

Regular Article

NORM Measurements and Radiological Hazard Assessment in the Gold Mining Areas of Eastern Cameroon

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The aim of the study is to assess natural radiation exposure to the public in the gold mining areas of Eastern Cameroon. For this purpose the sodium iodide (NaI) detector was used to determine activity concentrations of natural radionuclides in soil samples. External radiation dose to the public and radiological hazards were assessed. The average activity concentrations determined for ²²⁶Ra, ²³²Th, and ⁴⁰K were 40.1 Bq kg⁻¹, 29.4 Bq kg⁻¹, and 216.9 Bq kg⁻¹, respectively in agreement with the mean values given by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The mean external annual effective dose was found to be 0.34 mSv yr⁻¹. The mean external hazard index was less than unity while the radium equivalent activity of all soil samples have shown lower values than the limit of 370 Bq kg⁻¹ defined by the Organization for Economic Cooperation and Development (OECD). Results of this study pointed out that soils examined in the gold mining areas of Eastern Cameroon can be used for buildings construction. Some recommendations were drawn to strengthen the environmental protection in mining areas.

Key words: gold mining, soil, natural radioactivity, external hazard index, radium equivalent activity

1. Introduction

Radiological exposure to the public arises from two pathways: external and internal radioactive sources. Irradiation of the human body by external sources is mainly due to gamma radiation from ²³⁸U and ²³²Th decay series and ⁴⁰K¹. At the ground state, the level of natural environmental radioactivity is low compared to world average activity concentrations of ²³⁸U, ²³²Th, ⁴⁰K given by UNSCEAR¹ but, it can be increased by

anthropogenic activities. Exposure of the general public to emissions of naturally occurring radionuclides to the environment mainly arises from industrial activities using raw materials containing the naturally occurring radioactive materials (NORM). NORM are extracted, transported, and processed for further use¹. Among the potential sources of exposure to NORM, there are: burning coal, making and using fertilizers, oil and gas production and mining operations. Although several studies were carried out on natural radiological exposure in Cameroon²⁻⁸, environmental radioactivity in mining sites remains largely unknown justifying the present study in the gold mining areas of Eastern Cameroon. In other African countries, similar studies were carried out on different gold mining sites. Augustine Faanu *et al.*^{9, 10} studied the Assessment of public exposure to

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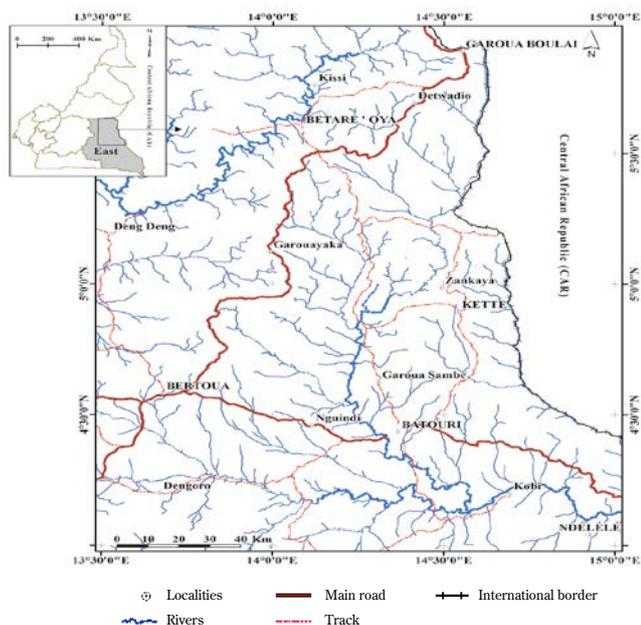


Fig. 1. Location of the gold mining areas of Bétaré Oya and Batouri (Kambélé) in Eastern Cameroon.

NORM from mining and mineral processing activities of Tarkwa Goldmine in Ghana. Sam *et al.*¹¹ have worked on radiological evaluation of gold mining activities in Ariab, Eastern Sudan. These studies reported average activity concentrations lower than the world average values given by UNSCEAR¹. Other reported studies in Nigeria¹²⁻¹⁴ concluded that average concentrations of ²³²Th found are relatively higher than the world average value¹. The objective of this study is to assess radiological exposure to the public around gold mining sites of Eastern Cameroon in order to contribute to the radiological mapping of the country and to better regulate mining activities in Cameroon. After soil sampling and conditioning, NORM measurements were carried out using Sodium Iodide detector. Sandy soils coming from mining activities are well used in the study areas for building construction. Thus external radiation dose to the public and radiological hazard associated with the use of these soils as building materials were assessed.

2. Material and methods

2.1. Study area

Bétaré Oya and Batouri are two towns located at about 176 km and 135 km respectively from Bertoua, the regional capital of East-Cameroon. The study was conducted in the gold mining sites of Bétaré Oya (latitudes 5° 35'00" to 5° 39'00"N; longitudes 14° 04'00" to 14° 07'00"E) and Batouri (latitudes 4° 35'00" to 4° 38'00" N; longitudes 14° 24'00" to 14° 26'00" E) as shown in Figure 1. The study areas are dominated by the remains of

Table 1. Air kerma conversion coefficients [(nGy h⁻¹) / (Bq kg⁻¹)] of radioactivity in soil

| Radionuclides | Air kerma conversion coefficient (nGy h ⁻¹)/(Bq kg ⁻¹) |
|--------------------------|--|
| ²³⁸ U series | 0.46 |
| ²³² Th series | 0.60 |
| ⁴⁰ K | 0.042 |

artisanal gold mining marked by the presence of several abandoned open pits scattered in areas of ancient or recent gold mining. These pits are of various sizes that can reach 200 m² and often contains stagnant water. This situation leads to a severe disturbance of the land surface and the disappearance of the cultivable land and gallery forest. The rejection of oil and fuel used for the engines, the vestiges of abandoned machines, rusty empty barrels and plastic tins, the production of large amounts of fined-grained material due to the digging of numerous pits in river sediments tends to clog up the soil and increases the risk of contamination¹⁵.

Batouri and Bétaré Oya are respectively subject to the influence of hot and humid equatorial climate Guinean classical and of tropical climate Guinean Sahelian. There is a transition between the equatorial forest to the south and the savannah to the north, height from tropical climate. The climatic conditions of the study areas are (i) a rainy season from April to October, (ii) dry season from November to March and (iii) absolute low water in March. The annual average rainfall at Bétaré Oya is 1590 mm^{15, 16}. The majority of the land is covered with sparse grass, shrubs and trees. According to the villagers, agricultural activities such as crop cultivation are being slowly abandoned; however, the remains of artisanal gold mining are much practiced.

2.2. Sampling and conditioning

Thirty two (32) soil samples were collected within selected areas of the gold mining sites of Bétaré Oya and Batouri using a random sampling basis because of the irregular form of the study areas. Each sample, with a total fresh mass of about 2 kg, was made up of a composite of material from 5 holes distributed at the 4 corners and centre of a 1 m² surface area with a maximum depth sampled from 5 cm. The soil samples were stored in sealed plastic containers, carefully labeled and taken to the Institute of Geological and Mining Research (IRGM) for analysis.

In laboratory the big particles were removed from the samples and they were dried in a UK Gallenkamp Hotbox Oven with fan size 3, for 2 days at 70°C. The samples were then pulverized using a German Fritsch Pulverisette with speed up to 60 rotations per minute (rpm), sieved at about 2 mm diameter. Each sample was weighed,

Table 2. Activity concentrations, external effective dose, radium equivalent activity and external hazard index in soil samples of gold mining sites of East Cameroon

| | Nuclides | Statistical parameters | BO1 | BO2 | BO3 | BO4 | BO5 | BA1 | BA2 |
|---|-------------------|------------------------|------------|-----------|------------|------------|-------------|------------|-------|
| Activity concentration (Bq kg ⁻¹) | ²²⁶ Ra | Range | 20.5–53.8 | 22.2–45.1 | – | 36.6–84.9 | 18.5–43.4 | 33–62 | – |
| | | Mean | 36.5 | 37.3 | 41 | 51.7 | 33.3 | 46.3 | 34.8 |
| | ²³² Th | Range | 14.7–42.5 | 18.8–34.2 | – | 27.9–58 | 11.8–30.5 | 22.4–39.6 | – |
| | | Mean | 25.1 | 28.5 | 30.4 | 40.7 | 22.3 | 31.7 | 27.03 |
| | ⁴⁰ K | Range | 80.3–443 | 221–279 | – | 182.3–582 | 152.3–437.3 | 40.6–114.6 | – |
| | | Mean | 249.5 | 255.9 | 228.9 | 339 | 217.2 | 75.5 | 152.8 |
| E_{ext} (mSv yr ⁻¹) | Range | 0.17–0.52 | 0.23–0.40 | – | 0.32–0.75 | 0.17–0.42 | 0.25–0.42 | – | |
| | Mean | 0.32 | 0.34 | 0.36 | 0.47 | 0.29 | 0.33 | 0.29 | |
| R_{aeq} (Bq kg ⁻¹) | Range | 47.7–148.6 | 66.1–113.6 | – | 93.6–212.5 | 47.1–117.6 | 73.8–123.1 | – | |
| | Mean | 91.7 | 97.8 | 102.1 | 135.9 | 81.9 | 97.4 | 85.2 | |
| H_{ext} | Range | 0.13–0.40 | 0.18–0.30 | – | 0.25–0.57 | 0.13–0.32 | 0.12–0.33 | – | |
| | Mean | 0.25 | 0.26 | 0.28 | 0.37 | 0.22 | 0.26 | 0.23 | |

homogenized and conditioned in 500 ml Marinelli beakers, completely sealed during at least 1 month to avoid radon escape. This ensures secular equilibrium between ²²⁶Ra and its daughters¹⁷.

2.3. Gamma spectrometry

2.3.1. Experimental set-up

Radioactivity measurements in soil samples were carried out using a Canberra NaI(Tl) detector (Model 802) with a crystal size of 7.6 cm × 7.6 cm, a multiple channel analyzer of 1024 channels and a resolution of 7.5% at 662 keV. The detector is mounted in a cylindrical lead chamber. This model 802 plugs directly into the Model 2007 Tube Base which provides power to the photomultiplier tube. Spectrum acquisition and analysis were carried out using GENIE 2000 software⁴.

2.3.2. Detector calibration and activity concentration calculations

The energy and efficiency calibrations of the detector were performed using a Multi-gamma ray standard MGS5M3, a customer supplied 500 ml Marinelli Beaker-resin containing ¹⁵⁵Eu, ⁵⁷Co, ¹¹³Sn, ¹³⁷Cs, ⁵⁴Mn and ⁶⁵Zn traceable to international standards and emitting gamma-rays in the energy range of 60–1115.5 keV. To measure radioactivity in soil samples, the same geometry was used with a counting time of 10⁵ seconds. The activity concentration (Bq kg⁻¹) of a given radionuclide in the spectrum was calculated using the following equation:

$$A_i = \frac{C_i}{\varepsilon(E) \cdot P \cdot M_s \cdot t_c} \quad (1)$$

Where C_i is the net peak area at energy E (keV), $\varepsilon(E)$ is the detection efficiency at energy E , P is the gamma ray emission probability at energy E , M_s is the mass of the sample in kg, and t_c is the counting time in seconds. Activity concentrations of ²²⁶Ra and ²³²Th were determined from their daughters assured to be at secular equilibrium. In this work, the gamma-ray of ²¹⁴Pb at 609.31 keV (44.8 %) was considered to evaluate activity concentrations of ²²⁶Ra. Gamma-rays of ²²⁸Ac at 911.2 keV (26.6%) and 968.97 keV (16.2 %) were used to evaluate activity concentrations of ²³²Th. The activity concentrations of ⁴⁰K were determined using the gamma-ray at 1460.8 keV (10.67%).

2.4. External annual effective dose

To estimate the external annual effective dose for adult persons in the gold mining region of East Cameroon, the conversion coefficients for ²³⁸U and ²³²Th series and ⁴⁰K listed in Table 1 were used¹. It can be obtained directly using the formula proposed by Saïdou *et al*¹⁸.

$$E_{ext} = F_c [(1 - F_{occ}) + F_{occ} F_b] \times \sum_{i=1}^3 A_i (KCF)_i \times t \quad (2)$$

Where F_c is the conversion coefficient of 0.7 Sv Gy⁻¹ used to determine the corresponding effective annual dose, F_{occ} is the indoor occupancy factor of 0.6, which implies that people spend 40% of the time outdoors. However, since the materials used in the construction of most of building contain radionuclides, the average factor F_b of 1.4 was applied to take into account their contribution and estimate the indoor rate. A_i are average activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K, $(KCF)_i$ are corresponding air kerma conversion factors showed in

Table 3. Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples of gold mining sites of East Cameroon compared to the world average value¹⁾

| Radionuclides | Meanactivity concentrations (Bq kg ⁻¹) | Range of activity concentrations(Bq kg ⁻¹) | World average value (Bq kg ⁻¹) |
|-------------------|--|--|--|
| ^{226}Ra | 40.1 | 18.5 – 85 | 33 |
| ^{232}Th | 29.4 | 11.8 – 58 | 45 |
| ^{40}K | 217 | 40.6 – 582 | 420 |

Table 4. Comparison of average activity concentrations with those of similar studies carried out in other African countries

| Activity concentrations(Bq kg ⁻¹) | | | Country | References |
|---|-------------------|-----------------|----------|----------------------------------|
| ^{226}Ra | ^{232}Th | ^{40}K | | |
| 2.38 | 32.36 | 383.8 | Nigeria | Abdullahi et al. ¹²⁾ |
| 2.39 | 52.0 | 390.9 | Nigeria | Abdulkarim et al. ¹³⁾ |
| 55.3 | 26.4 | 505.1 | Nigeria | Ademola et al. ¹⁴⁾ |
| 8.37 | 11.4 | 232.7 | Sudan | Sam et al. ¹¹⁾ |
| 15.2 | 26.9 | 157.1 | Ghana | Faanu et al. ⁹⁾ |
| 13.6 | 24.2 | 162.1 | Ghana | Faanu et al. ¹⁰⁾ |
| 40.1 | 29.4 | 217 | Cameroon | Present study |

Table 1 and t the number of hours in one year (8766 h yr⁻¹).

2.5. Radium equivalent activity

The health hazard associated with natural radioactivity in soil used as building materials was assessed using Ra_{eq} (radium equivalent activity). This hazard index is normally used to compare the uniformity of specific activity of materials containing different amounts of ^{226}Ra , ^{232}Th and ^{40}K . It can be calculated from the following formula^{19, 20)}:

$$Ra_{eq} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K \quad (3)$$

Where A_{Ra} , A_{Th} and A_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in Bq kg⁻¹, respectively. The radium equivalent activity is based on the fact that 370 Bq kg⁻¹ of ^{226}Ra , 259 Bq kg⁻¹ of ^{232}Th and 4810 Bq kg⁻¹ of ^{40}K produce the same gamma ray dose rate; this implies that a radium equivalent of 370 Bq kg⁻¹ in building materials will produce an external dose of 1.5 mSv yr⁻¹ ^{21, 22)}.

2.6. External hazard index

In order to assure that the external gamma-radiation dose from building materials is insignificant, H_{ext} (external hazard index) may be used. This index evaluates the level of gamma-radiation hazard associated with the natural radionuclides in the investigated specific samples ^{19, 20)}. It is given by the following formula:

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (4)$$

Where A_{Ra} , A_{Th} , and A_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in Bq kg⁻¹, respectively. The external hazard index must be less than unity so that the annual

effective dose due to radioactivity in the materials will be limited to 1.5 mSv yr⁻¹ ⁸⁾.

3. Results and discussion

Activity concentrations, external annual effective dose, radium equivalent activity and hazard indices in the soil samples for each site are summarized in Table 2. The corresponding comparisons with similar studies carried out in other African countries are displayed in Table 4.

3.1. Activity concentrations

Activity concentrations of ^{226}Ra in soil samples for the various sites varied from 18.5 to 84.9 Bq kg⁻¹, with an average value of 40.1 Bq kg⁻¹ as indicated in Table 3. The highest activity concentration of ^{226}Ra was recorded at the mining site located behind the Government High School of BO4 (Bétaré Oya) with the activity concentrations higher than the world average value¹ whilst the lowest was obtained at the town Centre of BO5 (Bétaré Oya). The mean activity concentrations of ^{226}Ra in the soil samples of all sites studied were slightly higher than the world average value¹⁾.

Activity concentrations of ^{232}Th for the various sites varied from 11.8 to 58 Bq kg⁻¹, with an average value of 29.4 Bq kg⁻¹ (Table 3). As shown in Table 2, the lowest and the highest activity concentrations of ^{232}Th were obtained at the same sites than those of ^{226}Ra . Except BO4 site, the activity concentrations of ^{232}Th in all the soil samples studied were lower than the world average value.

Concerning ^{40}K , the average activity concentration in soil samples for the various sites was 217 Bq kg⁻¹ in a range of 40.6 Bq kg⁻¹ from a soil sample taken at Batouri-

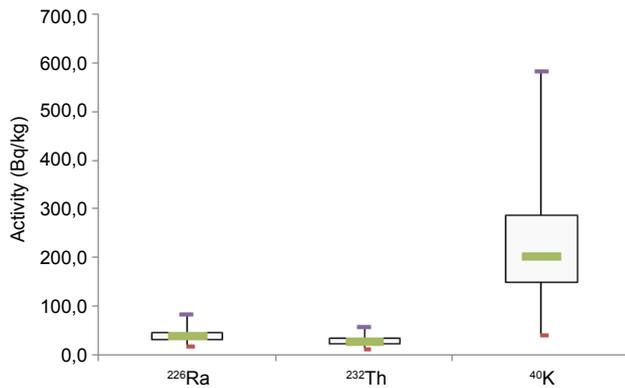


Fig. 2. Activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in soil samples of the gold mining sites of East Cameroon.

Kambelé, Metalicon Company site (BA1) to 582 Bq kg^{-1} from a soil sample of BO4 site (Table 2). The mean activity concentrations of ^{40}K in soil samples of all sites studied were relatively lower than the world average value¹). However the three sites of Bétaré Oya: BO1, BO4 and BO5 have shown the activity concentrations slightly higher than the world average value.

Figure 2 shows the distribution of activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples of the gold mining region of East Cameroon. A comparison between the average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the two towns, Batouri and Bétaré Oya is shown in Figure 3. Independently of location, it appears that the activity concentrations are very close for ^{226}Ra and ^{232}Th , whereas those of ^{40}K are very different. This can be explained by the difference in geochemical properties of radium, thorium and potassium that do not accumulate at the same degree in soil.

As indicated in Table 4, the average activity concentrations for the present study were compared with those of similar studies carried out in other African countries. Gold mining sites of Eastern Cameroon present relatively high values of ^{226}Ra compared to other gold mining sites in Africa except those reported by Ademola *et al.*¹⁴) and are slightly higher than the value given by UNSCEAR¹). The average activity concentration of ^{232}Th is relatively lower than those reported for two gold mining sites of Nigeria^{12, 13}) but higher than those of Sudan¹¹) and Ghana^{9, 10}). The mean activity concentration of ^{232}Th is found to be within the range given by UNSCEAR¹). The average activity concentration of ^{40}K is relatively lower than the world average value¹) and those observed in similar studies carried out in other African countries.

3.2. External annual effective dose

Table 5 gives the result of average E_{ext} (external annual effective dose) for soil also well used as building materials. It can be observed in Table 2 that soil from BO4

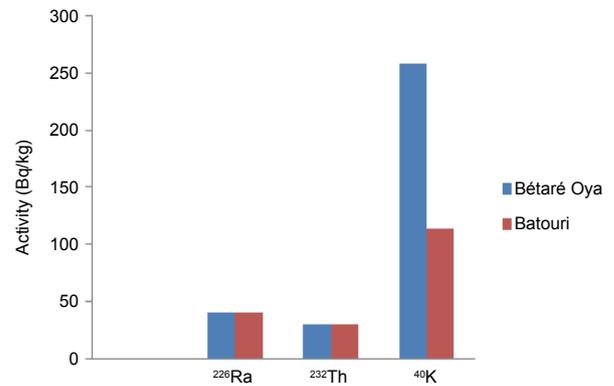


Fig. 3. Comparison between activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples of Batouri and Bétaré Oya.

Table 5. External annual effective dose E_{ext} , external hazard index H_{ext} and radium equivalent activity Ra_{eq} in soil samples of gold mining sites of Eastern Cameroon

| Statistical parameters | E_{ext} (mSv yr ⁻¹) | Ra_{eq} (Bq kg ⁻¹) | H_{ext} (mSv yr ⁻¹) |
|------------------------|--|---|--|
| Range | 0.17 – 0.75 | 47.1 – 212.5 | 0.12 – 0.57 |
| Mean values | 0.34 | 99 | 0.27 |

shows the highest value (0.75 mSv yr^{-1}) with an average of 0.47 mSv yr^{-1} , whereas the lowest value is found in soil collected from the Centre of Bétaré Oya town (BO5) (0.17 mSv yr^{-1}). Except for the values of 0.75 mSv yr^{-1} and 0.56 mSv yr^{-1} obtained respectively in soil samples from BO4 and BO1 sites, all the other values are lower than the outdoor average effective dose of 0.46 mSv yr^{-1} for soils²³). According to these results, ^{226}Ra is the main contributor to the annual effective dose from outdoor terrestrial radiation in the study areas.

3.3. Radium equivalent activity

The calculated values of the Ra_{eq} (Radium equivalent activity) for the investigated samples ranged from 47.1 to 212.5 Bq kg^{-1} , with the highest value recorded in the soil sample of BO4 site, while the lowest value was recorded in BO5 site (Table 2). As indicated in Table 1, all the calculated average Ra_{eq} values of the studied samples in the present work were lower than the recommended maximum value of 370 Bq kg^{-1} defined by the OECD for building materials²²).

3.4. External hazard index (H_{ext})

H_{ext} ranged from 0.12 to 0.57 with an average value of 0.27 as shown in Table 5. The highest value of external index was recorded in soil sample of BO4 site, while the lowest value was recorded in BO5 site. All estimated values of H_{ext} in this work were lower than the recommended limit (unity).

The values of $R_{a_{eq}}$ and those H_{ext} have revealed that soils examined in Eastern Cameroon may be used for buildings construction and may not carry considerable radiological hazard to the workers and local public.

4. Conclusion

The activity concentrations of NORM and the related radiation hazards in the soil samples investigated were determined by using NaI (TI) gamma spectrometer. The average activity concentrations determined for ^{226}Ra , ^{232}Th , and ^{40}K were 40.1 Bq kg^{-1} , 29.4 Bq kg^{-1} , and 217 Bq kg^{-1} , respectively. The comparison between the activity concentrations found in this study with those of similar studies undertaken in other African countries shows relatively high levels of ^{226}Ra in Eastern Cameroon, while ^{232}Th and ^{40}K are found to be almost at the same proportions. Moreover we have observed that the annual effective dose in all samples ranged between 0.17 and 0.75 mSv yr^{-1} within the safety limit of 1.5 mSv yr^{-1} defined by the OECD report. The external hazard index and radium equivalent activity have indicated lower values than the unacceptable level. Except the mining site located behind the Government High School of Bétaré Oya (BO4), the findings of this study indicate that the soil samples investigated in Eastern Cameroon can be used as building materials. Since only soil samples were investigated within the framework of this study, other data are required to make a whole environmental natural radiation monitoring of the region, particularly those of foodstuffs and of radon and thoron in dwellings. However, it is recommended to strengthen the monitoring of mining areas to better protecting people and environment against harmful effect of ionizing radiation. This should be accompanied by a strong regulation on radiation protection.

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All authors contributed equally in the preparation of this manuscript. Supplementary materials can be accessed by contacting the corresponding author.

Conflict of Interest Disclosure

The authors declare that they have no conflict of interest.

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