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## Abstract

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Over the decade many villages in Zamfara State Nigeria were known for artisanal gold mining which caused pollution of vast water bodies and area of land. This makes it imperative to analyze heavy metal contamination in wells and surface water. The present study evaluated the concentrations of heavy metals (iron, lead, cadmium, zinc, nickel, and chromium) in the ore processing water and well-water in some selected villages of Anka local government area in Zamfara State where mining is taking place. Anka town served as control due to absence of mining activities. The concentrations of heavy metals in the water samples were measured using atomic absorption spectrophotometry. The concentration of Fe, Pb, and Cd ranged from 23.3 - 921.46, 0 - 132.8, and 3.3 - 4.1 ppm respectively. While Zn, Ni, and Cr were not detected in the water samples. Virtually all the detected heavy metals in the water samples exceeded the international organizations (WHO, USEPA, and EPA-EUC) maximum permissible limit of 0.3 - 1.00, 0.01 - 0.05, and 0.03 - 0.05 ppm of Fe, Pb, and Cd in water samples respectively. Interestingly, all the well-water from all the study locations was not contaminated with Pb despite its high concentrations in the processing water. Meanwhile both the processing water and well water were contaminated with Fe and Cd. These revealed that artisanal gold mining is contributing to the pollution of surface water bodies (used for irrigation and drinking) with Pb, while Fe and Cd may be in addition to their abundance in natural soil deposits.

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#### Introduction

Water serves a vital role in all life processes for being an essential component of all forms of life on earth. There are two major natural sources of domestic water; surface water and groundwater (Awomeso et al., 2010). Most rural communities in Nigeria use surface water directly without undergoing any water treatments for drinking and domestic purposes including irrigation. Artisanal mining in many rural communities in Nigeria has resulted in the release of many heavy metals into soil and water bodies (Abdu and Yusuf, 2013; Nuhu et al., 2014; Yahaya et al., 2020). The persistence, non-biodegradability, and toxic nature of heavy metals have rendered them a major threat to the environment (Islam et

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al., 2017; Wuana and Okieimen, 2011; Yahaya et al., 2022). Although some heavy metals such as iron, copper, zinc, and nickel are essential to some life processes, they are required in small quantities, while at a higher concentrations are toxic to both plants and animals (Kumar and Trivedi, 2016). The heavy metals in the water bodies can move up the trophic levels in the food chain accumulating in human bodies. These heavy metals can then combine with DNA, proteins, and enzymes molecules to form stable and highly bio-toxic compounds, thus their normal affecting functions and preventing them from undergoing bioreactions, which leads to genotoxic, carcinogenic, organs failure, and mutagenic effects (Mishra et al., 2019).

The realization of the economic benefits of gold mining has increased the artisanal mining activities in Zamfara State Nigeria, especially Anka local government. The miners have no knowledge or regard for the harmful effects of illegal mining activities. It was reported that approximately 163 persons lost their precious lives to heavy metals contamination in some villages of Anka's local government of Zamfara State Nigeria (Ogelekaa and Alaminiokumab, 2020). This was attributed to the illegal mining of gold and other minerals like chromite, limestone, and gypsum. The environmental consequences of illegal mining are particularly due to the nature of the chemical processes involved in gold extraction. In virtually all the villages, the mercury amalgamation method of gold extraction was used. The process includes manual digging of the metal ore, grinding of the extracted ore, washing of the ground soil, amalgamation using mercury, and melting of the amalgamated product to extract the gold from the ore (Nnaji and Omotugba, 2016). This process normally degrades and pollutes the environment, hindering the process of sustainable development, producing a morass of hazardous waste, and using large water. Considering all these negative effects, the need to evaluate the quality of surface water in relations to heavy metal concentration in all the gold mining areas of Zamfara state becomes crucial since water from these sources has been used for irrigation, livestock activities, domestic uses, and drinking. In addition, information from this study is necessary for developing strategies remediation site and for urban environmental quality management. The main objective of this study is to assess the status of heavy metal pollution in waters (well and ore-processing water) in the chosen villages of Anka Local Government in Zamfara State, Nigeria.

### **Chemicals and reagents**

All experimental reagents used in this study were of analytical grade. This is to guarantee the accuracy of experimental data. Deionized distilled water was used in experimental processes. The all the chemicals used in this research were obtained from Sigma-Aldrich with an analytical grade without further purification. These include nitric acid, and distilled water.

## **Study location**

The study location is Anka Local Government Area in Zamfara State, Nigeria. It was made up of six (6) towns (

Figure 1) including Anka town (control site) with no record of mining activities. The sampling sites were selected due to their prominent intense artisanal mining activities. Since 2010, all the sites were confirmed to be contaminated areas (Joint and Unit, 2010). The area has a typically tropical climate with dry and rainy seasons. The dry season spans from late October to May while the rainy season starts in early May and terminates in October. The harmattan season is in the dry season December spanning from barely to February every year. The mean annual rainfall is  $899 \pm 16$  mm with a mean temperature of  $38.8 \pm 2.2$  <sup>0</sup>C (Ogelekaa and Alaminiokumab, 2020). The major work/employment of the people in the area includes farming, mining, trading, and agricultural processing. The abundance of many mineral ores attracted people to mining activities. The process of mining includes manual digging of the metal ore, grinding of the extracted ore, washing of the ground soil, amalgamation with mercury, and melting of the amalgamated product to extract the gold from the ore (Nnaji and Omotugba, 2016).

## Materials and Methods

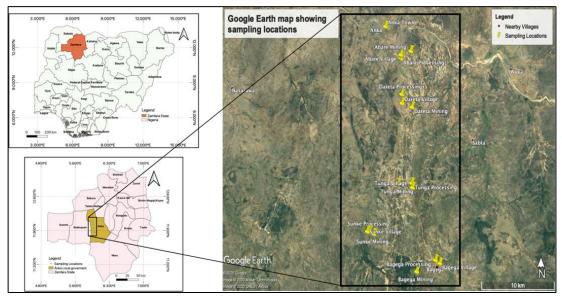


Figure 1. Map of Nigeria – Zamfara State – Anka Local government depicting the study locations

#### Sample collection

Sixteen sampling sites were identified with the coordinates and altitudes detailed in table 1. At each site, three (3) subsamples were randomly collected in a pre-cleaned 1liter sampling bottle. Each bottle was rinsed 3 times at the sampling point before sampling the water. The water collected was preserved before analysis by acidifying using 2 cm<sup>3</sup> of conc. HNO<sub>3</sub> to suppress microbial activities (Abdu and Yusuf, 2013)

# Determination of heavy metals concentration

Heavy metals concentration of water samples was determined using the digestion method as described by the APHA procedure (Nuhu *et al.*, 2014). In the procedure,  $1000 \text{ cm}^3$  of the preserved sampled water were put into a beaker followed by addition of  $50 \text{ cm}^3 \text{ conc. HNO}_3$ and heated to boiling. It was then evaporated to  $20 \text{ cm}^3$  on a hot plate. The addition of HNO<sub>3</sub> and heating were continued until digestion was completed through observation of colorless, and clear solution. The digest was then turned into plastic bottles, diluted to  $100 \text{ cm}^3$  mark, and cooled. The solution was then used for heavy metal determination.

The concentrations of metals in the sample solutions were measured using atomic absorption spectrophotometry. The heavy metals analyzed include lead (Pb), iron (Fe), Nickel (Ni), zinc (Zn), chromium (Cr), and cadmium (Cd).

|                   |               | GPS Coo                               | rdination                            |                 |
|-------------------|---------------|---------------------------------------|--------------------------------------|-----------------|
| Locations         | Sampling site | Latitude                              | Longitude                            | Altitude (masl) |
| Bagega Mining     | 1             | 11 <sup>°</sup> 51.494 <sup>1</sup> N | 5 <sup>0</sup> 56.299 <sup>1</sup> E | 386m            |
| Bagega Processing | 2             | 11 <sup>°</sup> 51.874 <sup>1</sup> N | 5 <sup>°</sup> 59.760 <sup>1</sup> E | 393m            |
| Bagega Village    | 3             | 11 <sup>0</sup> 51.486 <sup>1</sup> N | 5 <sup>°</sup> 54.394 <sup>1</sup> E | 390m            |
| Dareta Mining     | 4             | 11 <sup>°</sup> 59.196 <sup>1</sup> N | 5 <sup>°</sup> 59.507 <sup>1</sup> E | 386m            |

Table 1. Geographic coordinates and altitudes of the sampling points

| Dareta Processing | 5  | 12°02.331 <sup>1</sup> N              | 5 <sup>°</sup> 57.391 <sup>1</sup> E | 358m |
|-------------------|----|---------------------------------------|--------------------------------------|------|
| Dareta Village    | 6  | 12°02.322 <sup>1</sup> N              | 5 <sup>°</sup> 57.320 <sup>1</sup> E | 349m |
| Sunke Mining      | 7  | 11 <sup>°</sup> 53.180 <sup>1</sup> N | 5 <sup>0</sup> 56.656 <sup>1</sup> E | 389m |
| Sunke Processing  | 8  | 11 <sup>°</sup> 53.865 <sup>1</sup> N | 5 <sup>0</sup> 55.185 <sup>1</sup> E | 371m |
| Sunke Village     | 9  | 11°53.718 <sup>1</sup> N              | 5°55.339 <sup>1</sup> E              | 366m |
| Tunga Minning     | 10 | 11°57.303 <sup>1</sup> N              | 5 <sup>0</sup> 57.419 <sup>1</sup> E | 375m |
| Tunga Processing  | 11 | 11 <sup>0</sup> 53.865 <sup>1</sup> N | 5 <sup>°</sup> 55.185 <sup>1</sup> E | 371m |
| Tunga Village     | 12 | 11°56.932 <sup>1</sup> N              | 5 <sup>°</sup> 57.989 <sup>1</sup> E | 385m |
| Abare Mining      | 13 | 12 <sup>0</sup> 05.280 <sup>1</sup> N | 5 <sup>°</sup> 57.216 <sup>1</sup> E | 363m |
| Abare Processing  | 14 | 12 <sup>0</sup> 04.355 <sup>1</sup> N | 5 <sup>°</sup> 57.324 <sup>1</sup> E | 347m |
| Abare Village     | 15 | 12 <sup>0</sup> 04.696 <sup>1</sup> N | 5 <sup>°</sup> 57.324 <sup>1</sup> E | 349m |
| Anka town         | 16 | 12 <sup>0</sup> 06.648 <sup>1</sup> N | 5 <sup>°</sup> 56.443 <sup>1</sup> E | 318m |

## Risk assessments of heavy metals pollution

#### **Enrichment Factor (EF)**

The enrichment factor is an index used to assess the origin of heavy metals contamination. The origin the of contamination be either may from anthropogenic sources or from the natural processes in the environment. A reference element (most abundant element) is often used as a normalizer to calculate the EF. In this study, iron was used as the reference element due to its abundance in the soil of the study area (Yahaya et al., 2021). The EF is calculated using equation 1.

## EF= (Mcw/REcw)/(Mnc/REnc) Equation (1)

Mcw and REcw represent the concentration of heavy metal and reference heavy metal in contaminated water, and Mnc and REnc are the concentration of heavy metal and reference heavy metal in control water respectively.

An EF of < 2 indicates that there is no anthropogenic pollution due to the heavy metal, 2 - < 5 indicates moderate enrichment, 5 - < 20 indicates significant enrichment, 20 - < 40 indicates very high enrichment, and  $\geq 40$  indicates extremely high enrichment due to anthropogenic activities respectively (Sutherland, 2000).

#### **Contamination Factor (CF)**

The contamination factor (CF) is used determine severity of the to the pollution/contamination of a particular element using the fraction of the concentration of the heavy metal in the medium over the maximum allowable concentration of that particular heavy metal (equation 2) (Lacatusu, 2000).

CF= (Concentration of heavy metal in the medium)/ (Maximum allowable concentration of the heavy metal) Equation 2.

A CF of  $\leq 1$  indicates a low degree,  $1 \leq 3$ indicates moderate contamination,  $3 \leq 6$ indicates considerable contamination and  $\geq$  6 indicates very high contamination (Adewumi, 2020; Islam *et al.*, 2017).

#### **Pollution Load Index (PLI)**

This index determines the combined heavy metal contamination status of a given area. It is calculated as the geometric mean of the calculated contamination factors (Yahaya et al., 2021).

PLI=  $[(CF1 \times CF2 \times CF3 \times ..., CFn)]$ ^(1/n) equation (3)

The PLI value of < 1 indicates no pollution, 1 < 2 indicates moderate pollution, 2 < 3 indicates heavy pollution, and  $\geq$  3 indicates extreme pollution (Wang et al., 2010).

### **Statistical analysis**

Each water sample was analyzed in three replications, and a standard sample was used in between every three experimental samples. To ensure the accuracy of data, experimental blank and field blank samples were also analyzed.

Data obtained were subjected to analysis using Microsoft Excel (Version 2016) and SAS (Version 9.0).

#### **Results and Discussions**

## Heavy metals status in the water and the allowable concentrations

In this study, the concentrations of six heavy metals (iron (Fe), lead (Pb), cadmium (Cd), chromium (Cr), zinc (Zn), and nickel (Ni) in water samples from the wells and ore processing waters were determined. The samples revealed the presence of lead (figure 2), iron (figure 3), and cadmium (figure 4), and their concentrations were compared with the international organizations' standards such as United State Environment Protection Agency (USEPA), World Health Organization (WHO), and Environment Protection Agency European Commission (EPA-EUC). Heavy metal concentration in drinking water in the study area was assessed by (various) authors (Adewumi, 2020; Nuhu et al., 2014; Ogelekaa and

Alaminiokumab, 2020; Yahaya et al., 2021). Each author has stated the concentrations of the heavy metals with their maximum permissible limits. None of the samples showed the presence of chromium, zinc, and nickel. This is in contrast to the finding of Nuhu et al. (2014), where nickel was recorded in the well-water of some of the study locations. Adewumi (2020) recorded high chromium in some places and attributed it to the illegal mining of chromite deposits located in the area. Although zinc was recorded by Ogelekaa and Alaminiokumab (2020), it's far below the maximum allowable limits set by different organizations. The result obtained revealed that all the ore processing water contains a high concentration of iron, lead, and cadmium. Interestingly, none of the well-water recorded lead, but all have a high concentration of iron and cadmium. All the recorded concentrations of iron, lead, and cadmium was found to be above the permissible limits of WHO, USEPA, and EPA-EUC (EPA-EUC, 1998; USEPA, 2016; WHO, 2004).

The concentration of Pb found in the ore processing water ranged from 0.40 ppm to 132.80 ppm (Figure 2). These far exceeded the maximum permissible limit of 0.01 ppm (WHO) and 0.05 ppm (USEPA) for drinking water. The maximum permissible limit is of concern as Pb is a toxic metal that can cause impairment to nervous systems (particularly in children) which can result in brain and blood disorders (Metryka et al., 2021). Hematological damage is well noted with high Pb absorption, which is one the most serious and of important biochemical effects of Pb in drinking water (Ehi-Eromosele and Okiei, 2012). The presence of a high concentration of Pb in the ore processing water is a serious matter of worry and the potential for human contact, as the water is discharged to the main water bodies, and in some instances, the miners drink the water directly without any cautions (Mohammadi et al., 2019). This calls for immediate awareness and remediation studies as the accumulation of this metal has a cumulative fatal effect and

is a probable human carcinogen (Ghaffari *et al.*, 2021).

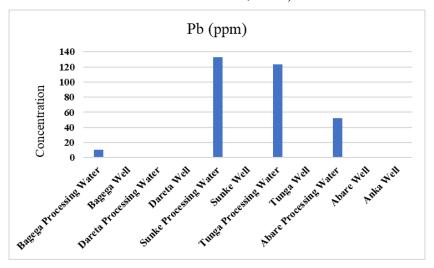


Figure 2. Mean lead (Pb) concentrations (ppm) in water samples of the study area

All the samples revealed a detectable concentration of iron and cadmium, and all have a concentration above the maximum permissible values for surface and drinking water. Cadmium ranged from 3.3 ppm to 4.1 ppm, while iron ranged from 23.3 ppm to 921.46 ppm respectively (figure 3 and figure 4). The maximum tolerable limit for Cd is 0.005 ppm (USEPA) and 0.003 ppm (WHO and EPA-EUC), and that of Fe is 0.3 ppm (USEPA), 1.0 ppm (WHO), and 0.2 ppm (EPA-EUC) respectively. Thus, the whole samples analyzed showed pollution of Fe and Cd including the control site (Anka town). Therefore, all the water

samples were not suitable for drinking purposes until further purified. The higher concentrations of Fe and Cd in both the ore processing water and the well-water may not be due to mining activities alone, as much literature (Amusan *et al.*, 2005; Solomon *et al.*, 2016; Yahaya *et al.*, 2021) reported that the soils of the study area including Anka town (control site) contain an abundant amount of Fe and Cd. Adewumi (2020) opined that the metals in the water samples are both from mining activities and geogenic, as geogenic sources contributed an important role in the discharge of toxic heavy metals.

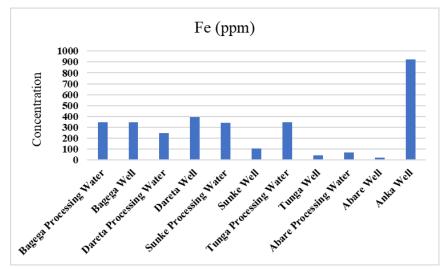


Figure 3. Mean iron (Fe) concentrations (ppm) in water samples of the study area

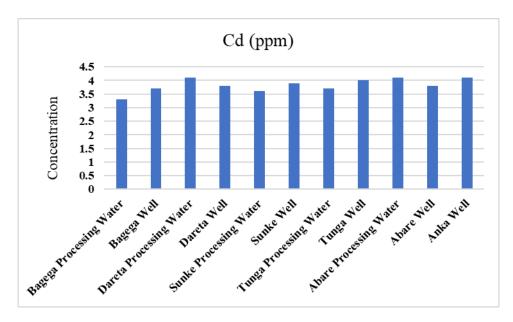


Figure 4. Mean cadmium (Cd) concentrations (ppm) in water samples of the study area

Among the detrimental effect of Cd is its accumulation in the human body (especially kidney and liver (Genchi et al., 2020)) throughout life. Its toxicity is primarily in the proximal tubular cells of the kidney, which is the main site of accumulation, causing bone demineralization directly or indirectly because of renal dysfunction ((Bernard, 2008)). Based on the epidemiological data, breast, kidney, pancreas, prostate, nasopharynx, and lung cancer were all associated with Cd toxicity (Genchi et al., 2020). Although Fe is an essential element for the growth and development of organisms such as electron transport, DNA synthesis, and oxygen transport, excess amounts beyond the permissible limits can cause tissue damage, anemia. iron overload, and neurodegenerative diseases (Abbaspour et al., 2014).

# Risk assessments of heavy metals in the water samples

The enrichment factor was used to evaluate the origin of the heavy metals in the water samples. **Error! Reference source not found.** shows the results of the enrichment factors of Pb and Cd. The results revealed that Bagega, Sunke, Tunga, processing and Abare waters have extremely high to very high enrichment of Pb. None of the well water shows any enrichment of Pb. This indicates that artisanal mining has a strong influence on the enrichment of the ore-processing water with Pb. This agrees with the finding of Nuhu et al. (2014) and Adewumi (2020) of some of the study locations. Therefore, based on the rating of Sutherland (2000), all the processing water except for Dareta, has an enrichment of Pb due to the anthropogenic activities in the study area. As for Cd, virtually all the water samples (except for Anka well (control site)) show a moderate to very high enrichment of Cd (table 2). The absence of Cd enrichment in Anka well suggests that enrichment of well waters in the mining communities is directly related to the mining activities. From the result of Ogelekaa and Alaminiokumab (2020), it was revealed that even after remediation of some of the study contaminated sites, Cd still recorded a very high enrichment factor, indicating that in as much as illegal mining is taking place, Cd will always record a high EF value, making it a more sensitive heavy metal.

| Locations               | Enrichment Factor of Pb |    | Enrichment Factor of Cd |    |
|-------------------------|-------------------------|----|-------------------------|----|
| Bagega Processing Water | 23.92                   | VE | 2.14                    | ME |
| Bagega Well             | 0.00                    | NE | 2.41                    | ME |
| Dareta Processing Water | 1.24                    | NE | 3.71                    | ME |
| Dareta Well             | 0.00                    | NE | 2.17                    | ME |
| Sunke Processing Water  | 296.87                  | EE | 2.36                    | ME |
| Sunke Well              | 0.00                    | NE | 8.08                    | SE |
| Tunga Processing Water  | 275.20                  | EE | 2.41                    | ME |
| Tunga Well              | 0.00                    | NE | 21.66                   | VE |
| Abare Processing Water  | 590.33                  | EE | 13.57                   | SE |
| Abare Well              | 0.00                    | NE | 36.65                   | VE |
| Anka Well               | 1.00                    | NE | 1.00                    | NE |

 Table 2. Enrichment factor of the heavy metals in water from all the study locations in Anka

 Local Government, Zamfara State

NE = no enrichment; ME = moderate enrichment; SE = significant enrichment; VE = very high enrichment; EE = extremely high enrichment.

Contamination factor and pollution load index were used to assess the severity and status of contamination of Fe, Pb, and Cd in the study area. Table 3 shows the CFs and PLI of the heavy metals. The CFs show that all the study areas were contaminated with Fe and Cd, and only Sunke, Tunga, and Abare processing waters were contaminated with Pb. This shows that the high concentration of the heavy metals in the soils of the study locations contributed to the contamination of the water samples with heavy metals through geogenic the processes (Adewumi, 2020). Remember as earlier reported that Fe and Cd were abundant in the soil of the study area including Anka the control site (Amusan et al., 2005; Solomon et al., 2016; Yahaya et The PLI revealed that al.. 2021). cumulatively all the ore-processing waters have heavy to extreme pollution of the heavy metals. Looking at the results, it can be concluded that artisanal mining activities greatinfluence have a over Pb contamination in the study location, while Fe and Cd are in addition to their abundance in the soil of the study area.

Table 3. Contamination factors and pollution load index of the heavy metals in water from all<br/>the study locations in Anka Local Government, Zamfara State

| Locations               | CF Fe   | CF Cd     | CF Pb   | PLI     |
|-------------------------|---------|-----------|---------|---------|
| Bagega Processing Water | 2.33 VC | 283.34 VC | 0.54 LC | 7.43 EP |
| Bagega Well             | 2.61 VC | 285.24 VC | 0.00 LC | 0.16 NP |
| Dareta Processing Water | 2.89 VC | 221.18 VC | 0.02 LC | 2.41 HP |

| Dareta Well            | 2.68 VC | 319.83 VC | 0.00 LC | 0.17 NP  |
|------------------------|---------|-----------|---------|----------|
| Sunke Processing Water | 2.54 VC | 304.71 VC | 6.64 VC | 17.45 EP |
| Sunke Well             | 2.75 VC | 119.95 VC | 0.00 LC | 0.11 NP  |
| Tunga Processing Water | 2.61 VC | 305.05 VC | 6.17 VC | 17.21 EP |
| Tunga Well             | 2.82 VC | 73.75 VC  | 0.00 LC | 0.08 NP  |
| Abare Processing Water | 2.89 VC | 102.20 VC | 2.61 CC | 7.84 EP  |
| Abare Well             | 2.68 VC | 58.67 VC  | 0.00 LC | 0.06 NP  |
| Anka Well              | 2.89 VC | 696.33 VC | 0.01 LC | 2.95 HP  |

 $\overline{CF}$  = Contamination factor; PLI = Pollution load index; LC = low contamination; CC = considerable contamination; VC = very high contamination; NP = no pollution; HP = heavy pollution; EP = extreme pollution.

#### Conclusion

The study was conducted to assess the consequence of artisanal gold mining on heavy metals exposure to surface water in Anka local government, Zamfara State. The lucrative nature of the gold mining business makes many people living in those areas to be directly involved in artisanal mining activities, resulting in the pollution of the vast water bodies and area of land. It was observed that three heavy metals (Fe, Pb, and Cd) were all above the maximum permissible limits of the heavy metals in drinking water fixed by different international organizations. The contaminated/polluted water poses a very high risk to Fe, Pb, and Cd toxicities. The findings revealed that illegal mining activities are contributing to the pollution of water bodies with Pb, while Fe and Cd are in addition to their abundance in natural soil deposits through geogenic processes. In the future, research should be conducted to carry out epidemiological studies on contaminated environmental media. Finally, we recommend that periodic monitoring and evaluation should be conducted in the area to assist the government and other policymakers in mitigation strategies

#### **Conflict of Interest**

No known conflict of interest for the research.

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