



## Impact of Pb-Zn Mining on Heavy Metal Levels in Soil from Arufu Mine Field, Wukari, Nigeria

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### Authors' contributions

This work was carried out in collaboration among all authors. Author GGY conceived the idea and designed the study. Author AMM performed the literature searches. Author RO performed the statistical analysis and wrote the first draft of the manuscript. Author BNH wrote the final manuscript and author JY assisted in the statistical analysis. All authors participated in the laboratory work, proofread and approved the final manuscript.

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### ABSTRACT

Analysis of soil around the vicinity of Arufu mine field was conducted in order to ascertain the impact of mining on the environment. Concentrations of trace metals associated with Pb – Zn – barite mineralization were determined in soils using atomic absorption spectrophotometer (AAS) Varian AA 240, after acid digestion with a mixture of 1:1 hydrofluoric acid and aqua regia. Mean concentrations ( $\mu\text{g/g}$ ) of metals ranged from 5.72 (As) – 451.44 (Pb) in dry season and 1.32 (Cd) – 504.61 (Pb) in wet season. Index of geoaccumulation were in the following order of increasing magnitude:  $\text{Cu} < \text{Zn} < \text{As} < \text{Pb} < \text{Ag}$  for dry season and  $\text{Cu} < \text{As} < \text{Zn} < \text{Cd} < \text{Pb}$  for wet season. On the other hand, soil enrichment ratio ranged from Cu (1.06) – Ag (126.18) and Cu (1.35) – Pb (34.10) in dry and wet season respectively. Levels of metals measured were higher than their corresponding values reported for soil in the upper continental crusts. Toxic metal (Pb) has its mean concentration above maximum tolerance level while Zn and As levels were below the maximum tolerable levels. The presence of Pb above maximum tolerance level may render the soil unsuitable for agricultural practices, hence high impacts of Pb – Zn mining in the study area.

*Keywords:* Trace metals; geoaccumulation; mining; Arufu, geochemical.

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## 1. INTRODUCTION

Nigeria is endowed with several mineral resources of economic importance and these minerals have directly or indirectly played an important role in social and economic development of the country through job and wealth creation. Mining and processing of minerals is associated with large number of environmental degradation issues and hazards at different stages [1]. Over 37 different mineral resources were found across Nigeria, but attention has mainly been paid to 9, including iron ore, tin, bitumen, coal, gold, columbite and tantalite, lead/zinc, and wolframite [2]. Lead/zinc mineralization along the Benue trough is often associated with other minerals and ores such as barite, copper gangue, and marcasite among others [3]. Impacts of mining activity on the environment has called for concern due to enormous amount of toxic substances released into soil, water and air, land degradation and deforestation. Difficulty in closure of pits created and disposal of rock waste and tailings means alteration of natural landscape [4]. These pits have been reported to be dead traps when filled with water and homes to dangerous reptiles like snakes as in the case of tin and columbite mines in Jos plateau [5] and Azara barite mines in Nasarawa State [6]. The transportation and distribution of the discharged toxic substances such as mercury, cyanide, arsenic, lead, and cadmium in the environment is aided by the action of wind and storm water on mining waste and tailing materials. The presence of heavy metals contaminants in soils is a serious environmental challenge on a global scale [7]. In Nigeria, the government policy on diversification of the economy is with focus on agriculture and exploitation of solid minerals. Contamination by heavy metals arising from mining and processing of all minerals will become of increasing concern with national development, industrialization and urbanization if not checked. From the knowledge of soil formations, soils naturally contain metals at trace levels as such their presence in soils is not necessarily an indication of contamination. A study conducted on Daretta soil in Zamfara state of Nigeria has attributed elevated level of Pb, Cd, Cu in the soil to the impacts of illegal gold mining by artisans in the state [8]. The effects of these chemicals have resulted to large number of death recorded and signs of neurological imbalance, and abnormal behaviors among the community members. Tailings, sands and waste rocks deposited around the mine area are main sources of heavy metals in the environment [9].

When these deposits contain sulfides (example pyrite), in the presence of oxygen and water, acid mine drainage results [10]. Transformation of sulphides and a shift to more acidic conditions enhances the mobility of elements such as manganese (Mn), iron (Fe), zinc (Zn), copper (Cu), and cadmium (Cd) [11]. Several research outcomes have also shown that heavy metals are potentially toxic to crops, animals and humans when contaminated soils and water as well as forages are used for animal and crop production [12,13,14,15]. Trace metals including lead, arsenic, mercury and cadmium have not been confirmed to perform any significant function in human biochemistry and physiology. The presence of these metals in human and other living organisms even at low concentration is a cause for concern because of their inherent toxicity. Once these contaminants get into human body, they disrupt the normal functions of vital organs through competition with and displacement of minerals of physiological importance. This study was therefore designed to assess the impacts of Pb-Zn mining on heavy metal levels in Arufu soil and its possible adverse effects on plant and animals including humans in that area.

## 2. MATERIALS AND METHODS

### 2.1 The Study Area

The studied area (Arufu) is located in the northwestern part of Wukari Local Government Area of Taraba State, North-Eastern Nigeria. It lies between Latitudes 7°50' 4"N and Longitudes 7°30' 1000" E. The area is endowed with mineral resources among which are economic deposit and lead/zinc-barytes mineralisation along the Middle Benue Trough. Galena mining is the predominant activities in the area.

### 2.2 Soil Sampling

The study area was broken into 10 sites and in each site, approximately 10–20 portions of soil samples were randomly collected at the depth of 0 – 15 cm with stainless steel hand trowel. The 10-20 soil portions were bulked together in a polyethylene bag to form composite sample to be prepared for analysis. The composite samples were thoroughly mixed, manually sorted to eliminate pebbles and coarse materials, and air-dried at less than 30°C over few days with occasional breaking of aggregated materials with wooden roller. This was followed by sieving through a nonmetallic sieve with mesh hole of 2 mm diameter to remove pebbles and debris and

obtain a homogenous representative samples ready for analysis.

### 2.3 Mineralization of Soil

This was carried out according to the method reported by Okoye et al. [14] briefly 0.5 g of each homogenized composite sample was accurately weighed into screw-capped Teflon bottle. A mixture of 5 ml HF acid and 5 ml aqua regia (1:3 HNO<sub>3</sub> / HCl V/V) was added to the bottle containing the soil. The resultant acid-soil mixture was digested in a water bath at 60°C for 3 hours during which complete decomposition of soil was achieved. The digest was allowed to cool to room temperature, followed by addition of 20 ml boric acid to form complex with residual hydrofluoric acid which would attack glassware. The digests were filtered into 50 ml standard flask using Whatman No 1 filter paper which had been profusely leached with 1:1 HCl, and the sample solution made up with the 1:1 HCl.

### 2.4 Determination of Trace Metals

The concentrations of metals in the sample solutions were determined using Varian AA240 Atomic Absorption Spectrophotometer equipped with Zeeman's background correction (SR-BDG).

Prior to the sample analysis, the flame conditions were optimised for maximum absorbance and linear response while aspirating known standards. The standards were prepared from individual 1000 ppm stock solution supplied by the manufacturer. 100 mL of the combined standards were prepared in 0.1M HNO<sub>3</sub>. The lamp current was automatically selected for each metal. The pre-spray time, integration time and response time were 3, 5, and 1 respectively. Reproducibility that is better than 1.0% was considered and all the metal concentrations were expressed on a dry weight basis.

In order to check the reliability of the analytical procedure employed for heavy metals determination, replicate digestion and analysis of the samples together with blank were carried out. Data obtained were subjected to statistical tests of significance using the one way analysis of variance (ANOVA) to assess significant variation in the levels of the trace metals in the soils from the studied area. Probability less than 0.05 ( $p < 0.05$ ) was considered statistically significant. Correlation coefficient was used to determine the association between the trace metals in the samples at  $\alpha = 0.05$ . All statistical analyses were done by SPSS 17.0 for windows.

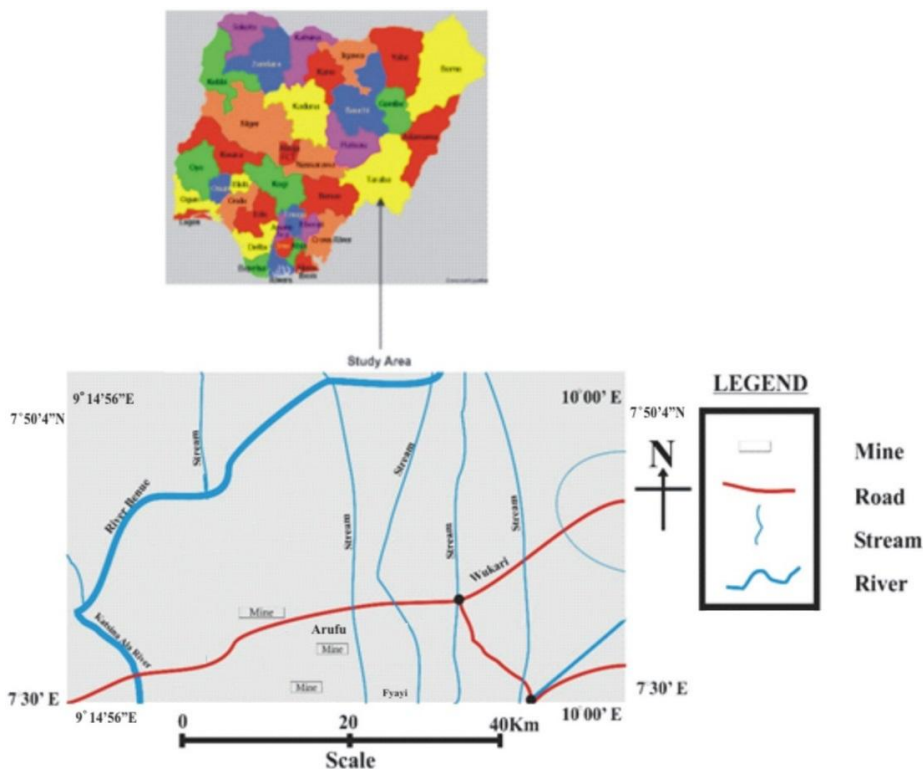


Fig. 1. Map of the study area showing sampling sites

### 3. RESULTS AND DISCUSSION

Results of the geochemical analysis of soil samples around Arufu mine field in dry and wet season were: Mean concentrations ( $\mu\text{g/g}$ ) and standard deviations in dry season; Ag ( $6.94 \pm 1.18$ ), As ( $5.72 \pm 2.89$ ), Cu ( $26.44 \pm 11.54$ ), Pb ( $451.44 \pm 87.69$ ), Zn ( $147.63 \pm 66.73$ ) and wet season; As ( $7.86 \pm 2.58$ ), Cd ( $1.32 \pm 0.52$ ), Cu ( $33.63 \pm 18.48$ ), Pb ( $504.61 \pm 375.43$ ), Zn ( $429.07 \pm 241.83$ ) while Cd and Ag were not detected in dry and wet seasons respectively. Metals in the soil around the mine field had higher concentrations in wet season than in the dry season.

Comparing the mean concentrations obtained in dry and wet seasons, test for equality of mean revealed the following: *P* – value of Zn (= 0.001) is less than 0.05 level of significance, implying the mean of Zn for wet season is significantly greater than for dry season. *P* – Values of Cu (= 0.311), As (= 0.098), Pb (= 0.613) are each greater than 0.05 level of significance as such, there is no significant difference in their means for wet and dry seasons. Multiple comparison of mean using Dunnett C, showed a significance difference ( $p < 0.05$ ) in the mean of Arufu mining field. The mean concentration of Cu in Arufu was significantly higher in magnitude ( $p < 0.05$ ). Duncan multiple test revealed that, the mean of As in Arufu mine field is significantly greater ( $p < 0.05$ ) than means of control area (Table 1).

There was an inter-element Pearson correlation matrix in the soil (Table 2). A very high positive correlation with high significant probability ( $p < 0.01$ ) was seen between Zn/Cu. The above observations reflected common association of these metals with Pb – Zn ores. Lead showed a special behavior in soil and correlate positively ( $p < 0.01$ ) with Ag and negatively with As ( $p < 0.05$ ). These observations tend to agree with the report that, Pb-Zn mineralization may contain traces of Ag responsible for its silver shining appearance.

The studied area is highly polluted with Pb and Zn, according to [16], which showed levels of metals such as Pb, Cd to be above maximum tolerance level and As, Cu, Zn below maximum tolerable level (Cd  $< 0.1$ , As 1 – 20, Cu 30 - 60, Pb 2 – 13.4, Zn 60 – 780 ppm [17]. The very high concentration obtained for Pb and Zn, indicates contributions from mining activity, dissolution of rock or sulphide minerals. Arsenic mean concentration both in dry and wet season was the least while Pb has highest concentrations in

dry and wet season. The mean concentration in order of decreasing magnitude were: Pb > Zn > Cu > Ag > As > Cd for wet season and Pb > Zn > Cu > As > Cd > Ag for dry season. In the control site, Ag were found below the detection limit (0.0001  $\mu\text{g/g}$ ).

The most common and frequent route of trace elements exposure in human is through ingestion and inhalation. Mean concentration of Ag (6.94  $\mu\text{g/g}$ ) was about 126 higher in magnitude than 0.055 ppm in the upper continental crusts and about 8.7 fold higher than reported for Luku mining area [9]. This generally revealed pollution of the study area and support the association of Ag with Pb-Zn mineralization in the middle Benue trough. Arsenic in the literature has been shown to occur in relatively large amount with Au, Cu, Ni and Ag bearing ores as well as in regions where these ores were found. Arsenic in the body can cause cancer of the skin, lungs, urinary bladder, kidney, and death from respiratory and cardiovascular failure in human [18,19]. Mean concentration of arsenic in this study is about 3 times greater than its concentration in upper continental crust [20].

The Mean concentration of Cu (26.44  $\mu\text{g/g}$ ) obtained in this study was 1 time higher in magnitude than its guideline in upper continental crusts, about 2 times higher than published for Luku mining field [9]. The concentration of lead ranged from 2.6 – 25  $\mu\text{g/g}$  in natural soil [21]. However, in this study, mean concentration of Pb was 451.44  $\mu\text{g/g}$  which is 30.50 higher in magnitude than in upper continental crusts [22] and about 2 fold in magnitude than 300  $\mu\text{g/g}$  EU regulation commission safe limits in soils [23]. This indicates high level of Pb in the mining fields and the Arufu area. Hence, the soils may not be suitable for agricultural practices. Lead gets accumulates in plants and animals through water and feeds which passes across to humans through the food chain, drinking water and ingestion of contaminated soil especially by children [24]. In humans, Pb has been reported to cause brain and kidney problems [25].

Zinc is a vital micronutrient identified to affects several metabolic processes in plants when present in a very small quantity but at elevated level Zn can lead to chlorosis in leaves, stunted plants growth and kidney failure in human. In the present study Zn mean concentration (147.63  $\mu\text{g/g}$ ) was 2.27 fold in magnitude than 65 ppm published for upper continental crust, 3.52 times higher than the world average of 42 ppm, 2 times

lower than value reported for Luku and Abakaliki abandoned mine [26]. The elevated Zn level above its value in the upper continental crust and soil worldwide indicates its pollutional status in the study area. The suspected pollution may be attributed to the mining activity, geology and mineralogical composition of the area.

### 3.1 Assessment of Trace Metal Accumulation and Pollution in the Environment

Index of Geological Accumulation and Enrichment Ratio were calculated to help in assessment of pollution levels and trace metals origin. These indices compared the observed concentrations and the background concentrations and are indicative of degree of anthropogenic pollution. The background concentration used in this study was the original geochemical background in the upper continental crust.

**Index of Geological Accumulation (Igeo):** The index of geoaccumulation was calculated and interpreted as follows [27]:

$$I_{geo} = \text{Log}_2 \left( \frac{C_{\text{metal soil}}}{1.5 \times C_{\text{metal background}}} \right) \quad (1)$$

Where  $C_{\text{metal soil}}$  is the metal concentration in the enriched sample and  $C_{\text{metal background}}$  is the background concentration of metal in unpolluted soil. A factor 1.5 was used to account for variations in the background or control values

which may be attributed to lithogenic variations and minimized over estimation of population level. The degree of metal pollution was assessed in terms of seven contamination classes based on the increasing numerical value of the index as follows:

$I_{geo} < 0$  means unpolluted,  $I_{geo} \geq 0 < 1$  means unpolluted to moderately polluted,  $I_{geo} \geq 1 < 2$  means moderately polluted,  $I_{geo} \geq 2 < 3$  means moderately to strongly polluted,  $I_{geo} \geq 3 < 4$  means strongly polluted,  $I_{geo} \geq 4 < 5$  means strongly to very strongly polluted,  $I_{geo} \geq 5$  means very strongly polluted.

Results obtained for degree of metals pollution in Figure 2 revealed that soil in the study area is not polluted by Cu (-0.63) in the two seasons with  $I_{geo}$  values less than zero. On the other hand, the soil is in the level of unpolluted to moderately polluted by Zn (0.42) and As (0.73) in dry season with  $I_{geo}$  values greater than zero but less than 1 and moderately polluted in wet season with  $I_{geo}$  value (Zn 1.97, As 1.29) greater than 1 but less than 2. Lead (1.78) with  $I_{geo}$  value greater than 1 but less than 2 means the soil was moderately polluted during the dry season but strongly to very strongly polluted in the wet season with  $I_{geo}$  value equals to 4. The soil was also found to be moderately polluted by Cd (3.00) and very strongly polluted by Ag (6.37). The extent of metals pollution in the soils can be summarized in a decreasing order of  $I_{geo}$  value as follows:  $Ag > Pb > As > Zn > Cu$  and  $Pb > Cd > Zn > As > Cu$  in dry and wet seasons respectively.

**Table 1. Test for equality of variances and means of metals in soils for dry and wet seasons**

	As	Cu	Pb	Zn
F	0.163	2.788	0.319	5.016
Sig.1 tailed*	0.691	0.112	0.579	0.038
Sig.2 tailed**	0.098	0.311	0.613	0.001
Df	18.00	18.00	18.00	18.00

\*test for equality of variances, \*\*test for equality of means

**Table 2. Inter-elements Pearson’s correlation for soil samples from Arufu mining field.**

ARUFU	Zn	Cu	As	Ag	Pb
Zn	1				
Cu	0.614**	1			
As	-0.067	0.318	1		
Ag	0.111	-0.334	0.286	1	
Pb	-0.213	0.059	-0.281	-0.382	1

\*Pearson’s correlation significant at  $p < 0.05$ ,

\*\*Pearson’s correlation significant at  $p < 0.01$

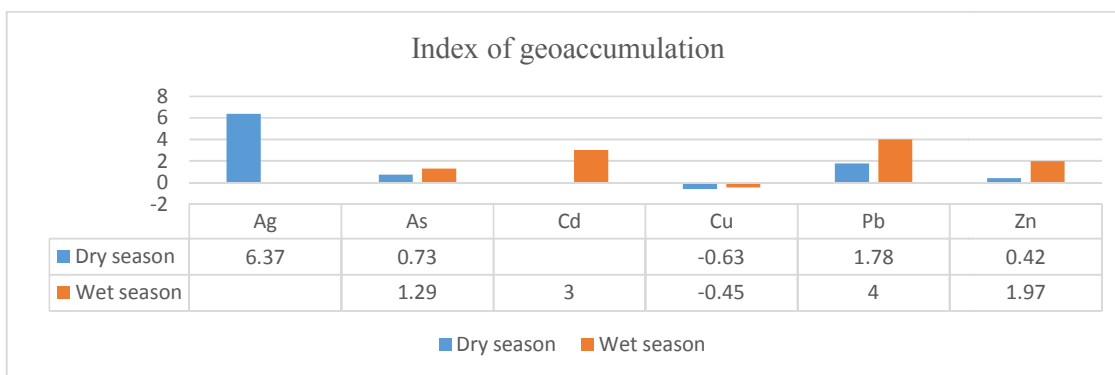


Fig. 2. Index of Geological accumulation of trace metals in soil samples from mine area

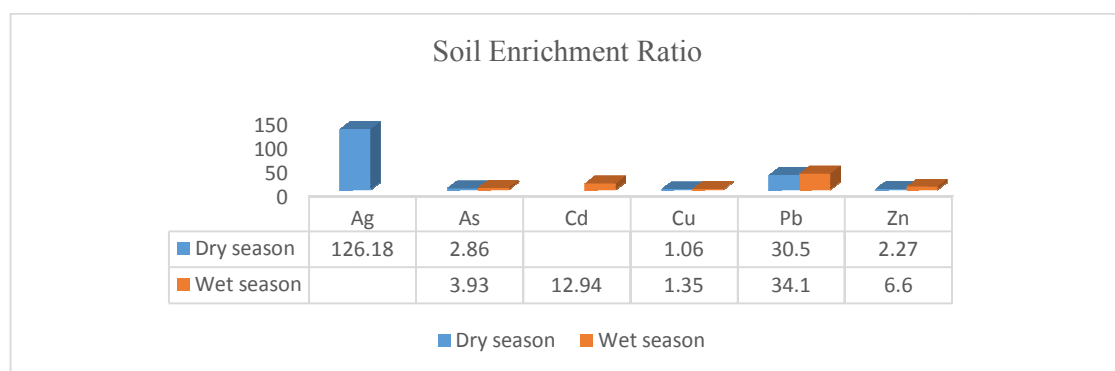


Fig. 3. Elements Enrichment Ratios (ER) in soil samples

**Element Enrichment Ratios (ER):** Element Enrichment Ratios were calculated in order to know the extent of enrichment or depletion of elements in the soils relative to the crustal concentration. Enrichment ratio (ER) was calculated using the equations:

$$ER = C_n / B_n \tag{2}$$

Where  $C_n$  is the concentration of an element measured in a sample and  $B_n$  is the background concentration, which was the standard concentration in the upper continental crust. The ER values obtained were interpreted as follows [7]:

ER < 2 (depletion to minimal enrichment), ER ≥ 2 < 5 (moderate enrichment), ER ≥ 5 < 20 (significant enrichment), ER ≥ 20 < 40 (very high enrichment), ER ≥ 40 (extreme enrichment).

The ER values obtained for Cu showed that Cu was minimally enriched compare to its background value, with enrichment ratio less than 2. As and Zn were found to be moderately enriched with enrichment ratios between 2 and 5 in dry season while soils collected in wet season

showed Zn and Cd to be significantly enriched with enrichment ratio greater than five. Cadmium was found to be significantly enriched with ER = 12.94. The result also showed that Ag is extremely enriched in the soil compared to its background values, with enrichment ratios above 40 while Pb is highly enriched relative to its background values, with enrichment ratio between 20 and 40. In view of the enrichment status of the soil, it can be suggested that Arufu soil is minimal to extremely pollute by the following elements: Cu, Zn, As, Cd, Pb and Ag. The elevated ER value obtained for soil in the study areas may be attributed to the geology of the area and the ongoing mining activity.

Pb is not an essential element to both plant and animal with humans inclusive and when present in even small quantity in any mineral or chemical form, it may be toxic. It is obvious from the ER value that, plants, animals and human in the study areas may be at risk of lead toxicity. The very high enrichment ratio calculated for Pb in this study was high both in dry and wet season. High concentration of Pb has been reported to

cause certain health risk in plants and animals [23].

Zn and As are moderately enriched relative to their background values in the study areas both in dry and wet season except for Zn which is significantly enriched in wet season. Copper is minimally enriched in the two seasons. Both As and Cu in Luku soil is about twice the value recorded in this study. Zn in Luku soil was found to be depleted while in this study, it was moderately enriched. The high concentration of Zn and As in the study area are probably related to the geology of the area and may not be controlled by the mining activity. The result of the present work agrees with high concentration of Pb, Cu and Cd reported for soils from the open cast mining of solid mineral in Nasarawa state, North-central Nigeria [28].

#### 4. CONCLUSION

The environmental impacts of Arufu mining fields was carried out and mining activity was confirmed to be a good source of trace metals in the environment which can pose severe threats to human health. The environmental pollution caused by trace metals could be for long-term. Pb, As and Cd are not required by the body and can be toxic even at low concentration. The present study has also confirmed the prevalence of Cd, Zn, Cu and Pb in the study area at alarming levels. All have their concentrations above their guideline in the upper continental crust. Wide distribution and high concentration of Pb above the tolerance levels identified the potential risks associated with lead-zinc mineralization and mining. The presence of Pb, As, Zn, Cu and Cd may therefore, affect soil quality and its ability to sustain plants and animals life.

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#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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