Trace metals in cocoa soils exposed to illegal gold panning in central west Côte d'Ivoire.

Abstract

Côte d'Ivoire, the world's leading cocoa producer, is facing the challenge of illegal gold washing, which is likely to jeopardise its goal of a sustainable cocoa farming system. This is due to the degradation of soil resources through trace metal contamination (TM). The aim of this study is to determine the levels of copper (Cu), cadmium (Cd), lead (Pb) and arsenic (As) contamination in cocoa growing soils in Bonikro.70 soil samples were collected using the toposequential method, processed and sent to the laboratory for analysis. The results show that TM levels vary significantly depending on the topographical position and depth of the soil. However, these levels are below tolerable limits. There are significant correlations between pH and TM, which vary with depth and topographic position. A strong positive correlation was observed at depth between pH and Pb ($r = 0.82$) and Cd ($r = 0.71$), whereas at the surface the correlations were negative for the same elements. This indicates that pH plays a crucial role in the evolution of TM concentrations. In conclusion, this study highlights the fact that cocoa-growing soils have TM levels below tolerable thresholds, but with significant variation as a function of depth and topographic position. Soil pH appears to be a determining factor in the mobility of TMs, which has important implications for the sustainable management of soils and the environment. Further research is needed to refine these observations and improve natural resource management.

Keywords: Gold panning, trace metal, cocoa, soil management, environment.

1. Introduction

Cocoa in Côte d'Ivoire generates more than 30% of export earnings and contributes more than 15% of gross domestic product (GDP) [1][2]. However, recent studies show a decline in the yields of cocoa plantations, partly due to the ageing of the plantations. This ageing of orchards, combined with climate change, is making certain regions increasingly marginal for cocoa production [3]. This is compounded by the state of soil fertility and soil acidification[4]. Soil acidification leads to the solubilisation of metals such as aluminium (Al), iron (Fe) and manganese (Mn), which then bind nutrients, particularly phosphorus (P) in tropical soils. In addition to these difficulties, for more than fifteen years cocoa farming has been facing a new challenge linked to the development of the mining sector.

Mining is known to be an activity that can lead to the degradation of soil resources through their contamination with trace metal (TM) such as lead (Pb), cadmium (Cd), arsenic (As) and copper (Cu) [5]. However, studies on the soil contamination levels of artisanal miners, who are generally illiterate farmers engaged in illegal and clandestine gold panning, remain patchy, and the case of the Bonikro mining area in west-central Côte d'Ivoire is illustrative. The remaining cocoa plantations are under attack from illegal gold panners, with the result that waste is dumped at various sites, which may contain chemical elements that accumulate

Comment [TR1]: I think it is better to modify the title "Trace Metal Contamination of Cocoa Soils Resulting from Illegal Gold Panning in Central West Côte d'Ivoire"

Comment [TR2]: •**Strengths**: The abstract effectively summarizes the study's objective, methods, key findings, and implications.
•**Recommendation**: Clarify the significance of the findings for practical soil and environmental management. For example, specify how the role of pH could inform future strategies.

Comment [TR3]: **Strengths:**
1.**Relevance of Topic:** The paper addresses an important issue at the intersection of agriculture and mining, particularly the impact of trace metal contamination on cocoa farming in Côte d'Ivoire, a country heavily reliant on cocoa exports.
2.**Clarity of Purpose:** The aim of the study is clearly stated, focusing on the assessment of soil contamination and its implications for sustainable cocoa farming and environmental management.
3.**Contextual Background:** The introduction effectively highlights the significance of cocoa farming to Côte d'Ivoire's economy and provides context about challenges such as plantation ageing, soil fertility issues, and the impact of illegal mining.

Comment [TR4]: **Suggestions for Improvement:**
2. Citation Placement and Integration:
•The placement of citations like "[1] [2]" can be improved for smoother readability. For example, instead of mentioning them at the end of the sentence, you could write: "Cocoa in Côte d'Ivoire, which contributes over 30% of export earnings and 15% of GDP, is a critical sector of the economy [1, 2]."
•Similarly, for "This ageing of orchards, combined with climate change, is making certain regions increasingly marginal for cocoa production [3]," a brief elaboration of how climate change impacts cocoa farming would strengthen the connection.
2 . Structure and Flow:
• . The transition between soil acidification issues and mining-induced contamination is somewhat abrupt. Consider adding a linking sentence to bridge these two points and provide a coherent narrative on how both issues converge to threaten cocoa farming sustainability.
3. . Terminology Clarification:
• . Terms such as "illegal and clandestine gold panning" could be briefly explained for international readers unfamiliar with artisanal mining practices in Côte d'Ivoire.
4.**Typographical Issues:**
1.Replace "The remaining cocoa plantations are under attack from illegal gold panners, with the result that waste is dumped at various sites, which may contain chemical elements that accumulate in the soil" with "Illegal gold panners are targeting the remaining cocoa plantations, leading to the disposal of waste at various sites, potentially causing the accumulation of chemical elements in the soil."

in the soil [6]. This situation could therefore jeopardise Côte d'Ivoire's goal of a sustainable cocoa farming system [7]. The aim of this study is to determine the level of TM contamination (Pb, Cd, As and Cu) in cocoa growing soils in Bonikro. In the current context, the aim is to assess the level of contamination of agricultural soils in this region, which is currently dominated by illegal gold panning, in order to identify the factors contributing to this contamination. The results will then be used to make recommendations for the sustainable management of agro-systems in mining areas and for environmental protection.

2. Material and Methods

2.1. the study site

The study was carried out in Bonikro, located in central westCôte d'Ivoire, in the Divo department. The department covers an area of approximately 3,577 km² and lies between latitudes 05°40' and 06°10' North and longitudes 05°30' and 04°40' West (Figure 1). The climate is transitional equatorial, with two rainy seasons and two dry seasons. The average annual rainfall is 1,400 mm and the average annual temperature is 25.3°C. The soils are predominantly ferralitic, highly desaturated and divided into three groups: plinthosols, ferralsols and gleysols. All these soils are the result of long and intense weathering, with a clay fraction composed mainly of kaolinite [8].

2.2. Soil sample collection and processing

Soil samples were collected from several cocoa plots in Bonikro using the toposequence method. A 30°N toposequence was opened and soil samples were drilled from 0-20 cm and 20-40 cm from the topographic positions to the shallows, with a rate of 10 samples per topographic level. The choice of sampling point per topographic position was based on the ability to report metal contamination of the soil. A total of 70 soil samples were collected from the study area. These samples were transported to the laboratory, crumbled by hand and then approximately 500 g of soil was dried in a room at room temperature according to ISO standard 11464:2006. After drying, each sample was sieved through a square mesh sieve (Ø = 2 mm) to collect the fine soil for laboratory analysis.

2.3. Chemical analysis of soils

Determination of soil pH

The fine soil obtained from each sample was divided into two parts. The first part was used to determine soil pH according to the method recommended in ISO 10390:2021. Specifically, 10 g of fine soil from each sample was placed in a 100 ml plastic beaker with 50 ml of distilled water. The mixture was stirred for 1 hour and then the pH was measured using a glass electrode in a dilute soil suspension. The reading was taken when the digital display of the pH meter had stabilised.

Comment [TR5]:
•**Geographic Information**: The geographical coordinates are well-presented, but it would be useful to include a clearer reference to a map or a figure (such as Figure 1) showing the exact location of Bonikro. This could enhance the reader's understanding of the site's location within Côte d'Ivoire.
•**Climate and Soil Characteristics**: The climate and soil information is well detailed. It might be useful to briefly explain how these climatic and soil characteristics could potentially influence the soil properties you are studying, especially regarding metal contamination.
•**Soil Composition**: The description of the soils and their weathering process is clear. However, it might be helpful to provide more specific information about how the soil properties (e.g., clay content or organic matter) could impact the distribution or retention of trace metals (TMs) in the area.

Comment [TR6]: •**Sampling Methodology**: The use of the toposequence method for sampling is appropriate. However, more details could be provided on the criteria used to select cocoa plots (e.g., age of trees, management practices, or geographical variation within the plots) to ensure the representativeness of the samples.
•**Sampling Depths**: It is good that both 0-20 cm and 20-40 cm depths are considered. It would be useful to justify why these specific depths were chosen for sampling. For example, are they representative of the rooting zone of the cocoa plants, or do they reflect the expected areas of metal accumulation?
•**Sample Size**: The total sample size (70 samples) is appropriate, but further clarification on how the 10 samples per topographic level were distributed across the study area would be helpful. Were there variations in sampling density depending on the topographic level, and if so, why?

Comment [TR7]: •**pH Determination**: The method for determining soil pH is clear. However, it would be beneficial to specify the type of pH meter used (e.g., brand/model) and any calibration steps undertaken to ensure accurate readings.
•**Trace Metal (TM) Content Analysis**: The use of ICP-AES after mineralization with aqua regia is an appropriate method for analyzing trace metals. Consider briefly justifying why aqua regia was chosen for the mineralization process over other methods (e.g., microwave-assisted digestion), especially in relation to the types of trace metals expected in the samples.
•**Quality Control**: It would be helpful to mention any quality control measures that were applied during the chemical analysis. For instance, did you use standard reference materials for calibration or conduct any replicate analyses to ensure precision and accuracy?

Determination of soil TM content

The TM content of the samples was determined by inductively coupled plasma atomic emission spectrometry (ICPAES) after mineralisation by hot acid etching of the samples with aqua regia (1/3 HNO3 and 2/3 HCl).

2.4. Statistical analysis

All analyses were performed using R software. After checking for homogeneity and normality within groups, a one-way ANOVA followed by a Student-Newman-Keuls (SNK) test comparison was used to identify significant differences between the means of ETM content by topographic level at the 5% p threshold.

3. Results

3.1. Spatial distribution of trace metals (TM)

Analysis of the vertical variation of TM content and pH shows that depth has a significant effect on the content of Pb, Cd, As and pH in the soil. Cd (0.003 $mgkg^{-1}$), As (6.21 $mgkg^{-1}$) and pH (6.46) in the soil are significantly lower at depth (20-40 cm) than at the surface (0-20 cm), while Pb (0.16 mgkg-1) is significantly higher at depth (20-40 cm). The Cu content shows no significant difference whatever the depth (Table 1).

Table 1. Average TM and pH levels as a function of depth and tographic position

Depth	TM ($mgkg^{-1}$)				Acidity
	Pb	Cd	As	Cu	pH
	Vertical variation				
0-20 cm	0,07b	0,04a	9,36a	6,65a	7,2a
20-40 cm	0,16a	0,003b	6,21b	6,46b	6,7b
Topographic position	Lateral variation				
	Pb	Cd	As	Cu	pH
Lowland (BF)	0,13ab	0,03b	7,71b	7,84b	6,9b
Low lying area (BV)	0,16a	0,05a	7,03b	7,05b	7,9a
Top of slope (HV)	0,05c	0,02b	14,11a	9,39a	6,5c
Mid-slope (MV)	0,10b	0,003c	4,61c	3,54d	7,2b
Summit (S)	0,12ab	0,015bc	5,46c	4,95c	6,5c
Tolerable limit values ($mgkg^{-1}$)	50	1,5	20	140	6,5-7,5

The letters assigned to the averages (a, ab, b and c) represent the statistical differences between the average levels, where the values sharing the same letter are not statistically different from one another.

3.2. TM distribution along the toposequence

Figures 1 to 5 illustrate the distribution of ETM values and soil pH along the toposequence around the gold mine. Analysis of the trends reveals a number of significant observations.

The elements As, Cd, Cu, Pb and pH show significant variations in content between positions BF, BV, HV, MV, S and depths 0-20 cm and 20-40 cm:

Comment [TR8]: •**Analysis Method**: The statistical methods (one-way ANOVA and SNK test) are appropriate for comparing trace metal content across topographic levels. However, it would be helpful to specify which assumptions were checked for the ANOVA (e.g., homogeneity of variances, normality of data) and how violations were addressed, if applicable.
•**Software**: Mentioning that R software was used is fine, but it would be helpful to specify the version of R and any relevant packages used for the statistical analysis, as this could affect the reproducibility of the analysis.
•**Significance Threshold**: The 5% p-value threshold for statistical significance is standard, but you might consider mentioning if any corrections were applied for multiple comparisons (e.g., Bonferroni correction), especially if a large number of comparisons were made.

Comment [TR9]: 1.**Depth vs. Topographic Position**: The distinction between vertical and lateral variation is valuable. However, the presentation of the data in the text could be more concise to avoid repetition.
2.**Tables and Figures**:
•Table 1 is informative but could include error margins for clarity.
•Figures 1-5 are relevant but lack detailed captions. Consider including statistical significance markers and brief explanations of the trends.
3.**Data Interpretation**: The results regarding the role of pH in trace metal mobility are critical. However, the interpretation could be strengthened with references to related findings in similar ecosystems.

- The mean pH value (Fig.1) varies between positions BF, BV, HV, MV and S at depth 0-20 cm with a mean of about 6.70 ± 0.326. At a depth of 20-40 cm, the average pH shows more pronounced variations, with an average of around 6.34 ± 0.58 at MV and 6.33 ± 0.578 at BF.

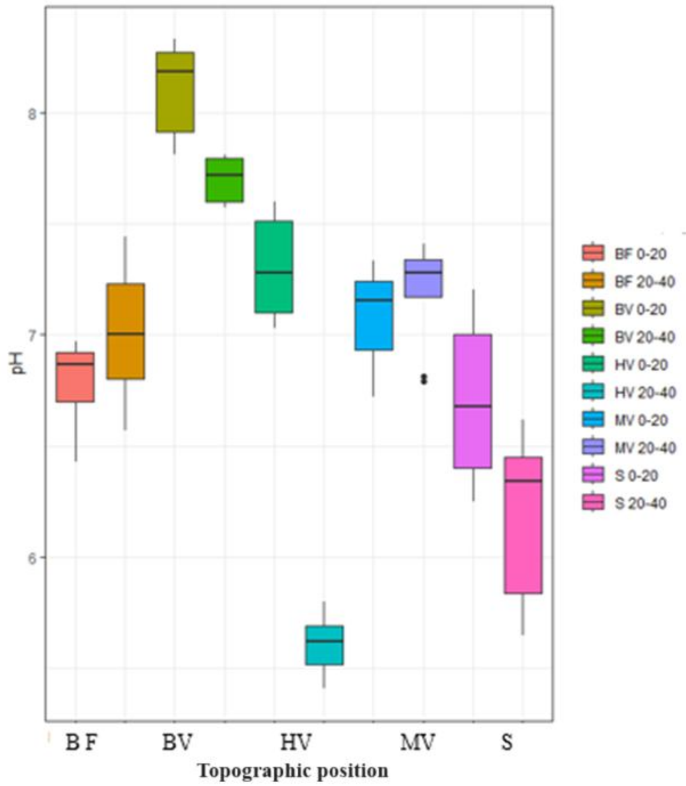

Figure 1 : Distribution of pH values according to topographic position

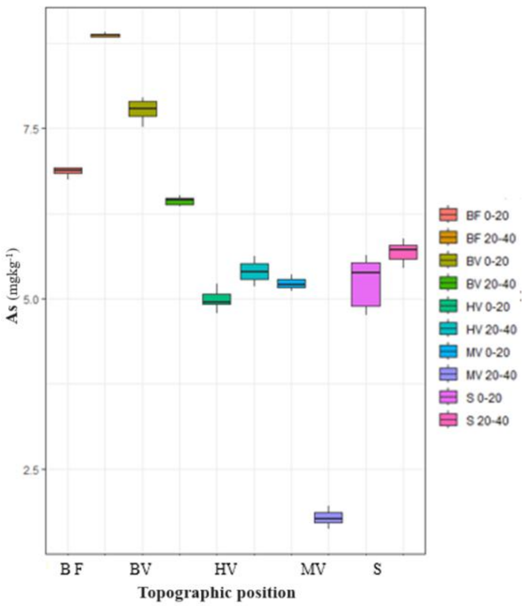

Figure 2 : Distribution of As content according to topographic position

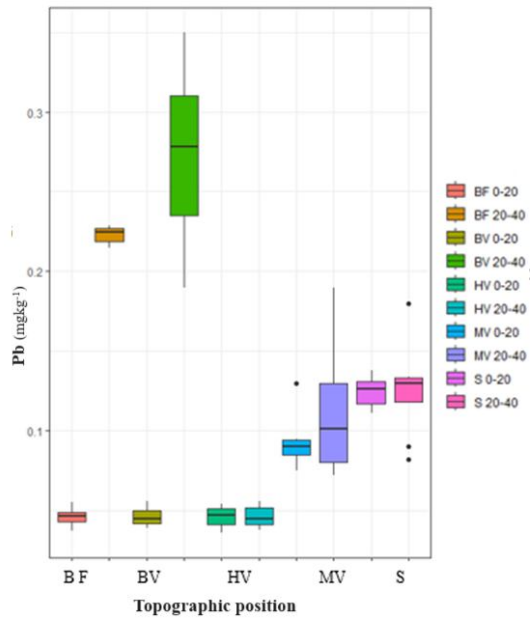

Figure 3 : Distribution of Pbcontent according to topographic position

- The mean As contents (Fig. 2) are relatively similar between the BF, BV, HV, MV and S positions at 0-20 cm depth, with a mean of about 5.25 mgkg-1 ± 0.32. However, at 20-40 cm depth, the mean As content decreases significantly to about 0.03 mgkg-1 ± 0.004, indicating a different distribution in the deeper soil horizons. However, at depths of 20-40 cm, the mean As content decreases significantly to around 0.03 mgkg-1 ± 0.004, indicating a different distribution in the deeper soil horizons.

- The average Pb content (Fig. 3) is very low at depths of 0-20 cm, with an average of about 0.002 mgkg-1 ± 0.0004 at MV. At depths of 20-40 cm the average Pb content increases significantly to around 4.80 mgkg-1 ± 0.19 at MV.

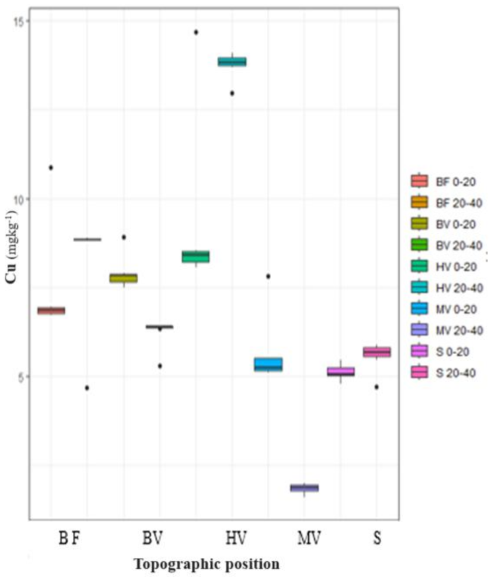

Figure 4 : Distribution of Cd content according to topographic position

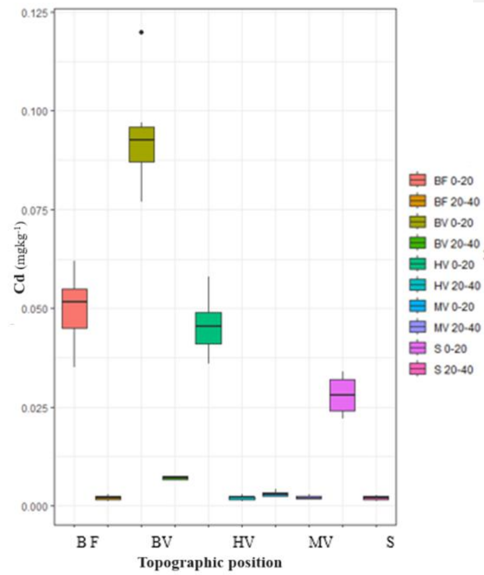

Figure 5: Distribution of Cu content according to topographic position

- Mean Cd contents (Fig. 4) varied between the BV and HV positions at depths of 0-20 cm, with a mean of approximately 5.10 mgkg-1 ± 0.18 at BV and 6.70 mgkg-1 ± 0.33 at HV. At depths of 20-40 cm, the mean Cd content decreased significantly at BV (approximately 0.12 mgkg-1 ± 0.009) while remaining relatively stable at HV.

- The mean Cu content (Fig. 5) shows a significant difference between BF and MV positions at depths of 0-20 cm, with a mean of 6.70 mgkg-1 ± 0.33 at BF and 5.68 mgkg-1 ± 0.13 at MV. At depths of 20-40 cm, mean Cu concentrations show similar variations for all positions, with a mean of around 4.80 mgkg-1 ± 0.19.

The trend analysis shows that the concentration of lead (Pb) in the soil varies significantly with depth. The coefficients for the different depths indicate that the Pb concentration tends to decrease with increasing depth. This trend is particularly pronounced for the HV position where the Pb concentration decreases significantly ($p < 0.001$). This suggests that lead levels in soil are higher at the surface (shallower) and decrease with depth.

As for cadmium (Cd), the trend analysis shows that the concentration of this metal also varies with depth. The depth BV coefficient indicates an increase in Cd concentration with depth, although this trend is not statistically significant. However, the MV depth shows a significant decrease in Cd concentration ($p = 0.00454$). This suggests that cadmium may accumulate more at the soil surface, while its concentration decreases with depth.

For arsenic (As), the results of the trend analysis show significant variations in concentration as a function of depth. The positive coefficient for the HV depth indicates a significant increase in arsenic concentration with depth ($p < 0.001$). In contrast, depths MV and S show opposite trends with significant negative coefficients. This suggests that arsenic may be more abundant at depth and less abundant at the soil surface.

For copper (Cu), the trend analysis shows significant variations in concentration as a function of depth. Depth BV shows a significant negative relationship with Cu, indicating that Cu concentration decreases with depth ($p = 0.0234$). Depth HV shows the opposite trend with a significant increase in copper concentration ($p < 0.001$). Similarly, depths MV and S show significant negative coefficients, also suggesting a decrease in copper concentration with depth.

For soil pH, the results of the trend analysis indicate significant variations with depth. Depth BV showed a significant positive relationship with pH, indicating that soil pH tends to increase with depth ($p < 0.001$). In contrast, depth HV showed a significant negative relationship with pH ($p = 0.00353$). Depths MV and S also show coefficients suggesting variations in pH with depth, although the trends are not as pronounced.

Table 2. Correlation between different variables as a function of depth

	0 – 20 cm					20 – 40 cm				
	Pb	Cd	As	Cu	pH	Pb	Cd	As	Cu	pH
Pb	1					1				
Cd	-0,641	1				0,715	1			
As	-0,812	0,241	1			-0,501	-0,117	1		
Cu	-0,893	0,692	0,851	1		-0,258	0,127	0,96	1	
pH	-0,546	0,728	0,218	0,612	1	0,821	0,642	-0,778	-0,597	1

Overall, there were average to strong positive correlations between Cd, Cu, As and pH at all depths, and average to strong negative correlations between Pb, As and Cu at all depths (Table 2). However, there were differences between surface (0-20 cm) and depth (20-40 cm) correlations. There was a strong positive correlation at depth between pH and Pb ($r = 0.82$) and Cd and Pb (0.71), whereas at the surface the correlation was negative ($r = -0.54$) and ($r = -0.64$). The same observation is made for As ($r = -0.77$) and Cu ($r = -0.59$) with pH, which show negative correlations at depth, while at the surface there are positive correlations.

4. DISCUSSION

4.1. Mobility of TM in soil

Comment [TR10]: 1.**Depth and TM Levels**: The discussion of the role of pH in influencing TM mobility is insightful. Expand on the mechanisms of metal mobility and how they relate to soil acidity.
2.**Practical Implications**: While the study mentions implications for sustainable soil management, it could be more specific about actionable recommendations.
3.**Comparison with Other Studies**: Include comparisons with similar studies conducted in other regions or crops to place the findings in a broader context.

The results of the spatial distribution of the concentrations of trace metals (As, Cd, Cu, Pb) and soil pH show that the concentrations vary significantly with topographic position and depth. The Cu content showed a slight variation with depth. This variation could be attributed to Cu migration and redistribution processes in the soil profile. In fact, [9] observed a gradual decrease in Cu content as a function of depth along a forest toposequence.

For cadmium, the results show that the content of this metal decreases with increasing depth. This is because cadmium tends to bind to soil particles, especially clay and humus. According to the work of [10], the content of organic matter in the soil could influence the retention of cadmium. This could explain the observed variation in cadmium levels with depth, as different depths could have different organic matter contents. Pb is a metallic element that is not very mobile in soil. Its high affinity with organic matter explains its high concentration in surface horizons [11]. In our case, the high concentration of Pb in the 20-40 cm horizon can probably be explained by the cultivation work in the cocoa plots.

The work of [12] showed a decrease in arsenic levels with increasing soil depth along a toposequence. According to this work, this trend could be attributed to vertical migration of arsenic from deeper geochemical sources. [13] showed that arsenic concentrations were higher in middle and upper slope areas.This distribution could be attributed to erosion and runoff processes of arsenic-rich particles from higher to lower areas of the toposequence.

4.2. Relationship between TM and pH

It is also important to note that soil pH can influence TME availability and mobility [14][15]. Our results show a strong relationship between pH and TMs. There is a strong positive correlation at depth between pH and Pb ($r = 0.82$) and Cd and Pb (0.71), whereas at the surface the correlation is negative ($r = -0.54$) and ($r = -0.64$). The same observation is made for As ($r = -0.77$) and Cu ($r = -0.59$) with pH, which show negative correlations at depth, while at the surface there are positive correlations. These results are in line with those of [16] and [17], who found either a decrease or an increase in the uptake of TM with pH, depending on the soil type. According to [18], metal concentrations are higher at $pH < 6$. This explains the low levels found in this study.

The correlations between the metals themselves give us information about the complex interactions that exist between the different MTEs in the soil. Among the MTs, we observe a relatively strong positive correlation ($r = 0.73$) between As and Cu, suggesting a certain consistency in their concentrations at different depths and positions. We also observed a very strong negative correlation ($r = 0.64$) between Pb and As, indicating that higher concentrations of one are associated with lower concentrations of the other.

5. Conclusion

Comment [TR11]: 1.The conclusion is concise and summarizes the findings well. However, it could emphasize the broader implications for policymakers, farmers, and environmental managers.
2.Suggest specific areas for future research, such as long-term monitoring of soil pH and TM levels or the impact of remediation efforts.

The aim of this study was to determine the level of contamination of cocoa growing soils by illegal gold washing. The various results show that the levels of TM (Pb, Cd, Cu and As) in the soil are below tolerable thresholds. However, the levels vary according to the different topographic segments and the depth of the horizons. The pH could play an important role in the evolution of the TM levels, as it is strongly correlated with them. These results may have important implications for the sustainable management of soils and the environment. They also provide important insights into the distribution and interactions of metals in soils, which may have important implications for ecosystem health and soil quality. Further studies with larger samples could help to refine these relationships and provide more detailed information for effective management of natural resources.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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Comment [TR12]: Use of Recent Literature: It is recommended to include more recent references to ensure that the literature review reflects the most current trends and findings. This could involve incorporating studies from the last 5-10 years, especially in rapidly evolving fields such as environmental contamination and sustainable agriculture.
Error Checking: Please ensure that all references are accurately cited. Some references, such as [1] and [5], may need a closer look to ensure they are correctly formatted and complete. For example, titles, page numbers, and other relevant details should be reviewed for accuracy.

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General Comments

1. **Clarity and Organization**: The paper presents an important and relevant topic on trace metal contamination in cocoa-growing soils affected by illegal gold panning. However, the organization of the manuscript could be improved to enhance readability and logical flow, particularly in the results and discussion sections.
2. **Novelty and Contribution**: The study addresses an issue of socio-economic and environmental importance. However, the novelty of the study compared to similar research conducted in other cocoa-producing regions could be better articulated.
3. **Language and Style**: The language is clear, but there are minor grammatical issues and redundancies in some sections (e.g., repetitive statements about pH correlation). Proofreading is recommended.

Minor Comments

1. **Abbreviations**: Some abbreviations (e.g., TM) are introduced without definition in the abstract and introduction. Ensure all abbreviations are defined upon first use.
2. **Formatting**: Ensure consistent formatting of units (e.g., mgkg-1 should be written as mg/kg for clarity).
3. **Redundancy**: Remove redundant statements, such as repeated observations about pH and Pb/Cd relationships in different sections.