

8.12 NATURAL RADIONUCLIDES AND TRACE ELEMENTS IN RICE PADDY SOILS IN RELATION TO FERTILIZER APPLICATION: STUDY OF A CHRONIC KIDNEY DISEASE AREA IN SRI LANKA

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ORIGINAL ARTICLE

Natural radionuclides and trace elements in rice paddy soils in relation to fertilizer application: study of a chronic kidney disease area in Sri Lanka

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Abstract The rising number of chronic kidney disease patients with no identifiable cause (CKD of uncertain aetiology), prevalent in some areas of the dry zone of Sri Lanka is suspected to be related to the environmental exposure to heavy metals. Agricultural soils are well recognized as being contaminated with potentially toxic metals from various forms of fertilizers and agro-chemicals, which could easily enter the human body through the food chain. The objective of this paper is to determine the content of heavy metals and activity concentration of background radionuclides such as K-40, Ra-226 and Th-232, in rice paddy soils. Rice farming is the most common agricultural practice in the affected region and possible heavy metal sources such as fertilizers are applied in abundance in the rice paddies. Soils collected from a rice paddy in a non-CKD region was used for the comparison. In dry zone soils, Ca, K, Ba, Pb and Zr contents were higher and Fe, Mn, Cr, Ni and Zn contents were lower compared to that of soils from the wet zone non-CKD region. However, the activity concentration of soils was mostly the same in all samples, except for the K-40 contents of the soils, which were higher in the rice paddy soils compared to the undisturbed forest soils and also to the world averages.

The mean U content was 3.6 mg/kg in the studied soils, although extremely high uranium contents were found in some fertilizer samples particularly in the triple superphosphates. Most uranium applied via fertilizer could contaminate the drinking water sources and even low uranium concentrations in drinking water may cause nephrotoxic effects.

Keywords Phosphate fertilizer·Radionuclides·Chronic kidney diseases·Dry zone·Uranium toxicity·Trace elements in paddy soils

Introduction

Concern over the entry of potentially toxic heavy metals into the human food chain via all aspects of agricultural production has been given increased attention in recent years. This is due to their possible health impacts as some trace elements present in agricultural soils may be transferred to the human body easily through food. Agricultural soils naturally contain trace elements that are essential for plant growth, but these soils are also particularly susceptible to degradation and pollution. Trace elements may be introduced into agricultural soils by cultivation practices such as fertilization, irrigation, pesticide application and organic waste disposal (Mortvedt 1996; Chen et al. 2007). Fertilizers, indispensable as they may seem, are nevertheless materials that clearly cause serious environmental contamination notably in the agricultural soils. In order to increase the agricultural productivity needed by the ever-increasing population, the application rates of chemical fertilizers have also been increased and this may lead to excessive accumulation of potentially toxic elements that are commonly present in soils as traces. High levels of such

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trace elements in agricultural soils become a problem because they can enter the food chain or leach into groundwater causing some serious health problems.

Sri Lanka, as a tropical island, presents a unique case for the study of the influence of natural environmental factors (i.e. soil, water, plants) on human and animal health. It has a wide range of environmental zones ranging from arid to wet and is rich in natural resources and groundwater. It is also noteworthy that nearly 75% of the entire population live in close association with the physical environment, obtaining their food and water directly from the immediate vicinity. The strong linkage of the incidence of some geographically distributed diseases and natural environmental factors is clearly seen in Sri Lanka (Dissanayake and Chandrajith 1999).

As in other countries of Asia, rice is the staple food of Sri Lanka and is deeply enmeshed into its traditions and culture. It was estimated that 8,350 km² of paddy lands are scattered throughout the island, which supply nearly 86% of the national annual demand of rice (Ganashan et al. 1996). Most importantly, more than 70% of the annual rice production in Sri Lanka originates from the dry zone and these lands have been used for rice cultivation for 1,000s of years.

Pollution of paddy soils by potentially toxic metals mainly due to fertilizer applications has often been reported, especially in those areas experiencing rapid economic development (e.g. Cao and Hu 2000; Wang et al. 2003). In Sri Lanka, rice is cultivated in two, or in some areas three, seasons per year and fertilizers are applied three times per season in general. The recommended amount for application is 270 kg of fertilizers such as urea, triple superphosphates (TSP) and potash per hectare per season. However, application of fertilizers has increased by more than a factor of two during the past few decades, exceeding the recommended levels due to the government granting of subsidies for fertilizer. The paddy soils in Sri Lanka are therefore highly modified by artificial fertilizer applications. During the period of waterlogging in rice cultivation, the chemical nature of the soil can be drastically changed and pollutants can be leached easily into the subsurface, which eventually contaminate the groundwater that rural people use as a drinking water source (Chandrajith et al. 2005a). Phosphate fertilizers, which are the most commonly used materials in rice fields in Sri Lanka, contribute to outdoor terrestrial natural radionuclides belonging to the ²³²Th and ²³⁸U series as well as radioisotope of potassium (⁴⁰K) (Ghosh et al. 2008).

Based on the average annual rainfall, Sri Lanka has been divided into two broad climatic zones: the wet zone in the southwest quadrant and the dry zone in the north and east (Fig. 1). In between the wet and the dry zones, an intermediate climatic zone has been recognized. The wet

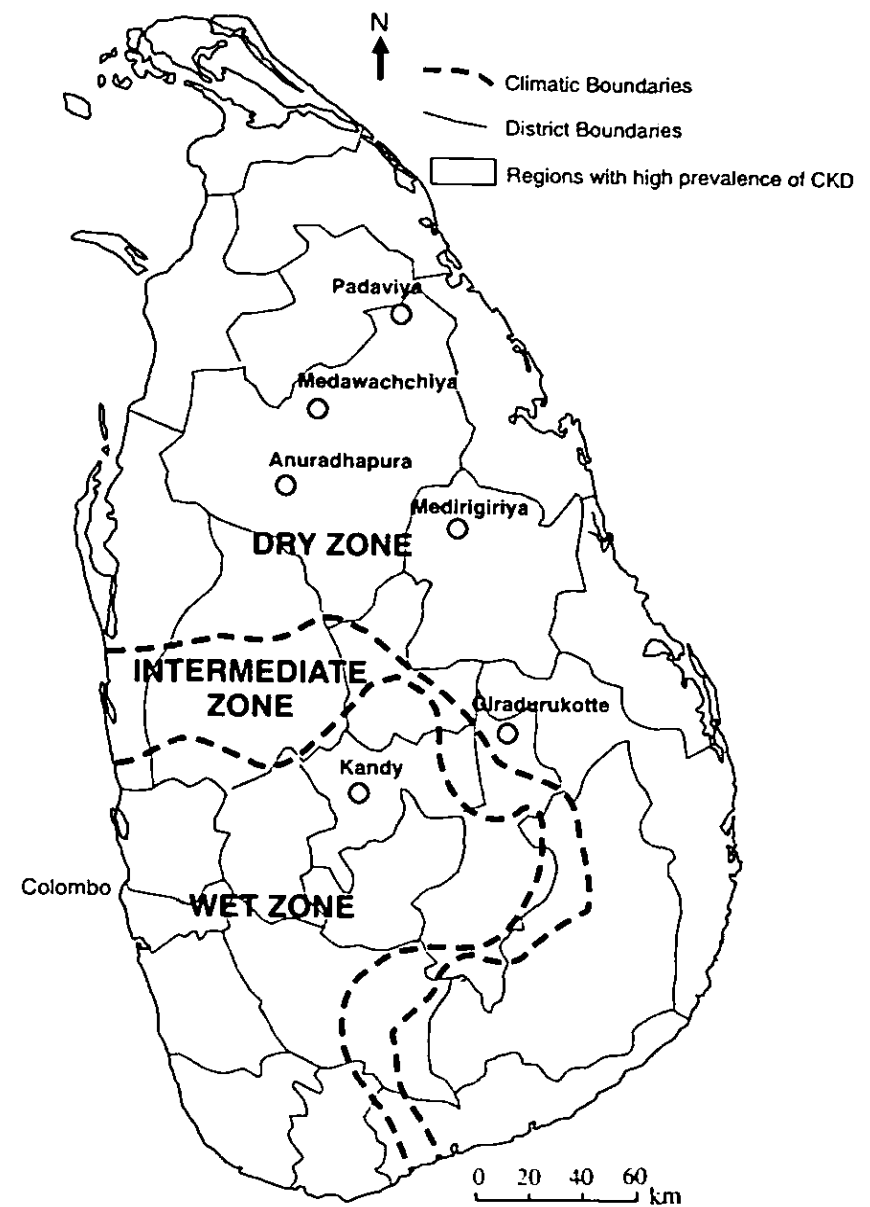


Fig. 1 Map of Sri Lanka showing the sampling locations and climatic boundaries

zone receives a mean annual rainfall of 2,500 mm, mostly during the two monsoon seasons. Geologically, 90% of the island is underlain by high grade metamorphic rocks of Precambrian age. The distribution of soil types in Sri Lanka, therefore, closely follows the climatic boundaries. The more important soil groups found in Sri Lanka are reddish brown earth, low humid gley soils, solodized solonetz, non-calcic brown soils, immature brown loams, soils on old red yellow latosols, grumusols, regosols, red-yellow podzolic soils, reddish brown latosolic soils, bog and half-bog soils (Panabokke 1996). Reddish brown earth is the most widespread soil group in Sri Lanka and occupies the largest area compared to all other soils. Because of the gently undulating landscape of the dry zone, the soil on the higher parts of the land is reddish brown, while the soil on the adjacent lower part of the land is greyish brown in colour (Panabokke 1996).

This study aims at investigating in detail the trace element geochemistry and natural radioactivity of paddy soils collected from the dry zone districts of Sri Lanka where people suffer from some form of endemic environmental

disease. Among such diseases, dental and skeletal fluorosis (Dissanayake 1996) and CKD of uncertain aetiology (Aturaliya et al. 2006, Dissanayake and Chandrajith 2006) are commonly observed in certain parts of the dry zone districts, particularly in the north central and eastern sector of the island. Over 6,000 patients with CKD with uncertain aetiology were reported since 2003 from the North Central Province (NCP) (Fig. 1) of Sri Lanka (Dr. Thilak Abeysekera, Teaching Hospital, Kandy, personal communication). The histological appearance of the kidney disease in the dry zone of Sri Lanka is tubulointerstitial, which is commonly observed in toxic nephropathies. The disease process appears to mainly affect the proximal tubules and the interstitium giving rise to characteristic, recognizable pathohistological and clinical features. Therefore, it is important to study the trace element composition and natural radioactivity in paddy soils in affected areas with respect to their possible sources such as fertilizer applications. The results that are obtained will form the baseline data for future changes in environmental monitoring, as such studies are lacking in Sri Lanka.

Materials and methods

In order to obtain a generalized idea of the geographic distribution of the heavy metals and the activity of radionuclides in the dry zone rice soils of Sri Lanka, samples were collected from regions of Anuradhapura, Medawachchiya, Medirigiriya, Giradurukotte and Padaviya. These areas are considered as regions with a high incidence of CKD of uncertain aetiology. Another set of samples were collected from the upland wet zone region of Kandy (Fig. 1). Composite samples from five locations in each region were also collected. A total of 30 samples were collected for radioactivity measurements from three locations and another two samples were collected from undisturbed forest areas as reference sites. The objective of collecting samples from reference sites is to compare the radioactivity in each site. All samples were air dried, disintegrated and passed through a 2 mm polyethylene sieve.

X-ray fluorescence (XRF) analysis of fused glass discs is one of the most commonly used methods for the determination of the content of major and trace elements in many geological materials. Subsamples of each soil sample were finely powdered using an agate ball mill and the glass discs were prepared fusing the sample aliquot with spectroscopic grade lithium tetraborate and lithium metaborate flux. The major and trace element analyses were determined by X-ray fluorescence at the Institute of Geology and Mineralogy, University of Erlangen-Nürnberg, Germany. The instrument was calibrated using appropriate international reference samples that are similar to the samples. A fused

glass disc was prepared using the reference material JSd-2 (Japan Geological Survey Sediment Standard) with samples and analysed as an unknown for quality controlling purpose. The percentage recoveries for the JSd-2 were between ± 0.05 and $\pm 1.5\%$ for major elements and ± 0.5 and $\pm 6.0\%$ for trace elements except for Cr and La that were given an RSD of 11 and 13, respectively.

The gamma spectrometry was utilized to measure the K-40, Ra-226 and Th-232 activity concentration in some selected samples from Anuradhapura ($n=10$), Giradurukotte ($n=10$) and Kandy ($n=10$). Two reference samples were collected from an undisturbed natural dry zone forest and from a wet zone upper montane forest area (Fig. 1). The gamma ray spectra of the prepared samples were measured using a high-resolution gamma spectrometer based on a shield p-extended type HPGe detector (model: Gx3020-Canberra) with a relative photopeak efficiency of 30% and energy resolution of 2 keV FWHM for 1,332 keV gamma ray line of Co-60. Genie 2000 Data Acquisition Canberra software was used to analyse the photopeaks. The HPGe Detector was calibrated for energy and efficiency over the proton energy range 20–3,000 keV using mathematical efficiency calibration option of LABSOCS software (model 5574C Canberra) for the measurement geometry and sample matrix. The counting time for gamma ray spectra measurement was 20,000 s for cylindrical geometry. The activity of ^{40}K was measured directly via its 1,461 (10.7%) keV peak of the gamma ray spectra. To determine the activity concentration of ^{226}Ra , the gamma ray lines 295.1 (19.2%) and 351.9 (37.1%) keV from ^{214}Pb , and 609.3 (46.1%) and 1764.5 (15.9%) keV gamma rays from ^{214}Bi were used. Activity concentration of ^{232}Th was determined using the gamma ray lines 238.6 (43.6%) keV from ^{212}Pb , 338.4 (12%), 911.1 (29%) and 583.1 (86%). Quality Control of analytical measurement was carried out using IAEA reference materials (IAEA RM soil 6, RGU-1 (U ore), RGTH-1 (Th ore) and RGK-1 (K_2SO_4)). The laboratory was accredited by the Sri Lanka Accreditation Board (SLAB) for radioactivity measurement under the requirement of ISO/IEC 17025.

Fifteen samples of fertilizers, urea, granulate triple superphosphate (TSP) and nitrogen phosphorus potassium (NPK) were collected from local markets in the sampling regions and analysed for their toxic trace element contents and also for their activity concentrations that enable the estimation of the inputs from fertilizers to the paddy soils. At 80°C, 5 g of fertilizer samples were digested with 25 ml of concentrated HNO_3 in a Teflon crucible, then filtered and transferred into 50 ml volumetric flask with 5% HNO_3 . The trace element contents were determined by inductively coupled plasma emission spectrometry. The natural radioactivity of some selected fertilizer samples was measured using the gamma spectrometry as described above.

Results and discussion

In this study, major elements Fe, Mn, Mg, K and Ca (given as oxide %), trace elements Ba, Co, Cr, Cu, Ni, Pb, Zn and rare elements Ce, La, Nb and Zr were measured in paddy soils from the dry zone regions where CKD is widespread. The concentration ranges and mean values for the studied elements are shown in Table 1, in which soils were categorized based on the collected locations. The measurements for the activity concentration (Bq/kg) in paddy soils are given in Table 2.

Figure 2 shows the regional distribution of the studied major and trace elements as box and whisker plots, which indicates that there are often significant differences in the elemental concentrations of different areas. The mean contents of Fe, Mg, Cr, La, Zn and Ni were higher, whereas Mn, Ca, K, Ba, Pb and Zr were lower in paddy soils collected from Kandy regions (low CKD) compared to that of the other regions. The mean contents of Ce, Co, Cu and Nb were similar in all studied regions possibly due to their geogenic origin. Chandrajith et al. (2005a,b) showed that the rice paddy soils of Sri Lanka were mainly inherited from the soil parent materials. Most of the trace elements, particularly La, Ce, Nb and Zr reside in accessory minerals such as zircon, monazite and garnet phases. The heavy rainfall associated with the Kandy region results in the prolonged submerging of paddy soils and extreme reducing conditions. This may facilitate the dissolution and subsequent loss of some trace elements. Conversely, dry zone paddy soils irrigated at rates that prevent prolonged submergence have significantly higher contents of some trace elements such as Ca and Mn. However, the contents of trace elements Ba, Cr, Ni, Pb and Zn are affected by geochemical redistribution as a result of both pedogenic and anthropogenic processes. The clay mineralogy of the two climatic regions of Sri Lanka is also different. The dry zone paddy soils are dominated by kaolinite with minor illite, while in the Kandy region, the soil consists mostly of gibbsite and traces of goethite (Navaratne et al. 1994). It is also possible that rare earth elements accumulate in paddy soils via phosphate fertilizer applications. Haleem et al. (2001) showed that superphosphate fertilizers contains 18.09 mg/kg of La and 8.55 mg/kg of Ce, and rare earth metal accumulation in soils due to phosphate fertilizer applications were in the order of $La > Ce > Sc > Eu > Hf$. Rare earth elements are highly immobile in the soil profile and therefore the higher mean contents of rare earth elements in paddy soils could be attributed to both mineralogical contribution and fertilizer applications.

As listed in Table 2, the activity values of K-40 obtained in this study ranged from 542 to 680, 308 to 637 and 296 to 846 Bq/kg in Anuradhapura, Giradurukotte and Kandy, respectively. The averages of K-40 were 604 ± 40 ,

484 ± 103 and 591 ± 151 Bq/kg in the above sites, respectively. The values are greater than the typical world average value of 370 Bq/kg (UNSCEAR 1993). The radioactivity of K-40 in two reference sites (undisturbed natural forests) from dry zone and wet zone were 258 and 138 Bq/kg, respectively (Table 2), and these K-40 values are lower than that of paddy soils, which are highly modified by anthropogenic activities such as puddling, submerging and artificial fertilizer applications (Fig. 3).

An index Ra_{eq} called the radium equivalent activity is also presented in Table 2 for comparison with the specific activities of materials containing different amounts of radium, thorium and potassium. This is a widely used radiological hazard index and was calculated according to Beretka and Mathew (1985):

$$Ra_{eq} = C(Ra) + 1.43C(Th) + 0.777C(K)$$

where $C(Ra)$, $C(Th)$ and $C(K)$ are the activities of ^{226}Ra , ^{232}Th and ^{40}K , respectively, in Bq/kg.

The average Ra_{eq} in the studied soil samples (109–142 Bq/kg) was below the world value (370 Bq/kg). The average dose rates due to the natural radionuclides at 1 m above the paddy soil were 54, 66 and 70 nGy/h in Anuradhapura, Giradurukotte and Kandy, respectively. The average values of outdoor external gamma radiation dose rate for 23 countries, representing about one-half of the world population were 24–85 nGy/h with a mean of 55 nGy/h, as reported by UNSCEAR (1993). The calculated annual effective dose due to terrestrial radiation was 1.5–1.8 mSv/year and the world average annual effective dose from terrestrial gamma rays is 0.46 mSv (UNSCEAR 1993).

As shown in Table 3, the TSP fertilizers contain extremely high K-40 counts and this contributes to the increasing K-40 activity in the dry zone rice paddy soils. The activity value of K-40 obtained from different fertilizers in this study varies from 103 to 15,606 Bq/kg. Ghosh et al. (2008) indicated that the phosphate fertilizer is one of the most important contributors of natural outdoor terrestrial radiation. Continued application of phosphate fertilizer, particularly TSP, into the rice paddy fields over a long period of time could increase the K-40 contents. The contribution of radionuclides in phosphate fertilizer to external radiation exposure in the agricultural areas is proportional to the intensity of fertilizer application (Sam and Holm 1995). In Sri Lanka, farmers use TSP twice a year (three times in some cases) for their rice cultivations and the average amount of TSP applied to rice paddy fields are 85 kg/ha per season. Although the dry zone rice paddy soils of Sri Lanka are characterized by a higher content of calcium and potassium levels compared to that of the wet zone, this might not contribute to the higher K-40 concentration in the investigated samples because even the wet zone paddy

Table 1 Summary of major and trace element data obtained from paddy soils (oxides are given in percentage and all other trace elements in mg/kg)

	Anuradhapura			Medawachchiya			Medirigiriya			Giradurukotte			Padaviya			Kandy		
	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum
Fe ₂ O ₃	7.71	4.81	14.61	7.32	4.57	9.28	8.70	6.46	9.54	8.96	6.82	11.59	8.61	4.75	11.24	9.98	6.45	11.01
MnO	0.14	0.08	0.30	0.15	0.08	0.32	0.10	0.08	0.13	0.25	0.15	0.38	0.23	0.09	0.40	0.11	0.08	0.19
MgO	0.84	0.65	1.22	0.70	0.40	0.91	1.03	0.57	1.28	0.65	0.29	1.13	0.87	0.66	1.20	1.66	0.83	1.99
CaO	1.85	1.24	2.25	1.86	1.03	2.58	1.38	1.17	1.63	1.75	0.79	2.46	1.76	1.34	2.08	1.43	0.89	1.89
K ₂ O	2.13	0.92	2.94	2.57	1.78	3.22	2.59	1.67	3.45	2.13	1.02	3.09	2.37	1.68	2.85	1.578	1.00	2.21
Ba	1,190	834	1,324	1,375	924	2,000	1,128	849	1,520	1,293	972	1,578	1,735	1,161	2,491	844	714	1,089
Ce	137	78	193	175	145	209	179	136	223	167	102	205	159	119	207	163	112	189
Co	23.4	15.0	41.0	25.2	14.0	37.0	29.8	22.0	38.0	47.2	36.0	62.0	31.4	13.0	49.0	34.6	26.0	39.0
Cr	89.8	70.0	136	89.4	69.0	115	94.8	64.0	115	94.0	66.0	134	104	74.0	124	111	88.0	118
Cu	50.2	14.0	102	18.0	6.0	27.0	26.6	20.0	39.0	42.4	26.0	63.0	36.0	16.0	67.0	48.4	12.0	103
La	67.6	43.0	93.0	87.2	63.0	109	103	76.0	149	63.2	23.0	100	71.0	44.0	83.0	100.8	46.0	119
Nb	17.4	10.0	23.0	20.6	13.0	26.0	14.0	9.0	21.0	14.2	7.0	21.0	17.8	10.0	32.0	13.8	10.0	16.0
Ni	33.2	19.0	53.0	30.0	18.0	48.0	41.2	29.0	56.0	35.2	26.0	45.0	47.2	26.0	66.0	55.4	36.0	68.0
Pb	17.4	10.0	29.0	20.4	10.0	26.0	23.4	13.0	35.0	22.0	13.0	29.0	21.6	12.0	27.0	15.8	7.0	34.0
Zn	86.2	66.0	132	84.0	57.0	126	91.2	67.0	121	86.2	64.0	118	95.8	71.0	120	136	70.0	190
Zr	759	525	1167	976	563	1569	628	360	984	711.4	416	1141	653	535	833	241	162	434
U	1.0	4.0	2.6	3.0	7.0	4.8	3.0	6.0	5.2	3.0	4.0	3.8	1.0	4.0	3.0	3.0	5.0	3.4

Table 2 Activity concentrations, Ra equivalent activity (Ra_{eq}) and dose rate in soil samples collected at Anuradapura, Giradurukotte and Kandy

Sample	Activity (Bq/kg)			Ra_{eq} (Bq/Kg)	Dose rate nGy/h ⁻¹
	²²⁶ Ra	²³² Th	⁴⁰ K		
Anuradhapura					
2-A-01	13± 1	49 ±2	542±16	124.80	61.40
2-A-02	12± 1	35 ±2	578±17	106.55	53.26
2-A-03	14± 1	42 ±1	607±16	120.79	60.00
2-A-04	16± 1	45 ±2	603±15	126.78	62.67
2-A-05	14± 1	31 ±2	642±18	107.76	54.23
2-A-06	14± 1	30 ±1	642±15	106.33	53.57
2-A-07	17± 1	43 ±2	580±14	123.15	60.78
2-A-08	7± 1	14 ±1	680±16	79.38	41.63
2-A-09	8± 1	21 ±1	584±16	82.99	42.55
2A-10	13± 1	40 ±2	584±14	115.168	57.26
Mean	13± 3	35 ±11	604±40	109±17	54.7±7.5
Giradurukotte					
3-G-01	19± 1	61 ±2	579±14	150.81	73.51
3-G-02	20± 2	53 ±2	485±15	133.135	64.58
3-G-03	41± 2	62 ±3	446±11	164	77.82
3-G-04	31± 1	51 ±2	420±11	136.27	65.14
3-G-05	15± 1	32 ±1	526±13	101.26	50.31
3-G-06	13± 1	44 ±2	436±12	109.49	53.51
3-G-07	17± 1	56 ±2	637±15	146.12	71.85
3-G-08	20± 1	64 ±2	608±15	158.33	77.17
3-G-09	13± 1	55 ±2	398±11	122.29	59.15
3-G-10	18± 1	63 ±3	308±9	131.8	62.70
Mean	20.7±9	54.1±10	484.3±103	135.35±20	66±10
Kandy					
1-P-01	23± 1	84 ±3	876±20	210.57	103.27
1-P-02	17± 1	52 ±2	645±16	141.02	69.55
1-P-03	16± 1	43 ±2	526±22	117.99	58.02
1-P-04	16± 1	52 ±2	668±19	141.79	70.11
1-P-05	14± 1	45 ±2	516±16	118.08	58.06
1-P-06	11 ±1	32 ±2	296±9	79.55	38.67
1-P-07	17± 1	53 ±2	543±14	134.6	65.8
1-P-08	22± 2	69 ±3	662±16	171.64	83.67
1-P-09	20± 1	72 ±2	661±16	173.85	84.76
1-P-10	16± 2	59 ±3	516±18	140.1	68.18
Mean	17.2±3.6	56±15.4	591±151	143±36	70±18
References					
Dry zone natural forest		18±1	258± 8	10 ±1	
Wet zone natural forest		125±55	138± 6	34 ±2	

soils from the Kandy region contain similar amounts of K-40, compared to that of the dry zone paddy soils.

Table 4 shows the toxic trace element contents measured in different fertilizers used in Sri Lanka. Significantly greater concentrations of most of the measured trace elements were found in TSP samples. Depending on the origin, TSP may contain different quantities of potentially

toxic heavy metals or compounds. These impurities in TSP are mainly derived from parent rock materials (Barbarick et al. 1995). TSP fertilizer is of special interest due to its high U and Cd content, which varies widely. The preliminary results obtained from this study indicate that the U content in TSP collected from the local market varies from 5.8 to 364 mg/kg. However, the content of U in rice field

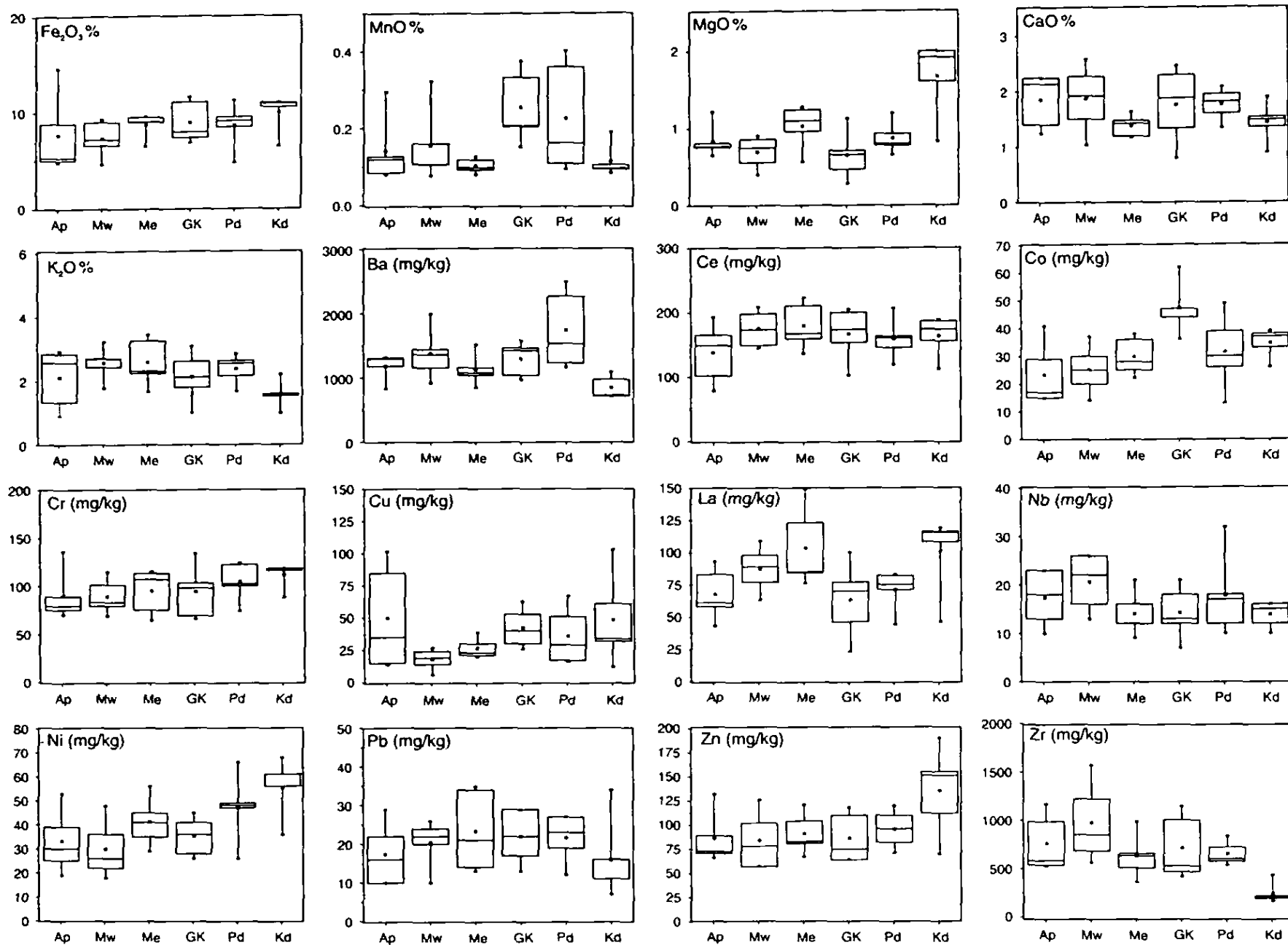


Fig. 2 Box plots showing trace element distribution in the rice paddy soil samples from study regions (outliers and far outliers are not shown). *Ap*Anuradhapura, *Mw*Medawachchiya, *Me*Medirigiriya, *GK*Giradurukotte, *Pd*Padaviya, *Kd*Kandy

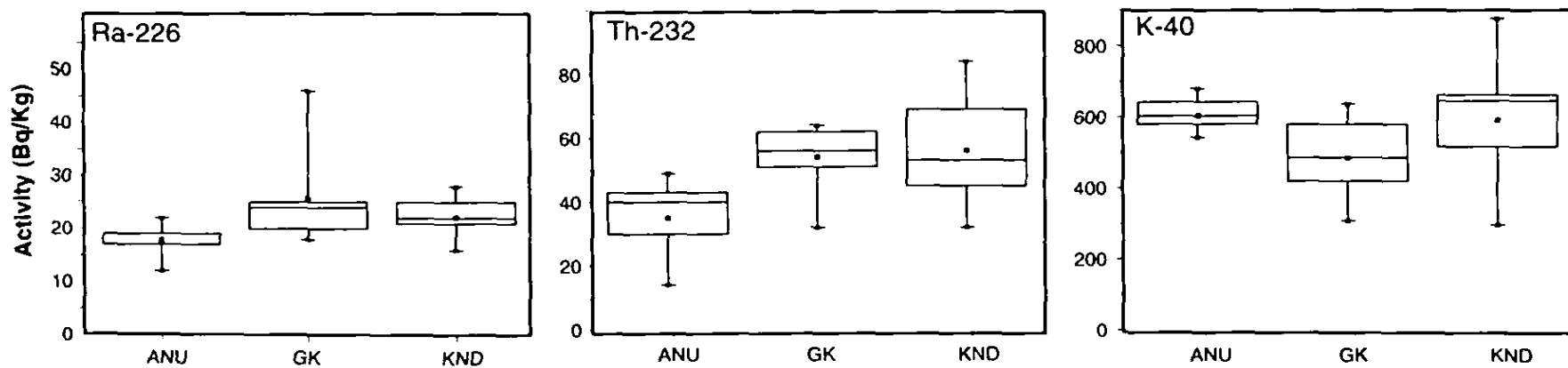


Fig. 3 Box plots showing activity concentrations of Ra-226, Th-232 and K-40 in the soil samples from three study regions. *ANU*Anuradhapura, *GK*Giradurukotte, *KND*Kandy

soils samples was below 7 mg/kg with an average of 3.6 mg/kg as measured by the XRF method. It has been reported that the global uranium concentrations in soils range from 0.3 to 11.7 mg/kg (UNSCEAR 1993), the average background concentration being 2 mg/kg (NCRP 1984).

The significance of application of uranium-bearing fertilizer was shown by Takeda et al. (2006). Large quantities of uranium contained in phosphate fertilizers are expected to enrich the uranium in agricultural soils after application of the fertilizer (Mermut et al.1996). Soil types, quality of fertilizer and application rates are among some of the

Table 3 Activity concentrations measured on TSP fertilizer samples

Sample reference	Activity (Bq/Kg)		
	K-40	Ra-226(eq)	Th-232
FER/01	109± 6	52 ± 2	10 ±1
FER/02	15,606±320		
FER/03	106± 6	72 ± 2	10 ±1
FER/04	103± 6	56 ± 2	11 ±1
FER/05	2,133±47	17± 1	11 ±1

Table 4 Content of trace elements (mg/kg) in different fertilizers collected from study regions

Collected location	Fertilizer type	Al	Cr	Ni	Cd	Pb	U
Anuradhapura	Urea	37	3.9	1.4	0.4	3.8	6.0
	NPK	203	3.9	1.4	0.4	3.8	20.1
	TSP	9,949	52.9	35.2	3.6	50.7	107
Medawachchiya	Urea	32	3.9	1.4	0.4	3.8	6.0
	NPK	262	3.9	1.4	0.4	3.7	6.5
	TSP	9,405	43.6	27.1	4.0	79.2	75.9
Medirigiriya	Urea	25	210.3	1.4	0.4	3.7	28.3
	NPK	135	23.7	1.4	0.4	3.8	6.1
	TSP	8,563	59.5	22.3	46.1	41.1	5.8
Giradurukotte	Urea	54	19.6	1.4	0.4	6.0	25.3
	NPK	143	22.8	1.4	0.4	3.8	6.1
	TSP	9,016	65.9	24.2	39.8	58.2	64.1
Giradurukotte	Urea	27	nd	1.6	nd	4.0	nd
	NPK	77	2.6	1.3	0.5	2.6	119
	TSP	5,177	19.2	10.6	2.3	67.2	364
Kandy	Urea	52	21.0	1.4	0.4	3.9	24.4
	NPK	140	22.1	1.4	0.6	3.8	6.1
	TSP	10,113	62.1	27.3	4.3	80.2	166

ndnot detected

factors that influence the accumulation of uranium in surface soils. In view of the fact that about 90% of uranium input to the field was attributable to superphosphates, Takeda et al. (2006) estimated that the annual input of uranium from the application of fertilizer material was 3.0 mg/m² per year. However, rice paddy soils are submerged most of the time and virtually all of the water-soluble uranium in fertilizers was absorbed readily after application (Tsumura and Yamasaki 1993) and highly mobile uranyl cation, therefore, hardly accumulates in the soil. The U applied with phosphate fertilizer could be leached into the groundwater draining from paddy fields and this would concentrate in the surface and groundwater used as drinking water sources by the local community. The mobility of fertilizer-derived uranium is dependent on fertilizer type, pore water chemistry, pH, mineralogy,

texture, etc. of soils (Zielinski et al.1997). Makweba and Holm (1993) showed that the radioactivity of superphosphates in Tanzania had activity concentrations as high as 400 Bq/kg. Currently, much of the required phosphate fertilizers to Sri Lanka are imported from other countries and no detailed measurements of the trace elements or activity of radionuclides in phosphate fertilizer have been reported so far.

van Kauwenbergh (2002) considered about 16 elements that are associated with phosphate rock and fertilizer, which are potentially hazardous to human health. Among these elements, Cd and U are the most important elements and uranium is particularly known for its nephrotoxic nature (Zamora et al. 1998; Kurttio et al. 2002). Kurttio et al. (2002). Zamora et al. (1998) observed renal effects in humans due to chronic exposure of U through drinking water. They concluded that uranium exposure is weakly associated with altered proximal tubular functions suggesting that even low uranium concentrations in drinking water can cause nephrotoxic effects. The increasing incidence of CKD in the dry zone regions of Sri Lanka may possibly be attributed to the high input of uranium-rich superphosphates that lead to increase in the U content in groundwater. Groundwater is the main source of drinking water in the CKD-affected regions. It is also important to note that the groundwater in the dry zone region is characterized by high fluoride levels (Dissanayake1996).

Conclusions

Due to the unavailability of a proper controlling mechanism, very low-quality fertilizers are imported to Sri Lanka, making the problem more severe. This study revealed that some phosphate fertilizers used in CKD-affected areas contain very high amounts of potentially toxic trace metals including U. Although the trace element geochemistry of rice soils in the affected and unaffected regions are not remarkably different, some toxic metals such as U can easily get leached and washed away from the rice fields and the surrounding environment due to heavy rains (exceeding 2,500 mm per annum), bearing in mind that the fluoride contents in groundwater of the dry zone regions are also much higher.

It is proposed that fertilizers applied to the rice fields, mainly phosphate fertilizers, should be investigated fully in terms of application, accumulation in soils and transfer into drinking water and finally into the human body. The causative factors for the CKD in dry zone regions of Sri Lanka are still speculative, but contributions from the contamination due to fertilizer could be possible, bearing in mind that fertilizers are added, in some cases three times annually into the rice fields.

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