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Dose assessment of ^{137}Cs in agricultural surface soil in Selangor, Malaysia

H. Muthu[1,2], K. Ramesh[1], S. Ramesh[1*], S. Bashir[1,3*]

[1]Centre for Ionics University of Malaya, Department of Physics, Faculty of Science, University Malaya, 50603, Kuala Lumpur, Malaysia
[2]Taylor's College, Taylor's Lakeside Campus, No. 1 Jalan Taylor's, 47500 Subang Jaya, Selangor, Malaysia
[3]Higher Institution Centre of Excellence (HICoE), UM Power Energy Dedicated Advanced Centre (UMPEDAC), Level 4, Wisma R&D, Universiti Malaya, Jalan Pantai Baharu, 59990, Kuala Lumpur, Malaysia

► ***Original article***

***Corresponding authors:**
Subramaniam Ramesh, Ph.D.,
***E-mail:** rameshtsubra@gmail.com*
Shahid Bashir, Ph.D.,
***E-mail:** shahidbashirbaig@gmail.com*

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ABSTRACT

Background: The activity concentration (AC) of cesium-137 (^{137}Cs) in the agricultural soil was measured in this study to set reference data and an indicator of the radionuclide fallout especially in Malaysia. ***Materials and Method***: Using the High Purity Germanium (HPGe) gamma-ray spectrometer, the AC of ^{137}Cs was employed to determine the radiological hazards to the public. ***Results***: Results revealed that the AC of ^{137}Cs in the soil samples ranged between 0.34 ± 0.09 to 3.21 ± 0.17 Bqkg^{-1}. Dose rate computed from the corresponding value of AC ranged from 0.01 to 0.10 nGyh^{-1}. The annual effective dose rate ranged between 1.25 to 11.8 μSv y^{-1}. The values of Excessive lifetime cancer risk, ELCR ranged between 0.47×10^{-5} to 4.45×10^{-5} is lesser than the safety threshold of 0.29×10^{-3}. The analysis of variance of this parameter is found to be at $p<0.05$ which is statistically significant in this study. ***Conclusion***: The outcomes from this study show that the analysed values are below than the recommended values by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and The International Commission on Radiological Protection (ICRP) and do not cause any radiological hazards to the general population.

Keywords: *Cesium-137, nuclear fallout, anthropogenic radionuclides, annual effective dose rate, excessive cancer lifetime risk, radionuclides.*

INTRODUCTION

The tragedy at the Fukushima Daiichi Nuclear Power Plant (FDNPP) began on March 2011, when an earthquake and subsequent tsunami struck Japan. Radionuclides, mainly ^{134}Cs and ^{137}Cs, were discharged into the natural ecosystem after the accident and deposited on soil as radioactive pollutants (1).

As an anthropogenic radionuclide, ^{137}Cs does not occur in the natural environment where its half-life is around 30 years, which is extremely lengthy. This specific isotope of caesium is both a beta and gamma emitter. Its presence in the environment is mostly due to nuclear weapons testing occurred between the 1950s and the 1970s, according to the Environmental Protection Agency. The Chernobyl nuclear power plant (CNPP) reactor tragedy in 1986 resulted an increased radioactive fallout entering the environment, with ^{137}Cs being one of the radionuclides found in the fallout. Testing of nuclear weapons, the CNPP disaster, which resulted the release of 15 and 30 $\times 10^{15}$ Bq of ^{137}Cs and ^{134}Cs, respectively, were the primary sources of radioactive contamination (2). It was discovered in the marine sand around Malaysian waters that this radioactive fallout had occurred (3).

Heavy fission products containing ^{137}Cs are the most common. The fission of a variety of thorium, uranium, and plutonium isotopes produces a 6 percent ^{137}Cs production in all situations. ^{137}Cesium is abundant in the nuclear fuel, and in areas contaminated by a fission by-product following nuclear disasters, as a result of the high yield of fission. A continuing concern is posed by the high quantities of ^{137}Cs created during fission events. ^{137}Cs has a long half-life to ensure that objects and regions contaminated by the radioactive element continue to be hazardous to population for a generation or more. It is, however, short enough to ensure that even little concentrations of ^{137}Cs emit harmful radiation doses (its specific radioactivity is 3.2×10^{12} Bqg^{-1})(4)

Exposure to gamma photons from ^{137}Cs buried in the ground is a major cause for alarm because of the external radiation dosages they may impart to the human body. There has been recent research done on the FDNPP catastrophe and its effects on the Fukushima prefecture in Japan (5,6). Similar inquiries have been recorded in Bryansk, one of the Russian cities worst hit by the CNPP disaster. In 2012, researchers in Minsk and Gomel, Belarus, and Chernobyl, Ukraine, measured recent exposures to

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radiation from the outside world. Reporting that even 26 years after the mishap, the projected effective doses near CNPP are higher than the ICRP's suggested public exposure threshold of 1 mSv yr^{-1}, the agency expressed concern. The aforementioned research was conducted mostly at various local and regional scales (7–9). There are no large-scale or national-level comparative studies in the literature on the soil dispersal of ^{137}Cs and corresponding absorbed doses in the human organs, despite the importance of organ dose calculation for cancer risk assessment (10, 11). Those who reside in close proximity to nuclear power stations have had their organ doses estimated (12).

Soil absorption of ^{137}Cs has been investigated since the 1960s, and this research has recently been revived to track the long-duration impacts of the Chernobyl disaster (13). The kind of soil has a significant impact on the amount of ^{137}Cs that may be absorbed. Within plants, it is transferred easily from leaves to seeds. Inhalation and skin contact may also cause a hazard to human health, however these are the most common routes of exposure (14).

Because Malaysia is situated in the south of South China Sea, radio-caesium from FDNPP events is expected to be carried to the country at low ACs or at a level that is only marginally significant, considering the country's geographic location (15). A study on the AC of ^{137}Cs in surface soil of Fraser's Hill, Malaysia, a popular hillside tourist destination, was found to be between 0.26 $Bqkg^{-1}$ and 5.15 Bq kg^{-1}.(16) A similar study was conducted at different slopes of a hill in Cameron Highlands, Malaysia. The ^{137}Cs ACs range detected at top locality was from 0.05 $Bqkg^{-1}$ to 1.53 $Bqkg^{-1}$, the centre was 0.22 $Bqkg^{-1}$ to 2.11 $Bqkg^{-1}$, bottom was from 0.00 $Bqkg^{-1}$ to 2.03 $Bqkg^{-1}$ and forestry was 0.00 $Bqkg^{-1}$ to 0.96 $Bqkg^{-1}$. The researchers concluded that the AC of ^{137}Cs at the top is lowest, while at the bottom is highest, showing that there is a downward transport of ^{137}Cs (17)

Several research projects have looked at NORM in various sediments, rocks, and soils. Multiple studies have looked at the agricultural soil in the Malaysian state of Selangor from a radiological perspective (18–21). Despite this, there is very little data available on the radiation threat posed by ^{137}Cs nuclear fallout in this area. Since there are few studies carried out on the agricultural soil in Malaysia, the significance of the ^{137}Cs study in this area could provide baseline data to assess the radiological hazards to the general population. Outcomes will be assessed with the recommended values by UNSCEAR(22) and ICRP (23) to evaluate the radiological hazards to the general public living around the vegetable farm.

MATERIALS AND METHOD

Study technique

Soil samples taken from a vegetable farm in Klang, Selangor, shown in figure 1, a Malaysian agricultural area about 50 kilometers southeast of Kuala Lumpur known as the Green Revolution Land, where the state government began pushing agricultural activities in the early 1990s. Vegetables such as Japanese Mustard, Water Spinach, Bitter Gourd, Spinach, Cucumber, Lady's Finger, Long Bean, Pumpkin, Tapioca, Sweet Potato and different kinds of herbs are grown in this area. The location of this farm at latitude 2°57'03.5'N and longitude 101°28'30.0'E and is shown in figure. 2. Tropical peat soil has been determined to be the soil type in this research area, and it is distinguished by the fact that it is exceptionally soft, damp, and unconsolidated deposits. It is made up of a mass of semi-decomposed woody debris that was formed from the forest detritus in this research region, and it is referred to as peat soil in the scientific community. A large portion of its composition is made up of organic material, which gives it a dark brown colour. It also has a high-water retention capacity, which can be as much as 15-20 times its dry weight. As a general rule, peat soil is deficient in nutrients and acidic, with an acidity range ranging from 3.0 to 4.5 on the pH scale (24).

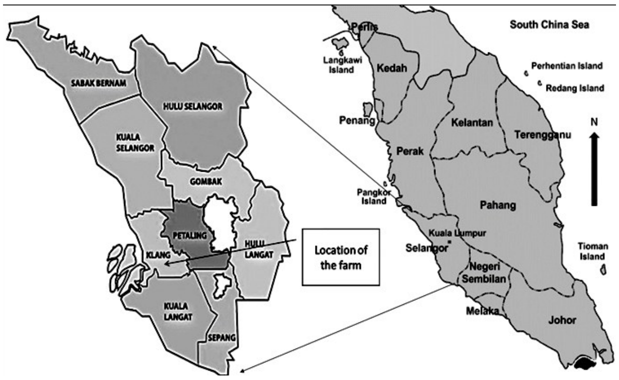

Figure 1. Location of district of Klang in state of Selangor, West Malaysia.

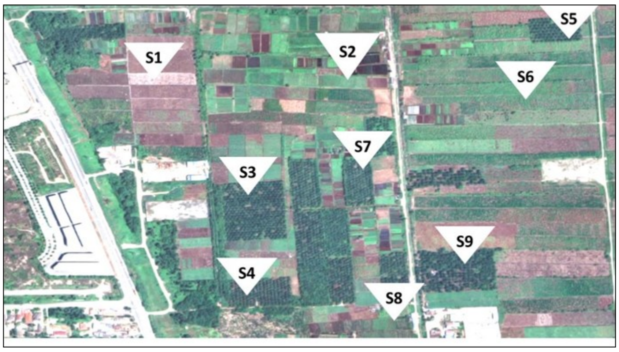

Figure 2. Location of the soil samples taken in the farm located in Klang, Selangor.

An overall total of nine soil samples (S1 to S9) were collected from the study site and transferred to the laboratory in a sealed plastic bag, with each plastic bag being labelled with the required information. All big stones in the soil samples were removed, and the entire sample was sieved in

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stainless steel. The sample of soils was then dry up in an furnace at 108°C for 24 hours before being crushed further[25]. The dried sample was then crushed into a fine grain and passed through a filter with a 2 mm opening to ensure homogeneity. Each soil sample was weighted using an electronic balance and then laid in a 250 ml Marinelli beaker, closed, and kept at room temperature for 28 days to allow secular balance between ^{226}Ra and its daughter nuclide before the gamma spectrometric analysis [12].

Radioactivity measurements

Using a closed-end coaxial HPGe gamma-ray spectrometer, procured from Mirion Technologies, (CANBERRA), USA, the radioactivity ACs of ^{137}Cs in the samples were analysed. This instrument has an efficiency of about 30 percent and an energy resolution of 1.8 keV-FWHM at the 1333 keV peak. A static bottomed lead shield with cylindrical dimension and a moveable lid covered the detector in order to minimise the amount of outdoor gamma-ray background present in the recorded spectra. The data collection system consisted of a 16k Multi Channel Analyser 2 (MCA2) linked to the HPGe detector. With the help of the Genie 2000 Canberra software, the gamma rays produced by the samples were examined. Standard calibration sources comprising the following were used to calibrate the spectrometer's energy and relative efficiency: ^{203}Hg (280 keV), ^{113}Sn (392 keV), ^{85}Sr (514 keV), ^{137}Cs (662 keV), ^{88}Y (1836 keV), and ^{60}Co (1173 and 1332 keV). Eckert & Ziegler, Isotope Products Laboratories from Valencia, California, 91355, USA, provided the standard source with a preliminary activity of 5.076 Ci. Each sample was analysed for 24 hours (86400s) and background data set to decrease counting error and background counts. The gamma-ray line at 662 keV was used to estimate the AC of ^{137}Cs as per International Atomic Agency (IAEA) recommendations [28,29]. Energy calibration and channel number calibration of the spectrometer are shown in figure 3.

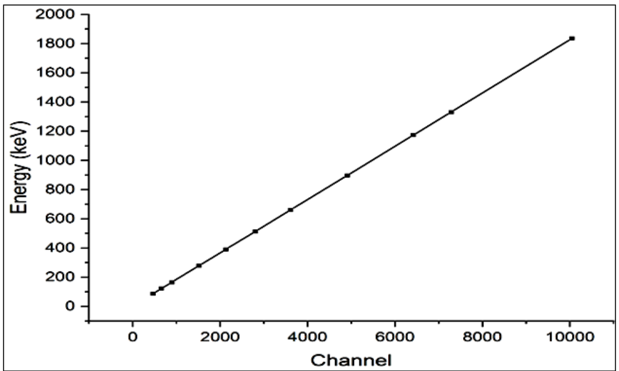

Figure 3. Energy versus channel number curve of the detector.

Measurement of the ^{137}Cs activity and minimum measurable activity

The activity of ^{137}Cs (Bq kg^{-1}), was determined using the equation 1.

$$\text{Activity}, A_{cs} = \frac{N-B}{C_{eff} \times t \times m \times \gamma} \quad (1)$$

where *N* is the net count of a peak at energy *E*, *B* is the background measurement, C_{eff} is the detection efficacy at energy *E*, γ is the proportion of the radionuclide *i*'s gamma emission probability for an energy transition *E*, *m* is the mass in kg of the calculated sample, and *t* is the count period [30].

The minimum measurable activity (MMA) (Bq kg^{-1}) was assessed applying Currie's method with 95% confidence level using equation 2.

$$\text{MMA} = \frac{4.66 \times \sqrt{B}}{C_{eff} \times t \times m \times \gamma} \quad (2)$$

Where *B* is the background measurement, C_{eff} is the detection efficiency at energy *E*, γ is the proportion of the radionuclide *i*'s gamma emission probability for an energy transition *E*, *m* is the mass in kg of the calculated sample, and *t* is the count period [31].

For ^{137}Cs, the dose rate, D_R (nGyh^{-1}) is calculated using the formula in equation 3.

$$D_R = 0.03 \times A_{cs} \quad (3)$$

Where A_{cs} is the AC of ^{137}Cs and 0.03 nGyh^{-1} per Bqkg^{-1} is the dose conversions factor for the ^{137}Cs.

Annual effective dose rates, E_{eff}

The efficient yearly dosage of gamma radiation absorbed by a person, E_{eff}, is determined by the length of time that the person is exposed to gamma radiations during the year. On the basis of a report issued by UNSCEAR in 2000, it is believed that the people of metropolitan areas spend 80% of their time throughout the daytime in an indoor setting, while only twenty percent of their time is spent outside. This means that, according to the UNSCEAR, the occupancy factor for indoor radiation exposure is 0.8 while the occupancy factor for outdoor radiation exposure is 0.20 [22]. Hence, to assess the yearly effective dose for an person, a transfer factor (TF = 0.7 SvGy^{-1}) is applied to cover the absorbed dose in air to the yearly effective dose [32]. Equation 4 was used to determine the annual effective dose E_{eff}.

$$E_{eff} = TF \times t \times O_f \times D_R \times 10^{-6} \quad (4)$$

Where *t* is the time in hours for a year, i.e., 8760 hours, D_R is the dose rate in nGyh^{-1} and O_f is the transfer factor for outdoor and indoor conditions. For indoor exposure transfer factor is taken as 0.8 while 0.2 for outdoor [22].

Excessive lifetime cancer risk (ELCR)

Various regions of the body may experience a variety of health repercussions as a consequence of exposure to radiation doses of differing amounts. Expanded cancer risk per administered dose is the accepted definition of a risk factor (R_f). Exposure to a carcinogen at any point in a person's life increases

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the likelihood that they may acquire cancer later in life. Equation 5 was used to calculate the ELCR [33].

$$ELCR = E_{eff} \times DL \times R_f \quad (5)$$

Whereas E_{eff} is the yearly effective dose rate, DL is the duration of life, 75 years, for Malaysian[34], and R_f is the risk factor that is 0.05 for the public according to the ICRP. The worldwide recorded ELCR value is 0.29×10^{-3} [23,35].

Statistical analysis

The statistical evaluation, analysis of variance (ANOVA), and correlation analysis were utilized to evaluate for statistical significance and correlation between the various radiological parameter analyses in these investigations were all performed in MS-Excel 365.

RESULTS

Soil samples S1 – S9 are taken from the respective vegetable farms as presented in table 1. The AC of ^{137}Cs in the vegetables samples was below the detection threshold. Hence the radiological hazards of ^{137} Cs were not able to determine in this analysis. The data for ^{137}Cs in the agricultural soil samples gathered from vegetables farms situated in the district of Klang, Selangor is given in table 1 indicates significant variability. AC of ^{137}Cs in the soil varies from 0.34±0.09 to 3.21±0.17 $Bqkg^{-1}$ where the highest AC of ^{137}Cs is found in soil sample S1 and the lowest AC is found in soil sample S6. Dose rate computed for the corresponding value of AC ranged from 0.01 to 0.10 $nGyh^{-1}$. The soil sample, S6 has the lowest dose rate compared to soil sample S1. The E_{eff} ranged between 1.25 to 11.8 $\mu Sv\ y^{-1}$. These determined values are comparatively insignificant when evaluated to the ICRP suggested yearly dose limit of 1.0 mSv and yearly external gamma radiation dose of 0.48 mSv y^{-1}, recommended by UNSCEAR [22, 23] The values of ELCR ranged between 0.47×10^{-5} to 4.45×10^{-5} which is lesser than the safety limit of 0.29×10^{-3}. The highest value of AC and the rate contributed higher value to the E_{eff} and ELCR as for soil sample S1. These high results might be attributed to anthropogenic radionuclide fallout from earlier worldwide nuclear tests, as well as other nuclear mishaps like the FDNPP and CNPP accident. Because of these nuclear activities, ^{137}Cs was released into the air, and radionuclides from fallout accumulated in the soil; more specifically, agricultural peat soil, which was the focus of this investigation.

The analysis of variance of the AC of ^{137}Cs is at $p<0.05$ are statistically significant ($p=0.002$), according to table 2. The strong positive correlation between the AC and other variables ($p<0.05$) suggests that the agricultural soil samples taken from this research site are endowed with ^{137}Cs and contributes to the gamma dose parameters, D_R, E_{eff} and ELCR respectively.

Table 1. AC, D_R, E_{eff} and ELCR for soil samples.

Vegetable Farm	Soil Sample	Activity Concentration (Bq kg^{-1})	Dose rate, D_R ($nGyh^{-1}$)	E_{eff} ($\mu Sv\ y^{-1}$)	ELCR $\times 10^{-5}$
Japanese Mustard	S1	3.21±0.17	0.10	11.8	4.45
Water Spinach	S2	0.56±0.17	0.02	2.06	0.77
Spinach	S3	0.55±0.17	0.02	2.02	0.76
Okra	S4	1.02±0.18	0.03	3.75	1.41
Long Bean	S5	0.88±0.18	0.03	3.24	1.22
Four Angles Bean	S6	0.34±0.09	0.01	1.25	0.47
Cucumber	S7	0.59±0.24	0.02	2.16	0.81
Brinjal	S8	1.87±0.17	0.06	6.88	2.58
Bitter Gourd	S9	0.37±0.09	0.01	1.37	0.51
	Mean	1.04±0.21	0.03	3.84	1.44

Table 2. Statistical significant test.

Groups	Count	Sum	Average	Variance		
Activity Concentration (Bq kg^{-1})	9	9.390	1.043	0.880		
Dose rate, D_R ($nGyh^{-1}$)	9	0.300	0.033	0.001		
E_{eff} ($\mu Sv\ y^{-1}$)	9	34.530	3.83	11.885		
ELCR$\times 10^{-5}$	9	12.980	1.442	1.690		
ANOVA						
Source of Variation	**SS**	**df**	**MS**	**F**	**P-value**	**F crit**
Between Groups	70.123	3.000	23.374	6.467	0.002	2.901
Within Groups	115.656	32.000	3.614			
Total	185.779	35.000				

DISCUSSION

The mean value of this study was evaluated with the mean value of ^{137}Cs presented in the literature for other countries given in table 3. Soil concentration at a given place may be affected by factors beyond source receptor distance, including metrological settings for radioactive fallout and soil property, as indicated in table 3. According to the past research carried out on these locations, the AC of ^{137}Cs in Minamisohma city, 15 km from FDNPP was 66.7 $kBqkg^{-1}$ while cities located relatively far away from FDNPP (>50km) was found to have AC of about 10 $kBqkg^{-1}$ [54, 55]. The highest AC of ^{137}Cs was determined in the town of Tomioka, Fukushima, Japan. According to the researchers, the sample was taken from a decontaminated agricultural area in 2016 [52].The decontamination exercises were carried out by eliminating the polluted surface soil layer (0-5 cm depth), resurfacing the soil with granitic sand and mixing fresh soil with subsurface soil, the AC of ^{137}Cs was reduced from 9100 $Bqkg^{-1}$ to 1200 $Bqkg^{-1}$. Similar experiments were carried out 32 years after the CNPP tragedy in the heavily polluted Plavsk radioactive hotspot in the Tula area of Central Russia. The AC of ^{137}Cs in soil samples collected from an agricultural area varies from 67 to 306 $Bqkg^{-1}$.

Less than 30 kilometres away from the CNPP,

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researchers discovered ACs of 250 kBqkg^{-1}. There was a concentration of above 20 kBqkg^{-1} within 100 km of the CNPP, although the pollution pattern was very uneven and anisotropic. Areas with maximal concentrations of 2.8-23 kBqkg^{-1} were found in Belarus, Russia, and Ukraine, covering a total of 7200 and 116000 km^2 [56].

Table 3. Assessment of the present results with data from other countries.

Country	Activity Concentration (Bq kg^{-1})	References
Turkey	171	(36)
Venezuela	5.0	(37)
Bangladesh	7.0	(38)
Spain	35	(39)
Egypt	10.4	(40)
Pakistan	3.2	(41)
USA	31.5	(42)
Yugoslavia	16	(43)
Sudan	9.3	(44)
Libya	1.3	(45)
Saudi Arabia	1.0	(46)
Taiwan	14.2	(47)
India	32.7	(48)
Indonesia	1.64	(49)
Thailand	2.30	(50)
Vietnam	0.88	(51)
Tomioka, Fukushima, Japan	1200	(52)
Chernobyl, Russia	284	(53)
Present Study	1.04	

It is noticeable that nuclear weapon possessed and tester countries such as United Stated of America (USA) and India had higher AC of ^{137}Cs which is about 32 -33 Bqkg^{-1}. This suggests that areas in this region have more ^{137}Cs fallout probably due to nuclear weapon testing and may not entirely due to FDNPP or CNPP disasters. However, researcher from Turkey reported the AC of ^{137}Cs is much greater than USA or India. The researcher conclusively explained that high measurement of ^{137}Cs in this region could be due to the CNPP event.

AC of ^{137}Cs obtained from this study in Malaysia was compared to data of similar research conducted in neighbouring countries (Indonesia, Vietnam, and Thailand) where the reading is below 3 Bqkg^{-1} but the AC of 137Cs in Taiwan is 14.2 Bqkg^{-1} since it is located closer to Japan compared to other South Asia countries.

The mean value AC in the current study is determined to be one of the lowest compared to data from other nations and the criteria set by the ICRP and the UNSCEAR, 2000 report [22,23].

Conclusion

The AC of ^{137}Cs was used to calculate the D_R, E_{eff} and the ELCR where it is seldom reported especially for agricultural soil in Malaysia. It was found that the exposure to the ^{137}Cs will not cause any radiological risks to the general residents. However, continuous monitoring of the radiological risk due to ^{137}Cs in this area should be regularly conducted to ensure the protection of the general population and consumers of these vegetables.

Highlights

- Nuclear fallout radionuclide, ^{137}Cs was found in the surface soil sample in a vegetable farm
- Activity concentration of ^{137}Cs was measured to determine the radiological hazards.
- Excessive Lifetime Cancer Risk was observed to be within the safety limit.

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CONFLICT OF INTEREST: The authors wish to state that there are known conflict of interest associated with this publication.

ETHICAL CONSIDERATION: Not Applicable.

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AUTHORS' CONTRIBUTIONS: The conceptualization, methodology, and formal analysis were made by all authors. Hariandra Muthu collected the samples, do the analyses, and collected the data and drafted the manuscript. Ramesh Kasi, and Ramesh T. Subramaniam did the editing and revised the original manuscript draft. Shahid Bashir participated in the results and discussion and made the original draft.

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