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Study of Soil around Goalundo Upazilla by Proton Induced X-ray Emission (PIXE) Technique

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Abstract

Heavy element content in the environment has been increasing at an alarming rate and some of them are highly toxic and can cause harmful effect that may even lead to death. In order to study the impact of toxicity of these heavy elements, soil samples collected from the surrounding area of Goalundo Upazilla in Rajbari has been analyzed by 3 MV Van de Graaff Accelerator at AECD using proton induced X-ray emission (PIXE) technique. The elements that were found to be present in these samples are Cl, K, Ca, Mn, Fe, Co, Zn, and Rb of the amount of 1120±272.59, 9706±626.73, 36936±255.84, 754.66±67.68, 57391±83.02, 98±10.11, 586.40±143.12, 105.93±38.90 ppm respectively. Except Cl and K all other elements Ca, Mn, Fe, Co, Zn and Rb are found to be of high concentration which may transfer to food chain and consequently may affect the human health. The experimental procedure provided comparable results with certified values of the standard sample. The search was made for certain toxic elements including Cd, Hg, Pb, etc because of their negative roles in enhancing infant mortality rates, but none of them was detected. Finally, the observed highest concentrations of these elements in soil could have an effect on human health to the surrounding study area, which may cause public health hazards. Hence, pretreatment process of waste water is necessary for reducing the amount of heavy metals before using it to the agricultural soil.

Keywords: PIXE; toxicity; pollution.

1. Introduction

Developmental activities e.g. industrial, agricultural, transportation, constructional work, etc. cause degradation and drastic changes in every component of environment such as soil, water and air through pollution. The role of heavy and trace elements in the soil system is increasingly becoming amatter of global concern at private as well as governmental levels, especially as soil constitutes a vital component of rural and urban environments [1]. Agricultural soil contamination with heavy metals through the repeated use of untreated or poorly treated wastewater from industrial establishments causes severe ecological problems in Bangladesh. Although some trace elements are essential in plant nutrition, plants growing in the close vicinity of industrial areas display increased concentration of heavy metals, serving in many cases as biomonitors of pollution loads [2]. Uptake of such metals and accumulates them in quantities high enough in vegetables, crops, fruits, etc. cultivated in this polluted soils may cause clinical problems both to animals and human beings [3-4].

Essential elements can become toxic if ingested or inhaled at a sufficiently high level and for a long enough period [5]. Any fluctuation, i.e., deficiency or excess in their normal level in living cells may cause the physiological disorders and may lead to various diseases like cancer, dental caries, heart disease, goiter, hypertension, gallstones, obesity, osteoporosis, osteomalacia, arthritis, anemia, etc. Heavy metals and trace elements are also a matter of concern due to their non biodegradable nature and long biological half-lives. The knowledge of elemental contents of soil is important, because these are the primary sources of minerals for all living beings and many diseases are caused in men and animals due to mineral deficiency.

Wastewater from industries or other sources carries significant amounts of toxic heavy metals such as Cr,

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Mn, Cu, Ni, Zn, Cd and Pb in surface soil which creates problem for safe rational use of agricultural soil. Long-term use of industrial or municipal wastewater in irrigation is known to have a significant contribution to the quantity of trace and heavy elements in surface soil. As a result, excessive accumulation of trace elements in agricultural soils through wastewater irrigation may not only result in soil contamination but also affect food quality and safety [6]. Also, over utilization of natural resources for development purposes cause imbalance in the environmental sustainability. So, to ensure sound health and quality of life, identification and regular monitoring of pollution sources are essential. The present work is aimed at investigating essential and toxic elements in agricultural soil of the study area, which will help us to understand the extent of environmental pollution of the area and their effects on human health. The data generated in this study may help to focus our attention to control pollution and to generate awareness among the general public and to warn the policy makers about the disastrous consequences that would follow if the appropriate measures are not taken to contain the environmental pollution in time.

2. Materials and method

2.1 Sample Collection and Preparation

The soil samples were collected from fifteen different locations of Goalundo Upazilla of Rajbari districts and were prepared at the Accelerator Facilities Division of Atomic Energy Center, Dhaka. The collected arable soils were labeled and dried in an oven at a temperature of 60^0 C for about 10 hours to make the samples water free. After cooling the samples to the room temperature in a dessicator, the weights were taken. The process of heating, cooling and weighing was repeated until a constant weight is shown by the balance which is a confirmation of zero water content. The dried weighted samples were then grinded in a grinder and made into pellets with a hand press pellet maker. These pellets were used as the targets for irradiation.

2.2 Method of Analysis

Concentration measurements of elements were made with PIXE technique using proton beam of energy 2.2 MeV obtained from the 3 MV Van de Graaff Accelerator at the Atomic Energy Centre, Dhaka (AECD). In PIXE measurements the characteristic X-rays emitted from the sample upon bombardment with 2.2 MeV proton beam were detected using a 30 mm^{2}Si(Li) detector with associated electronic setup. The detector resolution (FWHM) at X-ray energy of 5.9 keV was 165 eV. The PIXE count rate was reduced using a 170 m thick Mylar absorber to reduce the dead time of the measurements and the dead time was generally less than 5%. A 200 g/cm^2 gold diffuser foil was used to homogenize the beam onto the target samples and the current was in the range of 5-10 nA. The PIXE analysis detects elements such as S, Cl, K, Ca, Fe, Cu, Zn, Ga, Br, Ru, Pb etc. and gives the accurate concentration of the most of the elements present in the samples with high accuracy [7]. In PIXE set up MAESTRO software was used to display and store the X-ray emission spectrum. GUPIX with DAN32 interfacing software [8] were used for analyzing PIXE spectra to obtain the concentration value of each element detected in the sample.

2.3 Method of validation

The validity of the PIXE set up is done by analyzing IAEA standard 'soil-7'. Measurements were taken several times and the average obtained result agreed within ±5 % of the certified values, which conforms the validity of this method (Table 1).

Table 1. Elemental concentrations of the Standard Reference Material

Elements	Certified value (ppm)	Measured Value (ppm)	Certified Value/ Measured Value
K	12100	10925	1.107551
Ca	163000	174153	0.935959
Zn	104	112.51	0.8
Fe	25700	30000	0.856667
Co	8.9	10.05	0.885572
Rb	51	65.23	0.924362

3. Results and Discussions

In the present study 15 soil samples were analyzed and the studied elements were Cl, K, Ca, Mn, Fe, Co, Zn and Rb. The analytical results with % of errors are tabulated as in table 2. Large fluctuations in errors ~ 1 to 40 % in the measured values of concentrations of the elements were observed from table 2. There may be several reasons for these fluctuations in the errors. All the present experiments were done at fixed proton energy of 2.2 MeV, which was not high enough for exciting all the elements present in the samples with sufficient intensity for concentration determination below percentage level. Proton beam of higher energy (> 2.2 MeV) might be required to excite all the elements and isotopes present in the target samples.

Table 3 shows the comparison of elemental constituents/abundances of soil samples of different countries with that of the present work. Percent wise distributions of soil constituents are shown graphically as pie chart in Fig. 1. According to this diagram the soil constituents are: Ca 35%, Fe 53%, K 9%, Cl 1%, Mn 1%, Zn 1% and other elements Rb and Co 0%. Figure 2 represents PIXE spectra for standard reference material.

Table 2. Elemental constituents and % of errors of Soil samples

Sample code/ID	K (ppm)	Ca (ppm)	Mn (ppm)	Fe (ppm)	Co (ppm)	Zn (ppm)	Rb (ppm)	Cl (ppm)
S-1	11167 5.65%	20401 0.64%	1695 8.77%	73782 0.12%	68 10.55%	720 24.79%	227 40.18%	435 26.28%
S-2	7624 4.6%	42907 0.72%	754 9.05%	60730 0.16%	125 10.03%	294 15.73%	51 14.44%	238 28.99%
S-3	6526 5.31%	45719 0.69%	295 7.85%	64073 0.16%	50 10.80%	622 36.45%	171 35.18%	866 25.33%
S-4	8678 8.53%	19124 0.59%	1442 10.57%	74931 0.12%	225 10.92%	1074 21.10%	81 40.38%	626 20.23%
S-5	13167 6.35%	32401 0.64%	175 6.71%	46082 0.12%	328 9.95%	520 20.49%	127 40.18%	505 22.28%
S-6	9624 7.6%	54907 0.72%	324 9.55%	30624 0.16%	66 10.03%	194 16.23%	62 14.24%	240 21.99%
S-7	7926 5.41%	44719 0.69%	293 8.05%	65073 0.16%	104 11.00%	1402 35.41%	101 33.18%	906 25.03%
S-8	10678 8.13%	16124 0.59%	732 11.17%	40893 0.12%	49 11.02%	214 19.20%	64 40.20%	613 20.73%
S-9	12167 5.15%	79401 0.64%	1015 9.07%	63982 0.15%	98 10.55%	910 23.89%	227 36.58%	525 23.28%
S-10	7924 3.6%	43907 0.72%	934 9.15%	52720 0.16%	75 10.69%	1534 17.53%	75 17.14%	318 27.49%
S-11	5926 5.31%	45019 0.69%	505 7.35%	50073 0.13%	54 9.80%	672 34.43%	43 35.18%	766 25.73%
S-12	12608 10.53%	19124 0.59%	512 10.67%	41911 0.18%	55 10.92%	124 25.20%	51 32.98%	696 19.33%
S-13	7926 5.41%	46719 0.83%	1295 8.97%	63682 0.12%	78 8.35%	320 23.89%	97 37.26%	442 26.02%
S-14	11378 9.13%	15124 0.89%	354 9.95%	61730 0.15%	45 10.33%	94 16.33%	41 15.34%	201 27.99%
S-15	12267 6.15%	78451 0.75%	995 7.65%	70573 0.16%	50 9.80%	102 35.43%	171 33.15%	9416 24.53%
Mean	9706 ±626.73	36936 ±255.84	754.66 ±67.68	57391 ±83.02	98 ±10.11	586.4 ±143.12	105.93 ±38.90	1120 ±272.59

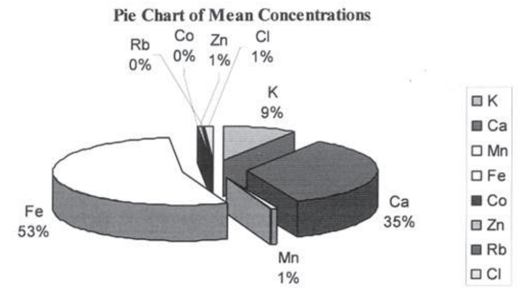

Fig.1. Percent-wise distribution of elemental concentrations

Table 3. Comparison of Elemental constituents/abundances of Soil samples in ppm of different countries with that of the present work.

Elements	Cl	K	Ca	Mn	Fe	Zn	Rb	Co	Reference study
Concentration in ppm	88.5	2976	371	129	32274	56.9	58.1	-	[9]
	-	37000	12000	70	46000	1300	-	-	[10]
	-	-	-	-	18000	62	-	63	[11]
	2521	884	16220	219	21040	-	-	-	[12]
		15000	24000	550	26000	60	67	9.1	[13]
	1120	9706	36936	754	57391	586	106	98	Present study

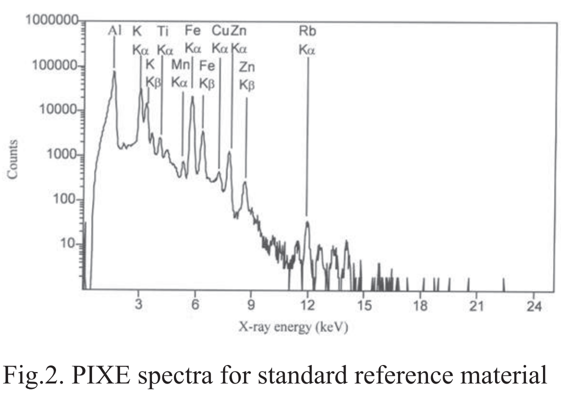

Fig.2. PIXE spectra for standard reference material

The average Cl content in soil as observed in the present work is 1120±272.59 ppm within the range of 201-9416 ppm. Another two studies found that the Cl content in soil are 2521 ppm [12] and 88.5 ppm [9] respectively, as shown in table 3and our obtained value of Cl concentration also falls within this range. The percentage of error varies between 19.33-28.99% and the Minimum Detection Limit (MDL) varies between112-238 ppm.

In the present soil study the concentration of K is dominant in most of the samples. Thus there is an adequate supply of K from the source to site. The average K observed in this study is 9706±626.73 ppm varying within 5926-13167 ppm. This result lies within the previous experimental values and close to 15000 ppm as mentioned in an earlier report [13]. The percentage of error varies between 3.6-10.53% and the MDL range is 486-1235 ppm, thus providing reliability in the K measurement.

The present study shows that average Ca content is 36936±255.84ppm (range 15124-79401 ppm) which is somewhat greater than the highest value of 24000 ppm as mentioned in the reference table 3[13]. The percentage of error varies between 0.59-0.89% and the MDL range is 392-940 ppm.

Mn has been observed to have the average 756.66±67.68 ppm within the range of 175-1695 ppm. The percentage of error varies between 6.71-11.17% and the MDL varies between 509-1460 ppm. It can be observed from table 3that the experimental result is more than the reference value, a study in U.S. [13].

The soil samples analyzed in this study were found to contain the average Fe concentration of 57391±83.02 ppm within the range of 30624-73782 ppm, which is also greater than the value as enlisted in the table 3. The percentage of error varies between 0.12-0.18% and the MDL varies between 288-1532 ppm.

Co has been observed to have the average 98±10.11 ppm within the range of 45-328 ppm. The percentage of error varies between 8.35-11.02% and the MDL varies between 3364-41543 ppm. From table 3, it can be observed that the experimental result is two times greater than the reference value.

In this study, the range of Zn concentration is found 94-1534 ppm with the average of 586.4± 143.12 ppm, which is inconsistent with the previous results as mentioned in table 3. The percentage of error varies between 15.73-36.45% and the MDL variation is in between 1074-8178 ppm. Although further study is necessary to reach a conclusion, probably, it may be considered that Zn concentration in soil varies place to place.

Rb has been observed to have the average 105.93±38.90 ppm within the range of 41-227 ppm. The percentage of error varies between 14.24-50.98% and the MDL varies between 743-73070 ppm. It can be observed from Table 3 that a study in U.S. observed the Rb content of 67 ppm [13].The experimental result is greater than the reference value.

4. Conclusions

Proton Induces X-ray Emission technique has been employed to examine soil samples to monitor some important trace elements. Since vegetables and food stuff are produced in soil so if soil is contaminated then it may affect human health. It is, therefore, necessary to have the information on the levels of both toxic and essential elements in soil. In the developed countries such analysis are done routinely. Keeping this in mind soil samples were collected from surrounding Goalundo Upazilla, and analyzed for major, trace and toxic elements present in those samples. The following order of decreasing concentration was observed: Fe> Ca> K> Cl>Mn> Zn>Rb>Co. The measured values of trace elements are compared with the certified values and found to be compatible. So far, no toxic elements such as As, Cd, Pb, etc were detected. Except Cl, K and Zn all other elements that are Ca, Mn, Fe, Co and Rb are found to be a high concentration with respect to reference study shown in Table 3, that may transfer to food stuff and consequently may affect the human health. Since the experimental results are interesting, further study is recommended.

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