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RADIOMETRIC AND CHEMICAL ANALYSIS OF SALINE SOIL SAMPLES OF PACCA ANNA, FAISALABAD, PAKISTAN

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Abstract: Some saline soils from Pacca Anna, District Faisalabad, Pakistan were analyzed for the naturally occurring radionuclides of K^{40}, the uranium and thorium series; and a fission product Cs^{137} was determined by the gamma ray spectroscopy using high purity Germanium (HPGe) detector. The soil samples were collected from the depths of plough layer i.e. 0-25 cm with 5cm increments. After crushing and drying, these samples were stored in radon impermeable plastic containers for at least forty days in order to bring Rn^{222} and its short-lived daughter products in secular equilibrium with Ra^{226}. The geometric dimensions of the samples were identical with those containing the reference materials from IAEA. The concentration range of different radionuclides in becquerel per kg (Bq kg^{-1}) of dry mass of soil samples were observed for K^{40} to lie in the range 563.24-619.57 Bq kg^{-1} 499.24-604.20 Bq kg^{-1} and 614.45-670.77 Bq kg^{-1} for the virgin, fertilized, and NIAB soils, respectively. The concentration values of Cs^{137} for virgin, fertilized, and NIAB soils ranged between 3.57-3.63 Bq kg^{-1}, 1.98-5.15 Bq kg^{-1} and 2.12-3.15 Bq kg^{-1}, respectively. For Th^{232} the results are given as 45.63-61.99 Bq kg^{-1}, 48.55-54.30 Bq kg^{-1} and 51.69-60.03 Bq kg^{-1} for the same soils. For the radioisotopes Ra^{226} the results are 27.34-32.66 Bq kg^{-1}, 24.29-28.61 Bq kg^{-1} and 28.61-32.66 Bq kg^{-1}, respectively.

Keywords: Natural radioactivity, radiation monitoring, radionuclides, soil radiation.

INTRODUCTION

The present human environment cannot be protected against contamination from radioactive nuclides [FAO 1988]. The human beings are exposed to natural and man-made radioactivity. The natural radioactivity is from the terrestrial radionuclides, which belong to uranium and thorium series and potassium–40. Artificial radionuclides in the environment were first observed after the first nuclear missile tests [Forsberg 2000]. However, at the present time a significant source of radioactive contamination is nuclear power engineering and application of radionuclides in science and technology [Rosen 1991].

Radiation contamination can result from either expected or accidental release of radioactive materials during the treatment of uranium and thorium ores, the operation of nuclear reactors, the processing of burnt fuel elements from nuclear reactors, or the application of radionuclides in medicine, research, industry and agriculture, as well as from radioactive fallout from the atmosphere [IAEA 1973, 1989, Coxm and Hauser 1984].

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On the average during nuclear fission as many as 200 radionuclides of 35 different elements with medium atomic numbers are formed. However, abundance of many of them in the fission mixture decreases rapidly because of their short half–lives; this also leads to a rapid decrease in the total activity of the fission mixture [Dejoung and Dettany 1982, Mollah *et al.* 1984].

Radionuclides in soil are a source of the contamination of forage and food with radioactive substances, of which potassium, cesium and radium isotopes are the most significant [IAEA 1989]. Radionuclides penetrate into plants either from the atmosphere as deposits on soil surface or through roots from the soil. Radioactively contaminated plants are a significant hazard to man, either directly (foods of plant origin), or indirectly (milk of animals which receive contaminated forage) [Delavne *et al.* 1986, Pan *et al.* 1984].

Measurement of radioactivity in soils can be used to determine the cumulative fallout from nuclear weapon test, nuclear accident and the natural radioactivity from the primordial radionuclides [Floro and Kritidis 1991]. Because of increased public concern and awareness about radioactive pollution, attention was paid in this task to study the occurrence of some gamma radionuclides and their determination in different soils.

MATERIALS AND METHODS

AREA UNDER STUDY

The area under study was located at a place called Pacca Anna, which is situated at a distance of 34 km in the north of Faisalabad. It is a Bio Saline Research Station (BSRS) of the Nuclear Institute for Agriculture and Biology (NIAB), Faisalabad. It has an area of 1000 acres (400 hectares). A number of salt tolerant plants, trees and bushes were grown and the outlook of a barren and waste landscape was changed into green crop cover in few years. It is a saline patch of the soil cultivated with brackish water having electrical conductivity of 4.8 dS m^{-1}. The annual rainfall is 310mm. The soil is medium texture with salinity ranging from 21 - 6 dS m^{-1}.

SAMPLE COLLECTION

Soil samples were collected by random sampling method (RSM) from ten fields of one hectare each; five of virgin soil and five of fertilized soil. The soil samples were collected at five different depths of 0-5, 5-10, 10-15, 15-20, and 20-25 cm. Finally, 15 samples were prepared after mixing various samples collected from different depths and fields. The samples were designated by names as BSRS-2V for virgin soils and BSRS-2F for fertilized soils where BSRS-2 stands for Bio Saline Research Station-2.

The soil samples were dried, sieved and kept moisture free, by keeping over night in the electric oven. After drying these samples were packed in airtight plastic bottles, which are similar to IAEA standards for natural radionuclides.

RADIATION MONITORING

Gross radioactivity was counted with a radiation monitor type 335 Airmec consisting of a Geiger Mueller Detector, which measures number of counts per second (CPS). Data were recorded for each soil sample and the same was recorded again after two days.

CHEMICAL ANALYSIS

The pH of soil samples was measured by a pH meter, PH-530, W-T-W Germany and the concentration of various metal ions was determined by Flame Photometer, PFP7 Jennway. Whereas the electrical conductivity (σ) of all the soil samples was measured by a conductivity meter, LF-538 W-T-W Germany.

Table 1: Readings of the soil samples by monitor (335).

Soil Type	Sample	Mean of Fresh Soil in CPS	Mean After 24 Hours in CPS
	F1	40.00	26.66
	F2	41.67	21.66
Cultivated	F3	43.50	23.33
	F4	33.33	20.00
	F5	40.00	20.00
	V1	33.33	21.66
	V2	41.67	16.66
Virgin	V3	40.00	21.66
	V4	40.00	20.00
	V5	38.33	18.33
	N1	40.00	18.33
	N2	43.33	11.66
Control	N3	36.67	16.66
	N4	33.33	16.66
	N5	33.33	13.33

RESULTS AND DISCUSSION

Radioactivity was monitored as number of counts per second. The results of measurements are shown in Table 1. From this data we observed that readings, which were taken after two days, were less than the readings taken for the fresh soils. This shows that soil radiate different radiations particularly radon gas to the surrounding environment.

The electrical conductivity (σ) of all the soil samples has been shown in Table 2. It is observed that σ of fertilized soil samples of BSRS-2 varied from 9.70-15.04 mS cm^{-1}; for virgin samples it varies from 4.02-15.30 mS cm^{-1} respectively. For NIAB it has value in the range of 2.53 - 4.38 mS cm^{-1}. The sodium concentrations (meq l^{-1}) are in the range 74-93, 76-99 and 26-32 for the same soils respectively (Table 2). The concentration of calcium plus magnesium as measured in meq l^{-1} varied from 0.4-1.0, 0.8-8.3, and 1.6-2.2 respectively (Table 2).

Table 2: Chemical analysis of soil samples.

Soil Samples	Depth (cm)	Electrical conductivity (σ) mS cm^{-1}	pH	Concentration of Ca + Mg (meq l^{-1})	Concentration of Na (meq l^{-1})
F1	0-5	15.04	7.80	1.0	89
F2	5-10	10.20	8.02	0.8	93
F3	10-15	9.70	8.21	0.4	78
F4	15-20	10.20	8.24	0.4	74
F5	20-25	9.70	8.32	0.4	82
V1	0-5	15.3	7.28	8.3	99
V2	5-10	7.15	7.50	7.9	76
V3	10-15	5.92	7.59	5.3	89
V4	15-20	4.50	7.64	2.0	90
V5	20-25	4.02	7.79	0.8	87
N1	0-5	3.17	7.7	1.6	32
N2	5-10	2.53	7.81	2.2	26
N3	10-15	3.10	7.74	1.6	27
N4	15-20	3.38	7.69	1.7	26
N5	20-25	4.38	7.72	1.6	28

In all soil samples, the concentration of the natural radionuclides of the uranium series, thorium and K^{40} and a fission product Cs^{137} have been determined by gamma ray spectrometry. The results of specific gamma ray activities due to Th^{232}, Ra^{226}, K^{40} and Cs^{137} are given in Tables 3 - 7 where the activity levels were found to be normally distributed except that of Cs^{137} (Table 6). The lower limit of detection (LLD) for K^{40}, Cs^{137}, Th^{232} and Ra^{226} are 59 Bq kg^{-1}, 1.27 Bq kg^{-1}, 3.31 Bq kg^{-1} and 3.26 Bq kg^{-1}, respectively. In fertilized soil samples of BSRS-2, specific activities for K^{40} (Table 5) ranged from 499.24-604.20 Bq kg^{-1} with an average of 558.12 Bq kg^{-1}; for Th^{232} (Table 3) from 48.55-54.30 Bq kg^{-1} with an average of 52.72 Bq kg^{-1}; for Ra^{226} (Table 4) from 24.29-28.61 Bq kg^{-1} with an average of 25.96 Bq kg^{-1} and for Cs^{137} (Table 6) from 1.98-5.15 Bq kg^{-1} with an average of 4.15 Bq kg^{-1}.

Table 3: Activity concentration of Th^{232} in soil samples of BSRS-2 and NIAB (LLD= 3.31 Bq kg^{-1}).

Sample	Abundance ratio (P_r)	Efficiency (ε)	Peak area counts	Back ground counts	Net counts	Activity (Bq kg^{-1})	Uncertainty (Bq kg^{-1})
F1	0.1126	0.0262	938	119	819	53.38	2.11
F2	0.1126	0.0262	867	119	748	48.55	2.04
F3	0.1126	0.0262	952	119	833	54.30	2.13
F4	0.1126	0.0262	943	119	824	53.71	2.12
F5	0.1126	0.0262	942	119	823	53.64	2.12
V1	0.1126	0.0262	1050	119	931	60.68	2.22
V2	0.1126	0.0262	976	119	857	55.86	2.15
V3	0.1126	0.0262	1070	119	951	61.99	2.24
V4	0.1126	0.0262	899	119	700	45.63	2.07
V5	0.1126	0.0262	1000	119	881	57.42	2.18
N1	0.1126	0.0262	2000	119	1238	5.05	2.19
N2	0.1126	0.0262	1910	119	1148	50.25	2.07
N3	0.1126	0.0262	1890	119	1128	51.69	2.09
N4	0.1126	0.0262	2050	119	1288	60.03	2.21
N5	0.1126	0.0262	1910	119	1148	55.86	2.15

Table 4: Activity concentration of Ra^{226} in soil samples of BSRS-2 and NIAB (LLD= 3.26 Bq kg^{-1}).

Sample	Abundance ratio (P_r)	Efficiency (ε)	Peak area counts	Back ground counts	Net counts	Activity (Bq kg^{-1})	Uncertainty (Bq kg^{-1})
F1	0.4460	0.0170	1720	762	958	24.29	1.26
F2	0.4460	0.0170	1790	762	1028	26.07	1.28
F3	0.4460	0.0170	1770	762	1008	25.56	1.27
F4	0.4460	0.0170	1760	762	998	25.31	1.27
F5	0.4460	0.0170	1890	762	1128	28.61	1.30
V1	0.4460	0.0170	1930	762	1168	29.62	1.31
V2	0.4460	0.0170	2040	762	1278	32.41	1.34
V3	0.4460	0.0170	1960	762	1198	30.38	1.32
V4	0.4460	0.0170	2050	762	1288	32.66	1.34
V5	0.4460	0.0170	1840	762	1078	27.34	1.29
N1	0.4460	0.0170	1010	762	891	31.40	1.33
N2	0.4460	0.0170	890	762	771	29.11	1.31
N3	0.4460	0.0170	912	762	793	28.61	1.30
N4	0.4460	0.0170	1040	762	921	32.66	1.34
N5	0.4460	0.0170	976	762	857	29.11	1.31

Table 5: Activity concentration of K^{40} in soil samples of BSRS-2 and NIAB (LLD=59 Bq kg^{-1}).

Sample	Abundance ratio (P_r)	Efficiency (ε)	Peak area counts	Back ground counts	Net counts	Activity (Bq kg^{-1})	Uncertainty (Bq kg^{-1})
F1	0.1070	0.00702	4610	2470	2140	547.88	21.54
F2	0.1070	0.00702	4420	2470	1950	499.24	21.25
F3	0.1070	0.00702	4590	2470	2120	542.76	21.51
F4	0.1070	0.00702	4800	2470	2330	596.52	21.82
F5	0.1070	0.00702	4830	2470	2360	604.20	21.87
V1	0.1070	0.00702	4750	2470	2280	583.52	21.75
V2	0.1070	0.00702	4930	2470	2460	629.81	22.02
V3	0.1070	0.00702	4890	2470	2420	619.57	21.96
V4	0.1070	0.00702	4860	2470	2390	611.89	21.91
V5	0.1070	0.00702	4670	2470	2200	563.24	21.63
N1	0.1070	0.00702	4980	2470	2510	642.61	22.09
N2	0.1070	0.00702	4870	2470	2400	614.45	21.93
N3	0.1070	0.00702	4930	2470	2460	629.81	22.02
N4	0.1070	0.00702	5050	2470	2580	660.53	22.20
N5	0.1070	0.00702	5090	2470	2620	670.77	22.26

Table 6: Activity concentration of Cs^{137} in soil samples of BSRS-2 and NIAB (LLD= 1.27 Bq kg^{-1}).

Sample	Abundance ratio (P_r)	Efficiency (ε)	Peak area counts	Back ground counts	Net counts	Activity (Bq kg^{-1})	Uncertainty (Bq kg^{-1})
F1	0.8460	0.0159	720	368	352	5.03	0.47
F2	0.8460	0.0159	687	368	319	4.55	0.46
F3	0.8460	0.0159	729	368	361	5.15	0.47
F4	0.8460	0.0159	652	368	284	4.05	0.45
F5	0.8460	0.0159	507	368	139	1.98	0.42
V1	0.8460	0.0159	618	368	250	3.57	0.45
V2	0.8460	0.0159	622	368	254	3.63	0.44
V3	0.8460	0.0159	420	368	52	N.D	N.D
V4	0.8460	0.0159	332	368	N.D	N.D	N.D
V5	0.8460	0.0159	318	368	N.D	N.D	N.D
N1	0.8460	0.0159	589	368	221	3.15	0.44
N2	0.8460	0.0159	548	368	180	2.57	0.43
N3	0.8460	0.0159	563	368	195	2.78	0.43
N4	0.8460	0.0159	537	368	171	2.44	0.42
N5	0.8460	0.0159	517	368	149	2.12	0.42

In virgin soil samples of BSRS-2 the specific activities for K^{40} ranged from 563.24 - 629.81 Bq kg^{-1} with an average of 601.48 Bq kg^{-1}; for Th^{232} from 45.63-61.99 Bq kg^{-1} with an average of 56.32 Bq kg^{-1}; for Ra^{226} (Table 4) from 27.34-32.66 Bq kg^{-1} with an average of 30.48 Bq kg^{-1} and for Cs^{137} only in soil samples of V1 and V2, the activity levels ranged from 3.57-3.63 Bq kg^{-1} with an average of 3.6 Bq kg^{-1}. The three most important primordial radionuclides investigated in the two types of soils are K^{40}, Ra^{226} and Th^{232}.

Table 7: Total activity concentration (Bq kg^{-1}) of K^{40}, Cs^{137}, Th^{232} and Ra^{226} in soil samples of BSRS-2 and NIAB.

Sample	K^{40}	Cs^{137}	Th^{232}	Ra^{226}
F1	547.88±21.50	5.03±0.47	53.38±2.11	24.29±1.26
F2	499.24±21.25	4.55±0.46	48.55±2.04	26.07±1.28
F3	542.76±21.51	5.15±0.47	54.30±2.13	25.56±1.27
F4	596.52±21.82	4.05±0.45	53.71±2.12	25.31±1.27
F5	604.20±21.87	1.98±0.42	53.64±2.12	28.61±1.30
V1	583.72±21.75	3.57±0.45	60.68±2.22	29.62±1.31
V2	629.00±22.02	3.63±0.44	55.86±2.15	32.41±1.34
V3	619.57±21.96	N.D	61.99±2.24	30.38±1.32
V4	611.89±21.91	N.D	45.63±2.07	32.66±1.34
V5	563.24±21.63	N.D	57.42±2.18	27.34±1.29
N1	642.61±22.09	3.15±0.44	58.05±2.19	31.40±1.33
N2	614.45±21.93	2.57±0.43	50.25±2.07	29.11±1.31
N3	629.81±22.02	2.78±0.43	51.69±2.09	28.61±1.30
N4	660.53 ±22.20	2.44±0.42	60.03±2.21	32.66±1.34
N5	670.77±22.26	2.12±0.42	55.86±2.15	29.11±1.31

Natural Potassium has three isotopes; K^{39}, K^{40} and K^{41}. Among them only K^{40} ($T_{1/2}$ = 1.3 x 10^9 years) possesses natural gamma radioactivity and its abundance in nature is 0.012% of all Potassium. During decay K^{40} produces two daughter isotopes; Ca^{40} and Ar^{40}, with the emission of beta and gamma radiation. Concentration of K^{40} in soil varies from 100-700 Bq kg^{-1} of soil. In the present work, high activity of K^{40} was determined in soil samples of cultivated soil of Pacca Anna. The mean activity of K^{40} observed in the present study is about two times higher than the world average [Floro and Kritidic 1991, NEA, OECD 1979, Hamid *et al*. 2002]. The average concentration of K^{40} in other parts of the world is also comparable with those of present study (Fig. 1).

As mentioned above, in addition to K^{40}, the other naturally occurring radionuclides, which were measured, were Ra^{226} and Th^{232}. Ra^{226} (U^{238} series) is considered as the highly radiotoxic natural radionuclides. The mean activity of Ra^{226} ($T_{1/2}$ = 1620 years) determined in the present study is nearly double of that of Th^{232} ($T_{1/2}$ = 1.4 x 10^{10} years) and is two times higher than the world average [Michel *et al*. 1981, Nurul Alam *et al.* 1990]. However, the average concentrations of Ra^{226} and Th^{232}, being comparable with those of soils of Bangladesh and India (Figs. 2, 3), can be justified by similarities in geographic locations and geological origin of soil as well as climatic, hydrological and agricultural history.

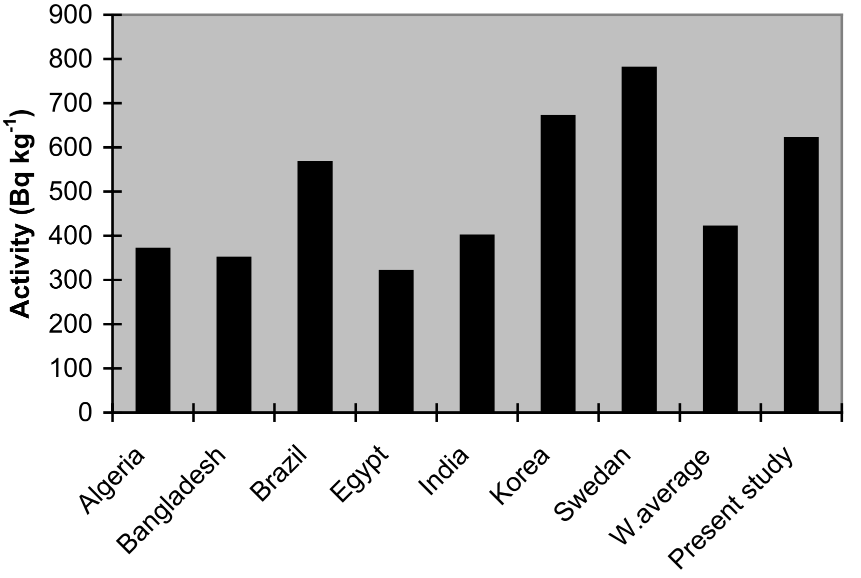

Fig. 1: A comparison of activity concentration of K^{40} corresponding to different soils of various countries.

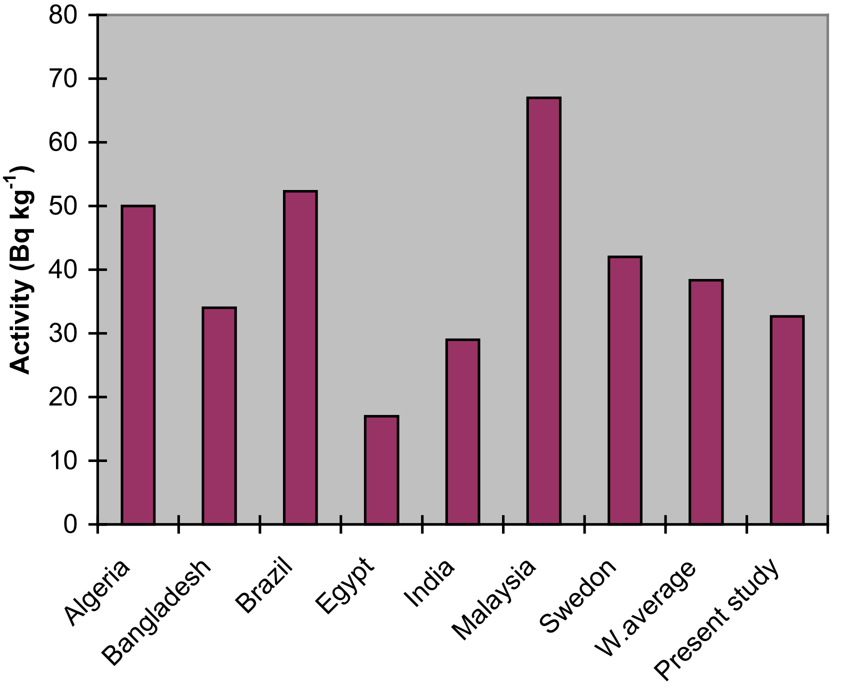

Fig. 2: A comparison of activity concentration of Ra^{226} corresponding to different soils of various countries.

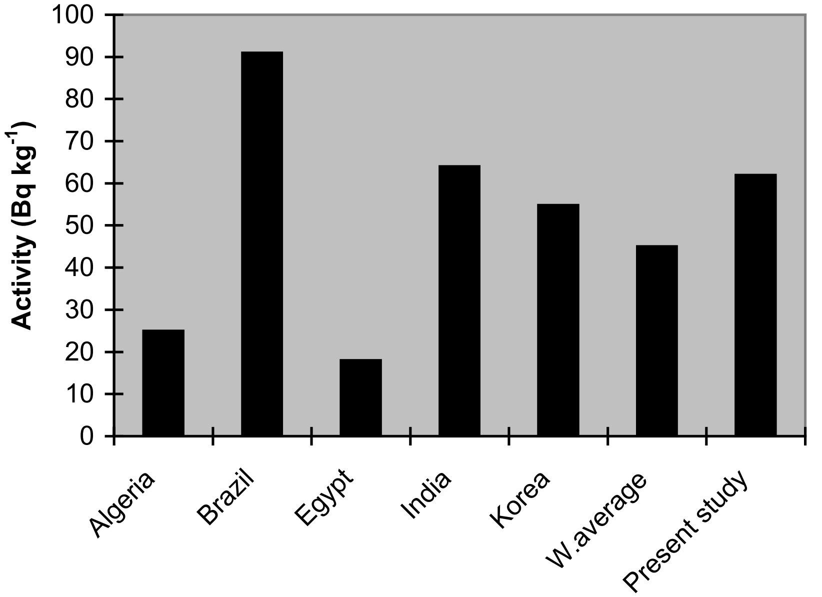

Fig. 3: A comparison of activity concentration of Th^{232} corresponding to different soils of various countries.

The concentration of natural radionuclides varies slightly in all soils of present study. Since high activity of K^{40}, Ra^{226} and Th^{232} have been observed. This may be attributed to the fact that the use of fertilizers in various agriculture situations have affected radio nuclides concentrations to a larger extent and the Potassium containing fertilizers are one of the main cause of presence of high activity of K^{40} in soils, particularly of cultivated soils of Pacca Anna.

It has also been known since early in this century that phosphate rocks contain substantial concentration of Uranium and its decay products Thorium and Radium. Since phosphate rock is an important raw material used for manufacturing different types of phosphatic fertilizers, therefore, when this rock is processed into phosphatic fertilizers, most of the Uranium and some of the Radium accompanies the fertilizers. It has also been estimated earlier that phosphatic fertilizers applied to the fields in recommended amounts could raise radioactivity level in soils [Bhatti and Malik 1994]. It is evident from the data that in addition to natural radionuclides, the soil samples of BSRS-2 [Bhatti and Malik 1994] also have a fission product Cs^{137}. This indicates that the soils of these areas might have received some fallout from Chernobyl accident or any other source, like open-air world wide nuclear explosions [Rosen 1991]. Cs^{137} is strongly absorbed and retained by soil particles, as are the natural radionuclides, which are found randomly distributed at different depths of the soil.

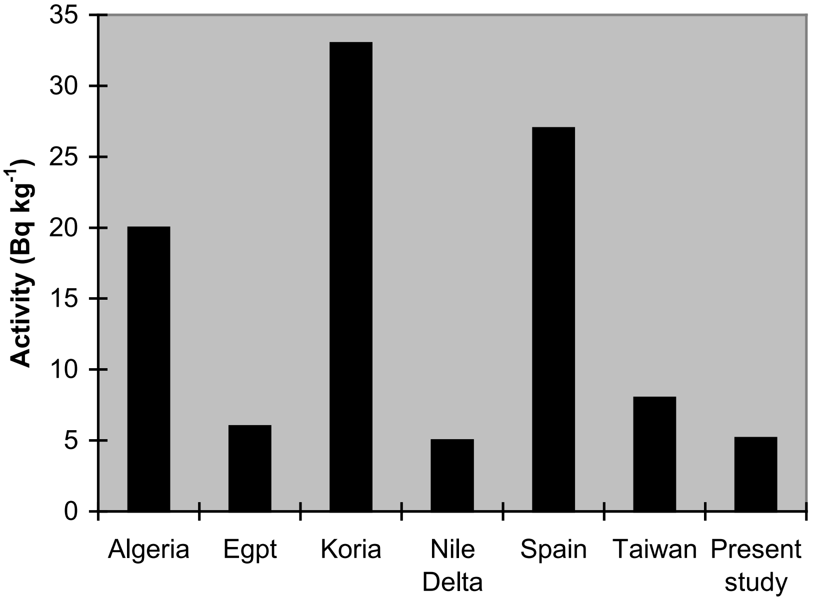

Fig. 4: A comparison of activity concentration of Cs^{137} corresponding to different soils of various countries.

Deposition of radioactive fallout including Cs^{137} at any given site is related to factors such as latitude, precipitation and local topography. Fixation of Cs^{137} in soil is dependent on clay content of the soil, which is affected by ploughing. Since it mixes the soil and associated radionuclides as evident from the present results of cultivated land.
The values of specific activities due to Cs^{137} were also compared with a similar work reported for other countries of the world as shown in Fig. 4. The graph shows that the highest value of world average activity of Cs^{137} was reported in Korean soils [FAO 1988], while the lowest was obtained in soils of present study and that of Egypt.

CONCLUSIONS

Following conclusions may be drawn from the present investigations:

1. Phosphatic rocks are used as raw material in the formation of fertilizers. These fertilizers are the main source of radioactivity in cultivated soils.
2. Cs^{137} has been indicated in upper layers of the virgin soils, it means that this area might have nuclear fall in the past.

3. Radioactivity is leached down in the down layers of the soil; this shows the leaching effect of nuclear fallout.

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