10908 ($<$ LD–35900)	–	285 $\pm$ 684 (0.2–2300)	9.8 $\pm$ 27.1 ($<$ LD–90)	18.3 $\pm$ 47.6 ($<$ LD–160)	16.3 $\pm$ 35.6 ($<$ LD–120)	85.8 $\pm$ 260 (1–870)	19.1 $\pm$ 96.2 (0.8–85)	62 $\pm$ 96 (0.02–340)	
Surface waters (n = 15)	Bandama River	Côte d'Ivoire				(89–1135)		(0.66–4.01)	($<$ LD–0.07)		(0.01–86.4)		($<$ LD–5.03)		Quattara et al. (2018)
Surface waters (n = 13)	AGSM area (Nigerten Lipiko)	Niger	6.4 $\pm$ 0.5 (5.7–7.2)	188 $\pm$ 141 (30.4–488)	206 $\pm$ 275 (7.52–821)	131 $\pm$ 186 (5.8–590)	30.7 $\pm$ 45.8 (1.01–161)	0.93 $\pm$ 0.90 (0.13–2.91)	0.08 $\pm$ 0.13 (0.003–0.46)	0.37 $\pm$ 0.38 (0.08–1.08)	2.35 $\pm$ 1.27 (1.06–5.67)	1.35 $\pm$ 0.82 (0.69–3.76)	0.16 $\pm$ 0.18 (0.03–0.52)	7.23 $\pm$ 3.95 (3.07–15.67)	El Farouk et al. (2022)
Groundwaters (n = 43)			6.5 $\pm$ 0.7 (5.2–8.7)	578 $\pm$ 380 (57.6–1605)	12.0 $\pm$ 19.9 (0.26–86.1)	12.6 $\pm$ 23.9 (0.29–103)	107 $\pm$ 237 (0.09–956)	10.3 $\pm$ 41.7 (0.005–203)	0.014 $\pm$ 0.025 (0.001–0.14)	0.51 $\pm$ 1.59 (8.82)	5.30 $\pm$ 22.8 (0.10–150)	2.14 $\pm$ 4.30 (0.05–24.2)	0.19 $\pm$ 0.58 (3.16)	14.4 $\pm$ 20.4 (0.56–94.7)	
Surface waters (n = 13)	AGSM and industrial mining regions	Burkina Faso	6.9 $\pm$ 0.9 (5.6–8.5)	189 $\pm$ 122 (57–406)	–	–	–	0.51 $\pm$ 0.16 (0.31–0.74)	–	–	–	–	–	–	Ouédrago and Anyot (2013)
Groundwaters (n = 36)	AGSM regions	Burkina Faso	7.1 (5.8–7.8)	–	18 (2.1–25)	0.008 ($<$ 0.005–0.69)	1.12 (0.20–98.9)	15.1 ($<$ 0.5–1630)	$<$ 0.05 ($<$ 0.05–0.12)	$<$ 0.5 ($<$ 0.31–5.78)	1.14 (0.5–4.65)	$<$ 0.2 (0.02–1.96)	0.1 ($<$ 0.1–2.3)	11 (3.1–46.10)	Smedley et al. (2007)
Groundwaters (n = 30)	AGSM area (Bomboré)	Burkina Faso	6.5 $\pm$ 0.5 (5.4–7.2)	466 $\pm$ 276 (58–946)	–	7017 $\pm$ 10304 ($<$ 100–44400)	288 $\pm$ 248 ($<$ 2–1250)	18 $\pm$ 26 ($<$ 0.5–100)	–	23.8 $\pm$ 34.6 ($<$ 1–161)	12.3 $\pm$ 14.0 (1–66)	17.0 $\pm$ 16.1 (3–71)	4 $\pm$ 3.9 ($<$ 1–11)	79 $\pm$ 47 ($<$ 5–160)	Sako et al. (2016)
Surface waters (n = 6)	AGSM area (Bonsa river)	Ghana	–	–	4160 $\pm$ 544 (3500–4980)	–	350 $\pm$ 122 (0.25–0.59)	1.47 $\pm$ 0.52 (0.69–2.14)	–	–	$<$ 10	–	–	–	Affum et al. (2016)
Groundwaters (n = 10)	AGSM area (Obuasi)	Ghana	5.0 $\pm$ 0.8	459 $\pm$ 391	–	513 $\pm$ 488	143 $\pm$ 97.0	8.81 $\pm$ 13.2	0.44 $\pm$ 0.27	1.0 $\pm$ 0.94	35.0 $\pm$ 7.80	7.31 $\pm$ 4.48	4.43 $\pm$ 3.34	81.5 $\pm$ 101	Bempah et al. (2016)
Groundwaters (n = 518)	AGSM regions	Ghana	6.8 $\pm$ 1.3 (4.1–11.3)	359 $\pm$ 321 (23.9–1854)	–	1426 $\pm$ 3330 (1–37630)	980 $\pm$ 2290 (0.1–26800)	340 $\pm$ 4320 (0.1–78000)	5 $\pm$ 13 (0.1–276)	–	–	–	30 $\pm$ 60 (1–497)	–	Armah (2014)

Table 4 (continued)

Sample name	Context	Country	pH	Cond µS/cm	Al µg/L	Fe µg/L	Mn µg/L	As µg/L	Cd µg/L	Cr µg/L	Cu µg/L	Ni µg/L	Pb µg/L	Zn µg/L	Reference
Groundwaters (n = 74)	AGSM area (Tarkwa)	Ghana	6.8 ± 0.7 4.9–7.7	399 ± 292 (150–1082)	–	–	274 ± 47.4 (4–1375)	2 ± 0.3 (0.1–15)	1 ± 0.2 (0–9)	3 ± 0.2 (1–8)	3 ± 0.2 (1–9)	55 ± 3.4 (2–118)	3 ± 0.2 (0.1–10)	2 ± 0.2 (1–6)	Gwira et al. (2024)
Surface waters (n = 9)	Industrial mining area (Morila)	Mali	6.3 ± 0.9 (4.6–7.4)	281 ± 267 (35–847)	87.2 ± 162 (5–508)	3914 ± 668 (10–20353)	423 ± 558 (4.36–1690)	14.9 ± 26.3 (0.8–82.4)	0.05 ± 0.05 (0.05–0.05)	2.56 ± 0.66 (2–4.1)	1.68 ± 1.14 (0.3–3.4)	4.99 ± 3.84 (1.9–13.9)	1.36 ± 3.51 (0.1–10.7)	50.6 ± 104 (4.3–326)	Bokar et al. (2020)

of Mn in three groundwater samples (GW1–GW3) are relatively high, exceeding the French drinking water guideline of 50 µg/L (JORF 2022). A monitoring of Mn levels in wells of the area would also be advisable given that Mn concentrations lower than the WHO guideline in drinking water are suspected to cause neurodevelopmental and neurotoxic effects in children, with greater detrimental impact in girls compared to boys (Bouchard et al. 2011; Kullar et al. 2019; Roels et al. 2012). For example, a Mn concentration of 78 µg/L and 185 µg/L would be associated with a decrease of 1% in Performance IQ score for girls and boys, respectively (Kullar et al. 2019). High concentrations of Mn are also found in groundwaters from other ASGM areas in West Africa (Table 4), such as in the Nigerien Liptako (Farouk et al. 2022), in Burkina Faso (Sako et al. 2016), in Ghana (Armah 2014; Bempah and Ewusi 2016; Gwira et al. 2024), and in Mali (Bokar et al. 2020) suggesting that the presence of geogenic Mn in groundwaters, in addition to As, may represent a public health issue in this region.

In surface waters, dissolved metals concentrations are generally low and comparable to the average composition of world and African rivers (Table 2, Gaillardet et al. 2003). Only Mn dissolved concentrations exceeded WHO guidelines in half of the samples. Aluminum, Fe, and Mn concentrations are generally higher in surface waters compared to groundwaters and it could be due to the presence of colloid material in the dissolved fraction (< 0.22 µm) of the surface waters in relation with the high amount of sediments in suspension.

The highest dissolved concentrations of metals (Al, Co, Cr, Cu, Fe, Mn, Ni, Pb, V, Zn) are found in a stream flowing through an abandoned cocoa plantation draining a cyanide treatment site (SW5w). During ore processing, CN likely leaches these metals in addition to complexing gold, leading to a greater mobility of these contaminants in case of uncontrolled discharge of cyanidation effluents. This is consistent with the significantly lower concentrations measured for Cu, Ni, Ti, and V in the cyanidation residues compared with the initial ore material and with XRD results which revealed the absence of titanium oxides in cyanidation residues while they were present in ore material. Although the low proportion of F-CN found in residues suggests a low long-term risk of transfer of F-CN to nearby streams and groundwaters, further investigation is required to assess the dispersion of F-CN and metal-CN complexes in water resources and to evaluate the effect of the CN and metal co-contamination on metal mobility. This should be investigated especially for Hg, as it may increase Hg mobility and methylation rate in the impacted streams (Malone et al. 2023). However, except for Mn, the low concentrations of metals in other surface water samples, including downstream cyanidation sites (SW4w, SW6w), suggest a low mobility of metals in downstream surface waters in a dissolved form. Indeed, the

conditions of the studied streams (i.e., near-neutral oxygenated waters, sediments containing a high proportion of iron oxy(hydr)oxides) promote the scavenging of metals with the solid phase through adsorption processes. This is supported by the correlation observed between Fe and As, Cu, Cr, and V (Spearman correlation coefficients $r^2 > 0.5$, p value < 0.05) in sediments (Table S4). In Kokumbo, artisanal gold miners primarily work with oxidized ore but are now increasingly exploiting the unaltered facies containing sulfide minerals. Therefore, these more recent mining practices could lead to the production of acid mine drainage and increase the risk of metal transfer towards the water compartment.

Conclusion

The aim of this study was to characterize the potential sources of contamination and to evaluate the environmental contamination by metals and metalloids potentially released by ASGM activities in Kokumbo, a historic mining area in central Côte d'Ivoire. It should be noted that the study was carried out during the rainy season and further investigation would be required to assess seasonal variations which can affect metal dispersion.

An enrichment, relatively to the average UCC composition, was found in alluvial ore material for different metals (Co, Cu, Cr, Fe, Mn, Ni, and V) and metalloids (As, Sb) revealing their high geochemical background in the studied area. However, enrichment factors ($EF_{\text{metal/ore}}$) in soils and sediments indicated that there was no exogenous contamination. These elements derive mainly from a geogenic source. Their concentrations remained moderate compared to other ASGM site across Africa but there is a possible environmental and health concern for several metals, including As, Cr, Hg, and Mn. Lead, Cd, and Sb showed a slight enrichment in cyanidation residues and in soils and sediments compared to alluvial ore and further research would be needed to determine its exact (anthropic) origin as it is not directly used for gold ore processing.

Mercury concentration in unprocessed alluvial ore material was low (0.06 ± 0.01 mg/kg) but reaches very high concentrations in cyanidation residues (up to 8.32 mg/kg) and in soils and sediments (up to 20.4 mg/kg) indicating that ASGM is the main source of environmental contamination with Hg. This may represent an important risk to local populations who consume fishes from the Bandama River, which drains the study area. Bandama sediments measured in this study also contained high Hg concentrations. Further investigations on the dispersion and speciation of Hg in this river are necessary to evaluate the potential risk of Hg transfer to the trophic chain and associated human exposure to Hg.

Except for Mn, dissolved As and other metal concentrations in surface water and groundwater were low, showing a limited mobility toward the aqueous phase under the field conditions, likely due to their adsorption on iron oxy(hydr)oxides. Manganese presented relatively high concentration in groundwater but remained below the WHO guideline. Long-term monitoring of As and Mn in the groundwater used for drinking supply is recommended since high geogenic background in West Africa is common and concentrations in waters can be highly variable in space and time.

Cyanidation residues contained up to 100 mg/kg of total cyanides but generally less than 3% were in the form of free cyanides. However, the cultivated soil next to cyanidation area had the highest proportion of F-CN (70.7%). The highest dissolved concentrations of metals were found in a stream draining a cyanidation site suggesting that they may enhance metal mobility. More work is needed to study the effect of cyanide-metal co-contamination (especially for Hg) on metal fate in impacted streams and groundwaters as well as the risk of CN dispersion and persistence.

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Author contributions All authors contributed to the study design and implementation. Material preparation and fieldwork were performed by K.B.K., E.R., K.E.A., Z.O., I.K., N.F. D.Ba., O.B., Data collection and analysis were performed by K.B.K., E.R. K.E.A., N.F., D.Bo., S.D., A.D., M.M. The first draft of the manuscript was written by K.B.K., E.R. and O.B., all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Declarations

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