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Radiological risk assessment of use of phosphate fertilizers in soil

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Background: The radiological impact of the use of phosphate fertilizers in soil is due to the internal irradiation of the lung by the alpha particles, short lived radon-thoron progeny and the external irradiation of the body by gamma rays emitted from radionuclides *in situ*. This paper describes the results of gamma spectrometric measurements of the concentration of the natural radionuclides namely ^{226}Ra, ^{232}Th and ^{40}K in the soil samples collected from the fields where a variety of phosphate fertilizers are being used by the farmers to enhance the crop yield. ***Materials and Methods:*** The experimental work utilizes actual measurements of ^{226}Ra, ^{232}Th and ^{40}K using gamma spectrometry and radon concentration and exhalation rates measurements using solid state nuclear track (LR-115, Type-II plastic) detectors to asses a first order exposure risk for the persons working in the fields where lot of fertilizers are being used to enhance crop yield in terms of occupational exposure. ***Results:*** The concentration of Radium, Thorium and Potassium in the mixed soil sample from crop fields is 16.2 ± 0.22, 68.1±1.44 and 875.0±9.68 Bq/kg, where as in barren soil sample is 9.1±0.13, 59.4±1.45 and 668.4±8.01 Bq/kg respectively. The radium equivalent activity (Ra_{eq}) in the mixed soil sample from crop fields is 225.9 Bq/kg, where as in barren soil sample is 193.1 Bq/kg. The values of absorbed dose and annual effective dose (indoors and outdoors) are found to vary from 90.87 nGyh^{-1} to 119.71 nGyh^{-1}, 0.45 mSv/y to 0.59 mSv/y and 0.11 mSv/y to 0.15 mSv/y respectively in soil sample from crop fields, whereas the value of absorbed dose and annual effective dose (indoors and outdoors) is 92.29 nGyh^{-1}, 0.45 mSv/y, 0.11 respectively in soil sample collected from barren land. The radon concentration and exhalation rates have also been reported. ***Conclusion:*** The activity concentration, exhalation rate and absorbed dose were found to increase substantially with the use of phosphate fertilizers and it varies from sample to sample. The radium equivalent activities in all the soil samples were found to be lower than the limit (370 Bq/kg) set in the Organization for Economic Cooperation and Development (OECD) report and the dose equivalent is within the safe limit of 1 mSv/y.
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Keywords: *Gamma spectrometry, soil samples, dose, radon, health risk assessment, exhalation rate, exposure.*

INTRODUCTION

We live in a milieu of radiation and exposed to ionizing radiation from natural sources. Natural radioactivity is wide spread in the earth's environment and it exists in various geological formations in soils, rocks, plants, water and air [1-3]. Naturally occurring radioactive materials (NORM) became the focus of regulatory interest with the publication of International Atomic Energy Agency SS115 and the subsequent publishing of European Council Directive 96/29/EURATOM. It has changed the profile of radiation protection completely, increasing the regulatory awareness of natural radiation and the industries involved. Three major industries have been identified in terms of their scope and the materials handled as industries requiring further attention. They are the Oil & Gas Industry, the Zircon Industry and the Phosphate & Fertilizer Industry. The Phosphate and associated Fertilizer industry has an added complication, because it has two distinct sources of raw material, e.g. being of igneous or sedimentary origin. The source material contains isotopes from the natural uranium and thorium decay chains that may or may not follow the gypsum in the process. The weathering of the parent igneous rock released the radionuclides by the process of leaching and finally they entered the appetite structure by adsorption and co-precipitation

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with calcium [4, 5]. In NPK fertilizers, potassium component augments the natural radioactivity because of the presence of radioactive ^{40}K, whose natural abundance in potassium is 0.00118%. In India, at present, about 10 million tons of rock phosphate is used for manufacturing different types of phosphate fertilizers. 15% of the requirement of rock phosphate is met by indigenous sources from Udaipur (Rajasthan), Jhabua and Hirapur (Madhya Pradesh) and rest 85% is imported from Jordan, Morocco, U.S.A., Senegal, Tunisia and China [6]. It is reported that approximately 80% of the Radium-226 follow the gypsum, whereas 86% of the uranium and 70% of the thorium are found in the phosphoric acid. Phosphogypsum is generated during the production process of phosphoric acid from phosphate rock. Simplified reaction equations are as follow:

$$Ca_3(PO_4)_2 + 3H_2SO_4 \rightarrow 2H_3PO_4 + 3CaSO_4$$
$$Ca_3(PO_4)_2 + 4H_3PO_4 \rightarrow 3Ca(H_2PO_4)_2$$
$$3Ca(H_2PO_4)_2 + 3H_2SO_4 \rightarrow 3CaSO_4 + 6H_3PO_4$$

Natural radioactivity in soils comes from ^{238}U and ^{232}Th series and ^{40}K, and lots of work has been done worldwide on the natural radionuclides contents in soil [7-9]. It is common practice to use phosphate fertilizers in soil to enhance the crop yield. As the phosphate fertilizers contain 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 plasterboards for houses etc. (Ceilings are typically made from plasterboard) has radiological impact on human health and the radiation exposure to the persons working in the fields is modified. It has been reported that there is a first order exposure risk for members of the public, when residing in a dwelling with phosphogypsum plasterboard as part of the structure [10]. The radiological impact of the use of phosphate fertilizers in soil is due to the internal irradiation of the lung by the alpha particles, short lived radon-thoron progeny and the external irradiation of the body by gamma rays emitted from radionuclides *in situ*.

In the light of the above mentioned facts, it is, therefore, fundamental to assess the radiological risk of use of phosphate fertilizers in soil in the crop fields from health and hygiene point of view. This paper emphasis on the public risk assessment in an integrated manner assessing doses to members of the public via the atmospheric aquatic and secondary pathways. The experimental work utilizes actual measurements of ^{226}Ra, ^{232}Th and ^{40}K using gamma spectrometry and radon concentration and exhalation rates measurements using solid state nuclear track detectors to asses a first order exposure risk for the persons working in the fields where lot of fertilizers are being used to enhance crop yield in terms of occupational exposure.

MATERIALS AND METHODS

Soil samples were collected from the fields in which different crops were sown and variety of fertilizers like, Urea, Di Ammonium Phosphate (DAP), Nitrogen Phosphate (NP) and Nitro Phosphate Potash (NPK) (all Indian products), as per requirement of the crop were used by the farmers to enhance the crop yield. Samples were also collected from the barren land where no crop was sown since long. The soil samples were collected from District Mathura (U.P.), India. After collection, samples were crushed into fine powder by using Mortar and Pestle. Fine quality of the sample was obtained using scientific sieve of 150 micron-mesh size. Before measurement samples were dried in an oven at about 110 °C for 24 hours. Each sample was packed and sealed in an airtight PVC container and kept for about four week period to allow radioactive equilibrium among the radon (^{222}Rn), thoron (^{220}Rn), and their short lived progenies. On an average 300 gram of soil was taken for each sample. For calibration of the low background counting system, a secondary standard was obtained, calibrated with the primary standard obtained from the International Atomic Energy Agency. The

concentration of ^{226}Ra was determined using a photon peak of 609 keV (46.1%) from ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of interfering peak of ^{235}U, with energy of 185.7 keV. ^{232}Th concentration was determined using the gamma transitions of 583 keV (86%) from ^{208}Tl [11] ^{40}K concentration was determined using the gamma transition of 1461 keV (10.7%).

MEASUREMENT TECHNIQUES

a) Measuring activity concentration of uranium, thorium, potassium

Using HPGe detector of high-resolution gamma spectrometry system, the activity of samples is counted. The detector is a co-axial n-type high purity germanium detector (Make EG&G, ORTEC, Oak Ridge, USA). The detector has a resolution of 2.0 keV at 1332 keV and relative efficiency of 20%. The output of the detector is analyzed using a 4K ADC system connected to PC, the spectrum is analyzed using the locally developed software "CANDLE (Collection and Analysis of Nuclear Data using Linux nEtwork)". The detector is shielded using 4" lead on all sides to reduce the background level of the system [12]. The efficiency calibration for the system is carried out using secondary standard source of uranium ore in geometry available for the sample counting. Efficiency values are plotted against energy for particular geometry and are fitted by least squares method to an empirical relation that takes care of the nature of efficiency curve for the HPGe detector. The samples were counted for a period of 72000 seconds and the spectra are analyzed of the photo peak of uranium, thorium daughter products and K-40. The net count rate under the most prominent photo peaks of radium and thorium daughter peaks are calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclides is calculated from the background subtracted area prominent gamma ray energies.

b) Measuring radon concentration and radon exhalation rates from soil samples

For radon concentration and exhalation rate measurements from soil samples "Can technique" was used [13, 14] as shown in figure 1. A known amount of given sample was kept in plastic Cans. LR-115 plastic track detector was fixed on the bottom of the lid of each Can with tape such that sensitive side of the detector always faced the specimen. The Can is tightly closed from the top and sealed (figure 1).

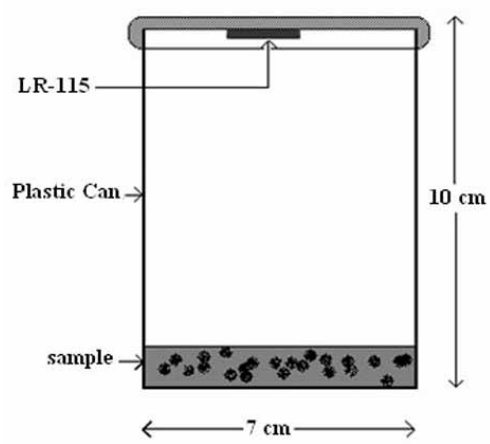

Figure 1. The Can used for the measurement of radon exhalation rate measurements

Following the exposure for a stipulated period (about 100 days), the LR-115 films (SSNTDs) are chemically etched in 2.5N, NaOH (sodium hydroxide) solution in an etching bath with a magnetic stirrer at a temperature of 60 °C for about one and half hour for developing the tracks recorded and registered in the films (figure 2). The etching process removes a bulk thickness of 4 µm leaving a residual detector thickness of 8 µm

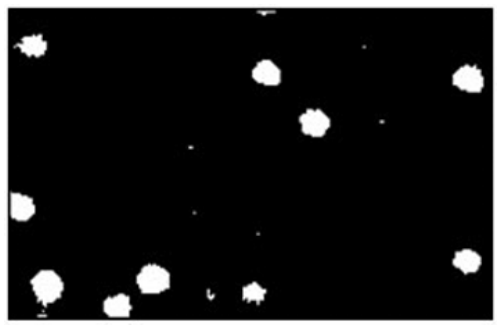

Figure 2. The tracks of alpha particles formed on LR-115 plastic track detector

and records alpha particles with 100% efficiency. The tracks produced by the alpha particles, were observed and counted under an optical Olympus microscope at 600×. Large numbers of graticular fields of the detectors were scanned to reduce statistical errors.

RESULTS AND DISCUSSION

a) Estimation of radium, thorium and potassium in building materials.

The concentrations of Uranium, Thorium and Potassium were calculated using the following equation:

$$\text{Activity(Bq)} = \frac{\text{CPS}\times 100\times 100}{\text{B.I}\times\text{Eff}} \pm \frac{\text{CPS}_{\text{error}}\times 100\times 100}{\text{B.I.}\times\text{Eff}} \quad (1)$$

Where, CPS - Net count rate per second,
B.I.- Branching Intensity,
Eff- Efficiency of the detector.

The gamma ray spectrum for one of the soil samples is shown in figure 3. Table 1 shows the concentration of the Radium, Thorium and Potassium as well as the ± value shows the corresponding standard deviation of σ, which is due to counting errors. The concentration of Radium, Thorium and Potassium in the mixed soil sample from crop fields is 16.2±0.22, 68.1±1.44 and 875.0±9.68 Bq/kg, where as in barren soil sample is 9.1±0.13, 59.4±1.45 and 668.4±8.01 Bq/kg respectively. The concentration of Radium, Thorium and Potassium varies considerably in different soil samples depending on the quantity and type of fertilizer used. From the data, it reveals that the activity concentration in the soil sample in which no fertilizer was used (barren soil), is significantly low than the soil samples collected from crop fields where fertilizers are used. Thus the use of phosphate fertilizers in the fields to enhance the crop yield enhances the activity concentration and hence the exposure of the farmers working in the fields.

b) Evaluation of radium equivalent activity

The distribution of natural radioactivity in the soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg to compare the specific activity of materials containing different amount of ^{226}Ra, ^{232}Th, ^{40}K. About 98.5% of the radiological implications are due to ^{226}Ra and its daughter products in ^{238}U series. The natural abundance of ^{235}U is only 0.72% of the total uranium content and hence was not considered in the present study, except its ^{226}Ra contribution at 186 keV. The gamma transitions of energy 609 keV or 1760 keV (due to ^{214}Bi) was used to determine the concentration of ^{226}Ra. The criterion for this

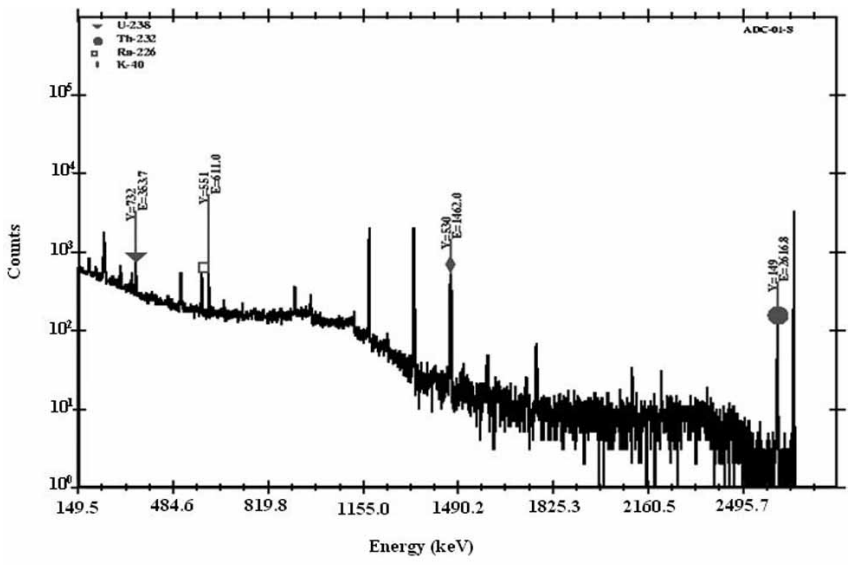

Figure 3. The gamma ray spectrum for soil sample.

Table 1. Activity concentration of uranium, thorium, potassium, radium equivalent.

Sr. No.	Sample	Activity Concentration (Bq/kg)			Radium Equivalent Activity Ra_{eq} (Bq/kg)
		Ra-226	Th-232	K-40	
1	Soil-1(Barren Land)	9.1 ± 0.13	59.4 ± 1.45	668.4 ± 8.01	193.1
Fertile Land (Soil with Fertilizers)					
2	Soil-2 (Potato)	13.8 ± 0.18	78.1 ± 1.77	769.2 ± 8.68	226.3
3	Soil-3 (Arhar)	14.8 ± 0.21	65.0 ± 1.42	698.5 ± 8.01	197.3
4	Soil-4 (Bajra)	14.0 ± 0.19	59.7 ± 1.32	685.1 ± 8.01	184.6
5	Soil-5 (Barsim)	12.8 ± 0.17	71.4 ± 1.49	790.9 ± 9.01	217.1
6	Soil-6 (Paddy)	10.8 ± 0.15	83.4 ± 1.83	665.4 ± 7.68	206.8
7	Soil-7 (Wheat)	15.6 ± 0.21	88.2 ± 1.91	820.6 ± 8.34	250.3
8	Soil-8 (Barley	12.5 ± 0.17	62.3 ± 1.35	762.2 ± 8.67	191.4
9	Soil-9 (Mustard)	16.4 ± 0.22	78.4 ± 1.57	698.5 ± 8.01	221.9
10	Soil-10 (Mixed)	16.2 ± 0.22	68.1 ± 1.44	875.0 ± 9.68	225.9

MDA (Minimum Activity Detection Limit) = 2Bq/kg, 2Bq/kg and 4Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K respectively.

model considers the external hazard due to gamma rays corresponds to a maximum radium equivalent activity of 370 Bq/kg for the building material. This Raeq is calculated using the assumption that 370 Bq/kg ^{226}Ra or 260 Bq/kg ^{232}Th or 4810 Bq/kg ^{40}K produces the same gamma dose rate [15].

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k \quad (2)$$

Where A_{Ra}, A_{Th}, and A_K are the concentrations of the three radionuclides ^{226}Ra, ^{232}Th and ^{40}K respectively, which is expressed in Bq/kg.

Using above equation [2], the radium equivalent activity found in the soil samples is given in table1. The radium equivalent activity (Ra_{eq}) in the mixed soil sample from crop fields is 225.9 Bq/kg, where as in barren soil sample is 193.1 Bq/kg. The radium equivalent activity varies considerably in different soil samples depending on the quantity and type of fertilizer used. From the data, it reveals that the radium equivalent activity in the soil sample in which no fertilizer was used (barren soil), is marginally below than the soil samples collected from crop fields where fertilizers are used. Thus the use of phosphate fertilizers in the fields to enhance the crop yield enhances the radium equivalent activity and hence the exposure of the farmers working in the fields. It is inferred that for all the soil samples analyzed, the radium equivalent activity value is well within the permissible limits of 370 Bq/kg [16].

c) Estimation of absorbed and effective dose

The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are converted into doses ($nGyh^{-1}$ per $Bqkg^{-1}$) by applying the factors 0.427, 0.662 and 0.043 for radium, thorium and potassium, respectively [17]. These factors are used to calculate the total absorbed gamma dose rate in air at one meter above the ground level using the following equation:

$$D\ (nGyh^{-1}) = 0.427C_{Ra} + 0.662C_{Th} + 0.043C_K \quad (3)$$

Where, C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq/kg) of radium, thorium and potassium respectively in the samples. To estimate annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. Annual

estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 $SvGy^{-1}$, which is used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors [18]. The annual effective doses are determined as follows.

Indoor (nSv) = (Absorbed Dose) $nGyh^{-1} \times 8760h \times 0.8 \times 0.7$ Sv Gy^{-1} (4)

Outdoor (nSv) = (Absorbed Dose) $nGyh^{-1} \times 8760h \times 0.2 \times 0.7$ Sv Gy^{-1} (5)

Table 2 shows the calculated absorbed and annual effective dose rates from soil samples using equations 3, 4, and 5 respectively. The values of absorbed dose and annual effective dose (indoors and outdoors) are found to vary from 90.87 $nGyh^{-1}$ to 119.71 $nGyh^{-1}$, 0.45 mSv/y to 0.59 mSv/y and 0.11 mSv/y to 0.15 mSv/y respectively in soil sample from crop fields, whereas the value of absorbed dose and annual effective dose (indoors and outdoors) is 92.29 $nGyh^{-1}$, 0.45 mSv/y, 0.11 respectively in soil sample collected from barren land. The absorbed dose and annual effective dose for the soil samples in which fertilizers were used are higher than the barren soil sample. The annual effective dose is marginally below the International Commission on Radiological Protection (ICRP) recommended the annual effective dose of 1 $mSvY^{-1}$ for the general public [19].

Table 2. Radiation absorbed dose and annual effective dose from soil samples.

Sr. No.	Sample	Absorbed Dose Rate ($nGyh^{-1}$)	Annual Effective Dose (mSv/y)	
			Indoor	Outdoor
1	Soil-1(Barren Land)	92.29	0.45	0.11
Fertile Land (Soil with Fertilizers)				
2	Soil-2 (Potato)	108.42	0.53	0.13
3	Soil-3 (Arhar)	96.73	0.47	0.12
4	Soil-4 (Bajra)	90.87	0.45	0.11
5	Soil-5 (Barsim)	106.74	0.52	0.13
6	Soil-6 (Paddy)	101.33	0.50	0.12
7	Soil-7 (Wheat)	119.71	0.59	0.15
8	Soil-8 (Barley)	94.92	0.47	0.12
9	Soil-9 (Mustard)	107.96	0.53	0.13
10	Soil-10 (Mixed)	111.43	0.55	0.14

d) Radon concentration and radon exhalation rate measurements

Calibration factor used was, 0.056 tracks cm^{-2} day^{-1}=1 Bq m^{-3} [20].

Exhalation rates (E_x) were calculated using the equations (6) and (7) used by various researchers [21, 22].

For mass exhalation rate,

and for surface exhalation rate

$$E_X = \frac{CV\lambda / M}{T + 1/\lambda(e^{-\lambda T} - 1)} \quad (Bqkg^{-1}h^{-1}) \qquad (6)$$

$$E_X = \frac{CV\lambda / A}{T + 1/\lambda(e^{-\lambda T} - 1)} \quad (Bqm^{-2}h^{-1}) \qquad (7)$$

Where, C = Integrated radon exposure (Bq $m^{-3}h^{1}$)

V = Volume of air in can (m^3)

T = Time of exposure (hrs)

λ = Decay constant for radon (h^{-1})

A = Area covered by the can or surface area of the sample (m^2)

The calculated values of radon concentration and exhalation rate for soil samples collected from fertile land and barren land in Mathura District (U.P.) are presented in table 3. The radon concentration varies from 214.22 Bq/m^3 to 471.57 Bq/m^3 and radon exhalation rate varies from 6.95 $mBqkg^{-1}h^{-1}$ to 13.33 $mBqkg^{-1}h^{-1}$ for mass exhalation rate and from 133.59 $mBqm^{-2}h^{-1}$ to 294.08 $mBqm^{-2}h^{-1}$ for surface exhalation rate in soil sample from crop fields, whereas the value of radon concentration, mass exhalation rate and surface exhalation rate is 107.92 Bq/m^3, 3.05 $mBqkg^{-1}h^{-1}$ and 67.3 $mBqm^{-2}h^{-1}$ respectively for soil sample collected from barren land. The calculated values of radon concentration and exhalation rate for the soil samples in which fertilizers were used are higher than the barren soil sample.

Table 3. Radon concentration and radon exhalation rates from soil samples.

Sr. No.	Sample	Radon Concentration (Bq/m^3)	Radon Exhalation Rates	
			Surface Exhalation Rate ($mBqm^{-2}h^{-1}$)	Mass Exhalation Rate ($mBqkg^{-1}h^{-1}$)
1	Soil-1(Barren Land)	107.92	67.3	3.05
Fertile Land (Soil with Fertilizers)				
2	Soil-2 (Potato)	342.45	213.56	9.68
3	Soil-3 (Arhar)	357.47	222.92	10.10
4	Soil-4 (Bajra)	471.57	294.08	13.33
5	Soil-5 (Barsim)	232.61	145.06	6.57
6	Soil-6 (Paddy)	419.16	261.39	11.85
7	Soil-7 (Wheat)	470.18	293.21	13.29
8	Soil-8 (Barley)	303.24	189.10	8.57
9	Soil-9 (Mustard)	214.22	133.59	6.05
10	Soil-10 (Mixed)	296.08	184.64	8.37

CONCLUSION

Thus we find that the application of fertilizers in the soil to enhance the crop yield enhances the activity concentration, radon and radium concentration and radon exhalation rates, however, well below the acceptable limits and all the soil samples found to satisfy the safety criteria. The results clearly indicate that the farmers are exposed to higher levels of radiation when they apply phosphate fertilizers to the fields and afterwards from the fertilizer mixed soil. Efforts should be made at national and international level to reduce Ra-226 activity in the fertilizers, like extracting uranium from phosphoric acid by solvent extraction method, so that the fertilizers are more eco-friendly.

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