Does Application of 'phosphogypsum Organic' to a Sandy BRIS Soil Has an Impact on Soil-plants-water Systems?

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**Does application of ‘Phosphogypsum organic’ to a sandy BRIS soil
has an impact on soil-plants-water systems?**

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38 **Abstract**

39 **Background:** Predominant sandy, Beach Ridges Interspersed with Swales (BRIS)
40 soil is a problematical soil for agriculture. It requires structure and capacity
41 improvement due to its weak composition, low water retention capacity and high
42 leaching of nutrients. However, there are hopes to improve it using different fertilizers
43 in combination with organic matter. Phosphogypsum organic (PG organic) is an
44 agricultural soil conditioner formulated from industrial by-products and organic filler
45 material. This research was carried out to investigate the accumulated levels of heavy
46 metals and radionuclides in water, soil and plants between BRIS soil under PG
47 organic conditioner treatment and normal BRIS soil.

48 **Results:** The result revealed that the PG organic's particles are as similar to the
49 reported pattern for calcium sulfate hydrate through powder X-ray diffraction (XRD).
50 The heavy metals measured in the surface and borehole water, and soils were below
51 the target values for raw drinking water and in the Dutch list, respectively. The values
52 for biological accumulation coefficients, contamination factors, I-geo index and
53 pollution load index (PLI) showed that there was no accumulation of metals in grain,
54 no contamination and no pollution in the BRIS soil that received from the PG organic.
55 Natural radioactivity concentrations, ^{226}Ra (^{238}U series), ^{228}Ra (^{232}Th series), and ^{40}K
56 in the collected samples were also measured. Naturally occurring radionuclide
57 concentrations, such as ^{226}Ra , ^{228}Ra , and ^{40}K , in soil and plant tissue were found to be
58 lower than the average value reported by several earlier studies in Malaysia. There
59 was no leaching of natural occurring radionuclides to the groundwater, nor was there
60 discharge to the nearest river. The calculated radium equivalent (Ra_{eq}) in soil for this
61 study was lower than the recommended value of 370 Bq/kg soil.

62 **Conclusion:** Therefore, the application of PG organic to the studied soil had no
63 impact on the soil, plants and water and suitable as a soil conditioner in BRIS soil.

64 **Keywords:** Sandy BRIS soil, Phosphogypsum organic, Heavy metals, Pollution
65 indices, Radiological hazard index.

66 **Background**

67 Phosphogypsum organic (PG organic) contains neutralization underflow (NUF)
68 residue, water leached purification (WLP) residue from rare-earth elements
69 processing, and organic filler material in a ratio of 2:1:7. The NUF residue is a solvent
70 extraction and product finishing raffinate neutralized with lime. It is also known as
71 Mg rich synthetic gypsum (MRSG) as it is primarily gypsum [1]. Although NUF is
72 classified by the Department of Environment (DOE), Malaysia as SW205, it is not a
73 hazardous material for use and does not negatively impact the environment [2-3].
74 Meanwhile, WLP residue is produced from the precipitation of post leaching liquor

75 with MgO. It is primarily iron phosphate, with minor iron hydroxide, aluminium
76 hydroxide and gypsum and contains thorium at activity concentration of about 6 Bq/g.
77 It is classified as radioactive material by the Atomic Energy and Licensing Board
78 (AELB), Malaysia and defined by the International Atomic Energy Agency (IAEA) as
79 a very low level radioactive waste [4].

80 PG organic contains no significant amounts of radionuclides other than naturally
81 occurring radionuclides (each radionuclide in the U or Th decay chain is < 1 Bq/g or
82 K-40 <10 Bq/g), ultimately is not a radioactive material as reported by Malaysia
83 Nuclear Agency (MNA) and approved by the AELB [5]. Furthermore, combination of
84 WLP and NUF has potential to improve soil nutrients (Ca^{2+} , SO_4^{2-} , Mg^{2+} , PO_4^{3-} , PO_4^{2-}
85 and SO_4^{2-}), reduce soil toxicity (SO_4^{2-} reduces Al toxicity), change soil mechanics
86 (Ca^{2+} promotes flocculation in soil hardening process) and act as liming agent to
87 increase pH of acidic soil for nutrient uptake.

88 The Beach Ridges Interspersed with Swales (BRIS) soil with more than 90% of sand
89 composition is a problematical soil in Southeastern Asia including Malaysia and
90 Indonesia [6]. It is unsuitable for agricultural purposes because of its weak structure,
91 deficient of nutrients, low capacity of water retention, high soil temperature, and
92 ultimately inadequate supports of plants to grow [7-9]. During monsoon seasons, the
93 steep cliffs layers erosion and deposition of residues and sand from the sea is
94 developed by moving sea water and create BRIS soil containing coarse sand
95 components [10]. In Malaysia, BRIS soil is widely distributed in Terengganu, the east
96 coast of Peninsular in Kelantan, Pahang and the west coast of Johor. This soil is

97 categorized into seven soil series based on drainage, depth and soil profile, including
98 Rhu Tapai, Rusila, Rudua, Rompin, Merchang, Jambu and Baging [11].

99 Although, BRIS soil is known as a problematic soil for agriculture, it is assumed that
100 fertilizer application helps to improve it. During this study, to develop specific soil
101 quality remediation and having soil with a great preference, which will be important
102 requisites in the development of agricultural product, PG organic has been applied to
103 improve BRIS soil.

104 One of the main sources of heavy metals in water (surface and ground water), soil and
105 plants is leachate migration [10-12]. Heavy metals can be transferred to plants
106 through polluted soil and contaminated waste water, and ultimately to the human's
107 foods by vegetables or animal's meat consumption [13-17]. The most challenging
108 heavy metals include chromium (Cr), lead (Pb), zinc (Zn), cadmium (Cd), copper
109 (Cu) and nickel (Ni) [17-19] contaminate natural environment. However, the toxic
110 effects of both individual and combined heavy metals on humans and other living
111 organisms are mediated by leached dosages, exposure duration and genetic factors
112 [20].

113 Technogenic and natural environmental radioactivity are the other problem in human
114 environment. Some of environmental radioactivity, like radioisotopes, including
115 strontium-90 (^{90}Sr) and technetium-99 (^{99}Tc) are results of human activity and only
116 found on Earth, some other, such as potassium-40 (^{40}K) are produced owing to the
117 natural processes and several isotopes like tritium (^3H) are the consequences of both
118 human activities and natural processes [21]. Studies of concentration of
119 environmental radioactive and their activities in various environmental systems are

essential to expect their radiological impacts on Earth. This study was conducted to investigate the accumulated levels of heavy metals (HMs) (e.g., As, Cd, Ce, La, Se, Sr, Th, Ra, Zn, B, Mn, Pb, Cr, Ag, Ba, Al, and Hg) in water, soil and plants between BRIS soil under PG organic treatment and normal BRIS soil. Furthermore, natural radioactivity concentrations e.g., ^{226}Ra , ^{228}Ra , ^{238}U and ^{232}Th in the collected samples were measured.

Materials and methods

Experimental Site

The field trial was conducted on 1.62 ha at Kampung Darat Sungai Ular, Pahang (GPS 3°50'51"N, 103°21'46"E) as shown in Fig. 1.

Fig. 1 The location of the experimental site and the Lynas Advance Material Plant (LAMP).

The implication of PG organic application to the environment was monitored for two planting seasons beginning in 2015 with 4-month cycles for each season. The residual effect of the PG organic conditioner was measured for three test crops, including corn (*Zea mays* L.), kenaf (*Hibiscus cannabinus* L.) and guinea grass (*Panicum maximum*).

Characterization of PG organic using Powder-X ray Diffraction

Powder X-ray diffraction (XRD) was performed on a Siemens D500 diffractometer, using the Cu K α radiation (1.5418 Å). The data collection was carried out from $2\theta = 10^\circ$ – 80° with a step of 0.02° and an acquisition time of 10s. Diffraction data were

142 analyzed by the Rietveld method, using the fully automated code RIETQUAN [22],
143 which is particularly useful for a quantitative evaluation of phases and reliable
144 structural and microstructural information, such as unit cell, average crystallite size
145 and lattice strain.

146 **Plot Design and Treatment**

147 The randomized complete block design (RCBD) was applied on the experimental site
148 with three replications for two seasons. There are seven treatments applied as shown
149 in Fig. 2.

150

151 **Fig. 2** The sub-plots receiving the PG organic conditioner treatment for 3 cropping
152 seasons for kenaf, maize, and guinea grass. Blue represents the sub-plot
153 receiving the soil conditioner for each cropping season, and yellow represents
154 the sub-plot receiving residual treatment.

155

156 The PG organic conditioner is a mixture of WLP: NUF: Composted oil palm empty
157 fruit bunch as filler in a ratio 1:2:7, which transferred to the site for immediate use.
158 The PG organic was applied at the top of soil surface and mixed thoroughly in the top
159 15 cm soil depth using a rotovator machine. There was allowed to equilibrate for 7
160 days before cropping of plants. Fertilizer treatments based on crop recommended rate
161 and frequencies of application were added to top of the soil under the plant canopy,
162 when permissible based on soil and foliar analyses. A drip irrigation water system was
163 applied to each plot using black plastic tape tubing. The water was supplied when
164 necessary to achieve the target moisture content of soil field capacity (FC). All soil,
165 plants and water characteristics were compared between control plots (without PG

166 organic and fertilizer) and those plots received maximum amount of PG organic as
167 conditioner (T6).

168 **Sampling of Soils, Water and Plants**

169 Soils were sampled on three occasions: before the PG organic application (base line),
170 after the PG organic application (one month after application) and at harvest. Topsoil
171 (0-30 cm) samples were taken randomly in a composite manner from the designated
172 plot to determine their physicochemical properties, the nutrient, heavy metal and
173 natural radionuclides contents. Approximately 2 kg of composite topsoil (0-30 cm)
174 samples was collected and kept in a plastic bag. Meanwhile, water was sampled from
175 boreholes within or near the plots, and surface water was sampled in the nearby
176 Sungai Ular. *In situ* water parameters, such as pH, salinity, dissolved oxygen,
177 electrical conductivity, temperature and total dissolved solids were examined in the
178 field using an YSI 556 MPS handheld multi-parameter water quality meter. All water
179 samples collected for laboratory analysis were kept in an ice box with a temperature
180 below 4°C and contained the addition of several drops of HNO₃ acid to control the pH
181 below 2. The aboveground mass of plants was cut and sampled for their stems, leaves
182 and grain for corn. The summarized of detailed information regarding the samples is
183 shown in Table 1.

184 **Table 1** **Locations of soil, plant and water sampling at UPM plots in Sungai**
185 **Ular.**

Season	Sample type	Sample ID	Note
2 nd Season	Soil	Kenaf-Soil-Control	Control sample without PG organic
		Kenaf-Soil-Control	
		Kenaf-Soil-PG organic	Soil treated with PG organic
		Guinea grass--Soil-PG organic	
	Water	BH-1	Underground water
		BH-2	
		BH-3	
		BH-4	
		BH-5	
		SW-1	Surface water (river)
		SW-2	
	Plant	Kenaf-stem-Control	Plant growing without PG organic
		Guinea grass-leaf-Control	
		Kenaf-stem-PG organic	Plant Growing with PG organic
		Guinea grass-leaf-PG organic	

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196 **Continued Table 1: Locations of soil, plant and water sampling at UPM plots in**
 197 **Sungai Ular.**

Season	Sample type	Sample ID	Note
3 rd Season	Soil	Kenaf-Soil-Control	Control sample without PG organic
		Kenaf-Soil-Control	
		Kenaf-Soil-PG organic	Soil treated with PG organic
		Guinea grass--Soil-PG organic	
	Water	BH-1	Underground water
		BH-2	
		BH-3	
		BH-4	
		BH-5	
		SW-1	Surface water (river)
		SW-2	
	Plant	Kenaf-stem-Control	Plant growing without PG organic
		Guinea grass-leaf-Control	
		Kenaf-stem-PG organic	Plant Growing with PG organic
		Guinea grass-leaf-PG organic	

198 **Characterization of Soil Physico-chemical Properties**

199 Soils were tested for their particle size distribution and organic matter content. The
 200 particle size distribution was determined using the pipette method together with dry
 201 sieving [23], whereas organic matter was determined by the loss on ignition method
 202 [23]. Soil chemical tests involved the pH (H₂O), nutrient contents (Ca, Mg, K, and P),
 203 organic carbon content, electrical conductivity and heavy metals. The soil pH in water
 204 was determined at soil: solution ratio (1/2.5) and measured using a pH meter. Organic
 205 carbon was measured with the Walkley and Black [24] method. Electrical

conductivity was measured in saturated gypsum extracts [25]. The total C, N, and S were measured using a CHNS analyzer (Model: LECO CHNS-932).

Analysis of Heavy Metal

Soils

Soil samples were dried at room temperature, ground and sieved to pass through a 2-mm sieve. For the determination of heavy metal contents, the soil was pulverized to pass through 63-micron sieves. Plant samples were cleaned in running water and rinsed with deionized water at least three times before being cut into small pieces, dried and ground using a micro-hammer mill. The total heavy metal contents in the soil was extracted with nitric acid and perchloric acid in a 3:1 ratio [26]. Heavy metal contents (As, Cd, Ce, La, Se, Sr, Th, Zn, B, Mn, Pb, Cr, Ag, Ba, Al, and Hg) were extracted by the wet digestion method. Heavy metals in solution were determined using the ICP-MS (Model: Perkin Elmers Elan 9000).

Water

The water analysis followed the standard methods proposed by American Public Health Association, APHA [27]. Heavy metal contents in water (e.g., As, Cd, Ce, La, Se, Sr, Th, Ra, Zn, B, Mn, Pb, Cr, Ag, Ba, Al, and Hg) were measured using ICP-MS.

Plants

The plant parts were separated into roots, stems, leaves and grain and analyzed for heavy metals and nutrient. The total heavy metal contents of the plants were extracted with nitric acid and hydrogen peroxide in a 3:1 ratio [28]. The heavy metal contents determined included As, Cd, Ce, La, Se, Sr, Th, Zn, B, Mn, Pb, Cr, Ag, Ba, Al, and Hg. Heavy metals in solution extracts were determined using ICP-MS.

229 **Natural Radioactivity Analysis**

230 *Sample preparation*

231 Each sample was prepared accordance to the International Atomic Energy Agency
232 (IAEA) guidelines [29]. The soil and plant samples were cleaned and dried in the
233 oven at 105°C for 72 hours, until a constant weight was achieved. Soil and plant
234 samples were then left to cool at room temperature, ground using a grinder and sieved
235 with a 500 µm sieve size to ensure the homogeneity of the samples. Meanwhile, water
236 samples were filtered out from impurities using a clean dried sieve cloth. The samples
237 were then packed into Marinelli counting beaker, sealed and for 30 days to attain
238 secular equilibrium between the parents' radionuclides (^{238}U and ^{232}Th) and their
239 respective progenies.

240 *Gamma spectrometry analysis*

241 Natural radioactivity in samples was determined after 12 hours of counting using a
242 gamma spectrometry system with a Canberra High-purity Germanium Detector
243 (HPGe) coupled to a multichannel analyzer. The HPGe detector was shielded inside
244 CANBERRA model-747 lead shielding coated with tin and copper to provide a low
245 background environment. The radioactivity concentrations of ^{226}Ra , ^{232}Th and ^{40}K
246 were determined through gamma-ray photo peaks of 1764 keV (^{214}Bi), 2614 keV
247 (^{208}Tl) and 1460 keV (^{40}K) with the corresponding emission probability of 15.2, 99.8
248 and 10.7%, respectively. The gamma spectrometry system was calibrated using a
249 standard source with a mixture of ^{22}Na , ^{51}Cr , ^{57}Co , ^{60}Co , ^{85}Sr , ^{88}Y , ^{109}Cd , ^{113}Sn , ^{137}Cs ,
250 ^{123}Te and ^{241}Am radionuclides. The analysis of gamma spectra was performed using
251 Genie-2000 software supplied by CANBERRA. The IAEA Soil-375 (certified

reference material), IARMA-004 (certified reference material) and 0802 Silica Standard (Multinuclide Standard Mixed with Silica) were used as reference in this research. The counting errors were quoted at 1-sigma absolute. Natural radionuclide concentrations in soil and plant samples were determined using Equation 1 and Equation 2, respectively, as follows [30-31].

$$W_s = \frac{M_{rm} \times A_s}{M_s \times A_{rm}} \times W_{rm} \quad (1)$$

$$W_s = \frac{A_s}{\epsilon \theta M_s} \quad (2)$$

The W_s and W_{rm} are radionuclide concentrations for sample and reference materials in $Bq\ kg^{-1}$, M_s and M_{rm} are sample and reference material masses (g), while, A_s and A_{rm} are the activities (net count per second, cps) for sample and reference materials, respectively. The ϵ and θ are the energy efficiency and abundance of the interest gamma energy peak, respectively. The accuracy of the procedure was checked by measuring the IAEA Soil-375 under the same condition and samples. The lower limit detection (LLD) and the measured activity of ^{226}Ra , ^{232}Th and ^{40}K are summarized in Table 2.

Table 2 Comparison between the measured radioactivity concentrations and the recommended values by IAEA.

Radionuclides	IAEA CRM-375		
	Recommended value ($Bq\ kg^{-1}$)	Measured value ($Bq\ kg^{-1}$)	LLD ($Bq\ kg^{-1}$)
^{226}Ra (^{238}U series)	24.4* (19 – 29.8)**	20.5	0.3
^{228}Ra (^{232}Th series)	20.5 (19.2 – 21.9)	21.7	0.2
^{40}K	424 (417 – 432)	430.6	1.2

Note: * Certified value; ** Certified value within 95% confidence level.

272 **Determination of Pollution Indices**

273 The guidelines used as a comparison to the heavy metal contents in water were the
274 available standard, such as the new Dutch List [32], Malaysian Guidelines for Raw
275 Water Quality Criteria [33] and the National Water Quality Standard (NWQS). The
276 metal contents in soil were compared with the new Dutch List [32], and the metal
277 contents in the grain were compared with values from the Malaysian Food Act [34]
278 and Food Regulation [35] standard.

279 **2.8.1 *Biological accumulation coefficient***

280 The biological accumulation coefficient (BAC), which is an indicator of the ability of
281 a plant to accumulate a specific metal in contrast to the concentration of the metal in
282 the soil substrate [36], was calculated as follows:

$$283 \quad BAC = \frac{\text{Metal concentration in plant}}{\text{Metal concentration in soil}}$$

284 Where, the heavy metal concentrations in the edible parts of the plant and the soil are
285 represented by metal concentration in the plant and metal concentration in the soil,
286 respectively.

287 **2.8.2 *Geo-accumulation index***

288 Established by Muller [37], the geo-accumulation Index (I-geo) was used to discover
289 how much metal has built up in sediments and has been employed in a number of
290 applications and for a number of research purposes. The I-geo is mathematically
291 expressed as:

292 $I\text{-geo} = \log_2(C_n/1.5B_n)$, where C_n = Concentration of element in the soil sample, and
293 B_n = the geochemical background value.

Factor 1.5 is used in the equation to take into consideration the chance of disparities in background data because of lithogenic factors. The I-geo scale is made up of seven levels (0 – 6), from no contamination to heavy pollution.

2.8.3 *Pollution load index*

The pollution index (PLI) is composed of contamination factors (CF), which are the quotients given by the division of the concentration of each metal. The CF is made of four levels (0 – 3, low CF to high CF) [38]. The PLI of the site is estimated by gathering the n-root from the CFs, which were found from the metals the PLI gathered from each location [39]. The PLI was created by Tomlinson et al. [40] using the formula below:

$$CF = C_{\text{metal}} / C_{\text{background value.}}$$

$$PLI = \sqrt[n]{(CF_1 \times CF_2 \times CF_3 \dots \times CF_n)}$$

Where, CF = contamination factor, n = number of metals, C metal = the metal concentration in soil samples and C Background value = the background value of that metal.

Determination of radiological hazard Index

A radium equivalent (Ra_{eq}) was used to estimate radiological hazard index (RHI). The significance of ^{226}Ra , ^{232}Th , and ^{40}K concentrations, with respect to radiation exposure, was defined in terms of Ra_{eq} activity in Bq kg^{-1} . The Ra_{eq} was calculated using the following equation [41]:

$$Ra_{eq} = C_{Ra} + 1.47C_{Th} + 0.077C_K \quad (4)$$

Where, C_{Ra} , C_{Th} , and C_K are the activity concentrations of ^{226}Ra , ^{228}Ra , and ^{40}K in Bq kg^{-1} , respectively. This equation is based on the estimation that 10 pCi g^{-1} of ^{226}Ra , 7 pCi g^{-1} of ^{232}Th and 130 pCi g^{-1} of ^{40}K will produce the same gamma dose rates [41-42]. The value of Ra_{eq} should be less than 370 Bq kg^{-1} to be safe for agricultural applications or building materials, as recommended by Nuclear Energy Agency (NEA) and Organization for Economic Co-operation and Development (OECD) [43].

Determination of radionuclide uptakes by plant

The uptake of ^{226}Ra , ^{228}Ra , ^{238}U , ^{232}Th and ^{40}K from soil to plants was determined using a parameter known as the transfer factor (TF), as follows [44]:

$$TF = \frac{C_p}{C_s} \quad (5)$$

Where, C_p and C_s are the concentrations of radionuclides of interest in plants and soil (Bq kg^{-1}), respectively. The transfer factor is used to describe the soil-to-plant transfer of radionuclides through the plant roots.

Quality assurance of heavy metal analysis

Quality assurance/quality control (QA/QC) was addressed by including blanks and replicates for the water, soil, and plant tissue samples. Standard reference materials (SRM) of the National Institute and Technology (NIST, 1640a for water, SRM-2711a Montana II soil, SRM 1568b rice flour and CRM-LGC7162 strawberry leaf) and internal reference materials were used for precision, quality assurance and control (QA/QC) for selected metal measurements. The average values of three replicates were taken for each determination. The precision of analytical procedures was expressed as the relative standard deviation (RSD), which ranged from 5-15% and

337 was calculated from the standard deviation divided by the mean. The recovery rates of
338 the studied metals were within $85\pm15\%$. Chemicals, stock solutions, and reagents
339 were obtained from Sigma/Fluka/Merck and were of analytical grade. All glassware
340 was washed with distilled water, soaked in nitric acid (30%) overnight, rinsed in
341 deionized water and air dried before use.

342 **Statistical Analysis**

343 All the data were analyzed using the Statistical Analysis System (SAS) version 9.4
344 software. All data were subjected to an analysis of variance (ANOVA) statistical
345 analysis to detect the variation of metal contents at different sampling times, which
346 coincided with the PG organic application. A comparison of means was performed
347 using the least significant difference (LSD) test for other crops and Student Newman
348 Keuls test for teak trees. The seedling growth model using linear regression was tested
349 for significance in the software.

350 **Results and discussion**

351 **Characterization of PG organic**

352 The XRD pattern (Fig. 3) matches well with the reported pattern for the gypsum
353 (Calcium sulfate hydrate) (PDF 00-033-0311) and display the monoclinic geometry
354 (SG: *C2/C (15)*).

355 **Fig. 3** The XRD diffractogram of PG organic conditioner using PXRD method.

356

357 The pattern indicates that the sample is crystallized, and the lattice parameter was
358 calculated to be, $a = 6.284$, $b = 15.20$, $c = 5.677$. However, there are other heavy metals

existing on a sample, including thorium, magnesium, and carbon. The XRD peaks of ThO₂ are in good agreement with the cubic geometry (SG: *Fm3m*) with the lattice constants $a = 5.597 \text{ \AA}$, which is a good agreement with reported in the literature for bulk ThO₂ (PDF00-042-1462).

Physico-chemical Properties of Soil

Physico-chemical properties of baseline soil before and after PG organic application

The soil is a sandy type, with particles dominated by more than 80% sand (Table 3).

Table 3 Soil properties before and after PG organic application at Sungai Ular research plots.

Parameter	Sampling frequency							
	22-05-2015	22-01-2016	27-09-2016		08-12-2016		24-04-2017	
	before (baseline)	after PG organic application	Control	Max	Control	Max	Control	Max
Sand (%)	87	90	ND	ND	90	91	84	85
Silt (%)	8	4	ND	ND	7	5	9	8
Clay (%)	5	6	ND	ND	3	4	7	7
Texture	Sand	Sand	ND	ND	Sand	Sand	Sand	Sand
Organic matter (%)	6.14	5.45	5.02	5.25	6.10	5.78	5.60	5.64
pH water	4.11	4.63	5.13	5.34	4.15	4.25	5.79	6.08
EC (mS/cm)	2.04	2.05	1.87	1.96	1.94	1.94	1.99	2.10
Exch Al (cmol ⁺ /kg)	0.47	0.04	ND	ND	0.01	0.01	0.05	0.04
Exch H (cmol ⁺ /kg)	0.33	0.01	ND	ND	0.02	0.01	0.04	0.03
ExchCa (cmol ⁺ /kg)	0.34	0.59	ND	ND	1.53	1.70	0.16	0.22
Exch Mg (cmol ⁺ /kg)	0.34	0.37	ND	ND	0.86	1.01	0.71	0.69
Exch Na (cmol ⁺ /kg)	0.19	0.11	ND	ND	1.52	1.18	0.22	0.36
Exch K (cmol ⁺ /kg)	0.09	0.30	ND	ND	0.45	0.46	0.09	0.09
CEC (cmol ⁺ /kg)	1.75	1.41	ND	ND	4.38	4.36	1.20	1.42
Available P (mg/kg)	2.44	5.35	ND	ND	1.60	1.39	1.90	2.04
Total C (%)	1.70	3.17	2.92	3.05	16.80	15.17	2.30	2.48
Total N (%)	1.16	1.05	0.16	0.02	0.28	0.28	0.10	0.22
Total S (%)	0.13	0.13	0.01	0.02	0.12	0.09	0.02	0.02
C/N ratio	1.47	3.02	18.25	152.50	59.36	54.40	22.35	13.02

Note: ND: not detected; Max: maximum treatment.

372 The organic matter content remained unchanged throughout the study period. Soil pH
373 was strongly acidic but improved significantly at the last harvest from 4.11 to 5.64.
374 The electrical conductivity (EC) and cation exchange capacity (CEC) remain
375 unchanged throughout the study period, with value of ≤ 2.0 mS/cm and <2.0 cmol⁺/kg
376 soil, respectively. The CEC value was extremely low, which is typical for BRIS soil.
377 The available P showed no significant difference before and after the PG organic
378 treatment. Total C significantly increased from 1.7 to 3.47% after PG organic
379 application; however, total N and S showed the opposite trend.

380 ***Effects of PG organic application on water surface and boreholes in the study area***

381 The pH values of surface water and water in the boreholes were acidic (Table 4). The
382 pH of water in the borehole was within Class IV of the National Water Quality
383 Standard (Environmental Quality Report, 2013). The pH of surface water was below
384 the Class IV range in the NWQS standard. Other *in situ* parameters fluctuated during
385 different sampling times. This *in situ* parameter could be influenced by the amount of
386 water in the borehole and stream due to changing seasons and the rising and falling of
387 the tide.

388 **Table 4 Physico-chemical properties and macronutrients contents in boreholes and surface water before and after PG organic**
389 **application.**

Parameters	Baseline (22-05-2015)		After PG organic application (22-01-2016)		After PG organic application Harvest 1 (17-09-2016)		After PG organic application Harvest 2 (08-12-2016)		After PG organic application Harvest 3 (10-05-2017)		Class IV NWQS
	Borehole	Surface Water	Borehole	Surface Water	Borehole	Surface Water	Borehole	Surface Water	Borehole	Surface Water	
pH	4.70	5.08	6.10	4.44	5.89	4.29	5.69	3.96	4.93	5.04	5-9
Temp (°C)	-	-	24.96	24.35	25.70	25.23	25.23	24.58	29.47	30.12	-
EC mS/cm)	0.06	6.52	0.04	0.08	0.05	82.00	0.07	0.40	0.11	0.30	6
TDS (g/L)	-	-	26.00	51.50	84.40	237.50	0.05	0.26	65.60	176.50	4000
Salinity (%)	-	-	0.02	0.04	0.06	0.20	0.03	0.20	0.05	0.13	2
DO (mg/L)	-	-	2.98	4.53	4.52	5.45	5.37	2.93	5.61	7.58	< 3
Al (µg/L)	37.43	804.64	12.42	631.10	97.93	420.54	48.35	554.07	261.59	607.21	NL
Ca (µg/L)	2681.89	33978.22	2229.5	1827.4	1149.86	2626.9	1074.8	707.21	1285.22	781.52	NL
Mg (µg/L)	667.20	193666.9	684.64	1151.9	1904.50	3679.5	766.83	537.17	10595.62	6958.86	NL
K (µg/L)	1543.86	42289.70	1736.7	1656.0	2614.78	5648.2	1289.2	828.94	5989.34	3481.78	NL
Na (µg/L)					4041.91	6010.9	1335.1	2012.1	2700.6	94031.1	NL

390Note: ND: not detected; Source of standard: *Malaysian Standard for Raw Drinking Water Quality [33]; the new Dutch list [32]; within rows,
391means followed by a common letter are not significantly different using LSD test at the 5% level of significance.

392 The salinity and EC readings in the boreholes were lower than the limit in the Class
393 IV standard. Surface water recorded a high EC reading at the baseline compared to
394 the Class IV standard. A high DO reading of more than 3 mg/L indicated that both
395 boreholes and surface water were much better than Class IV in the NWQS standard.
396 The cation contents (Al, Ca, Mg and K) in the surface water and in the boreholes
397 varied significantly with time of sampling.

398 *Effects of PG organic conditioner on nutritional values of plants*

399 Nutrient contents in plants and corn grain in three harvests are presented in Table 5.
400 Nutrient contents in the plants of the plot treated with PG organic were compared with
401 the control plot.

402 Macronutrients in control plants (Table 5) showed a significantly higher content
403 ($p < 0.05$) or were not significantly different ($p > 0.05$) from the PG organic treated
404 plant. Total N, C, and S contents also showed a similar level in treated and control
405 plants. The macronutrient contents in corn grains treated with PG organic were not
406 significantly different ($p > 0.05$) or significantly lower than ($p < 0.05$) the control (Table
407 5).

408 Carbon, N and S follow a similar pattern. There were significant increases in K and P
409 in the third harvest season compared to the second harvest season. An increase in
410 macronutrients is considered normal since they are extensively used as conventional
411 fertilizer in agriculture.

412

413 **Table 5 Nutrient contents of cultivated plants and corn grain in Sungai Ular research plots.**

Parameter (Plants)	Sampling frequency					
	27-09-2016		08-12-2016		24-04-2017	
	Control	Max	Control	Max	Control	Max
Total Ca (mg/kg)	1934.67a	914.70b	1981.95a	1909.94a	NPG	1651.62a
Total Mg (mg/kg)	1306.91c	1966.74b	10862.66a	9402.54a	NPG	1987.68b
Total K (mg/kg)	5164.62a	3433.87b	6877.75a	2271.21c	NPG	1745.17d
Total Al (mg/kg)	-	-	92.86a	53.00b	NPG	78.06a
Total P (mg/kg)	945.30b	829.45b	1968.21a	2043.75a	NPG	791.64c
Total C (%)	-	-	40.30a	40.23a	NPG	39.70a
Total N (%)	-	-	1.26a	1.28a	NPG	1.27a
Total S (%)	-	-	0.39a	0.39a	NPG	0.38a
C/N ratio	-	-	31.89a	31.52a	NPG	31.26a
Parameter (corn grain)	27-09-2016		08-12-2016		24-04-2017	
	Control	Max	Control	Max	Control	Max
	Control	Max	Control	Max	Control	Max
Total Ca (mg/kg)	-	-	1879.24a	1449.51b	27.50c	33.01c
Total Mg (mg/kg)	-	-	1964.66a	1954.59a	735.95b	820.60b
Total K (mg/kg)	-	-	2333.09b	1658.17c	15301.32a	16092.46a
Total Al (mg/kg)	-	-	75.67a	79.06a	28.27b	11.04c
Total P (mg/kg)	-	-	1587.28b	695.60c	2759.50a	3240.87a
Total C (%)	-	-	40.21a	39.61a	33.25b	34.98b
Total N (%)	-	-	1.26a	1.27a	0.75c	0.93b
Total S (%)	-	-	0.39a	0.39a	0.08c	0.12b
C/N ratio	-	-	31.88	31.14	44.53	37.48

414 Note: ND: not detectable; max: maximum treatment; NPG: no plant growing; within rows, means followed by a common letter are not
 415 significantly different using LSD test at the 5% level of significance.

416 **Heavy metal measurements**

417 *Heavy metals concentrations of baseline soil before and after PG organic*
418 *application*

419 The content of cytotoxic (As, Cd, Cr, Pb, Hg and Se) heavy metals in soil after PG
420 organic application did not show a significant increase from the metal contents before
421 PG organic applications throughout the study period (Table 6). All values were well
422 below the target values stipulated in the New Dutch List [32]. Phytotoxic metals
423 showed a significant ($p<0.05$) increase in Zn and Mn contents, but not B (Table 6).
424 Zinc and Mn are plant micronutrients normally added to fertilizer as trace elements to
425 improve plant growth and yield. The Zn content in the soil was lower than the target
426 values provided by the New Dutch List [32].

427 For the content of other metals, Ce, Sr and La showed significantly higher contents in
428 the maximum PG organic treatment at the second harvest compared to the baseline
429 data but decreased significantly at the third harvest.

430

431 **Table 6 Heavy metals content of soil before and after PG organic application at Sungai Ular research plots.**

Heavy metals (mg/kg)	Sampling frequency								Target value* (mg/kg)
	22-05-2015	22-01-2016	27-09-2016 Harvest 1		08-12-2016 Harvest 2		24-04-2017 Harvest 3		
	Before (baseline)	After PG organic application	Control	Max	Control	Max	Control	Max	
Total As	4.03b	2.97bc	2.24cd	1.49d	ND	ND	3.15b	5.54a	29
Total Cd	0.09c	0.07c	0.14b	0.06c	0.29a	0.25a	0.08c	0.08c	0.8
Total Cr	-	11.04a	8.53b	5.52c	BDL	BDL	6.61c	7.96b	100
Total Pb	-	10.41c	27.48a	16.65b	20.72b	19.76b	11.22c	11.71c	50
Total Hg	-	0.06a	ND	ND	ND	ND	0.03b	0.02b	0.5
Total Se	0.27c	0.85a	ND	ND	0.44c	0.34c	0.41c	0.41c	0.7
Extractable B	0.14d	0.09d	1.33c	1.26c	7.30a	3.35b	0.11d	0.12d	-
Total Zn	21.16d	18.53d	10.48e	9.30e	35.09b	26.80c	34.47b	40.31a	140
Total Mn	21.43e	41.98c	32.39d	33.89d	54.69b	87.74a	22.55e	44.47c	-
Total Ce	4.03f	-	12.73e	116.19b	20.84d	197.89a	11.06e	87.92c	-
Total La	1.96e	-	6.46d	70.80b	12.60c	137.43a	3.82d	53.95b	-
Total Sr	8.38c	12.57c	15.73c	20.73b	14.45c	29.91a	8.19d	11.74c	-
Total Ag	-	0.02b	0.06a	0.03b	BDL	BDL	0.07a	0.03b	-
Total Ba	-	33.00a	13.33c	13.14c	8.82d	21.71b	4.30e	28.55a	200

432 Note: Source of standard: *the new Dutch list [32]; Max: maximum treatment; within rows, means followed by a common letter are not
433 significantly different using LSD test at the 5% level of significance.

434 ***Effects of PG organic application on boreholes and surface water***

435 Data for heavy metal contents in water were separated between the boreholes and
436 surface water (Table 7). This was to give a better data comparison to their respective
437 values before PG organic application. Concentrations of cytotoxic heavy metals (As,
438 Cd, Cr, Pb, Hg, and Se) in water boreholes were generally significantly lower than
439 those in the surface water (Table 7). Their concentrations fluctuated with the time of
440 sampling. All values were lower than the target value stipulated in the Malaysian
441 Standard for Raw Drinking Water Quality [33]. Phytotoxic heavy metal (Zn, Mn, and
442 B) concentrations fluctuated with the time of sampling. The contents of these metals
443 in the boreholes were significantly lower than in the surface water. Their contents in
444 the borehole water were not significantly different with the baseline data and lower
445 than the target values in the Malaysian Standard for Raw Drinking Water Quality
446 [33]. The other metal concentrations in boreholes also fluctuated with the time of
447 sampling. Their concentrations in the boreholes were significantly lower than in the
448 surface water. The application of PG organic to the trial plot in Sungai Ular could
449 influence the metal concentrations in the boreholes but could not influence the metal
450 contents in the river due to the proximity of the trial plot. Any changes in metal
451 contents in the river were a direct contribution of the natural input and anthropogenic
452 input from the surrounding environment. Evidence from the data gathered in this
453 monitoring program showed that the application of PG organic did not accumulate
454 and contaminate the borehole water.

455 **Table 7 Heavy metals contents in boreholes and surface water before and after PG organic application.**

Heavy metals (µg/L)	Baseline (22-05-2015)		After PG organic application (22-01-2016)		After PG organic application Harvest 1 (17-09-2016)		After PG organic application Harvest 2 (08-12-2016)		After PG organic application Harvest 3 (10-05-2017)		*Target value (µg/L)
	Borehole	Surface Water	Borehole	Surface Water	Borehole	Surface Water	Borehole	Surface Water	Borehole	Surface Water	
As	0.30	4.08	0.05	0.85	0.21	0.99	0.86	2.81	0.18	1.40	10
Cd	0.01	0.17	0.02	1.04	0.04	0.42	0.04	0.03	0.05	0.02	0.3
Cr	-	-	0.61	3.59	0.38	4.06	0.19	1.84	2.02	3.02	50
Pb	-	-	0.39	3.56	0.70	4.52	ND	ND	7.27	0.68	10
Hg	-	-	0.06	0.05	ND	ND	ND	ND	1.15	1.09	1
Se	0.24	4.18	0.25	0.15	0.42	1.73	0.17	0.22	3.25	1.85	10
B	34.83	272.73	18.82	24.76	11.31	50.63	7.31	19.60	18.20	49.67	-
Zn	45.10	101.70	51.97	81.99	54.67	198.75	19.70	51.11	29.61	80.79	3000
Mn	19.44	82.80	5.08	32.25	26.70	398.57	8.94	25.92	23.65	80.61	200
Th	0.03	0.26	0.68	36.75	0.68	105.85	0.01	0.33	0.17	0.87	-
Ce	0.11	0.38	10.80	226.73	0.34	43.83	0.16	1.36	5.37	5.23	-
La	0.04	0.23	10.14	183.78	46.68	119.34	0.08	0.56	1.81	1.66	-
Sr	22.32	725.65	27.13	16.44	0.00	1.59	24.08	9.90	62.64	58.37	-
Ag	-	-	0.03	0.17	0.00	0.05	0.00	0.00	ND	ND	-
Ba	-	-	99.77	102.13	79.70	163.48	35.87	53.44	90.49	168.62	50

456 Note: ND: not detected; Source of standard: *Malaysian Standard for Raw Drinking Water Quality [33]; the new Dutch list [32].

457

458

459 *Effects of PG organic treatment on metal contents of cultivated plants and corn*
 460 *grain in the experiment area*

461 Metal contents in plants and in corn grain in three harvests are presented in Tables 8,
 462 respectively.

463 **Table 8 Heavy metals contents of cultivated plants and corn grain in Sungai**
 464 **Ular research plots.**

Heavy metals (Plants)(mg/kg)	Sampling frequency						
	27-09-2016		08-12-2016		24-04-2017		
	Control	Max	Control	Max	Control	Max	
Total As	0.68a	0.62a	ND	ND	NPG	0.10	
Total Cd	0.07b	0.09b	0.21a	0.22a	NPG	0.03c	
Total Pb	0.86b	0.64c	2.22a	0.98b	NPG	0.24d	
Total Cr	0.38b	0.33b	ND	ND	NPG	1.94a	
Total Hg	0.02a	0.01a	ND	ND	NPG	ND	
Total Se	0.18a	0.13b	0.05c	0.04c	NPG	0.06c	
Extractable B	29.81a	15.55b	26.64a	33.96a	NPG	6.25c	
Total Zn	142.33a	71.47b	35.37c	39.79c	NPG	9.17d	
Total Mn	15.59d	4.17e	92.98a	63.99b	NPG	49.64c	
Total Ce	1.47b	2.08b	0.34c	0.30c	NPG	5.39a	
Total La	0.97a	1.54a	0.34b	0.35b	NPG	0.16c	
Total Sr	16.63b	9.21c	58.49a	68.06a	NPG	11.18c	
Total Th	0.13b	0.12a	ND	ND	NPG	0.08c	
Total Ag	0.02a	0.00b	ND	ND	NPG	ND	
Total Ba	2.21c	1.11d	10.28a	8.29b	NPG	1.08d	
Heavy metals (Corn grain)(mg/kg)	27-09-2016		08-12-2016		24-04-2017		*Limit mg/kg
	Control	Max	Control	Max	Control	Max	
Total As	-	-	0.02c	ND	0.37a	0.22b	< 1
Total Cd	-	-	0.00	0.36a	0.01a	0.01a	< 1
Total Pb	-	-	0.07c	0.30a	0.44a	0.30a	< 2
Total Cr	-	-	1.43c	ND	3.17b	4.55a	-
Total Hg	-	-	ND	ND	ND	ND	-
Total Se	-	-	0.01b	0.03a	0.04a	0.02ab	< 0.05
Extractable B	-	-	2.82b	2.24b	23.88a	20.49a	-
Total Zn	-	-	10.63c	18.63b	40.91a	38.12a	-
Total Mn	-	-	7.52b	5.67c	6.69bc	9.91a	-
Total Ce	-	-	0.29c	0.20c	1.38a	0.69b	-
Total La	-	-	0.07d	0.23c	0.70a	0.47b	-
Total Sr	-	-	1.47a	0.80d	1.07b	0.94c	-
Total Th	-	-	0.02	ND	ND	ND	-
Total Ag	-	-	ND	ND	ND	ND	-
Total Ba	-	-	ND	ND	1.06a	0.97a	-

465 Note: ND: not detected; max:maximum treatment; NPG: no plant growing; source of
 466 standard:*Malaysian Food Act [34] and Food Regulation [35]; within rows, means
 467 followed by a common letter are not significantly different using LSD test at the 5%
 468 level of significance.

469 Metal contents in the plants of the plot treated with PG organic were compared with
470 the control plot. The metal contents in plants were also compared with the standard
471 given in Kabata-Pendias [45] and compared with the Malaysian Food Act [34] and
472 Food Regulation [35] for grain.

473 Cytotoxic heavy metal (As, Cd, Cr, Pb, Hg and Se) contents in plants treated with PG
474 organic were not significantly different or significantly lower than ($p < 0.05$) the
475 control. Phytotoxic metals (B, Zn, and Mn) in plants treated with PG organic were
476 significantly lower or not significantly different compared to the control plants. These
477 metals were also micronutrients for plants; therefore, their high contents in the plants
478 are desirable. There was no significant difference in the contents of other metals
479 between the plants treated with PG organic and the control plants. There was no
480 indication of metals accumulating with season of sampling.

481 Cytotoxic heavy metals (As, Cd, Pb, Cr, Hg and Se) recorded a low value compared
482 to the limit stipulated in the Malaysian Food Act [34] and Food Regulation [35]
483 standards. Phytotoxic heavy metals B, Mn and Zn in the plots treated with PG organic
484 were significantly higher than the control, which suggested that PG organic
485 application can contribute to a higher micronutrient supply to the plant.
486 Concentrations of the other metals were low, and in general, the contents of metals in
487 grain from plots treated with PG organic were significantly lower ($p < 0.05$) than the
488 control. Data of heavy metals in grains showed that there was no accumulation of
489 metals and lower contents of heavy metals compared to the standard. Therefore, PG
490 organic is safe for use as a soil conditioner.

491 **Heavy metals uptake by plants**

492 The uptake of heavy metals in grain is shown by its biological accumulation
 493 coefficient (BAC), and the accumulation and contamination of heavy metals in soil
 494 are shown by its geo-accumulation Index (I-geo), contamination factor (CF) and PLI
 495 in Table 9.

496 **Table 9 Biological accumulation coefficients of heavy metals in grains and soil**
 497 **contamination levels following treatment with PG organic at Sungai**
 498 **Ular.**

	Metal		SungaiUlar UPM plot	Notes
	As	BAC	0.19	Moderate
501		I-geo	-2.89	Uncontaminated
		CF	0.20	low CF
	Cd	BAC	0.75	Moderate
502		I-geo	-1.38	Uncontaminated
		CF	0.58	low CF
	Pb	BAC	0.05	Low
503		I-geo	-0.36	Uncontaminated
		CF	0.45	low CF
	Cr	BAC	0.03	Low
504		I-geo	-3.86	Uncontaminated
		CF	0.10	low CF
505	Se	BAC	0.00	Weak
		I-geo	0.00	Uncontaminated
		CF	0.00	low CF
506	Zn	BAC	0.61	Moderate
		I-geo	-3.08	Uncontaminated
507		CF	0.18	low CF
	Mn	BAC	1.76	Active
		Igeo	-4.29	Uncontaminated
508		CF	0.07	low CF
	B	BAC	20.88	Active
509		Igeo	-6.87	Uncontaminated
		CF	0.01	low CF
		PLI	0.25	Not polluted

510 The BAC factors >1 indicate that the plant species has the ability to store metals from
 511 the soil in the shoots [46]. In contrast, BAC factors <1 indicates that the plant species
 512 do not store metals from the soil in the shoots. The corn grain in Sungai Ular showed

513 BAC values of less than 1, except for B and Mn, which meant that all metals do not
514 have the potential to be accumulated in the plant. Boron and Mn have a BAC of more
515 than 1, indicating very active uptake. These metals are micronutrients normally used
516 by plants for their growth.

517 The I-geo index is a quantitative measure of the degree of pollution in aquatic
518 sediments or soils. It consists of seven grades ranging from unpolluted to a very
519 extremely polluted environment. The I-geo index showed that all heavy metals in
520 Sungai Ular fall in grade 0 (Table 9). This suggests that the BRIS soils at the research
521 plot at Sungai Ular were not contaminated.

522 The PLI was calculated from the contamination factors (CF). This CF is the quotient
523 obtained by dividing the concentration of each metal by its background concentration.
524 The PLI values of the plot were calculated by obtaining the n-root from the n-CFs that
525 were obtained for all metals from each plot. Pollution severity and its variation among
526 the trial plots were determined with the use of the PLI. This index is a quick tool in
527 order to compare the pollution statuses of different places. A PLI value = 0 is
528 'Perfection', $PLI=1$ indicates baseline levels of pollutants in the examined sample, and
529 $PLI>1$ denotes a steady increase of pollution in the specimen [40, 47]. The results of
530 the present study showed that the CF values of all of the metals in the study area were
531 low (<1) indicating that there was no increase of heavy metals in the soil. The PLI
532 values for all heavy metals in Sungai Ular were very low, indicating that the soils
533 were not polluted by the application of PG organic.

534 **Radiological Environmental Monitoring Program**

535 *Quality control of the analysis data*

536 The measured radioactivity concentrations for ^{226}Ra (^{238}U series), ^{228}Ra (^{232}Th series)
537 and ^{40}K for the gamma spectrometry system were in the range of the 95% confidence
538 level, as proposed by the IAEA for reference material CRM-375 (Table 10).
539 Moreover, the MDA analysis indicated that the lower limits of detection for ^{226}Ra ,
540 ^{228}Ra and ^{40}K were 0.7, 0.2 and 1.2 Bq kg⁻¹, respectively. Based on both analyses, it
541 can be judged that the gamma spectrometry system was in good condition, and the
542 results were acceptable during the analysis period.

543 *Natural radioactivity concentrations in soil*

544 The analysis revealed that the radioactivity concentrations of ^{226}Ra , ^{228}Ra , and ^{40}K in
545 all samples (Table 10) were found to be much lower than the average concentrations
546 in Malaysia's soil ($^{226}\text{Ra} = 67.0 \text{ Bq kg}^{-1}$; $^{228}\text{Ra} = 82.0 \text{ Bq kg}^{-1}$; $^{238}\text{U} = 66 \text{ Bq kg}^{-1}$; and
547 $^{232}\text{Th} = 82.0 \text{ Bq kg}^{-1}$), as reported by the United Nations Scientific Committee on the
548 Effect of Atomic Radiation [48].

549 The comparison between the previous studies performed in Malaysia showed that the
550 results obtained in this study were in the normal range, as shown in Table 10. The
551 differences in natural radionuclide concentrations in soil are related to several factors,
552 such as geographical factors and geological conditions, as well as to the extent that
553 fertilizer is used on the land in the agricultural industry [49].

554 In addition, the findings showed that ^{40}K had the highest concentrations compared to
555 ^{226}Ra and ^{228}Ra . Potassium (or ^{40}K) is a mobile nutrient that tends to move and
556 accumulate on the soil surface [50].

557 **Table 10 Comparison of radioactivity concentrations of ^{226}Ra , ^{228}Ra and ^{40}K in**
558 **soil samples obtained in this study with other literature.**

Season	Sample ID		Activity Concentration (Bq/kg)		
			²²⁶ Ra	²²⁸ Ra	⁴⁰ K
2 nd Season	Kenaf-Soil-Control	Range	13.8 - 17.6	15.7 - 16.8	44.5 - 47.0
		Average	15.7 ± 2.7	16.3 ± 0.8	45.7 ± 1.8
	Kenaf-Soil-Condi	Range	14.0 - 16.0	18.0 – 19.9	46.7 - 49.7
		Average	15.0 ± 1.4	19.0 ± 1.4	48.2 ± 2.1
	ANOVA Analysis		0.77 ^{ns}	0.13 ^{ns}	0.12 ^{ns}
	Guinea grass-Soil-Control	Range	14.4 - 16.8	15.6 - 17.1	59.6 - 72.8
		Average	15.6 ± 1.7	16.3 ± 1.1	66.2 ± 9.3
	Guinea grass-Soil-PG organic	Range	15.4 - 17.5	27.6 - 34.7	54.8 - 65.4
		Average	16.5 ± 1.5	31.2 ± 5.0	60.1 ± 7.5
	ANOVA Analysis		0.70 ^{ns}	0.06 ^{ns}	1.00 ^{ns}
3 rd Season	Kenaf-Soil-Control	Range	13.3 - 14.7	16.5 - 17.0	49.3 - 55.5
		Average	14.0 ± 1.0	16.8 ± 0.4	52.4 ± 4.4
	Kenaf-Soil-PG organic	Range	16.7- 17.6	19.2 - 21.0	47.8 - 57.7
		Average	17.1 ± 0.6	20.1 ± 1.3	52.8 ± 7.0
	ANOVA Analysis		0.07 ^{ns}	0.07 ^{ns}	0.96 ^{ns}
	Guinea grass-Soil-Control	Range	12.2 - 14.8	16.0 - 16.7	47.7 - 53.2
		Average	13.5 ± 1.8	16.3 ± 0.4	50.4 ± 3.9
	Guinea grass-Soil-PG organic	Range	13.7 - 15.1	16.8 - 18.1	60.6 - 61.4
		Average	14.4 ± 1.0	17.6 ± 0.7	61.0 ± 0.6
	ANOVA Analysis		0.60 ^{ns}	0.05*	0.06 ^{ns}
Malaysia’s Soil [48]			67.0	82.0	310
Palm Oil Plantation, Malaysia [51]			21.5 ± 8.4	29.2 ± 8.9	56.8 ± 36.3
Paddy Plantation, Malaysia [52]			83.6 ± 40.4	108.1 ± 28.4	403.8 ± 224.8
Kuala Krai, Kelantan [53]			40.2 – 264.0	29.2 – 312.9	491.1 – 1184.2

559 Note: ns: not significant; * and **: significant at $p \leq 0.05$ and $p \leq 0.01$, respectively.

561 The concentrations of ^{40}K in soil do not contribute significantly to the total
562 radioactive concentration in soil because ^{40}K is not from the uranium or thorium
563 decay series [53]. Potassium-40 exists in nature, but the activity concentration may
564 increase due to the use of fertilizer in agriculture.

565 Even though there were variations in the concentrations of ^{226}Ra , ^{228}Ra and ^{40}K in soil
566 before and after the application of PG organic for each season, the statistical analysis
567 (ANOVA) revealed that there were no significant differences ($p > 0.05$) in the natural
568 radionuclide concentrations due to the PG organic treatment. Based on this finding, it
569 can be concluded that the addition of PG organic as a soil conditioner for BRIS soil
570 did not increase radionuclides in soil.

571 *Natural radioactivity concentrations in water*

572 The activity concentrations of ^{226}Ra , ^{228}Ra and ^{40}K were different for each location
573 (Table 11). For example, the activity concentration of ^{228}Ra for both seasons ranged
574 from 0.3 to 3.9 Bq/L and 0.6 to 3.9 Bq/L, respectively. Even though the
575 concentrations of natural radionuclides in samples look different for each sampling
576 point (BH1 to BH5), the statistical analysis revealed that there were no significant
577 differences ($p > 0.05$) in ^{226}Ra , ^{228}Ra and ^{40}K concentrations in water for both seasons.

578 The activity concentrations of ^{226}Ra , ^{228}Ra and ^{40}K in water recorded from this study
579 were lower than the levels reported by Almayahi et al. [41] in water samples sampled
580 along the Northern Peninsula of Malaysia.

581

582

583 **Table 11 Activity concentrations of ^{226}Ra , ^{228}Ra and ^{40}K in water obtained from**
584 **studied location.**

Season	Sample ID		Activity Concentration (Bq/L)		
			²²⁶ Ra	²²⁸ Ra	⁴⁰ K
2 nd Season	BH-1	Range	0.5 - 0.9	1.1 – 3.9	6.9 - 8.0
		Average	0.7 ± 0.3	2.5 ± 2.0	7.4 ± 0.8
	BH-2	Range	0.4 - 0.5	0.9 - 1.1	1.8 - 9.8
		Average	0.5 ± 0.1	1.0 ± 0.1	5.8 ± 5.6
	BH-3	Range	BDL - 0.3	0.4 - 0.6	1.4 - 1.8
		Average	0.3 ± 0.0	0.5 ± 0.1	1.6 ± 0.3
	BH-4	Range	0.3 - 0.5	0.7 - 0.8	0.4 - 7.2
		Average	0.4 ± 0.2	0.8 ± 0.1	3.4 ± 5.4
	BH-5	Range	BDL	0.3 - 0.6	3.3 - 5.1
		Average	DBL	0.5 ± 0.2	4.2 ± 1.3
	Average for BH		0.4	1.1	4.5
	SW-1	Range	BDL - 0.3	0.6 - 0.9	2.5 - 2.9
		Average	BDL	0.8 ± 0.2	2.7 ± 0.3
	SW-2	Range	0.3 - 0.7	1.3 - 1.8	2.9 - 6.5
		Average	0.5 ± 0.3	1.5 ± 0.4	4.7 ± 2.6
	Average for SW		0.4	1.2	3.7
3 rd Season	BH-1	Range	BDL - 0.3	1.6 - 3.2	6.5 - 11.6
		Average	0.2 ± 0.1	2.4 ± 1.1	9.0 ± 3.6
	BH-2	Range	0.3 - 0.4	0.6 - 2.3	7.6 - 8.0
		Average	0.3 ± 0.0	1.5 ± 1.2	7.8 ± 0.3
	BH-3	Range	0.3 - 0.4	0.9 - 1.7	2.2 - 3.6
		Average	0.4 ± 0.0	1.3 ± 0.5	2.9 ± 1.0
	BH-4	Range	0.4 - 0.6	1.8 - 2.0	5.4 - 5.8
		Average	0.5 ± 0.2	1.9 ± 0.1	5.6 ± 0.3
	BH-5	Range	BDL	0.7 - 1.1	2.2 - 5.8
		Average	0.1* ± 0.0	0.9 ± 0.3	4.0 ± 2.6
	Average for BH		0.3	1.6	5.9
	SW-1	Range	0.3 - 0.4	1.7 - 2.9	6.1 - 10.1
		Average	0.4 ± 0.0	2.3 ± 0.9	8.1 ± 2.8
	SW-2	Range	BDL	0.5 - 1.3	4.7 - 5.8
		Average	BDL	0.9 ± 0.6	5.2 ± 0.8
	Average for SW		0.3	1.5	6.6
ANOVA analysis for BH for both seasons			0.31 ^{ns}	0.27 ^{ns}	0.38 ^{ns}
ANOVA analysis for SW for both seasons			1.00 ^{ns}	0.62 ^{ns}	0.24 ^{ns}
16 water sample at Norther Peninsular Malaysia [41]			0.9 – 7.0	0.6 – 8.6	53.0 – 222.0
Tap, stream, lake, river water at Seberang Perai, Malaysia [54]			3.3 – 4.3	0.9 – 2.2	134.8 – 172.4
10 drinking water samples at Malaysia [55]			0.02 – 0.09	0.02 – 0.08	1.7 – 8.8

585 Note: ND: Not Detected; ns: not significant; Source of standard: *The reported value is
586 below the detection limit; Lower limit detection (LLD): ^{226}Ra (0.3 Bq/L); ^{228}Ra (0.1
587 Bq/L); ^{40}K (1.2 Bq/L).

588 This finding also suggested that there was no leaching of the radionuclides from soil
589 to groundwater. The activity concentrations of ^{226}Ra , ^{228}Ra and ^{40}K in groundwater
590 depended greatly on the type of minerals derived from aquifer rocks and soil
591 compounds [56]. In addition, the surface water analysis from the nearest river also
592 suggested that there is no accumulation (discharge) of natural radionuclides in the
593 water due to the application of PG organic. Based on this finding, it can be concluded
594 that the applications of PG organic did not contribute to groundwater and surface
595 water contamination during the studied period.

596 *Natural radioactivity concentrations in plants*

597 There was a significant difference in the activity concentration for ^{40}K in Guinea grass
598 (2nd season) and kenaf (3rd season) between the control sample and treated sample
599 (PG organic sample) (Table 12). The average concentrations of ^{40}K in the control
600 samples for Guinea grass leaf (2nd season) and kenaf stem (3rd season) were recorded
601 to be 19.9 ± 8.5 and 44.8 ± 3.1 Bq/kg, respectively. On the other hand, the average
602 activity concentrations of ^{40}K in the samples treated with PG organic (Guinea grass
603 leaf and kenaf stem) increased to 225.3 ± 1.2 and 111.7 ± 1.2 Bq/kg, respectively. In
604 addition, most plant samples contain higher ^{40}K concentrations compared to the
605 concentrations of ^{40}K in soil. These findings are in agreement with previous studies
606 [52, 57-60].

607 This finding is not the expected result, since the activity concentrations of ^{40}K in the
608 soil samples treated with PG organic were much lower than the concentrations of ^{40}K
609 in the plant samples.

610

611 **Table 12 The radioactivity concentrations of ^{226}Ra , ^{228}Ra and ^{40}K in plant**
612 **samples.**

Season	Sample ID		Activity Concentration (Bq/kg)		
			²²⁶ Ra	²²⁸ Ra	⁴⁰ K
2 nd Season	Kenaf-stem-Control	Range	8.3 – 8.5	1.1 - 1.5	116.8 - 146.4
		Average	8.4 ± 0.1	1.3 ± 0.3	131.6 ± 20.9
	Kenaf-stem-PG organic	Range	10.2 - 18.5	0.3 - 0.7	192.9 - 321.8
		Average	10.6 ± 11.3	0.5 ± 0.3	257.3 ± 91.1
	ANOVA Analysis		0.19 ^{ns}	0.10 ^{ns}	0.20 ^{ns}
	Guinea Control	grass-leaf- Range	1.4 - 1.9	0.1 - 0.3	13.9 – 25.9
		Average	1.6 ± 0.4	0.2 ± 0.2	19.9 ± 8.5
	Guinea organic	grass-leaf-PG Range	1.8 - 1.9	0.1 - 0.6	224.4 - 226.2
		Average	1.9 ± 0.0	0.4 ± 0.3	225.3 ± 1.2
	ANOVA Analysis		0.51 ^{ns}	0.57 ^{ns}	0.0009 ^{**}
3 rd Season	Kenaf-stem-Control	Range	7.2 - 12.4	0.6 - 1.2	42.7 - 47.0
		Average	9.8 ± 3.7	0.9 ± 0.4	44.8 ± 3.1
	Kenaf-stem-PG organic	Range	5.0 - 9.5	0.1 - 0.4	110.9 - 112.5
		Average	7.2 ± 3.2	0.2 ± 0.2	111.7 ± 1.2
	ANOVA Analysis		0.54 ^{ns}	0.16 ^{ns}	0.001 ^{**}
	Guinea Control	grass-leaf- Range	4.8 – 8.5	1.2 - 2.7	321.0 - 384.4
		Average	6.7 ± 2.6	1.9 ± 1.1	352 ± 44.9
	Guinea organic	garss-leaf-PG Range	2.9 – 5.1	0.4 - 0.5	204.9 - 227.6
		Average	4.0 ± 1.6	0.4 ± 0.1	216.2 ± 16.0
	ANOVA Analysis		0.34 ^{ns}	0.18 ^{ns}	0.06 ^{ns}
Vegetables (Malaysia) [57]			1.3 – 7.8 [#]	0.4 – 4.1 ^{##}	-
Vegetable leaves (Malaysia) [60]			0.6 – 5.64	-	398.9 – 1072.6
Corn (Malaysia) [61]			0.1 – 19.2	0.1 – 3.2	26.1 – 129.0
Paddy grain (Malaysia) [52]			0.5 – 2.8	0.5 – 1.6	43.5 – 108.5
Vegetables (Iraq) [59]			4.0 – 7.0 [#]	2.2 – 10.6 ^{##}	109.0 – 319.2

613 Note: [#] Assume as ^{226}Ra (^{238}U decay series) in secular equilibrium; ^{##} Assume as ^{228}Ra (^{232}Th
614 decay series) in secular equilibrium. ns: not significant; * and **: significant at $p \leq 0.05$ and p
615 ≤ 0.01 , respectively.

617 This finding suggests that the increase in the ^{40}K concentration in plants (compared to
618 the soil) might be contributed by other or additional sources, such as fertilizers,
619 pesticides or herbicides rather than PG organic. It should be noted that the uptake
620 mechanism will differ for each plant species. Several factors, such as the kinetic
621 parameters of plants to absorb specific radionuclides, metabolic behavior as well as
622 plant tolerance for a specific radionuclide will influence the uptake of radionuclides
623 by plants [62]. In addition, International Atomic Energy Agency, IAEA [63] reported
624 that the level of radioactivity concentrations for ^{226}Ra and ^{238}U in NPK fertilizer can
625 reach up to 450 and 1280 Bq kg⁻¹, respectively. Therefore, further investigation is
626 recommended to verify the sources that contribute to the increase of ^{40}K in plants.

627 *Radiological hazard index and soil-to-plant transfer factor*

628 A radium equivalent (Ra_{eq}) is defined as the radiation hazard index (RHI), which is
629 used to assess the radiation hazard due to the natural radionuclides. The calculated
630 Ra_{eq} for soil was much lower than the 370 Bq kg⁻¹ recommended limit of Ra_{eq} in soil
631 (Table 13). For non-hazardous materials, the calculated Ra_{eq} should not exceed a
632 maximum value of 370 Bq kg⁻¹ for soil [64]. Therefore, the radiological concern due
633 to the ^{226}Ra , ^{228}Ra and ^{40}K in soil can be ignored; hence, PG organic is suitable for
634 agriculture applications. The TFs of ^{226}Ra , ^{228}Ra , ^{238}U , ^{232}Th and ^{40}K obtained in this
635 study were comparable to those observed in several types of plants from other studies
636 (Table 13). The highest radionuclide TF was ^{40}K , which ranged from 0.85 to 6.98.
637 Minor dissimilarities of the TF among the samples were perhaps due to transport
638 processes as well as different absorption capacities of the vegetables itself [65]. It may
639 also depend on the type of soil, soil texture, clay content, pH and organic matter
640 content [66].

641 **Table 13 Radium equivalent and soil-to-plant transfer factor obtained in this study.**
642

Season	Sample	Ra _{eq}	Transfer factor soil-to-plants			Note	643
			²²⁶ Ra	²²⁸ Ra	⁴⁰ K		644
		Bqkg ⁻¹					
2 nd season	Kenaf-Soil-Control	43.2	0.54	0.08	2.88	This research	645
	Kenaf-Soil-PG organic	46.6	0.71	0.03	5.34		
	Guinea grass-Soil-Control	44.7	0.10	0.01	0.30		
	Guinea grass -Soil-PG organic	67.0	0.12	0.01	3.75		
3 rd season	Kenaf-Soil-Control	42.7	0.70	0.05	0.85	This research	646
	Kenaf-Soil-PG organic	50.7	0.42	0.01	2.12		
	Guinea grass -Soil-Control	41.3	0.50	0.12	6.98		
	Guinea grass -Soil-PG organic	45.0	0.28	0.02	3.54		647
Rice (Malaysia)		-	0.04 – 0.20	0.02 – 0.15	0.09 – 4.14	[67]	
Rice (Malaysia)		9.9 – 27.7	0.01 – 0.40	0.01 – 0.60	0.10 – 1.30	[55]	
Vegetables		-	0.08 – 0.93	0.15 – 0.56	1.27 – 3.7	[68]	648
Vegetables (Malaysia)		-	0.03 – 0.09	0.001 – 0.01	1.59 – 5.20	[65]	
Peninsular of Malaysia		63.2 – 414.4	-	-	-	[41]	649

650 **Conclusion**

651 The heavy metal contents in borehole were significantly lower than the heavy metal
652 contents in surface water. Heavy metal contents in the borehole after PG organic
653 application were below the target value standard in the Malaysian Standard for Raw
654 Drinking Water Quality [33]. Heavy metal contents in soil in the plot treated with PG
655 organic were significantly lower or not significantly different compared to the heavy
656 metal contents in the control. All metal concentrations were below the target values
657 stipulated in the new Dutch list [32]. Heavy metal concentrations in the corn grain
658 were lower than the standards in the Malaysian Food Act [34] and Food Regulation
659 [35]. The analysis of the naturally occurring radionuclide concentrations in soil
660 samples indicated that the radioactivity concentrations of ^{226}Ra , ^{228}Ra , and ^{40}K in all
661 samples were much lower than their average concentrations in Malaysia's soil, as
662 reported by United Nations [48]. The determined radioactivity concentrations of
663 ^{226}Ra , ^{228}Ra and ^{40}K were also comparable with those of another study conducted in
664 Malaysia. The analysis of the natural radioactivity concentration in several plants also
665 showed that the results obtained in this study were comparable with those other
666 studies. The analysis of the groundwater indicated that no leaching of the
667 radionuclides from soil to groundwater occurred. In addition, the analysis of the
668 surface water (nearest river) suggested that no accumulation (discharge) of natural
669 radionuclides due to the application of PG organic occurred in the surface water. The
670 calculated Ra_{eq} for soil was less than the 370 Bq kg^{-1} recommended limit of Ra_{eq} in
671 soil. Based on these findings, it can be concluded that the application of PG organic
672 had no impact on the environment (in soil and plants) at the studied location. The

673 biological accumulation coefficient (BAC), contamination factor (CF), I-geo index
674 (Igeo) and PLI showed that there was no accumulation of metals in grain, and no
675 contamination and no pollution occurred in borehole water and the BRIS soil that
676 received applications of PG organic. These data indicated that the use of PG organic
677 as a soil conditioner in BRIS soil is safe for the soil-plant-water.

678

679 **Declarations**

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684

685 **Ethics approval and consent to participate**

686 Not applicable.

687 **Consent for publication**

688 Not applicable.

689 **Availability of data and material**

690 All data generated or analyzed during this study are included in this published article.

691 **Competing interests**

692 The authors declare that they have no competing interests.

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695 **Author's contributions**

696 MMH: Conceptualization, investigation, experimental analysis, data evaluation and
697 carrying out the experiment. PA: Writing manuscript and editing. ARS, IWMR and
698 IAF: carrying out the experiment. All authors read and approved the final manuscript.

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Figures

Figure 1

The location of the experimental site and the Lynas Advance Material Plant (LAMP).

Figure 2

The sub-plots receiving the PG organic conditioner treatment for 3 cropping seasons for kenaf, maize, and guinea grass. Blue represents the sub-plot receiving the soil conditioner for each cropping season, and yellow represents the sub-plot receiving residual treatment.

Figure 3

The XRD diffractogram of PG organic conditioner using PXRD method.