Study of radon exhalation rate and natural radioactivity in soil samples collected from East Singhbhum Shear Zone in Jaduguda U-Mines Area, Jharkhand, India and its radiological implications

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Received 22 April 2010; accepted 11 May 2010

In the present study, Sealed Can Technique was adopted for radon exhalation measurements in soil samples collected from some areas around the East Singhbhum shear zone in U-mining area of Jharkhand state of India. Radon activity varies from 3794.3 to 4891.4 Bq m\(^{-3}\) with an average value of 4368.6 Bq m\(^{-3}\) while radon exhalation rate varies from 1364.1 to 1758.6 mBq m\(^{-2}\) h\(^{-1}\) with an average value of 1573.8 mBq m\(^{-2}\) h\(^{-1}\). Activity concentrations of naturally occurring radionuclides \(^{238}\text{U}, \text{\(232^{\text{Th}}\)}\) and \(^{40}\text{K}\) were also measured in these soil samples using high resolution \(\gamma\)-ray spectroscopic system. Activity concentrations were found to vary from 6.1 ± 0.2 to 826.3 ± 8.5 Bq kg\(^{-1}\) for \(^{238}\text{U}\), 8.7 ± 0.3 to 236.7 ± 3.2 Bq kg\(^{-1}\) for \(^{232}\text{Th}\) and 291.5 ± 4.4 to 1391 ± 14.3 Bq kg\(^{-1}\) for \(^{40}\text{K}\). From the activity concentration of \(^{238}\text{U}, 232^{\text{Th}}\) and \(^{40}\text{K}\) in these soil samples, the radium equivalent activity (Ra\text{eq}) due to the presence of radionuclides is calculated and it varies from 34.0 to 924.5 Bq kg\(^{-1}\) with an average value of 260.4 Bq kg\(^{-1}\). Total absorbed gamma dose rates in the surrounding air are found to vary from 15.1 to 402.0 nGyh\(^{-1}\) with an average value of 120.8 nGyh\(^{-1}\). The indoor and outdoor annual effective dose rates from these soil samples are determined from 0.1 to 1.9 mSv y\(^{-1}\) and 0.03 to 0.8 mSv y\(^{-1}\), respectively. External hazard index, \(H_{\text{ex}}\) for the soil samples studied in this work ranges from 0.14 to 2.46 with a mean value of 0.70. The internal exposure to \(^{222}\text{Rn}\) and its radioactive progeny are controlled by the internal hazard index \(H_{\text{int}}\). Computed values of \(H_{\text{int}}\) vary from 0.2 to 4.8 with an average value of 1.02.

**Keywords:** Natural radioactivity, HPGe low level radiation counting system, Soil, Radium equivalent activity, Absorbed gamma dose rates

1 Introduction

Radioactive toxic elements uranium, thorium and potassium are found in tracers in almost all types of rocks, sands and waters. Uranium forms deposits in the earth’s crust where the geological conditions become favourable. Distribution of naturally occurring radionuclides mainly \(^{238}\text{U}, 232^{\text{Th}}\) and \(^{40}\text{K}\) and other radioactive elements depends on the distribution of rocks from which they originate and on the processes through which they are concentrated. Jharkhand state of India is rich in minerals and is called the store house of minerals. The measurement of activities of naturally occurring radio nuclides \(^{238}\text{U}, 232^{\text{Th}}\) and \(^{40}\text{K}\) is important for the estimation of radiation risk and has been the subject of interest of research scientists all over the world. Human beings are exposed to ionizing radiation from natural sources of \(^{238}\text{U}\) and \(^{232}\text{Th}\) present in solids like rocks, sand, soil etc., cosmic rays and the internal exposure from radioactive elements through food, water and air. Natural radioactivity is wide spread in the earth’s environment and it exists in soils, rocks, water and sand etc\(^{1,2}\). Radium and its ultimate precursor uranium are the source of radon, an \(\alpha\)-radioactive inert gas. As an inert gas is having sufficiently long lifetime (3.8 days), it can move freely through the materials like soil, sand, rock etc. Short lived radon progenies have been established as causative agents of lung cancer\(^1\). Radiation doses vary depending upon the concentrations of the natural radionuclides \(^{238}\text{U}\) and \(^{232}\text{Th}\) and their daughter products and \(^{40}\text{K}\), present in the soils and rocks which in turn depend upon the local geology and region in the world\(^3\).
of this study was focused on determining the natural radio nuclides (238\textsuperscript{U}, 232\textsuperscript{Th} and 40\textsuperscript{K}) concentration in soil samples collected from some areas around the East Singhbhum shear zone and U-mining area of Jharkhand state of India. Radon exhalation rates were also measured from these samples to estimate effective radiation dose to assess the radiation risk to the inhabitants.

2 Experimental Details

2.1 Sealed Can technique

Sealed Can technique\textsuperscript{4-6} was adopted for radon exhalation measurements from the soil samples. LR-115 Type II plastic track detector (2 cm × 2 cm) was fixed on the top inside the cylindrical Can of 4.5 cm height and 7.0 cm diameter. Equal amount of each sieved (100\textmu m grain size) sample (100 gm) from some areas around the East Singhbhum shear zone and U-mining area of Jharkhand state India was placed at the base of the Can. The Cans were sealed for 100 days. Thus, the lower sensitive part of the detector is exposed freely to the emergent radon from the sample in the Can so that it could record the tracks of alpha particles resulting from the decay of radon in the remaining volume of the Can and from \textsuperscript{218}Po deposited on the inner walls of the Can. Radon and its daughters reach in equilibrium in about 4 hours and hence the equilibrium activity of emergent radon can be obtained from the geometry of the Can and the time of exposure. After the exposure, the detectors were etched in 2.5 NaOH at 60°± 1°C for a period of 90 min in constant temperature water bath for revelation of tracks. The resulting \textit{α}-particles on the exposed face of the track detector were counted using an optical microscope (magnification 400×). The activity was obtained from the track density in the etched detectors using the calibration factor of 0.056 track cm\textsuperscript{-2} d\textsuperscript{-1} obtained from an earlier calibration experiment\textsuperscript{7}. The exhalation rate of radon is obtained from the expression\textsuperscript{6,8,9}.

\[ Ex = \frac{CV\lambda}{A} \left[ T + \frac{1}{\lambda} \left( e^{-\lambda T} - 1 \right) \right] \]  

...(1)

where \( Ex \) is the radon exhalation rate (Bq m\textsuperscript{-2} h\textsuperscript{-1}); \( C \) the integrated radon exposure as measured by LR-115 type II plastic track detector ( Bq m\textsuperscript{3} h); \( V \) the effective volume of Can; \( \lambda \) the decay constant for radon (h\textsuperscript{-1}); \( T \) the exposure time (h) and \( A \) is the area covered by the Can (m\textsuperscript{2}).

2.2 Estimation of \textsuperscript{238}U, \textsuperscript{232}Th and \textsuperscript{40}K

All soil samples collected around the East Singhbhum shear zone and U-mining area of Jharkhand state were dried for 24 h in an air-circulation oven at 110°C to ensure that moisture is completely removed. These samples were packed and sealed in an impermeable airtight PVC container to prevent the escape of radiogenic gases radon (\textsuperscript{222}Rn) and thoron (\textsuperscript{220}Rn). About 300 g sample of each material was used for measurements. Before measurements, the containers were kept sealed for about 4 weeks in order to reach equilibrium between the \textsuperscript{238}U and \textsuperscript{232}Th and their respective progenies. After attainment of secular equilibrium between \textsuperscript{238}U and \textsuperscript{232}Th and their decay products, the samples were subjected to high resolution gamma spectroscopic analysis.

Measurements were carried out at Inter-University Accelerator Centre, New Delhi with a coaxial \textit{n}-type HPGe detector (EG&G, ORTEC, Oak Ridge, USA). The detector having a resolution of 2.0 keV at 1332 keV and a relative efficiency of 20% was placed in 4° shield of lead bricks on all sides to reduce the background radiation from building materials and cosmic rays. The detector was coupled to a PC based 4K multi channel analyzer and an ADC for data acquisition.

The calibration of the low background counting system was done with a secondary standard which was calibrated with the primary standard (RGU-1) obtained from the International Atomic Energy Agency (IAEA). The efficiency for the system was determined using secondary standard source of uranium ore in the same geometry as available for the sample counting. The samples were counted for a period of 72000 sec for activity measurements. The activity concentration of \textsuperscript{40}K (\( C_K \)) was measured directly by its own gamma ray of 1461 keV. As \textsuperscript{238}U and \textsuperscript{232}Th are not directly gamma emitters, their activity concentrations (\( C_U \) and \( C_{Th} \)) were measured through gamma rays of their decay products. Decay products taken for \textsuperscript{238}U were \textsuperscript{210}Pb: 295 and 352 keV and \textsuperscript{214}Bi: 609,1120 and 1764 keV whereas for \textsuperscript{232}Th were \textsuperscript{228}Ac: 338, 463, 911 and 968 keV, \textsuperscript{212}Bi: 7 27 keV, \textsuperscript{212}Pb : 238 keV and \textsuperscript{234}Pa: 1001 keV gamma ray by assuming the decay series to be in equilibrium\textsuperscript{10}. To estimate the activity concentrations of \textsuperscript{238}U and \textsuperscript{232}Th weighted averages of several decay products were used. The spectra were analyzed using the locally developed software CANDLE (Collection and Analysis of Nuclear Data using Linux Net work).
Respective count rates after subtracting the background counts of the spectrum obtained for the same counting time were used to calculate the net count rate under the most prominent photo peaks of radium and thorium daughters. Then, the activity of the radionuclide was calculated from the background subtracted area of prominent gamma ray energies. Gamma ray spectra of one typical soil sample is shown in Fig. 1. The concentration of uranium, thorium and potassium was calculated using the following equation:

\[
\text{Activity (Bq.kg}^{-1} = \frac{(S \pm \sigma) \times 100 \times 1000 \times 100}{E \times W \times A} \quad \ldots (2)
\]

where \( S \) is the net counts/sec (cps) under the photo peak of interest, \( \sigma \) the standard deviation of \( S \), \( E \) the counting efficiency (%), \( A \) the gamma abundance or branching intensity (%) of the radionuclide and \( W \) is the mass of the sample (kg). Minimum activity concentration which can be measured by the system is \( ^{238}\text{U} \) 2 Bq kg\(^{-1} \), \( ^{232}\text{Th} \) 2 Bq kg\(^{-1} \) and \( ^{40}\text{K} \) 3 Bq kg\(^{-1} \). As soil is the basic ingredient used in construction materials in India, estimation of the radiation risk to the population is quite important and can be computed from the activities of the radio-nuclides present in these samples.

3 Estimation of Dose Rates

Outdoor air gamma absorbed dose rate \( (D) \) in nGy h\(^{-1} \) due to terrestrial gamma rays at 1 m above the ground surface which can be computed from the specific activities, \( C_U, C_{Th} \) and \( C_K \) of \( ^{238}\text{U}^{226}\text{Ra}, ^{232}\text{Th} \) and \( ^{40}\text{K} \) in Bq kg\(^{-1} \), respectively by Monte Carlo method\(^{11} \) (UNSCEAR, 2000):

\[
D \text{ (nGy h}^{-1} = 0.462C_U + 0.604C_{Th} +0.0417C_K \quad \ldots (3)
\]

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose, 0.7 Sv Gy\(^{-1} \) was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor and implying that 20\% of time is spent outdoors, outdoor occupancy factor of 0.2 proposed\(^{11} \) by (UNSCEAR, 2000) were used.

Fig. 1 — Spectra of the soil samples from East Singhbhum shear zone U-mining area of Jharkhand state of India.

The legends are: Ra-226 (V), Pb-214 (●), Bi-214 (●)
effective dose rate \((E)\) in units of mSv y\(^{-1}\) was calculated by the following formulae:

Indoor effective dose \(E\) (mSv y\(^{-1}\)) = \(D\) (nGy h\(^{-1}\)) \times 8760 h \times 0.8 \times 0.7 S v Gy\(^{-1}\)\times 10\(^{-6}\) \(\ldots\) (4)

Outdoor effective dose \(E\) (mSv y\(^{-1}\)) = \(D\) (nGy h\(^{-1}\)) \times 8760h \times 0.2 \times 0.7 S v Gy\(^{-1}\)\times 10\(^{-6}\) \(\ldots\) (5)

The distribution of \(^{238}\)U/\(^{226}\)Ra, \(^{232}\)Th and \(^{40}\)K in soil samples is not uniform. Uniformity with respect to exposure to radiation can be defined in term of radium equivalent activity (Ra\(_{eq}\)) in Bq kg\(^{-1}\) to compare the specific activity of materials containing different amounts of \(^{226}\)Ra, \(^{232}\)Th or \(^{4810}\)K. It can be calculated from the following relation\(^{12,13}\).

\[
Ra_{eq} = C_U + 1.43 C_{Th} + 0.07 C_K \) \(\ldots\) (6)

It has been assumed here that 370 Bq kg\(^{-1}\) of \(^{238}\)U or 259 Bq kg\(^{-1}\) of \(^{232}\)Th or 4810 Bq kg\(^{-1}\) of \(^{40}\)K produce the same gamma dose rate.

Beretka and Mathew\(^{14}\) defined two other indices that represent external and internal radiation hazards. The external hazard index is obtained from Ra\(_{eq}\) expression through the supposition that its allowed maximum value (equal to unity) corresponds to the upper limit of Ra\(_{eq}\) (370 Bq kg\(^{-1}\)). The external hazard index \((H_{ex})\) can then be defined as:

\[
H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \) \(\ldots\) (7)

Internal exposure to \(^{222}\)Rn and its radioactive progeny is controlled by the internal hazard index \((H_{in})\) as given below\(^{15}\).

\[
H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \) \(\ldots\) (8)

This index value must be less than unity in order to keep the radiation hazard to be insignificant. The knowledge of radio nuclides distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and extraterrestrial sources.

### 4 Results and Discussion

In India, it is a common practice to use phosphate fertilizers in soil to enhance the crop yield. As the phosphate fertilizers contains naturally occurring radioactive materials (NORM), the use of phosphate fertilizers in soil to enhance the crop produce and phosphogypsum in the building process, such as in the manufacture of bricks and plasterboards for houses etc. Ceilings which are typically made from plasterboard, has radiological impact of human health due to radiation exposure. The radiological impact from the soil is due to the internal irradiation of the lung by the \(\alpha\)-particles from short lived radon-thoron progeny and the external irradiation of the body by \(\gamma\)-rays emitted from radionuclides.

The results for the values of the radon activity and radon exhalation rates from the soil samples are presented in Table 1. Radon activity varies from 3794.3 to 4891.4 Bqm\(^{-3}\) with an average value of 4368.6 Bqm\(^{-3}\) while radon exhalation rate varies from 185 to 1758.6 mBqm\(^{-2}\) h\(^{-1}\) with an average value of 1573.8 mBqm\(^{-2}\) h\(^{-1}\). Natural radio nuclides \((^{238}\)U, \(^{232}\)Th and \(^{40}\)K) activity concentrations from the soil samples are presented in Table 2. Radium equivalent, absorbed gamma dose rate, annual effective dose rate and external hazard.

### Table 1 — Radon activity, radon exhalation rate and indoor inhalation exposure (radon)-effective dose in in soil samples collected from around the East Singhbhum shear zone and U-mining area of Jharkhand state, India

<table>
<thead>
<tr>
<th>Radon activity (Bqm(^{-3}))</th>
<th>No. of samples</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Average value</th>
<th>S D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radon exhalation rate (mBqm(^{-2}) h(^{-1}))</td>
<td>12</td>
<td>3794.3</td>
<td>4891.4</td>
<td>4368.6</td>
<td>337.9</td>
</tr>
</tbody>
</table>

### Table 2 — Natural radioactivity in soil samples collected from around the East Singhbhum shear zone and U-mining area of Jharkhand state

<table>
<thead>
<tr>
<th>(^{238})U (Bq kg(^{-1}))</th>
<th>No. of samples</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Average value</th>
<th>S D</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{232})Th (Bq kg(^{-1}))</td>
<td>12</td>
<td>6.1 ± 0.2</td>
<td>826.3 ± 8.5</td>
<td>119.1 ± 1.5</td>
<td>224.9 ± 2.2</td>
</tr>
<tr>
<td>(^{40})K (Bq kg(^{-1}))</td>
<td>12</td>
<td>8.7 ± 0.3</td>
<td>236.7 ± 3.2</td>
<td>64.5 ± 1.4</td>
<td>64.9 ± 0.9</td>
</tr>
</tbody>
</table>

*BDL- Below Detection Limit.*
index for the soil samples are given in Table 3. There are wide variations in the activity concentrations from $6.1 \pm 0.2$ to $826.3 \pm 8.5$ Bq kg$^{-1}$ for $^{238}$U, $8.7 \pm 0.3$ to $236.7 \pm 3.2$ Bq kg$^{-1}$ for $^{232}$Th and BDL to $1399.1 \pm 14.3$ Bq kg$^{-1}$ for $^{40}$K. One sample from near the Jaduguda U mine has the highest value of uranium activity with small value for thorium activity which is expected in uraniferous regions. Variations in natural radioactivity levels in the soil samples collected from different sampling sites are due to variation of concentrations of these elements in geological formations. From the activity concentration of $^{238}$U, $^{232}$Th and $^{40}$K in these soil samples the radium equivalent activity ($Ra_{eq}$) is calculated and varies from $34.0$ to $922.5$ Bq kg$^{-1}$ with an average value of $256.0$ Bq kg$^{-1}$. Total absorbed gamma dose rates in the surroundings air are found to vary from $15.2$ to $425.9$ nGy h$^{-1}$ with an average value of $120.6$ nGy h$^{-1}$. Figure 2 shows a plot of the variation of $^{238}$U, $^{232}$Th, $^{40}$K and absorbed dose rate in these soil samples.

<table>
<thead>
<tr>
<th>Radium equivalent activity ($Ra_{eq}$)</th>
<th>Absorbed gamma dose rate (D)</th>
<th>Indoor effective dose rate (mSv y$^{-1}$)</th>
<th>Outdoor effective dose rate (mSv y$^{-1}$)</th>
<th>External Hazard Index ($H_{ex}$)</th>
<th>Internal Hazard Index ($H_{in}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bq kg$^{-1}$</td>
<td>nGy h$^{-1}$</td>
<td>mSv y$^{-1}$</td>
<td>mSv y$^{-1}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Minimum</td>
<td>34.0</td>
<td>0.07</td>
<td>0.02</td>
<td>0.09</td>
<td>0.15</td>
</tr>
<tr>
<td>Maximum</td>
<td>922.5</td>
<td>2.08</td>
<td>0.52</td>
<td>2.46</td>
<td>4.76</td>
</tr>
<tr>
<td>Average Value</td>
<td>256.0</td>
<td>0.59</td>
<td>0.15</td>
<td>0.70</td>
<td>0.96</td>
</tr>
<tr>
<td>S.D.</td>
<td>261.2</td>
<td>0.58</td>
<td>0.15</td>
<td>0.70</td>
<td>1.31</td>
</tr>
</tbody>
</table>

Fig. 2 — Bar diagram showing activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K and absorbed gamma dose rate in different soil samples.
Table 4 — Comparison of $^{238}$U, $^{232}$Th, $^{40}$K activity concentrations, $Ra_{eq}$ and radon exhalation rates from different parts from India

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Location</th>
<th>$^{226}$Ra/$^{238}$U (Bq kg$^{-1}$) Range</th>
<th>$^{232}$Th (Bq kg$^{-1}$) Range</th>
<th>$^{40}$K (Bq kg$^{-1}$) Range</th>
<th>$Ra_{eq}$ (Bq kg$^{-1}$) Range</th>
<th>Radon exhalation rate ($\text{mBq m}^{-2} \text{h}^{-1}$) Range</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Upper Siwaliks, Northern India</td>
<td>28.3-81.0</td>
<td>61.2-140.3</td>
<td>363.4-1002</td>
<td>149.4-351.8</td>
<td>—</td>
<td>$^{15}$Joga Singh et al. (2009)</td>
</tr>
<tr>
<td>2</td>
<td>Some cities of Uttar Pradesh</td>
<td>13.6 - 35.8</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>573.2 – 851.2</td>
<td>$^{10}$R Kumar et al. (2006)</td>
</tr>
<tr>
<td>3</td>
<td>Some cities of Rajasthan</td>
<td>11.1-21.0</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>583.8 – 748.4</td>
<td>$^{10}$R Kumar et al. (2006)</td>
</tr>
<tr>
<td>4</td>
<td>Some cities of Punjab</td>
<td>35.9-64.8</td>
<td>49.5-95.3</td>
<td>80.4-165.1</td>
<td>113.0-213.1</td>
<td>—</td>
<td>$^{3}$S Singh et al. (2005)</td>
</tr>
<tr>
<td>5</td>
<td>Some cities of Himachal Pradesh</td>
<td>18.2-90.3</td>
<td>34.8-124.7</td>
<td>81.9-181.4</td>
<td>74.3-281.9</td>
<td>—</td>
<td>$^{3}$S Singh et al. (2005)</td>
</tr>
<tr>
<td>6</td>
<td>Delhi</td>
<td>30</td>
<td>20</td>
<td>200</td>
<td>—</td>
<td>12.4</td>
<td>$^{19}$Sonkawade et al. (2008)</td>
</tr>
<tr>
<td>7</td>
<td>Some places of Jharkhand</td>
<td>6.1 - 826.3</td>
<td>8.7 -236.7</td>
<td>BDL-1399.1</td>
<td>34.0 - 922.5</td>
<td>364.1-1758.6</td>
<td>Present Study</td>
</tr>
</tbody>
</table>

Indoor and outdoor annual effective dose rate from these soil samples are determined from 0.7 to 2.08 mSv y$^{-1}$ and 0.02 to 0.52 mSv y$^{-1}$, respectively. External hazard index, $H_{ex}$ for the soil samples studied in this work range from 0.09 to 2.46 with a mean value of 0.70. The internal exposure to $^{222}$Rn and its radioactive progeny is controlled by the internal hazard index $H_{in}$. Computed values of $H_{in}$ vary from 0.15 to 4.76 with an average value of 0.96. Table 4 presents the comparison of $^{238}$U, $^{232}$Th, $^{40}$K activity concentrations, $Ra_{eq}$ and radon exhalation rates from different parts from India taken from literature. Since most of these values of $H_{ex}$ are less than unity except for two soil sample no. –1 and 6 (collected from near the Jaduguda uranium mines) showing higher values than unity, the use of soil from this region can be used as construction material without posing significant radiological threat to population.

5 Conclusions

The radium equivalent activity ($Ra_{eq}$) in soil samples less than 370 Bq kg$^{-1}$, which are acceptable for safe use $^{10}$(OECD, 1979). Only two (No. 1 and 6) soil samples show high values of radium equivalent activity, this may be due to higher concentration of uranium in these soil samples. These two samples collected from near the uranium mines Jharkhand State of India. There are huge deposits of bauxite, mica and coal along with iron, copper, chromites, tungsten, lime stone, feldspar and quartz etc.

Acknowledgement

The authors thank, Chairman, Department of Applied Physics, Aligarh Muslim University, Aligarh, for providing the facilities for this work. Sincere thanks are also due to Dr Amit Roy, Director, Inter-University Accelerator Centre, New Delhi, for providing HPGe spectrometry system for the analysis of natural radionuclides and constant encouragement. Dr Rajesh Kumar also acknowledges the University Grants Commission (UGC), Govt. of India, New Delhi, for providing financial assistance to carry out this research work.

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