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2015

# Elemental Analysis of Some Selected Environmental Samples and Their Effect on Public Health

Bari, A. S. M. Shohidul

University of Rajshahi

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**Elemental Analysis of Some Selected Environmental Samples and  
Their Effect on Public Health**



*A Thesis*  
*Submitted to the Institute of Environmental Science (IES)*  
*University of Rajshahi*  
*for*  
*the Degree of*  
**DOCTOR OF PHILOSOPHY**  
**IN**  
**ENVIRONMENTAL SCIENCE**

**BY**  
**A. S. M. Shohidul Bari**

**INSTITUTE OF ENVIRONMENTAL SCIENCE**  
**UNIVERSITY OF RAJSHAHI**  
**RAJSHAHI-6205**  
**BANGLADESH**

**SEPTEMBER**  
**2015**

**DECLARATION**

I hereby certify that the work presented in this entitled “Elementary Analysis of Some Selected Environmental Samples and Their Effects on Public Health” in fulfillment of the requirements for the award of the Degree of Doctor of Philosophy in Environmental Science submitted to the Institute of Environmental Science, University of Rajshahi, Bangladesh is an authentic record of my own work under the supervision of Dr. Md. Redwanur Rahman, Associate Professor, Institute of Environmental Science, University of Rajshahi. The work embodied in thesis has not been submitted for the award of any other degree.

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## CERTIFICATE

The thesis entitled “Elementary Analysis of Some Selected Environmental Samples and Their Effects on Public Health” Submitted by A. S. M. Shohidul Bari, embodies the result of research carried out by him under my direct supervision and guidance. I certify that this work has not been presented for any degree or prize elsewhere.

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## ABSTRACT

Particle Induced X-ray Emission (PIXE) and Particle Induced  $\gamma$ -ray Emission (PIGE) technique have been employed to perform essential and trace elemental analysis of some environmentally important samples including water, soil, fruits and vegetables from Rajshahi district of north-western part of Bangladesh by Tandem accelerator.

Fluorine (F) has attracted serious attention in recent years because of the apparent role it plays in human health. The F content of ground water in the study area lies in the range of 0.96 to 2.01mg/L with a mean of 1.44 mg/L in monsoon season and winter season vary from 1.05 to 2.45mg/L with a mean 1.27mg/L. The maximum recommended value for Bangladesh being 1.00mg/L. The study present that 80% of the samples contain higher F than the recommended maximum which indicated the probability of F problem in drinking water for the population of Rajshahi region of Bangladesh. To find the source of F in ground water soil samples were analyzed. F content of soil in the study area lies in the range of 243 to 415mg/Kg with a mean of 312.5mg/Kg in monsoon season and concentration vary 200 to 401mg/Kg in winter season. Most of the investigated soil samples have values for F content above 300mg/kg, which is maximum permissible value for the content of this element in agriculture soil. It indicates that the F pollution of ground water may be caused by F bearing minerals. To know the F uptake by the population of the study area vegetables and fruits samples were analyzed but no samples contain F. So, it can be concluded that vegetables and fruits are not a significant source of F uptake of the population of this region.

Manganese (Mn) content of ground water in the study area lies in the range of 0.11 to 0.33 mg/L in monsoon season and 0.23 to 0.58 mg/L in winter season. It shows that about 100% of the surveyed samples have Mn concentrations exceeding the Bangladesh drinking water standard of 0.1 mg/L but do not exceed the WHO health-based guideline value of 0.4 mg/L.

Iron (Fe) content of ground water in the study area lies in the range of 0.95 to 2.03mg/L in monsoon season and 0.86 to 1.85 mg/L in winter season. The Bangladesh standard value being 1 mg/L and WHO guideline value 0.3mg/L. From the present study it is found that 80% of the studied samples contain higher Fe than the recommended maximum.

Calcium (Ca) content of ground water in the study area lies in the range of 80 to 115mg/L in monsoon season 89 to 120mg/L in winter season. The standard WHO guideline value 75mg/L. From the present study it is found that 100% of the studied samples contain higher Ca than the recommended maximum

Copper (Cu) content of ground water in the study area lies in the range of 0.29 to 1.1mg/L in monsoon and 0.49 to 1.13mg/L in winter season. The Bangladesh standard value being 1.3mg/L and WHO guideline value 1 mg/L. From the present study it is found that 100% of the studied samples contain lower concentration of Cu than the recommended maximum.

Zinc (Zn) content of ground water in the study area lies in the range of 1.20 to 4.52mg/L in monsoon and concentration vary from 2.20 to 4.25mg/L in winter season. The WHO guideline value 5.0mg/L. From the present study it is found that 100% of the studied samples contain lower Zn than the recommended maximum.

Soil samples of the study area were found to be rich in Cu, Mg, Mn, Ca and B but deficient in K, P and Zn. In the present study P was found in only two samples (567 & 409 mg/kg in monsoon season and 560 & 398 mg/kg in winter season) out of ten samples which are much lower than the optimum value (800 mg/kg). Concentration of K varies from 47 to 245 mg/kg in monsoon season and 55 to 180 mg/kg in winter season which is lower than the global range (78-273 mg/kg). So, P enriched fertilizer and potash fertilizer should be used in the study area and the farmer of this area should be made aware about the P and K deficiency of their lands. Soil samples of the study area are also found to be poor in Zn. Zinc-deficient soils can be easily treated with zinc fertilizers to provide an adequate supply of Zn to crops. Several different zinc compounds are utilized as fertilizers but zinc sulphate is by far the most widely used.

PIXE technique has been employed to perform essential and trace elemental analysis of Zn, Ca, Fe, K, Cl, Cu, Mn, Br in some fruits and vegetables of Rajshahi region of Bangladesh. The selected fruits and vegetables are commonly consumed by all income groups of the locality. The concentration of the elements in each fruit and vegetable and their role for the nutrition of human being are discussed. It is clear that K and Ca are the most abundant detected elements in the samples. It is observed that in fruits Guava (*Psidium guajava*) contains the highest amount of Ca (66800mg/kg) and Carambola (*Averrhoa carambola*) contain highest of K (28950mg/kg). In vegetables highest amount of Ca and K are exhibited by Turnip (*Brassica rapa*) (30541mg/kg) and Potato (*Solanum tuberosum*) (31098mg/kg) respectively. The Fe content fairly good in all fruit samples (varying from 250.30mg/kg in Amla (*Emblica officinalis*) to 545mg/kg in Papaya (*Cavica Papaya*)) and all vegetables samples (varying from 319 mg/kg in Bottle Gourd (*Lagenaria siceraria*) to 9230 mg/kg in Carrot (*Daucus carota*)). Zn content in the studied samples is highest in Jackfruit (*Artocarpus heterophyllus*) (65mg/kg) in fruit and Carrot (*Daucus carota*) 110.05mg/kg in vegetables. Mn is found in all fruits and vegetables samples. Among them in vegetables Carrot (*Daucus carota*) contains highest amount (10mg/kg) of Mn and in fruits Olive contain highest amount (9mg/kg) of Mn. Copper content in the studied samples is found only in Pumpkin (*Cucurbita moschata*) (26.87mg/kg) in vegetables and Carambola (*Averrhoa carambola*) (20.56mg/kg) in fruit which probably plays a role in prevention of inflammation, given that Cu is a useful anti-inflammatory agent. Highest Cl content is found in vegetables in Potato (*Solanum tuberosum*) (7890mg/kg) and Carambola (*Averrhoa carambola*) (4567mg/kg) in fruit. The minimum chloride requirement for a healthy adult is 700mg/day. The deficiency of chlorine causes anorexia, weakness, growth failure, severe convulsions. Bromine only found in tomato (*Lycopersicon esculentum*) (118mg/kg) in vegetables. No toxic heavy metals were detected.

The data from this study provide a preliminary assessment of the background levels of selected trace and essential elements of Rajshahi district. Therefore such studies will help us to understand the environmental pollution and its effects on human

health. In the developed countries such analysis are done routinely to ensure the good health and quality of life of their citizens.



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## ABBREVIATIONS

Abbreviation	Meaning
PIGE	Proton Induced Gamma Emission
PIXE	Proton Induced X-ray Emission
0°C	Degree Centigrade
<i>e.g.</i>	<i>example graita</i> (for example)
BAEC	Bangladesh Atomic Energy Commission
AERE	Atomic Energy Research Establishment
App.	Approximate
cm	Centimeter
gm or g	Gram
L	Liter
IAEA	International Atomic Energy Agency
kg	Kilogram
mg	Milligram
TA	Tandem Accelerator
ppm	Parts per million
ppb	Parts per billion
Conc.	Concentration
IBA	Ion Beam Analysis
RBS	Rutherford Backscattering Spectrometry
WHO	World Health Organization
EPA	Environmental Protection Agency
MCL	Maximum Contaminant Levels

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# REVIEW OF LITERATURE

## REVIEW OF LITERATURE

The main objective behind the literature review is to avoid duplication of previous research works. On the other hand, knowledge of previous research work done in the related areas of the study helps in developing ideas of interests and enhancing the researcher's knowledge. In carrying out this study, the works of other scholars have also provided the insights and background. Some relevant works done in home and abroad are discussed as follows:

Jelliffe and Patrice, 1979 have reported the proper feeding of the mother during pregnancy and lactation- preferably with locally available foods- will optimize the volume and composition of breast milk (both as the sole food for the young baby and as a supplement for the transitional). This approach will avoid the economic, infective and distributive complexities of introducing cow's milk and bottle-feeding unnecessarily, and will also help to retard the decline in breast-feeding on a community basis.

Hyvonen-Dabek, 1981 have used PIGE method for determining the light elements in the human bone samples. The mean concentration for the fifteen dense bone samples were found to contain boron 8 mg/kg, fluorine 639 mg/kg and sodium 5763 mg/km, *etc.*

Hall and Navon, 1986 have reported a PIXE- PIGE analysis scheme for simultaneous multi-elemental analysis of lithium through uranium in biological samples. The method has been applied to analyze routinely lithium-uranium in human colostrums, spermatozoa, follicular fluids, teeth, bone, fetal tissues, tree-rings and lichen. The best sensitivity obtained with PIGE was for sodium, fluorine and lithium where the

MDLs were less than 0.07ppm. Boron, magnesium (Mg<sup>25</sup>) and aluminium have sensitivities less than 10ppm. For silicon (Si<sup>28</sup>), phosphorus and nitrogen the detection limits are less than 50ppm while carbon and oxygen have sensitivities less than 1.5%. Analysis of human spermatozoa, where sample size are often less than 5mg, allows to ascertain the detail of the contributions of multi-elements (essential and toxic) in the sperm quality and fertility. Analysis of human colostrums allows one to determine the maternal to infant transfer of essential and toxic elements.

Zschau *et al.*, 1990 have reported the profiling of fluorine in the mild teeth using PIGE. They have reported that the relatively high amount of F in the mild teeth results from the environment, especially from drinking water and food stuffs. Zschau has tested the carries preventing gel together with Elmex on children and have reported that the application of each of the F containing gel led to distinct fluorine enrichment in teeth within the depth of interest.

Balogun *et al.*, 1994 have been employed Instrumental Neutron Activation Analysis (INAA) and Proton Induced X-ray Emission (PIXE) analysis to determine the concentration of 13 elements in human breast milk, various infant formulas, and locally produced cereals from Nigeria, as well as from various infant formulas and natural cow and goat milk available in the UK. The study shows that if the locally produced cereal is to be used on a regular basis for babies in Nigeria, then their diet must be supplemented with essential trace elements. Furthermore, parents should be discouraged from giving their infant's cow and goat milk because of the high concentration of major elements compared to human breast milk

Olabanji *et al.*, 1996 have carried out a qualitative and quantitative determination of the elemental composition of human breast milk in

Nigeria with a view to identifying the deficiencies of the essential elements and the presence of toxic elements in them. Determination of the elemental concentration of human milk constitutes may reveal the kind of nutrients that are passed from mothers to babies. The elements that were determined using PIGE techniques are sodium, magnesium, aluminum and chlorine. All of these elements are essential for the growth and development of babies and it has been found that most of the samples contain these elements at a beneficial level.

Chen *et al.*, 2000 have reported the measurements of P, K, Fe, Cu, Zn in soil from South Florida and found the concentration 256, 212, 5100, 2800 and 60.3 mg/kg respectively, which were 2.6 to 15.8 times greater than those of Florida soils

Rodriguez *et al.*, 2000 determined the concentrations of iron, copper and zinc in 56 samples of mature human milk from Canadian women and 5 samples of powdered infant formula. The mean concentration of Fe, Cu and Zn of powdered infant formula was significantly higher than those concentrations found in the human milks. Significant differences among the concentrations of the studied metals for the milks of considered mothers were observed. The Fe, Cu and Zn intakes of infants fed with human milk are lower than the requirements recommended by the Food and Nutrition Board (1989). However, the infants fed with powdered infant formula had consumed an adequate intake of Fe and Cu. A progressive decrease of the metal concentrations with the lactation stage was observed. The human milk obtained in spring presented Fe and Zn concentrations lower than in autumn, which could be due to changes in nutritional habits of the mothers. Age of

mother and number of previous children seem to influence the Zn and Cu concentrations of human milk.

Alam *et al.*, 2003 reported the measurements of fluorine in many vegetables and food samples by PIGE technique. They produced 197 keV gamma ray in F with proton beam energies in the range of 2.8 to 3.5 Me V and the Sensitivity which, they obtained was less than 1 ppm.

Alloway *et al.*, 2003 in this study they analysis Occurrence of manganese in groundwater of Bangladesh and its implications on safe water supply. Data obtained from the national hydro-chemical survey show that about 42% of tube wells have manganese concentrations exceeding the WHO health-based guideline value of 0.4 mg/l. High manganese concentrations in groundwater have been found in the central, northern, and western regions of Bangladesh; groundwater in the north-eastern region of Bangladesh contain relatively less manganese. Deeper wells (>150m) have been found to contain relatively lower concentrations of manganese. Distributions of arsenic and manganese concentrations are not similar in groundwater of Bangladesh. Areas with low arsenic in groundwater have been found to contain high manganese concentrations, and vice versa. Nationwide about 32% of wells, which contain safe level of arsenic (*i.e.*, < 0.05 mg/l) have been found to contain unsafe level of manganese (*i.e.*, > 0.4 mg/l). This would significantly increase the population exposed to unsafe water, beyond that estimated for arsenic alone. Detection of high concentrations of manganese in groundwater has added a new dimension to the already difficult safe water supply scenario in Bangladesh. However, manganese issue has attracted relatively less attention so far in the water supply sector. Currently iron and arsenic-

iron removal plants are being used in many regions of the country. In view of the widespread presence of manganese in groundwater in addition to arsenic and iron, it is important to raise awareness among the stakeholders about the manganese issue. It is also very important to identify areas unacceptable levels of arsenic and/or manganese and to develop water treatment technologies accordingly.

Akpanyung, 2006 analyzed six commercial brands of powdered milk produced in Nigeria for their major and trace element composition. The products were found to be good source of mineral nutrients except for Sodium. This paper suggests the need for regular intake of this brand of processed milk so as to derive optimal benefits from its reservoir of major and trace elements.

Khatir *et al.*, 2006 studied on protein content and the status of elemental composition in human milk from Sudanese subjects. The protein content was derived by multiplying the nitrogen content by a factor 6.25. The nitrogen was determined using a 14 MeV neutron generator. The median values for crude protein and the total dry matter found in this study were 1.23% (volume) and 104 g/l, respectively. Some minor and trace elements of biological significance namely, Cl, K, Ca, Mn, Fe, Cu, Zn, Cr, Co and Mo were determined using energy dispersive X-ray fluorescence spectrometry. The results obtained show good compatibility with the data reported by the WHO on elemental composition of human milk from different geographical regions.

Farid and Kinsara, 2007 have studied the concentrations of human milk and cow's milk samples available in and around Jeddah city. The experiment was done using atomic Absorption Spectrometer. The mean concentrations of Zn, Cu, and Fe in human milk are higher than the



corresponding values in cow's milk while the mean concentrations of Cr, Mn, Pb and Cd in human milk are lower than the corresponding values in cow's milk. The concentrations of all elements in human milk collected in the afternoon are higher than those in samples collected in the morning. The experimental results show that there is an apparent decline in the mean elemental concentration levels as the stage of lactation progressed.

Antoaneta *et al.*, 2009 worked on Particle-Induced X-ray Emission (PIXE) technique for the determination of minor constituents of some metallurgical and environmental samples has been done. The minor elements identified in the metallurgical samples (steels) using PIXE were: K, Ca, V, Cr, Mn, Fe, Co, Cu, Ni, Zn, W, Ga, As, Pb, Mo, Rb, In, Rh, Zr, Pd, Nb, Sn and Sb. In the investigated environmental samples (vegetal leaves, soil and mosses) PIXE analysis allowed determination of S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Sr, Se, Br, Hg and Pb in concentrations between 0.05-290 mg/kg.

Ahmed *et al.*, 2009 analyzed soil and crops samples from then regularly cultivated agriculture fields from Sonarga of Bangladesh using PIXE method. The elements measured in crops were K, Ca, Ti, V, Fe, Zn, As, Pb *etc.* Vegetables have shown the presence of all the elements above their detection limits. They found the deficiency of Cu in some crops and level of Co in soil were found to be the same as in vegetables.

Adnan *et al.*, 2011 recoded the heavy metals (Ni, Cu, Cd, Cr and Pb) in agricultural soils and their uptake in spring seasonal plants being irrigated by industrial waste water. For this purpose three periurban agricultural areas of Central Punjab Province were selected where irrigation is usually done by waste water or industrial sewage. Ten samples each of soil, waste water and plants were collected from the selected areas and were analyzed for pH, EC, Cr, Cd, Pb, Cu, and Ni by pH meter, Conductivity meter and atomic absorption spectrophotometer respectively. The results were compared with international standards. The spring seasonal plants did

not show any sign of contamination regarding the selected metals, except chromium. This may be due to the higher pH and fine texture of soils which do not support the uptake of selected metals to plants, making the plants safe for consumption.

Princewill *et al.*, 2011 determined the concentration of five heavy metals zinc (Zn), iron (Fe), manganese (Mn), chromium (Cr) and cadmium (Cd) in soil and woody plants species (*Azadiractha indica*, *Mangifera indica*, *Andropogon gayanus*, *Uvarea chamae* and *Gmelina arborea*) at NIGERCEM, Nigeria. Soil samples were collected at 40, 80, 120, 160 and 500m sampling positions. The highest soil concentrations of Zn (212.40 mg/kg), Fe (186.30 mg/kg), and Mn (68.10 mg/kg) in soil were obtained at 40m while Cd (3.02 and 2.96 mg/kg) and Cr (26.0 and 24.68 mg/kg) concentrations in soil was statistically equal ( $P > 0.05$ ) at 40 and 80 m. In plants, the highest concentrations of Zn (18.48mg/kg) and Fe (33.61mg/kg) were observed in *Andropogon gayanus*; Cd (0.13 mg/kg) was in *Mangifera indica*; while Cr (2.152mg/kg) and Mn (7.27mg/kg) was in *Azadiractha indica*. The concentrations of heavy metals in soil and plants were below toxicity level except for Cd in soil. There were significantly positive correlations between heavy metals in soils and plants.

Chiroma *et al.*, 2012 reported the content of Cu, Zn, Pb, Fe and Cr in Bushgreen and Roselle vegetable plants, soil irrigated with treated and untreated urban sewage water were evaluated using atomic absorption spectrophotometer. The concentration of Cu (0.078 µg/ml), Zn (1.065 µg/ml), Pb (1.034µg/ml), Fe (2.512µg/ml) and Cr (0.081µg/ml) in untreated sewage water were reduced by 58%, 46%, 27%, 70% and 33% respectively, after treatment with Alum. The mean concentration of Pb, Cu and Cr in treated and untreated sewage waters are above the maximum permissible values of 0.2µg/ml, 0.01µg/ml and 0.05µg/ml respectively for irrigation waters used on all types of soils. The levels of Zn in soils irrigated with sewage water and Pb in soils irrigated with

treated sewage water are above the maximum tolerable levels of 300 $\mu\text{g/g}$  and 100 $\mu\text{g/g}$  respectively. The contamination of Zn and Cr in leaves (unwashed), leaves (washed), stem and roots of Bush green irrigated with sewage water are 2.5, 1.7, 1.3, 3.2 and 2.1, 1.7, 1.9 and 1.1 times respectively higher than the maximum permissible level in plants sets by World Health Organization (WHO).

Bahrampoo, 2012 study heavy metal contamination of soil and plants was widely studied in different countries of the world. According to the conducted studies, heavy metals enter the food chain through uptake and accumulation in soil and water has negative influences on physiological and metabolic activities of living organisms. Principally the first step in evaluation and measurement of pollutants is identification of their sources. Therefore, this study was conducted during 2008 in Moghan, Iran, with 131 samples of arable land, non-arable land and irrigation water of 9 places in Moghan. Soil samples were dried and sieved in laboratory. Then, prepared samples were extracted and determined their concentration of Cu, Zn, Fe, Mn, Pb, Cd and Ni using Atomic Absorption Spectrometer. The results showed that there was statistically significant difference ( $p=0.01$ ) between arable land, non-arable land of different areas in term of metals mentioned. Concentration of heavy metals was more in arable lands than non-arable lands. The accumulation of Cu, Zn, Fe, Mn, Pb, Cd and Ni in arable lands was 5, 3.5, 10, 14, 3, 20, 5 times more than those of non-arable lands, respectively. It can conclude that increasing pollution and decreasing soil fertility may be attributed to irregular use of pesticides and fertilizers on arable lands and the use of contaminated irrigation water.

Having reviewed the literature, we conclude that no study has covered the study of essential and toxic elements in soil, water, vegetables and fruits by PIXE technique and its effects on human health. So, the current research work will fill up these gaps.

# CHAPTER ONE

## INTRODUCTION

## INTRODUCTION

### 1.1 INTRODUCTION

The development of analytical methods has led to detect elements in very low concentrations (Keen *et al.*, 2003). The use of ion beam analysis techniques (IBA) (Anderson *et al.*, 2008) has grown recently since they combine the advantages of being non-destructive and multielemental analytical techniques, they can be used singly or in combination to obtain more information about the system being studied. Fruit and vegetable materials are well suited to PIXE analysis owing to its great advantages. Ali *et al.*, 1985 and Johansson and Campbell, 1998 by making use of the high resolution Si(Li) photon detector and using MeV proton beams, showed that PIXE is highly sensitive method for the multielemental analysis. They demonstrate that many elements could be detected simultaneously at the 10-12g level. PIXE has the advantage over other IBA techniques that the cross-section for X-ray production is large and the background contribution from bremsstrahlung is low. Most important minor and trace elements can be determined basically in one single run in large number of samples during, relatively, short time. In addition, it is very easy to prepare the samples for analysis (i.e. it is sufficient to dry the sample and mount it in the target chamber). The use of PIXE technique in the analysis of fruit and vegetable materials has been quite few in the past due to its expensive operation. However, the spread of ion beam accelerators around the world and the recent development of IBA techniques have encouraged researchers to employ these powerful techniques in different analytical applications, including the

investigation of fruits and vegetables. The IBA techniques have been utilized successfully in the past ten years to perform elemental analysis in medicinal plants (Manuel *et al.*, 2008). Environment and development are the two sides of a coin. Developmental activities e.g. industrial, agricultural, transportation, constructional work, etc., cause degradation and drastic changes in every component of environment namely, hydrosphere (water), lithosphere (soil) and atmosphere (air) through pollution. In order to recognize and predict hazardous effects of pollutants on biosphere, the monitoring of these environmental components are essential. For this purpose, the quantitative characterization of water, soil, air and vegetation is a prerequisite. It will give us an idea about the gravity of the pollution problem in the different environmental components. This, in its turn, would help us to focus our attention to pollution control 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.

The effect of an ecological factor on plant life can be direct or indirect or remote. The direct environmental factors are those which influence the plants growth and their geographical distribution. Soil, water and soil minerals fall into this category (Underwood, 1997).

Human activities that disturb the conditions suitable for sustaining life, cause pollution. That is any undesirable change in the physical, chemical or bio-logical characteristics of soil, water and air that may create a hazard to health, safety or welfare of any living species is called pollution. In other words, pollution means the direct or indirect change in one or more components of the biosphere, that are harmful to

living entities. In a broad sense pollution means the presence of any substance (solid, liquid or gas) or agent in the atmosphere in such a concentration that may be injurious to environment affecting the living or nonliving things such as our natural cultural assets. Population explosion along with the rapid urbanization and expeditious industrialization greatly aggravated the environmental pollution (Ananda *et al.*, 1976).

Prior to the agricultural and industrial revolutions, the main fertilizers applied to the land were animal products, such as cow dung, blood and bone meal, farmyard manure human faces, but seaweed was also applied in coastal areas. A large fraction of trace elements removed from the soil was thus returned and the rate of depletion was relatively slow. The system of agricultural production was therefore, to a large extent, self-regenerating, in the sense that the balance between the major and minor nutrients were maintained naturally and proportionately. Although the problem of dispersion of non-essential elements did not exist, soil fertility by modern standards was sustained by this system only at a low level (Miller and Donahue, 1997).

The nineteenth century was the start of the final break in the United Kingdom with this age-old system of production. The need to increase agriculture productivity to meet the demands of the population explosion was met by supplementing home produced organic fertilizers by importing mineral salts from the stassfruit deposits in East Germany and rock phosphate for the manufacture of superphosphate fertilizers. Large quantities of South American deposits, derived from bird and bat faeces known as guano, were also imported. Global human population is rapidly increasing. In order to satisfy the basic human need of the



growing population increasing amounts of resources are required. Besides food, the majority of other human needs can only be met with goods and services produced by industry. Industry has the capacity to improve as well as to degrade the environment. Industry converts raw materials, which have been taken from the natural resource base, into products. During the production process it often releases pollutants to the environment. The negative effects of industrial activity on the environment are apparent in water, air, and soil pollution. The industrial development in Bangladesh is taking place without giving proper consideration given to its negative impacts on the environmental components such as air, water and soil (Walter, 1961).

The effect of an element on human body is concentration dependent. Essential elements can become toxic if ingested or inhaled at a sufficiently high level and for a long enough period (Mireles *et al.*, 2004). The knowledge of elemental contents of soil, water, foods, vegetables, fruits, drinks, etc. are important, because these are the primary sources of mineral for all living beings and many diseases are caused in men and animals due to mineral deficiency. The possibility that contamination of water supply and food items with trace elements, arising from geological bearing minerals, modern agriculture and industrial practices may have detrimental effects on the long term health and welfare of the human population. The nature and extent of the of the actual and potential deficiencies and toxicities of this sort shall have to be considered seriously. An essential requirement of such a comprehensive study is to primarily quantify the elements of interest in different components of the environmental system.

Recently some serious environmental problems are encountered in Bangladesh related to the presence of some toxic elements like arsenic in ground water and lead in air. Physicians have found lead in bloods of some babies in Dhaka Shishu Hospital and Zinc deficiency in arable soil has also become a serious problem.

Elemental analysis of any material from its physical, chemical, biological or environmental origin provides a holistic view about the intrinsic quality of the material to be used and / or consumed in different sectors of national economy, sustainable for desired quality of life.

In the present day of global economic activity, and the global concerns of environmental degradation with ever increasing industrialization and introduction of newer practices in agricultural developments, the need for ensuring material qualities in relation to economic developments and human health, has become an important issue. For such an issue to be addressed in right perspective, what is essential is to perform chemical compositional analysis of any material does fulfill the primary requirement to study the properties of a material, whenever it is a product from an industrial manufacturing process or environmental and biological specimens.

The chemical elements in any material are grouped as light and heavy elements in terms of mass and major, minor and trace components and in terms of concentrations. In any physical, some of the elements are considered as impurities at very low concentrations but are important to control its properties, for example Mo shows super-conducting transition subject to purification and the transition temperature has been found to depend on the exact state of purity. In biological systems, some of the transition elements like Co, Ni, Cu, Zn, Fe, *etc.* are recognized to be essential for regulatory physiological functions at very trace concentrations such as 10<sup>-6</sup> g/kg, while the bulk elements like carbon, nitrogen, hydrogen, etc. constitute the body of the biological system.

In recent years the study of both major and trace elements, in biological, environmental and agricultural materials has assumed increasing importance

because of their vital role in human health and diseases, Trace elements such as Cu, Zn, Co, *etc.* may be an integral part of the enzyme-protein molecule or they may be activators of the enzyme. The essential biological trace metals are also involved in hormone, vitamins and protein, DNA and RNA synthesis. Any fluctuation like deficiency or excess in their normal level in living cells may lead to physiological disorders causing various disease like hypertension, dental carries, goiter, cancer, heart disease, gallstones, obesity, osteoporosis, Osteomalacia, arthritis, anemia *etc.* To ensure proper diagnosis and treatment planning, the information of elemental analysis of body fluids and tissues such as blood, hair, urine, nail *etc.* is essential. Growing population, intensification of agriculture, increase in trading industrial and other economic activities are affecting the quality of life in many different ways. As a result deterioration of the environment and its consequent effect on human health has emerged as a major concern all over the world. From the increasing of motorization, urbanization, modern agriculture and industrial practices the possibility of contamination of the soil, air, water, vegetables and foods with trace elements may have deleterious effects on the long term health and welfare of human population.

The major pathway of the elements to the human body is the food, drinks and air. So from the point of view of environmental safety assessment and health care, it is necessary to have the informant to the levels of both toxic and essential elements in food and drinks as well as the agricultural products and other environmental resources such as air , water, soil, *etc.*

The depletion of the soil nutrients and its effect on productivity is also a major concern in today's world. With the intensification of agriculture Zinc deficiency in soil has already become a problem in Bangladesh and this could happen for other trace elements. The elements like N, Zn, P, and B Play vital roles in the fertility of the land and this productivity. To ensure the yield and also the food value of the crops presence of proper nutrients of the soil should be ascertained.

## **1.2 Water Analysis**

### **1.2.1 Fluorine (F) in ground water**

F is an univalent poisonous gaseous halogen, it is pale yellow-green and it is the most chemically reactive and electronegative of all the elements. F readily forms compounds with most other elements, even with the noble gases krypton, xenon and radon. It is so reactive that glass, metals, and even water, as well as other substances, burn with a bright flame in a jet of F gas. In aqueous solution, F commonly occurs as the fluoride ion. Fluorides are compounds that combine fluoride with some positively charged counterpart.

### **1.2.2 Fluorine in the environment**

Annual world production of the mineral fluorite is around 4 million tonnes, and there are around 120 million tonnes of mineral reserves. The main mining areas for fluorite are China, Mexico and Western Europe. F occurs naturally in the earth's crust where it can be found in rocks, coal and clay. Fluorides are released into the air in wind-blown soil. F is the 13th most abundant element in the Earth's crust: 950 ppm are contained in it. Soils contain approximately 330 ppm of fluorine, ranging from 150 to 400 ppm. Some soils can have as much as 1000 ppm and contaminated soils have been found with 3500 ppm. Hydrogen fluorides can be released into air through combustion processes in the industry. Fluorides that are found in air will eventually drop onto land or into water. When fluorine is attached to very small particles it can remain in the air for a long period of time. In the atmosphere 0.6 ppb of F are present as salt spray and organic fluoride compounds. Up to 50 ppb has been recorded in city environments.

### **Zinc (Zn)**

Zn is an essential mineral that stimulates the activity of about 100 enzymes in the body. It also supports your healthy immune system, is necessary to synthesize DNA, is essential for wound healing, supports the healthy growth and development of the body during adolescence, childhood and pregnancy. Though the actual amount of Zn necessary to support the human body is quite small, its effects on the body are astronomical.

### **Sources of Zn**

Zn is present in a variety of foods that many people consume daily. The food with the most Zn per serving is oysters, but most Americans receive the greatest portion of their zinc intake from red meat and poultry. Some other food sources that contain Zn are some seafood, whole grains, fortified cereals, beans, nuts and dairy products.

The absorption of Zn tends to be higher in diets high in animal protein, as opposed to those rich in plant protein. An element present in whole grains, breads, cereals and legumes called phytate can also work to decrease Zn absorption.

### **Zn Deficiencies**

Generally, when someone is suffering from a Zn deficiency, it is because the intake is inadequate, because it is being poorly absorbed into the body or their need for zinc increases.

Zn deficiency can be identified by: growth retardation, diarrhea, hair loss, delayed sexual maturation and impotence, loss of appetite, eye and skin lesions, white spots on the fingernails

### **1.3 Soil Analysis**

All soils provide the same basic growth factors to plants: support, oxygen for roots, adequate temperatures, water, and nutrients. But some soils do a better job than others in providing the optimum amounts of these growth factors. The number of elements considered essential for the growth of higher plants now varies from 16 to 20 or more, depending upon the definition of essentiality. Plants require 16 nutrients. Of these 16 nutrients, 13 are supplied from the soil. Carbon and Oxygen are supplied from carbon dioxide and oxygen in the air; and hydrogen is derived from water (Miller and Donahue, 1997).

#### **1.3.1 Balancing the soil's nutrients**

Fertilizing crops usually has as its major purpose to produce better and/or larger yields, often at decreasing cost per unit of production. To accomplish these goals, it is essential to know how much of the critical nutrients the crop will need, The amount of nutrients the soil will supply subtracted from the total plant needs and losses is the amount of fertilizer to add. The "excess" fertilizer to add to accommodate the losses is difficult to calculate. Although the losses may not be known, the total amount to be added for a certain response in a given field is measured in the field trials used for correlation to yields.

When a soil is under cultivation, losses of major elements caused by crop removal, chemical fixation, drainage to waterways and leaching down the soil profile, are normally replaced by regular liming, which replaces calcium and the application of synthetic compound fertilizers containing nitrogen, phosphorus and potassium. Normally, no attempt is made to replace corresponding losses with the majority of soil, which contains substantial total reserves of essential trace elements.

### 1.3.2 Environmental effects of fluorine

When fluorine from the air ends up in water it will settle into the sediment. When it ends up in soils, fluorine will become strongly attached to soil particles. In the environment fluorine cannot be destroyed; it can only change form. Fluorine that is located in soils may accumulate in plants. The amount of uptake by plants depends upon the type of plant and the type of soil and the amount and type of fluorine found in the soil. With plants that are sensitive for fluorine exposure even low concentrations of fluorine can cause leaf damage and a decline in growth. Too much fluoride, whether taken in forms the soil by roots, or adsorbed from the atmosphere by the leaves, retards the growth of plants and reduces crop yields. Animals that eat fluorine-containing plants may accumulate large amounts of fluorine in their bodies. Fluorine primarily accumulates in bones. Consequently, animals that are exposed to high concentrations of fluorine suffer from dental decay and bone degradation. Too much fluorine can also cause the uptake of food from the paunch to decline and it can disturb the development of claws. Finally, it can cause low birth-weights.

### 1.3.3 Trace-element contamination of soils

Plants and the animals are dependent on the soil for their supply of nitrogen and mineral elements; their internal biochemistry and associated composition is therefore a reflection of the composition of the soil. In view of the rate at which the chemical environment is at present being altered, it has become a matter of some importance to determine the composition of existing uncontaminated soils with respect to their nutritionally significant components. Atmospheric pollution is now having global effects on the composition of soils, and soils which can reasonably be described as uncontaminated will be progressively more difficult to find.

### **1.3.4 Concentration of Fluorine (F), Phosphorus (P), Copper (Cu), Manganese (Mn), Potassium (K), Calcium (Ca), Zinc (Zn), Boron (B)**

Fluorine is a widely distributed element and a member of the halogen family. It is naturally found in the rocks, coal, clay, and soil. In air it is found in the form of hydro fluoride gas. During rainy season this gas mixes with water and turns into hydrofluoric acid. In water it reacts with some of the other elements present in water and turns into salts and gets deposited on the soil as sediments. Some times approximately 1ppm fluorine in the form of fluorides is added to drinking water supplies. In turn this added fluorine increases its concentration in soil. Use of fluorine in toothpaste also increases its concentration in the soil around the mega cities. Use of fluorine and its compound in pharmaceutical industries, ceramic factories, nuclear power plants etc. is a major cause of its increase in concentration in the soil around these installations. This high concentration of fluorine affects all of the life forms in the soil. So periodically measurement and control of the concentration of fluorine is very important to avoid both biological and environmental damage.

Several analytical techniques are used to measure the concentration of trace elements like fluorine, chlorine, bromine etc. PIGE technique is suitable for the measurement of the concentration of fluorine because of its low atomic number. This method is based on the prompt gamma rays emitted from excited nuclei of the elements following the charged particle irradiation of the sample.

Phosphorus is the second key plant nutrient; it is the second most often deficient nutrient. Phosphorus is an essential part of nucleoproteins in the cell nuclei; these molecules carry the inheritance characteristics of living organisms. Phosphate to phosphate bonds are the major energy storage and energy transfer bonds as ATP and ADP. In its many compounds phosphorus has roles in cell division, in stimulation of early root growth, in hastening plant maturity; in energy transformations within the cells, and in fruiting and seed production. For people and other animals eating the plants, phosphorus is critical for growth of 'bones and teeth, which ate mostly calcium phosphates. Plants differ in their ability to compete for soil phosphorus. Young plant absorbs phosphorus rapidly, if it is available. Winter wheat absorbs about 70% of its phosphorus between tailoring and flowering. A cold, wet spring usually results in a retardation of plant growth, often



because of inadequate phosphorus absorption. The peak phosphorus demand for corn is just 3 weeks into the growing season, when the root system is still relatively small (Miller and Donahue, 1997) .

Large proportion of P present, ranging from 0.2 to 0.8% of the total dry substance in the plants, are found in the fruits and seeds. P is abundant in the meristematic regions of actively growing plants. Large quantities of P are taken up by the plants during the maturation of seeds. The increased demand for P during the flowering time is met by applying phosphate fertilizer to the soil near the base of the plant.

Cu is a very common substance that occurs naturally in the environment and spreads through the environment through natural phenomena. Humans widely use copper. For instance it is applied in the industries and in agriculture. The production of Cu has lifted over the last decades. Due to this, Cu quantities in the environment have increased.

The world's Cu production is still rising. This basically means that more and more Cu ends up in the environment. Rivers are depositing sludge on their banks that is contaminated with Cu, due to the disposal of Cu -containing wastewater. Copper enters the air, mainly through release during the combustion of fossil fuels. Copper in air will remain there for an eminent period of time, before it settles when it starts to rain. It will then end up mainly in soils. As a result soils may also contain large quantities of Cu after Cu from the air has settled.

Cu can be released into the environment by both natural sources and human activities. Examples of natural sources are wind-blown dust, decaying vegetation, forest fires and sea spray. A few examples of human activities that contribute to Cu release have already been named. Other examples are mining, metal production, wood production and phosphate fertilizer production. Because Cu is released both naturally and through human activity it is very widespread in the environment. Cu is often found near mines, industrial settings, landfills and waste disposals.

When Cu ends up in soil it strongly attaches to organic matter and minerals. As a result it does not travel very far after release and it hardly ever enters groundwater. In surface water Cu can travel great distances, either suspended on sludge particles or as free ions.

Cu does not break down in the environment and because of that it can accumulate in plants and animals when it is found in soils. On copper-rich soils only a limited number of plants have a chance of survival. That is why there is not much plant diversity near Cu -disposing factories. Due to the effects upon plants copper is a serious threat to the productions of farmlands. Cu can seriously influence the proceedings of certain farmlands, depending upon the acidity of the soil and the presence of organic matter. Despite of this, Cu -containing manures are still applied.

Cu can interrupt the activity in soils, as it negatively influences the activity of microorganisms and earthworms. The decomposition of organic matter may seriously slow down because of this. When the soils of farmland are polluted with copper, animals will absorb concentrations that are damaging to their health. Mainly sheep suffer a great deal from copper poisoning, because the effects of copper are manifesting at fairly low concentrations.

The behavior of Mn in soil is very complex and is controlled by different environmental factors, of which pH-Eh conditions are the most important Under cold climate conditions, Mn is removed from the weathering zone and soil by acid solutions as bicarbonate or as a complex with organic acids derived from decaying plants. Mn occurs in soils and minerals mainly in the forms of  $Mn^{2+}$ ,  $Mn^{3+}$ , and  $Mn^{4+}$  (McBride, 1994; Kabata-Pendias, 2001) but only  $Mn^{2+}$  is absorbed by plants (Tisdale *et al.*, 1985).

K is an essential plant nutrient and is required in large amounts for proper growth and reproduction of plants. K is considered second only to nitrogen, when it comes to nutrients needed by plants, and is commonly considered as the “quality nutrient.”

It affects the plant shape, size, color, taste and other measurements attributed to healthy produce.

K has many different roles in plants:

In Photosynthesis, K regulates the opening and closing of stomata, and therefore regulates CO<sub>2</sub> uptake. It triggers activation of enzymes and is essential for production of Adenosine Triphosphate (ATP). ATP is an important energy source for many chemical processes taking place in plant tissues.

K plays a major role in the regulation of water in plants (osmo-regulation). Both uptake of water through plant roots and its loss through the stomata are affected by potassium. Potassium is also known to improve drought resistance.

Potassium deficiency might cause abnormalities in plants, usually they are growth related. Potassium is an important growth catalyst in plants; potassium deficient plants will have slower or stunted growth some crops exhibit characteristic deficiency symptoms when adequate amounts of K are not available for growth and development. Potassium is mobile in plants and will move from lower to upper leaves. The entire leaf has a very distinct light green appearance when viewed from a distance.

Ca is present in adequate amounts in most soils. Ca is a component of several primary and secondary minerals in the soil, which are essentially insoluble for agricultural considerations. These materials are the original sources of the soluble or available forms of Ca. Ca is also present in relatively soluble forms, as a cation (positively charged Ca<sup>++</sup>) adsorbed to the soil colloidal complex. The ionic form is considered to be available to crops. Ca is essential for many plant functions. Some of them are: Proper cell division and elongation, Proper cell wall development, Nitrate uptake and metabolism, Enzyme activity, Starch metabolism.

Ca deficiency symptoms can be rather vague since the situation often is accompanied by a low soil pH. Visible deficiency symptoms are seldom seen in agronomic crops but will typically include a failure of the new growth to develop properly. Annual grasses such as corn will have deformed emerging leaves that fail to unroll from the whorl. The new leaves are often chlorotic. Extremely acid soils can introduce an entirely new set of symptoms, often from different toxicity's and deficiencies. Many fruits and vegetables demonstrate dramatic symptoms such as Black heart in celery and broccoli, Tipburn in lettuce and cabbage, White heart or Hollow heart in cucurbits, Blossom end rot in tomatoes and peppers, and Pops in peanuts. Tree fruit with low calcium will exhibit increased storage problems such as bitter-pit in apples, cork-spot in apples and pears, cracking in cherries, and other degradation of the fruit while in storage. Deficiency in all crops often also impairs root growth and lead to additional symptoms as a secondary effect. Ca deficient conifer trees will have exhibit yellowing then death and dropping of the needles on the new growth. The new growth may also be deformed.

Zn is a trace element found in varying concentrations in all soils, plants and animals and it is essential for the normal healthy growth of higher plants, animals and humans. Zn is needed in small but critical concentrations and if the amount available is not adequate, plants and/or animals will suffer from physiological stress brought about by the dysfunction of several enzyme systems and other metabolic functions in which zinc plays a part.

Zn is essential for the normal healthy growth and reproduction of plants, animals and humans and when the supply of plant-available zinc is inadequate, crop yields are reduced and the quality of crop products is frequently impaired. When the

supply of zinc to the plant is inadequate, one or more of the many important physiological functions of zinc is unable to operate normally and plant growth is adversely affected (Kabata-Pendias, 2001).

Zn deficient soils can be identified by soil testing, or the analysis of the crop plants (usually leaves) growing on them. The results obtained from soil and/or plant analysis can be compared against the lower critical values for zinc in local soil types for specific crops and a decision made on whether or not zinc fertilizer applications to the soil or crops are required.

Boron is essential for the growth of new cells. It is not readily mobile in the plant, and a boron deficiency causes the terminal bud to cease growth, followed by death of young leaves. Without adequate boron, the number and retention of flowers is reduced and pollen germination and pollen tube growth are less. The result is that less fruit develops.

#### **1.4 Fruits and Vegetables analysis**

Mineral elements play a crucial role for the nutrition of human being. Fruits and vegetables are one of the main sources of mineral elements. Excess or lack of mineral elements in fruits and vegetables may cause various disorders in human health and the people may suffer from various diseases. Increased fruits and vegetables consumption can improve the mineral regulation and reduce cardiovascular diseases and certain cancer risks. Therefore evaluation of micronutrients and essential trace elements levels of fruits and vegetables is a growing trend in nutritional studies throughout the world. Trace elements do not provide any calorie but they maintain the body pH, osmotic regularity and used as coenzyme which regularize the metabolic reactions. Trace elements are required in a diet in amounts less than 100 mg/day (Warldlaw, 1999) and which is vital for health. The deficiency of Zn in

the soil and the excess of As in water have already become two established problems in Bangladesh. Therefore such studies will help us to understand the environmental pollution and its effects on human health. In the developed countries such analysis are done routinely to ensure the good health and quality of life of their citizens.

### **1.5 Objective of the study**

The main objective of the present research is to find out the toxicity or the deficiency of some major, minor and trace elements in some selected environmental samples (Water, Soil, Fruit and Vegetables) which are commonly consumed by both rural and urban population of Bangladesh. So the main objectives are:

- (i) Analysis of major, minor and trace elements concentration in ground water in monsoon and winter season.
- (ii) Analysis of major, minor and trace elements concentration in arable soils in monsoon and winter season of selected land to study the effect of the elements on the crops.
- (iii) Analysis of major, minor and trace elements concentration in some fruits and vegetables.
- (iv) The concentration of the elements in each fruits, vegetables and their role for the nutrition of human being will be discussed.

# CHAPTER TWO

## MATERIALS AND METHODS

## **Materials and Methods**

2 MeV proton beam irradiation was carried out using the 3MV Tandem Accelerator at the Institute of Nuclear Science and Technology (INST), Atomic Energy Research Establishment (AERE), Bangladesh Atomic Energy Commission (BAEC). 3MV Tandem Accelerator has enormous facilities. Two separate ion sources of latest technology namely (i) Duoplasmatron ion source and (ii) Negative sputter ion source have been installed in association with the special T-shaped accelerator capable of double acceleration of the Ions. Almost all the elements in the periodic table ( $z = 1$  to 92) can be ionized by the two ion sources. The produced ions are then accelerated electro-statically which hits the target specimen in the experimental chamber after a guided travel of the ion beam with the help of a number of complex electronics collision phenomena and to detect the produced signals such as X-rays and gamma-rays coming from the target specimen. By a special set of nuclear electronics this information is collected and stored in the computer which is then analyzed by suitable software and shown in plate 2.1 to 2.4 (Tesmer and Nastasi, 1995).

### **2.2 Basic principle of Tandem Accelerator**

- Ion is produced from ion source externally from the accelerator tank.
- Then guided to the tank where accelerated to high speed and high energy.
- This high energy ion then strike the specimen and x-ray/Y-ray produced with particular energy.
- The emitted radiation is detected by detector and analyzed by MCA.
- The analyzed spectrum then brought to the computer to make human readable format.

### **2.3 System Description**

#### **MAIN SYSTEM:**

- (1) 3 MV Tandem Accelerator



- (2) Two Ion Sources :
  - (i) Duoplasmatron Ion Source
  - (ii) Negative Sputter Ion Source

(3) Data Acquisition system

**SUBSYSTEMS:**

- (1) High Vacuum system
- (2) Automatic Air Conditioner
- (3) Closed loop Cooling Water Supply System
- (4) Dedicated Electrical Sub-Station back up by UPS & Diesel Generator to ensure 24 hours uninterrupted power for the facility.

(5) Available Ions sources:

(i) Model 358 Ion source

$^1\text{H}_1^+$  around 10 uA from  $\text{H}_2$  gas upto 6 MeV.

$^4\text{He}_2^+$  around 1 uA from He gas upto 9 MeV.

(ii) Model 860C ion source

$^{12}\text{C}_3^+$  around 25 uA from C upto 12 MeV.

$^{28}\text{Si}_3^+$  around 30 uA from Si upto 12 MeV.

$^{197}\text{Au}_2^+$  around 20 uA from Au upto 9 MeV.

Measured in a Faraday cup after the high energy switching magnet.



Plate : 2.1 Tandem Accelerator at Bangladesh Atomic Energy Commission, Saver, Dhaka, Bangladesh.



Plate: 2.2 Ion Sources of Tandem accelerator



Plate : 2.3 Experimental Chamber of Tandem accelerator



Plate : 2.4 Tandem accelerator Control Room

#### 6) Data Counting System

- i) This is an Industrial PC
- ii) Various Software like TOS, RUMP, MPANT and GUPIX are running in this computer.
- iii) During operation user can access this place only.

#### **2.4 Selection of Analytical Technique**

Analytical technique, such as, atomic absorption, atomic emission, chemical and electro analytical methods, mass spectrometry in different modes, neutron activation technique and ion beam technique (IBA) which offer sufficiently low detection limits for a variety of matrices, are now available. The selection of a particular methodology in trace element analysis depends on the nature of the problem. The rapidity, sensitivity, accuracy and reproducibility, multielement or single element character, amount of sample needed, etc. limit the applicability of a given method. The spectrum of analytical methods used in trace analysis is indeed very broad. Among them multi elemental techniques are useful in obtaining

elemental composition profiles of a given specimen. The nondestructive methods offer possibilities of generating data for several elements in a multielement trace analytical scheme with minimum contamination. In a carefully designed study, multielement and nondestructive assays can provide useful information about a large number of elements at a relatively low cost.

### **2.5 Ion Beam Material Analysis**

From the very beginning of the invention, particle accelerators have become a major tool for research in many areas of science and technology. Accelerator based techniques are now routinely applied in science, industry, medicine, environmental studies and in many other fields. While they come in various range of sizes and types, accelerators that produce relatively low energy beams have become the most powerful nuclear analytical tools. Among the practical applications of low energy accelerators are the highly sensitive analyses of trace elements in environmental studies or in healthcare and treatment of patients.

Ion Beam Analysis (IBA) is a well developed field which uses the accelerator based methods of elemental analysis. A large number of applications of accelerator based nuclear techniques are found in the fields of Human health, Environmental Sciences, Agriculture, Biology, Industry, etc. A very comprehensive account of the applications of low energy accelerators and IBA techniques can be found in the proceedings of the international conferences (**Valkovic and Zyszkowski, 1994**) and books and review articles. The attractive features of the IBA methods are the small low energy accelerators which are widely available and the relatively simple detection systems that are required.

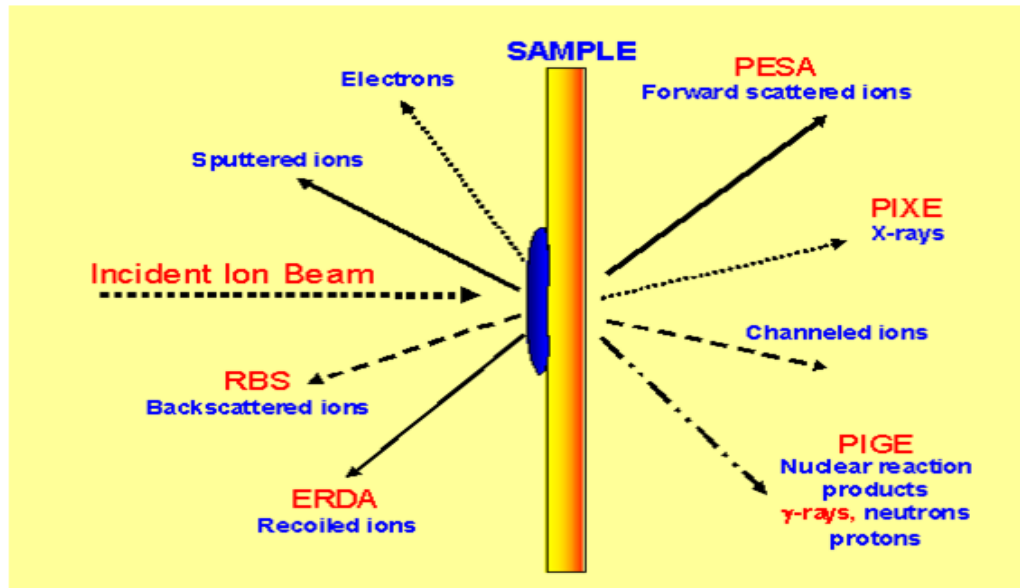


Plate: 2.5: Different methods based on ion beam and material interaction

The best way of determining many elements simultaneously on a typical sample is to use Ion Beam Analysis (IBA), since it is multielemental, sensitive, and virtually nondestructive (Johansson and Campbell, 1998). Under favorable conditions, it is possible to determine most of the elements contributing significantly to the total mass loading, thus allowing the additional constraint of mass closure to be imposed in statistical source apportionment methods. The fact that the IBA methods are virtually non-destructive means that collected samples may be archived as a part of an environmental specimen bank, or held for future work or for regulatory purposes.

In many areas, several powerful analytical techniques based on the accelerator technology are available (Plate 2.5). The IBA techniques include:

**PIXE:** Proton Induced X-ray Emission (PIXE) is based on atomic fluorescence created by energetic protons and the analysis is performed by measuring the characteristic X-rays emitted from the sample. PIXE is well adapted to measure major, minor and trace elements in different sample matrices such as, biomedical, environmental, agricultural and industrial samples. PIXE is a multielemental

analytical technique and is capable of measuring elements from Aluminum (Al) up to Uranium (U) in a single experiment.

**PIGE:** Proton Induced Gamma Emission (PIGE) is based on nuclear reaction and the analysis is performed by measuring the characteristic Gamma-rays emitted from the elements present in the sample. PIGE is particularly useful for measurements of light elements such as F, B, Mg, Na, etc. in different matrices such as, biomedical, environmental, agricultural and industrial samples, which are inaccessible by PIXE. PIGE is capable for isotopic speciation of the sample.

**RBS:** Rutherford Backscattering Spectrometry (RBS) is based on nuclear scattering of particles upon bombardment on the sample and the analysis is performed by measuring the scattered particles. RBS has proved its efficacy in identifying and localizing thin layers. RBS has the capability to measure the thickness, depth profiling and composition of thin films of technologically important and layered samples.

Using these techniques it is possible to identify the elements present in any sample and quantify the amount in the range as low as 1 ppm depending on the sample matrix and the element to be measured with accuracy: Within  $\pm 5 - 15\%$  , and precision: Within  $\pm 5\%$ .

#### **Available Software**

MPANT, TOS, GUPIX, RUMp, SIMNRA

PIXE has the advantage over other IBA techniques that the cross-section for X-ray production is large and the background contribution from bremsstrahlung is low. Most important minor and trace elements can be determined basically in one single run in large number of samples during, relatively, short time. In addition, it is very easy to prepare the samples for analysis (*i.e.* it is sufficient to dry the sample and mount it in the target chamber). The use of PIXE technique in the analysis of fruit and vegetable materials has been quite few in the past due to its expensive operation. However (Johansson and Campbell, 1998) the spread of ion beam accelerators around the world and the recent development of IBA techniques have encouraged researchers to employ these powerful techniques in different analytical applications, including the investigation of fruits and vegetables (Smit, 2005).

## **2.6 Methodology and Measurement**

### **2.6.1 Study Area**

The main object of the present study is to measure the concentration of element in the environmental samples soil, water, fruits and vegetables for the assessment of population exposure. Under the limited scope of thesis research, we have selected Rajshahi district from Northern part of Bangladesh for the study area. Samples are collected from different season during monsoon and winter season of different places of this district. The geographical location of sampling site is shown in Map 1.

### **2.6.2 Sampling and Sample Preparation**

10 water samples, 10 soil samples, fresh locally produced 10 fruits and 10 vegetables samples were collected from various areas of Rajshahi district namely Tanore, Mohanpur, Paba, Puthia, Godagari, Bagmara, Durgapur, Boalia, Chargat, Bagha in different season.

### **2.6.3 Collection and Preparation of Water Samples**

Clean plastic bottle has been used for sampling. These have securely fitting stoppers or caps with nontoxic, liners, clear of the screw thread to ensure that the risk of contaminating the water sample is minimized. The bottles are strong and durable, so that it did not break in transit and the cap also did not leak once the cap is secured. Cotton wool plugs and paper caps have been avoided as they tend to fall off during and after sampling and increase the risk of contamination. The stopper is loosely inserted into the bottle, and a brown paper or aluminum foil cover is tied to the neck of the bottle to prevent dust from entering (Hightower, 1995).

#### **Acid Washing**

This step serves to oxidize and solubilize any oxidizable materials. Bottles were rinsed with 8 M nitric acid ( $\text{HNO}_3$ ). Then rinsed the bottle twice with tap water and rinsed the inside of the bottle with 1.2 M hydrochloric acid (HCl) once again being sure to wet all internal surfaces. This acid was reused until it is visibly discolored. Then the bottle rinsed with tap water three times

#### **Collecting water samples for analysis**



Before collecting sample both hands have been washed with soap and water. 200 mL water samples were collected in plastic bottle. Plate 2.6 shows sampling from a tap or pump outlet or tube-well as described below.

**A. Clean the tap:**

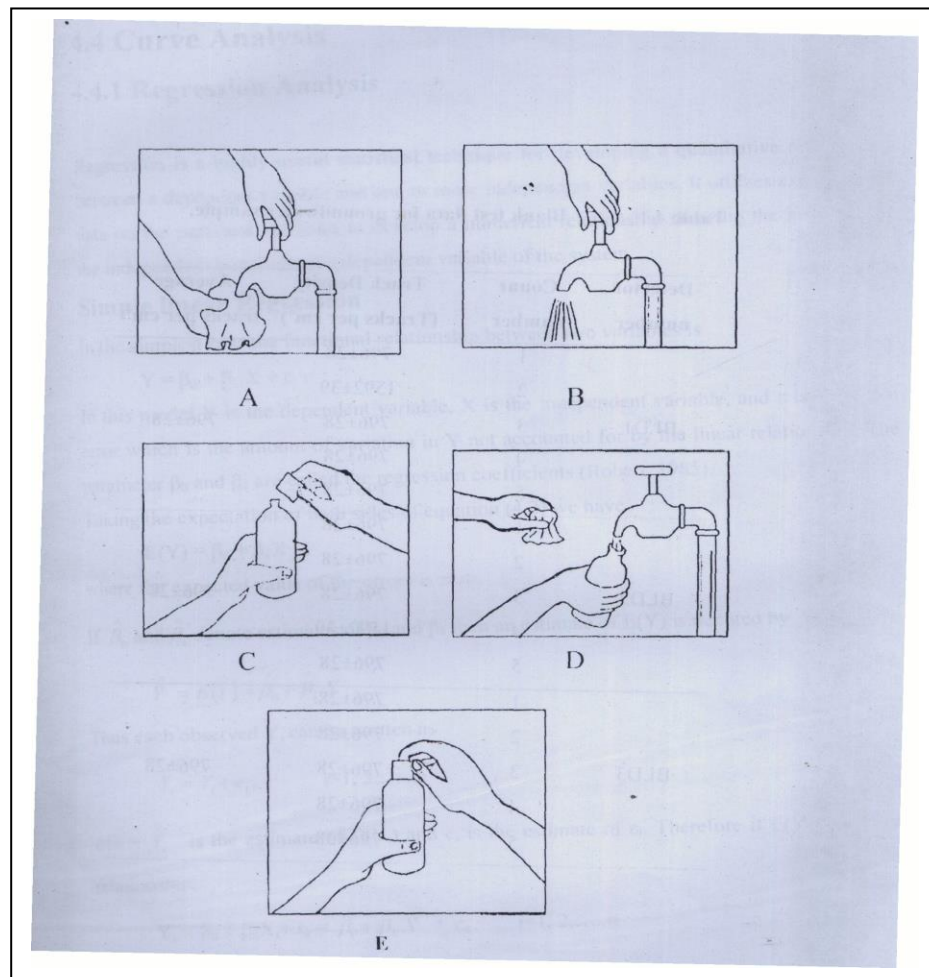
Using a clean cloth, wiped the outlet of the tap to remove any dirt.

**B. Open the tap:**

The tap turned on at maximum flow and let the water run for 1-2 minutes.

**C. Open the sterilized bottle:**

To avoid contamination while taking the sample, the bottle held near the bottom with one hand, the top of the cap with the other hand. Then the cap has been unscrewed.



**Plate 2.6** Sampling from a tap or pump outlet or tube-well (USEPA, 1987).

- A. Clean the tap    B. Open the tap    C. Open the sterilized bottle  
D. Fill the bottle    E. Cap the bottle

**D. Fill the bottle**

The bottle rinsed at least 4 times with the water to be sampled. While holding the cap and protective cover face downwards (to prevent entry of any dust, which may contaminate the sample), immediately hold the bottle under the water jet, and filled. A small air space was left to make shaking before analysis easier. After the sample bottle was filled to the correct level then removed it from the flow and immediately capped and screwed it down tightly.

**E. Stopper or cap the bottle**

The stopper is placed in the bottle and fixed the brown paper protective cover in place with the string. The bottle is adequately labeled so it can be identified at all stages of the transport and analytical process.

A Test Order Form (TOF), recording any relevant field information which would assist in the interpretation of the results was prepared. TOF carry all the information such as water system ID number, water system name, collection date and time the sample was taken, type of sample, sample location. If anything unusual during the sample collection, it was noted in the TOF. Sample bottle was send as soon as possible with TOF by express courier which delivers directly to the laboratory. Samples were analyzed within 48 hours of collection to avoid and absorption loss and to ensure reliable results.

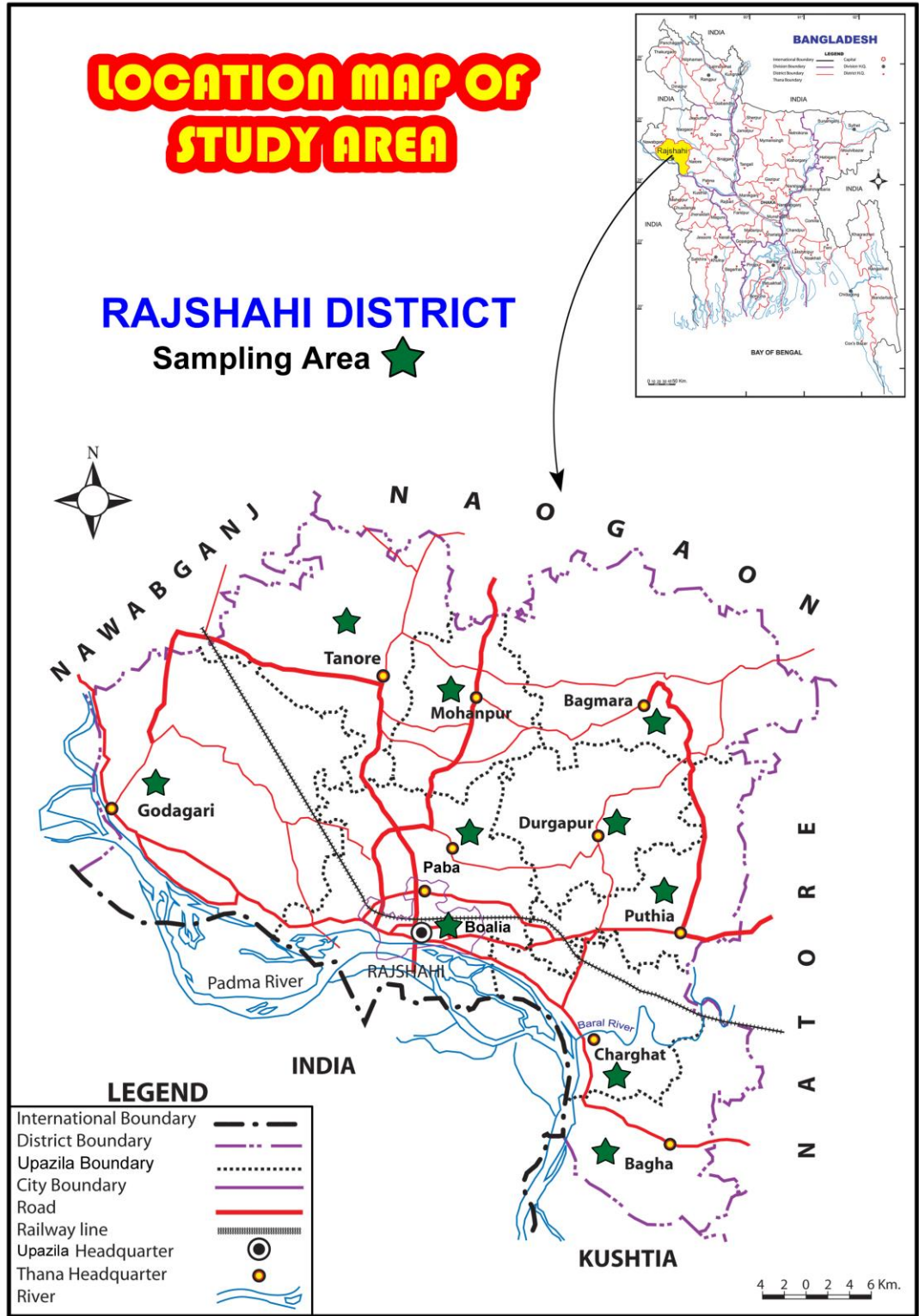
A 200 ml water samples together with 100 mg of ashless cellulose powder was evaporated to dryness in a porcelain dish on a steam bath and then under an infra-red lamp. The residue was cooled at room temperature in a desiccator for one hour. The powder sample was then weighted and sealed in a polythene bag and stored in a desiccator, the dried sample was milled by milling machine uniformly & very carefully and made it into powder. After milling the sample was taken into aluminum fuel so that no moisture could into the sample. Then it was

transferred to hydraulic pressure machine for making tablet. 2.6.4 Collection and preparation of the Soil Samples *Sample collection, preparation and measurement procedure are considered very vital for PIXE and PIGE techniques. The sampling area need to have the following criteria*

:

- (i) the collection area should be undisturbed flat open terrain preferably covered with grass but without any sheltering vegetable plants or housing structures
- (ii) there should be very little or no runoff soil during heavy rain particularly in rainy season.
- (iii) there should be a minimum of earthworm or rodent activity in the area, and
- (iv) the area should be free from termite heaps

10 soil samples from the different thanas of Rajshahi district were collected, using a soil sampler (a corer) at a depth of 0 to 10 cm, following standard procedures such as cleaning the grass surface. All the samples were air dried under laboratory temperature and relative humidity conditions and cleaned to remove the grass roots, biological matters, stones and gravels and dried in an electric oven at a temperature of 120°C for about four hours to make the samples water free. After cooling the samples to the room temperature in a dessicator, the weight was taken. The process of heating, cooling and weighting was repeated until a constant weight is shown by balance which is a confirmation of zero water content. After cooling the samples to room temperature,



Map 1: Location of the study area in Rajshahi district.

the dried sample was milled by milling machine uniformly and very carefully and made it into powder. Some of these prepared samples inside the containers are presented in a photograph in shown in plate 2.7 After milling the sample was taken into aluminum fuel so that no moisture could into the sample. Then it was transferred to hydraulic pressure machine for making tablet.



Plate : 2.7 Some of the prepared soil samples.

### 2.6.5 Vegetables and Fruits Samples Preparation

Locally produced 10 fruits and 10 vegetables samples were taken for the analysis.

#### Fruits Samples

Sample no.	Bengali name	English name	Scientific name
AF-1	Kamranga	Carambola	<i>Averrhoa carambola</i>
AF-2	Papay	Papaya	<i>Carica papaya</i>
AF-3	Peyara	Guava	<i>Psidium guajava</i>
AF-4	Kola	Banana	<i>Musa sapientum</i>
AF-5	Kalo Jam	Black Berry	<i>Syzygium cumini</i>
AF-6	Anaras	Pine-Apple	<i>Ananus comosus</i>
AF-7	Jalpi	Olive	<i>Olea europea</i>
AF-8	Aamalaki	Amla	<i>Emblica officinalis</i>
AF-9	Kathal	Jackfruit	<i>Artocarpus heterophyllus</i>

AF-10	Aam	Mango	<i>Mangifera indica</i>
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### Vegetables Samples

Sample no.	Bengali name	English name	Scientific name
AV-1	Misti kumra	Pumkin/ Sweet gourd	<i>Cucurbita moschata</i>
AV-2	Begun	Bringal	<i>Solanum melongen</i>
AV-3	Dherosh	Lady's finger/Orka	<i>Abelmoschus esculentus</i>
AV-4	Alu	Patato	<i>Solanum tuberosum</i>
AV-5	Tomato	Tomato	<i>Lycopersicon esculentum</i>
AV-6	Lau	Bottle Gourd	<i>Lagenaria siceraria</i>
AV-7	Gajor	Carrot	<i>Daucus carota</i>
AV-8	Phul kopi	Coliflower	<i>Brassica oleracea</i> var. <i>botrytis</i>
AV-9	Bandhakapi	Cabbage	<i>Brassica oleracea</i>
AV-10	Shalgom	Turnip	<i>Brassica rapa</i>

All the fruits and vegetables samples were dried in an electric oven at a temperature of 120°C for about four hours. After cooling the samples to room temperature, the dried sample was milled by milling machine uniformly & very carefully and made it into powder. After milling the sample was taken into aluminum fuel so that no moisture could into the sample. Then it was transferred to hydraulic pressure machine for making tablet.

#### 2.6.6 Making tablet by the sample (powder)

Some of the powder samples are presented in a photograph is shown in plate 2.8. Some portion (about 0.200gm) from powder samples was taken for making tablets by electrical balance during this time the electrical balance machine was calibrated by tearing. The sample was taken into



Plate: 2.8 Sample of Carrot (*Daucus carota*) Powder.

weighting papers and specula were used for taking sample. The machine was totally error free and gave accurate results shown in plate 2.9

The tablet was made by Hydraulic pressure machine that's diameter was 10mm. This work was done by giving 5 (five) tones pressure. After making tablet it was removed from the hydraulic pressure machine by just giving hand pressure and some of these prepared tablets are presented in a photograph is shown in Plate 2.10.



Plate 2.9: Hydraulic pressure machine

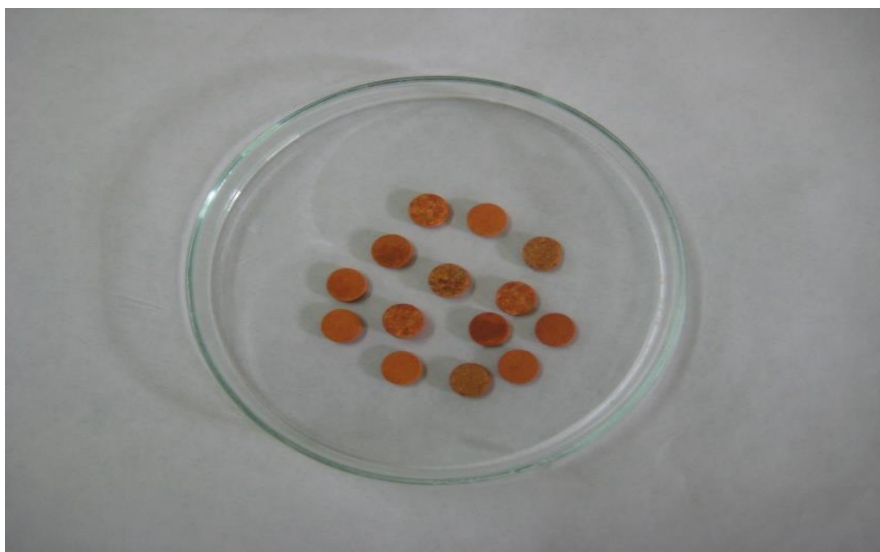


Plate 2.10 Sample of Carrot (*Daucus carota*) tablets

### **2.6.7 Storage of tablet**

After being made tablets, it was storage at  $-200^{\circ}\text{C}$  temperature into a refrigerator for analyzing essential contents by Tandem accelerator. It was kept into a deep freeze so that it could not absorb moisture from the outer sides.

### **2.6.8 Analysis by Tandem Accelerator**

Finally tablets were transferred to Tandem Facilities Division (TFD) for analyzing essential elements content of tablets by Tandem Accelerator (TA).

#### **(i) Irradiation and Data Acquisition**

The prepared samples and a Standard Reference Material soil (SRM 2586) sample from National Institute of Standards & Technology (NIST), Department of Commerce, USA were irradiated with proton beam of energy of 2.5 MeV at the 3 MV Tandem Accelerator, Atomic Energy Research Establishment, Dhaka, Bangladesh. The integrated charge irradiation on the sample was kept in the range of 5-10 micro Coulombs and the irradiation was performed for 5-10 min to get sufficient X-rays counts. X-ray emission analysis involves both a means of exciting the atoms of specimen so that they emit characteristic X-rays and a means of detecting and identifying their energies so that their intensities can be converted to elemental concentrations. Individual irradiation was performed for the each sample. The X-rays emitted in the irradiation were counted using the High Purity Germanium (HPGe) detector associated with the necessary electronics, A thin



Mylar foil was placed between detector and sample for the elimination of unwanted low energy X-rays to reduce noise in counting system and also to improve the spectrum quality. The obtained X-ray spectrum were analyzed using the GUPIX software

(Maxwell *et al.*, 1989). GUPIX is a program for the non-linear least-squares fitting of PIXE spectra. The concentration of the K, Ca, Ti, Cr, Mo, Fe, Co, Ni, Cu and Zn elements in the investigated samples were determined from the area under their characteristics well defined peaks at energies of 3.313, 3.691, 4.509, 5.412, 5.895, 6.399, 7.652, 8.266, 8.905 and 9.571 keV, respectively.

**(ii) Data analysis**

The PIXE spectral data were analyzed by using GUPIX software that can simply, automatically and quickly fit the spectra to obtain the elemental concentrations. Elemental concentration calculation of each element Cz is based on the following equation:

$$C_z = Y/Y_t Q \epsilon T H \dots\dots\dots(2.1)$$

where  $Y_t$  is the X-ray theoretical intensity (*i.e.* the yield per micro- coulomb of charge per unit concentration per steradian),  $Y$  is the X-ray experimental intensity or yield,  $Q$  is the measured proton beam charge,  $\epsilon$  is the efficiency of the detector and  $T$  is the transmission through any filters or absorbers between the target and the detector.  $H$  is an instrumental constant equivalent to the product of the geometric solid angle of the X-ray detector and any systematic normalization factor present in the charge integration system.  $H$  values as a function of X-ray energies can be measured experimentally using wide range of pure single-element standards emitting both K and L X-rays in the energy region 3-26 keV (Campbell *et al.*, 1993).

# CHAPTER THREE

## RESULTS AND DISCUSSION

### **3.1 Elemental Analysis of Water, Soil, Fruits and Vegetables Samples**

Elemental analysis is a process where a sample of some material (*e.g.*, soil, waste or drinking water, bodily fluids, minerals, chemical compounds) is analyzed for its elements and compounds and sometimes its isotopic composition. Elemental analysis can be qualitative (determining what elements are present), and it can also be quantitative (determining how much of each type are present).

#### **3.2 WATER ANALYSIS**

Water samples were analyzed and the concentrations of the elements are given below:

##### **3.2.1 Concentration of Fluorine (F), Manganese (Mn), Iron (Fe), Calcium (Ca), Copper (Cu) and Zinc (Zn)**

###### **Fluorine (F)**

The F content of ground water in the study area lies in the range of 0.95 to 2.01mg/L with a mean of 1.44mg/L in monsoon season shown in appendix table I and winter season vary from 1.05 to 2.45mg/L with a mean 1.27mg/L shown in appendix table II. The highest concentration of fluorine is found in the ground water of Godagari thana of Rajshahi district in monsoon season and Chargat thana in winter season. The lowest concentration of F is found in the ground water of Chargat thana of Rajshahi district in monsoon season and Bagha thana in winter season. The maximum recommended value for Bangladesh being 1.00 mg/L (Bangladesh Gazette, 1998). From the present study it is found that 80% of the studied samples contain higher F than the recommended maximum shown in graph 3.1 and 3.2 respectively.

Fluorine has attracted serious attention in recent years because of the apparent role it plays in human health. Plants, especially from the acidic soils readily absorb fluoride ions, but it is highly toxic for several species of plants (IAEA, 1980).

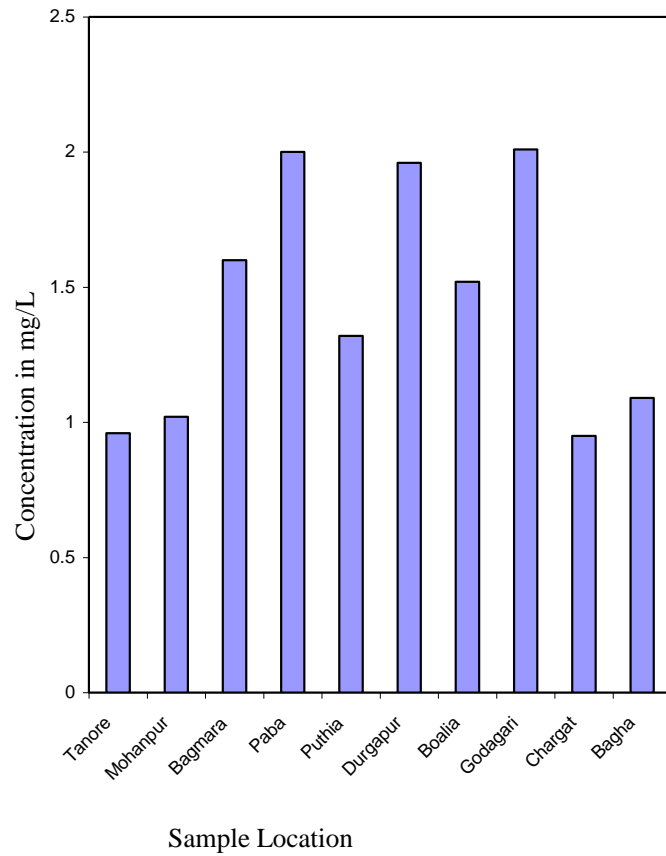
Fluoride in natural waters may originate from the solution of F, apatite, or more commonly from the solution of fluoride-bearing micas and amphiboles. A common sink for fluoride is adsorption by kaolinite. This is an example of anion exchange. The adsorption is highest at pH 6, and is negligible at pH 4 (and below), and above pH 7.5. As a result the alkaline waters are commonly high in fluoride (Hounslow, 1995).

### **Manganese (Mn)**

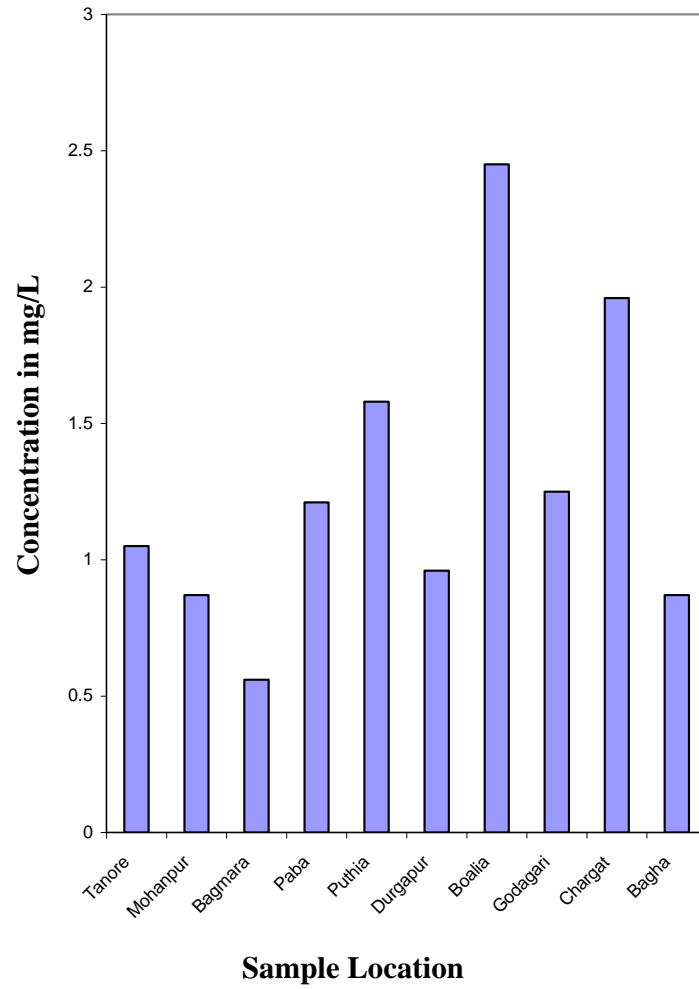
The Mn content of ground water in the study area lies in the range of 0.11 to 0.33mg/L shown in appendix table III in monsoon season and 0.23 to 0.58mg/L in winter season shown in appendix table IV. The highest concentration of Mn is found in the ground water of Tanore thana of Rajshahi district in monsoon and Boalia thana in winter season. The lowest concentration of Mn is found in the ground water of Durgapur thana of Rajshahi district in monsoon and Puthia thana in winter season. The Bangladesh standard value for Mn being 0.1 mg/L and WHO guideline value 0.4 mg/L (WHO, 2006).

It shows that about 100% of the surveyed samples have Mn concentrations exceeding the Bangladesh drinking water standard of 0.1 mg/L and do not exceed the WHO health-based guideline value of 0.4 mg/L shown in graph 3.3 and 3.4 .

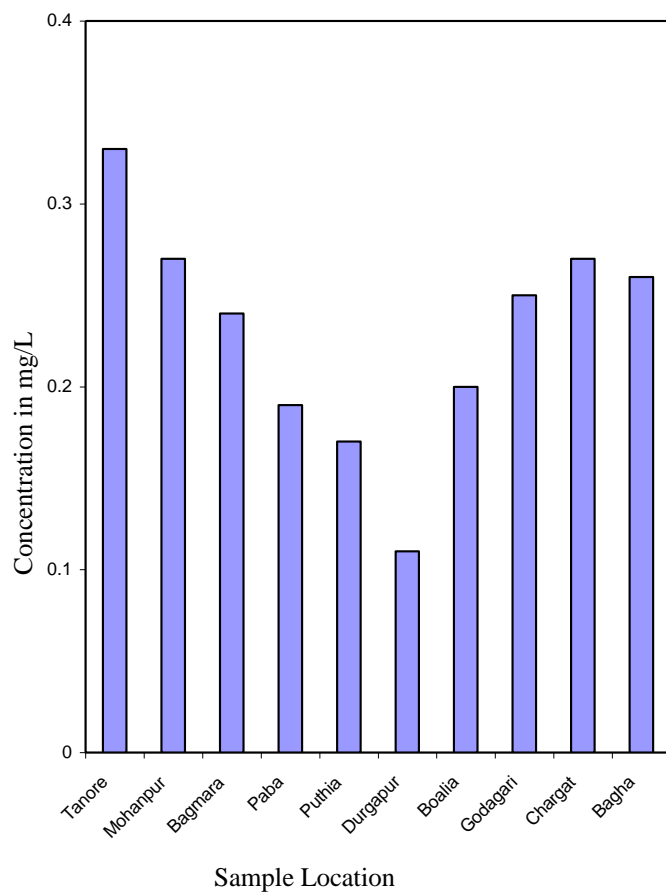
Detection of high concentrations of Mn in groundwater has added a new dimension to the already difficult safe water supply scenario in Bangladesh. However, Mn issue has attracted relatively less attention so far in the water supply sector. It is important to raise awareness among the stakeholders about the Mn issue. It is also very important to identify areas unacceptable levels of Mn and to develop water treatment technologies accordingly.



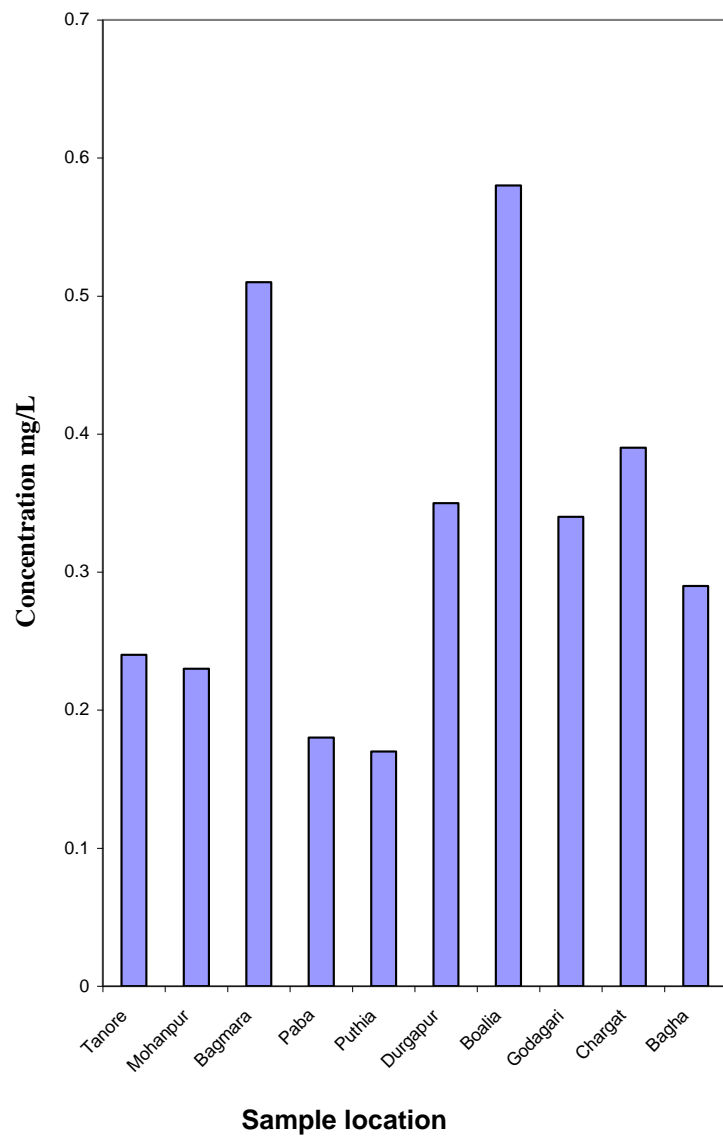
Graph 3.1 Sample wise distribution of F in the water of Rajshahi district in monsoon season.



**Graph 3.2** Sample wise distribution of F in the water of Rajshahi district in winter season.



Graph 3.3 Sample wise distribution of Mn in the water of Rajshahi district in monsoon season.



**Graph 3.4 Sample wise distribution of Mn in the water of Rajshahi district in winter season.**

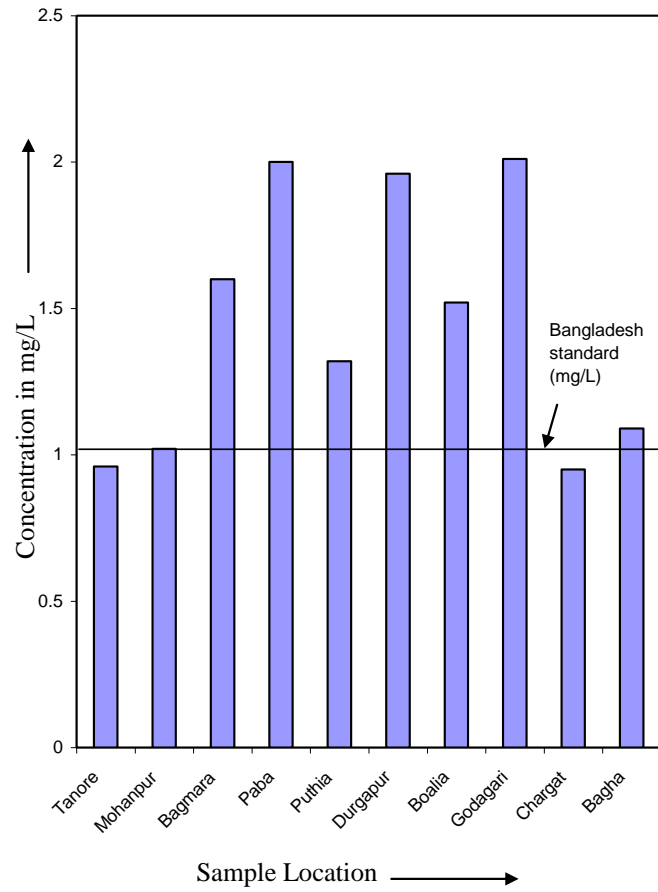


**Iron (Fe)**

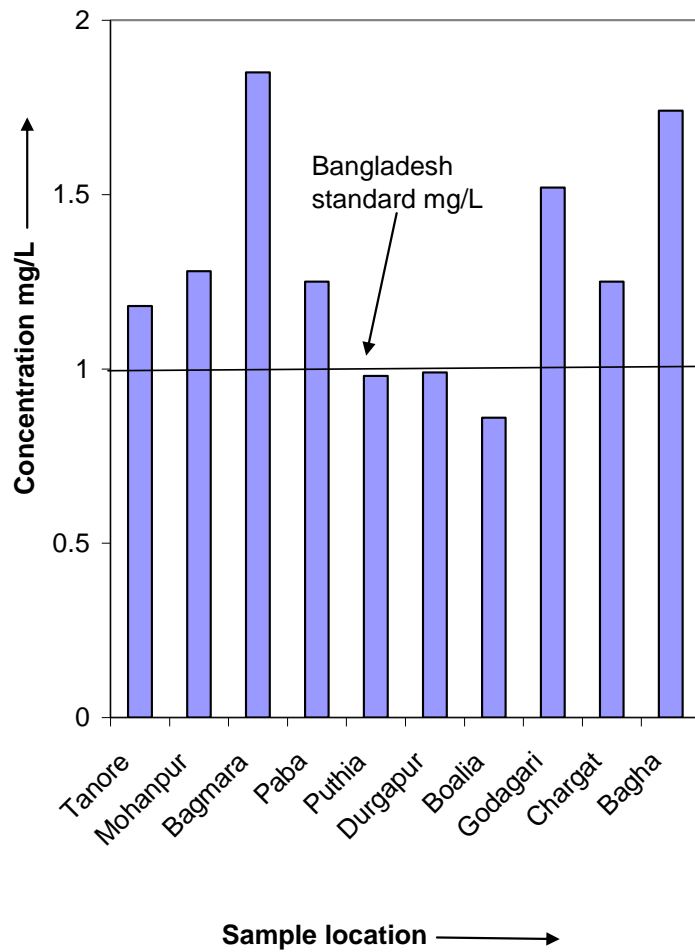
The Fe content of ground water in the study area lies in the range of 0.95 to 2.03mg/L in monsoon season shown in appendix table V and 0.86 to 1.85mg/L in winter season shown in appendix table VI. The highest concentration of Fe is found in the ground water of Bagmara thana of Rajshahi district in monsoon and also Bagmara thana in winter season. The lowest concentration of Fe is found in the ground water of Paba thana of Rajshahi district in monsoon and Boalia thana in winter season. The Bangladesh standard value being 1mg/L and WHO guideline value 0.3mg/L (WHO, 2006). From the present study it is found that 80% of the studied samples contain higher Fe than the recommended maximum shown in graph 3.5 and 3.6.

Groundwater contributes substantially to daily iron intake of rural Bangladeshi women and currently represents an under-assessed potential source of dietary Fe.

High levels of Fe and Mn do not pose any known adverse health risks. The U.S. Environmental Protection Agency (EPA) has not set maximum contaminant levels (MCL) for iron and manganese in the National Primary Drinking Water Regulation.



Graph 3.5 Sample wise distribution of Fe in the water of Rajshahi district in monsoon season.



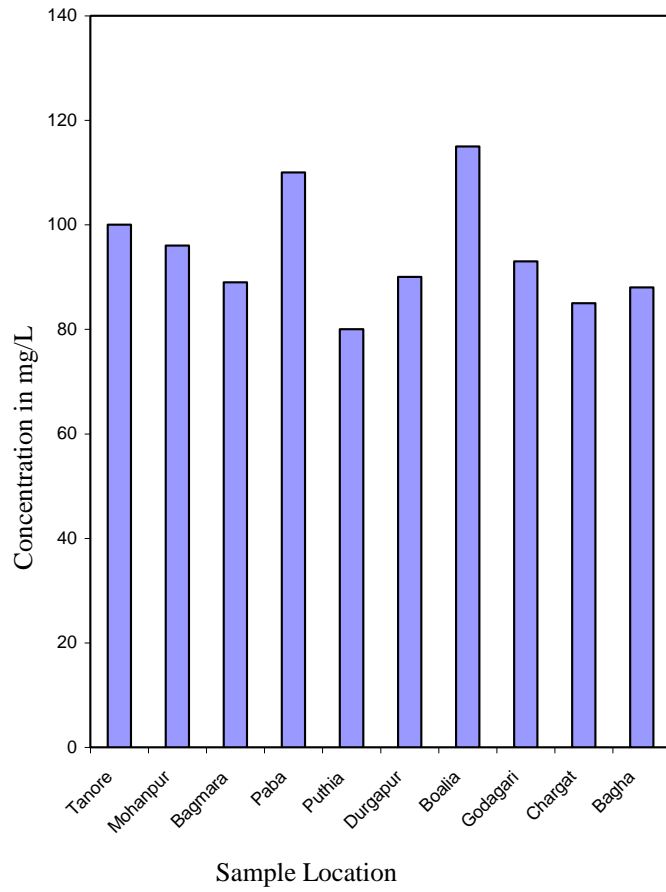
**Graph 3.6 Sample wise distribution of Fe in the water of Rajshahi district in winter season.**

### **Calcium (Ca)**

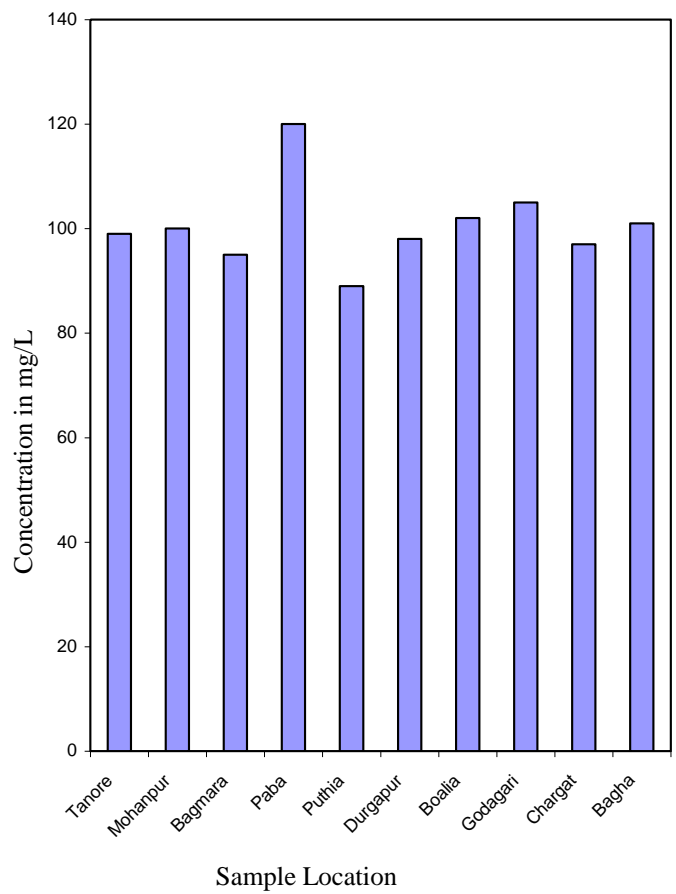
The Ca content of ground water in the study area lies in the range of 80 to 115mg/L shown in appendix table VII. The highest concentration of Ca is found in the ground water of Boalia thana of Rajshahi district in monsoon and Paba thana in winter season. The lowest concentration of Ca is found in the ground water of Puthia thana of Rajshahi district in monsoon and also Puthia thana in winter season shown in appendix table VIII. The standard WHO guideline value 75mg/L (WHO, 2006). From the present study it is found that 100% of the studied samples contain higher calcium than the recommended maximum shown in graph 3.7 and 3.8.

### **Copper (Cu)**

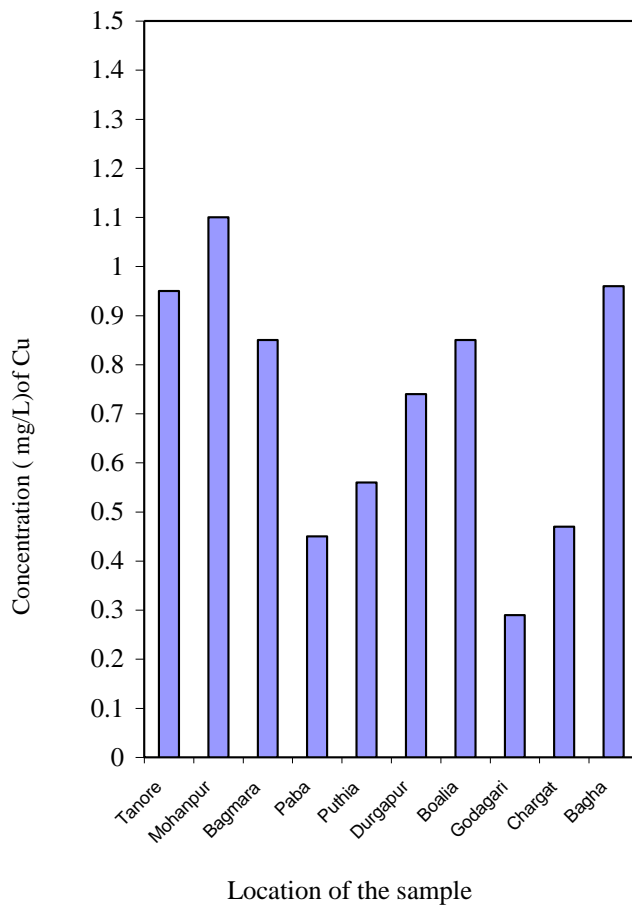
The Cu content of ground water in the study area lies in the range of 0.29 to 1.1mg/L in monsoon shown in appendix table IX and concentration vary from 0.49 to 1.13mg/L in winter season shown in table X. The highest concentration of Cu is found in the ground water of Mohanpur thana of Rajshahi district in monsoon and Bagha thana in winter season. The lowest concentration of Cu is found in the ground water of Godagari thana of Rajshahi district in monsoon and Chargat thana in winter season. The Bangladesh standard value being 1.3 mg/L and WHO guideline value 1mg/L (WHO, 2006). From the present study it is found that 100% of the studied samples contain lower concentration of Cu than the recommended maximum shown in graph 3.9 and 3.10.



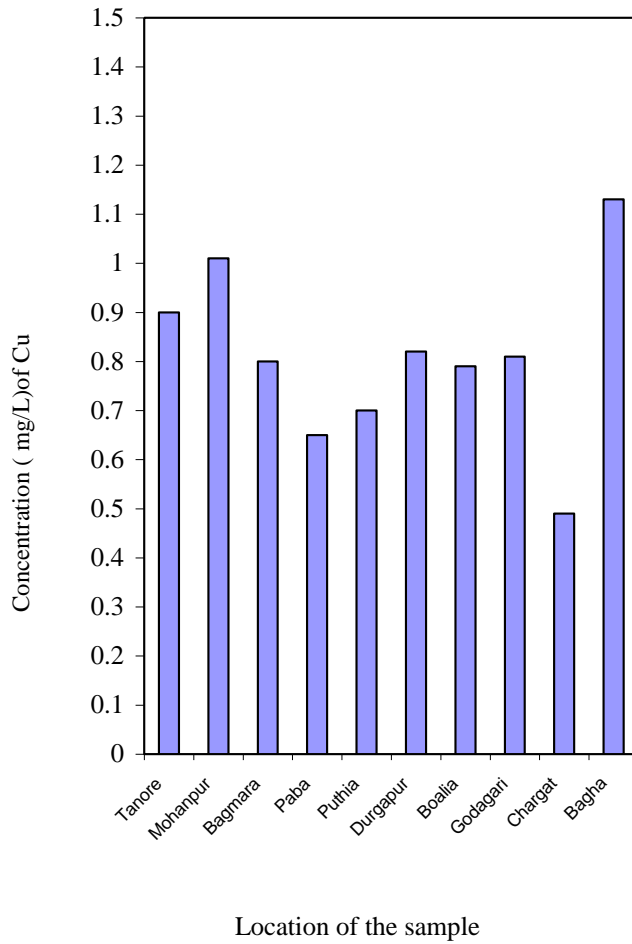
Graph 3.7 Sample wise distribution of Ca in the water of Rajshahi district in monsoon season.



Graph 3.8 Sample wise distribution of Ca in the water of Rajshahi district in winter season.



Graph 3.9 Sample wise distribution of Cu in the water of Rajshahi district in monsoon season.



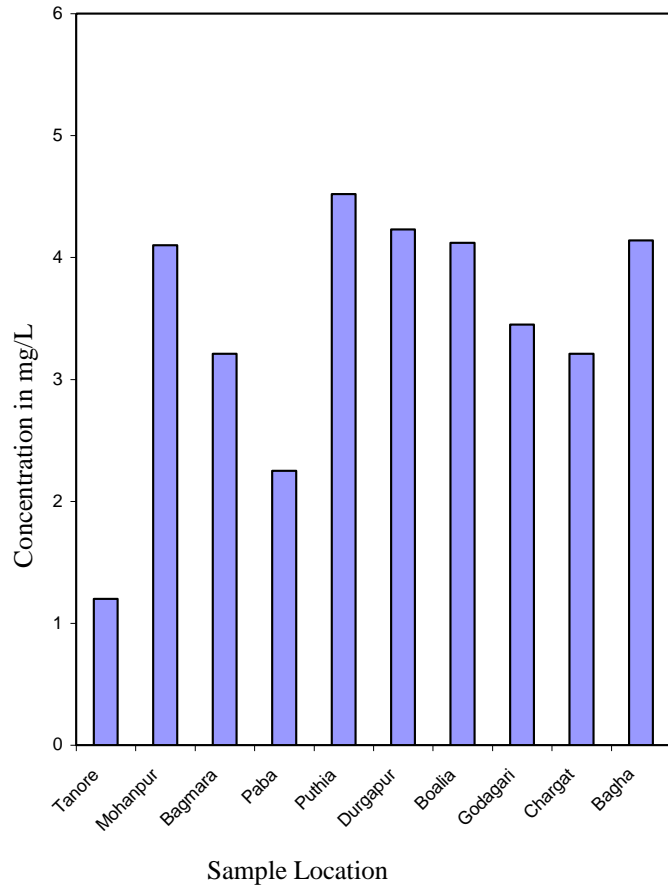
Graph 3.10 Sample wise distribution of Cu in water of Rajshahi district in winter season.



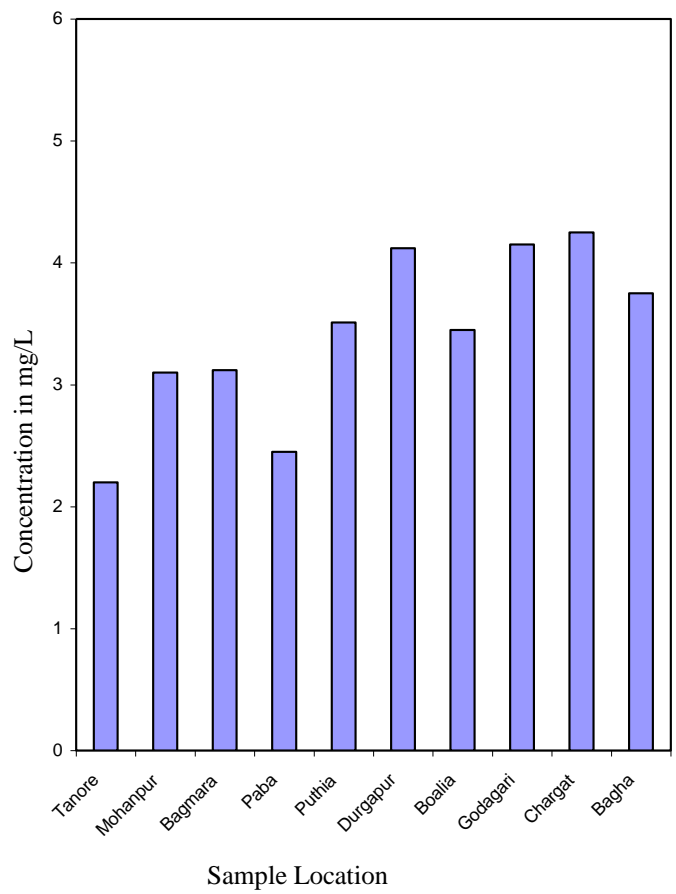
### **Zinc (Zn)**

The Zn content of ground water in the study area lies in the range of 1.20 to 4.52mg/L in monsoon shown in appendix table XI and concentration vary from 2.20 to 4.25 mg/L in winter season shown in appendix table XII. The highest concentration of Zn is found in the ground water of Puthia thana of Rajshahi district in monsoon and Chargat thana in winter season. The lowest concentration of Zn is found in the ground water of Tanore thana of Rajshahi district in monsoon and Tanore thana in winter season. The WHO guideline value 5.0mg/L (WHO, 2006). From the present study it is found that 100% of the studied samples contain lower Zn than the recommended maximum shown in graph 3.11 and 3.12 respectively.

In an average human body, there is about 2 to 3 grams of Zn. Most of this is found in the muscle tissue and the bone. Generally, most adults do not require zinc supplementation if they eat a healthy diet with vegetables, fruit and some protein sources. Males require more Zn than women, as the mineral is released when a man ejaculates. The more sexually active a male, the more Zn that is required, as semen has the highest levels of Zn in the body



Graph 3.11 Sample wise distribution of Zn in the water of Rajshahi district in monsoon season.



Graph 3.12 Sample wise distribution of Zn in the water of Rajshahi district in winter season.

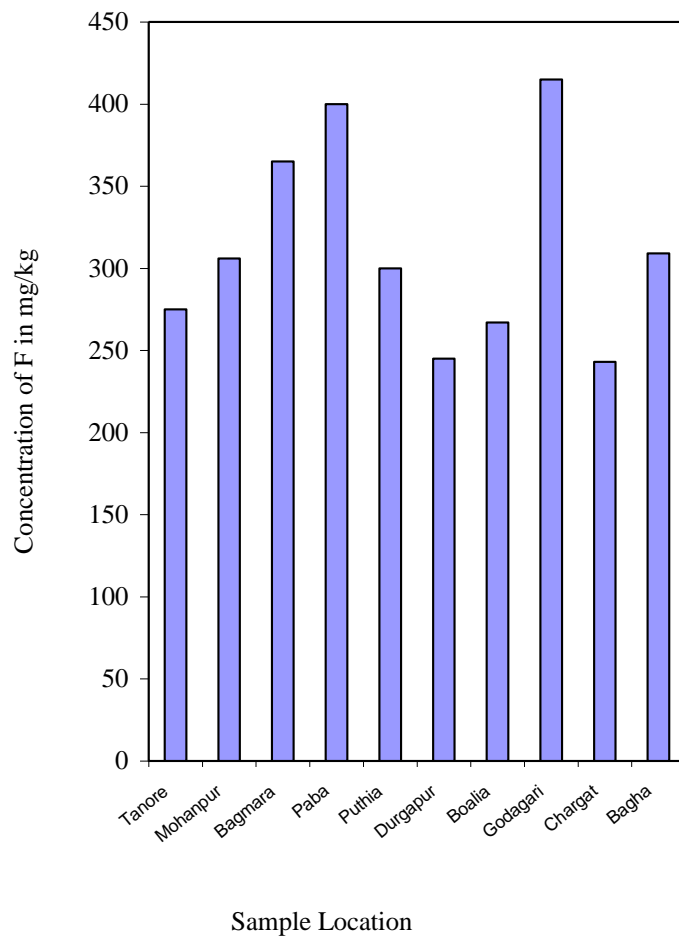
### **3.3 Soil Analysis**

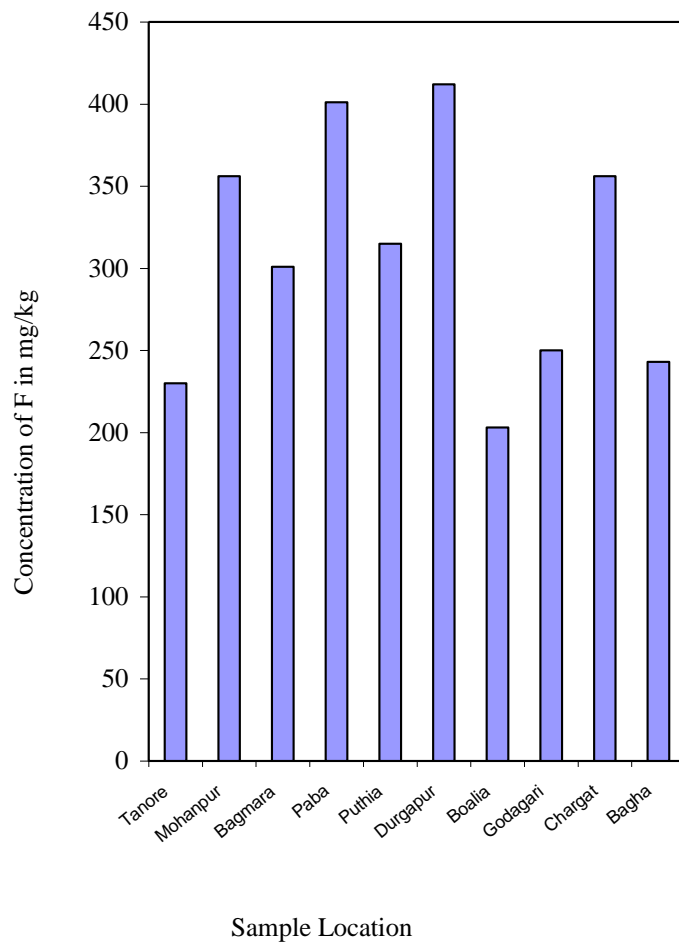
Soil samples were analyzed and the concentration of the elements Fluorine (F), Phosphorus (P), Copper (Cu), Manganese (Mn), Magnesium (Mg), Potassium (K), Calcium (Ca), Zinc (Zn), Bromine (B) were measured and are given in appendix table XIII to XXX. Sample-wise concentrations of the elements are also presented in graph 3.13 to 3.30 Significance of the elements studied are discussed below.

#### **3.3.1 Concentration of Fluorine (F), Phosphorus (P), Copper (Cu), Manganese (Mn), Magnesium (Mg), Potassium (K), Calcium (Ca), Zinc (Zn), Bromine (B)**

##### **Fluorine (F)**

The F content of soil in the study area lies in the range of 243 to 415 mg/Kg with a mean of 312.5mg/Kg in monsoon season shown in appendix table XIII and concentration vary 200 to 401mg/Kg in winter season shown in appendix table XIV. The highest concentration of F 415mg/Kg is found in the soil of Godagari thana of Rajshahi district in monsoon season and 412mg/Kg is found in the soil of Durgapur thana of Rajshahi district in winter season. The lowest concentration of F 243mg/Kg is found in the soil of Chargat thana of Rajshahi district in monsoon season and 203mg/Kg is found in the soil of Boalia thana of Rajshahi district in winter season. Most of the investigated soil samples have values for F content above 300mg/kg, which is maximum permissible value for the content of this element in agricultural soil (Pravilnik, 1990). Graph 3.13 and 3.14 depicts that the concentrations of F in a few samples are under optimum range and most of them are outside of the optimum range.





Trace elements fluorine is incidental impurities in rock phosphate used in the manufacture of phosphate fertilizer. As a result of the long-term application of phosphate fertilizer, there is the potential for accumulation of fluorine in agriculture soils over time, especially at sites where there are relatively large inputs of phosphate fertilizers.

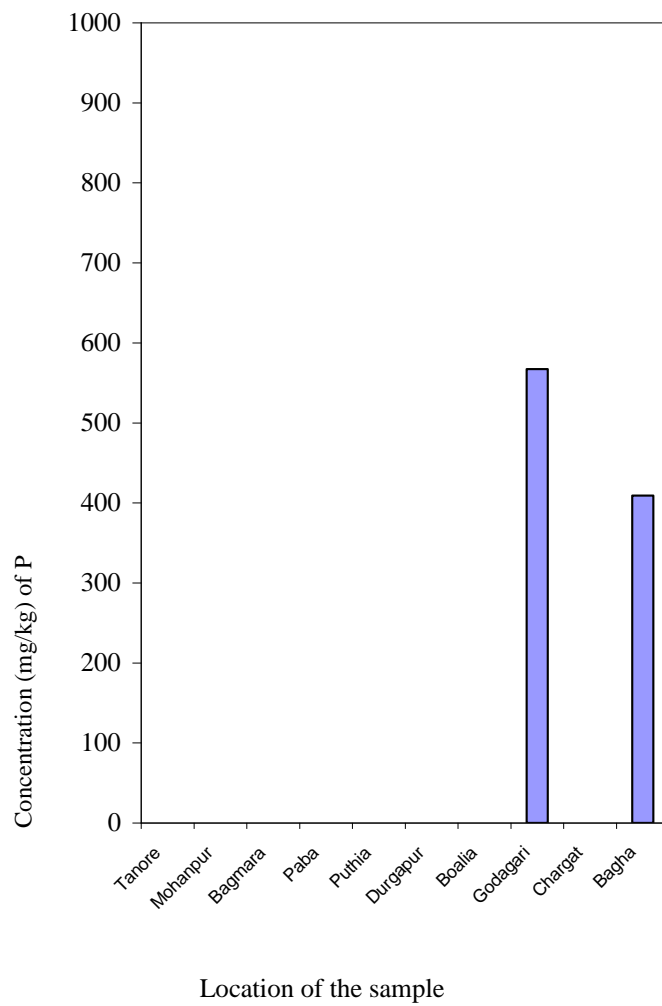
The microorganism, plants, and animals that exist in the biotic environment would be affected in the long run due to this high concentration of F. High concentration of fluorine will also affect the ecosystem of the study area which may bring heavy sufferings for the inhabitants of the area for a long span of time. Microorganisms, insects and even the birds and reptiles would be affected resulting in various diseases. Grasses, vegetables and plants deposit a huge amount of fluorine and animals depending on these suffer from different musculoskeletal diseases, heart diseases, bone fractures, stomachaches, kidney and lung damages.

Since F toxicity has harmful effects on microorganisms, insects, plants and animals, its concentration should always be routinely measured to keep the concentration within the optimum range.

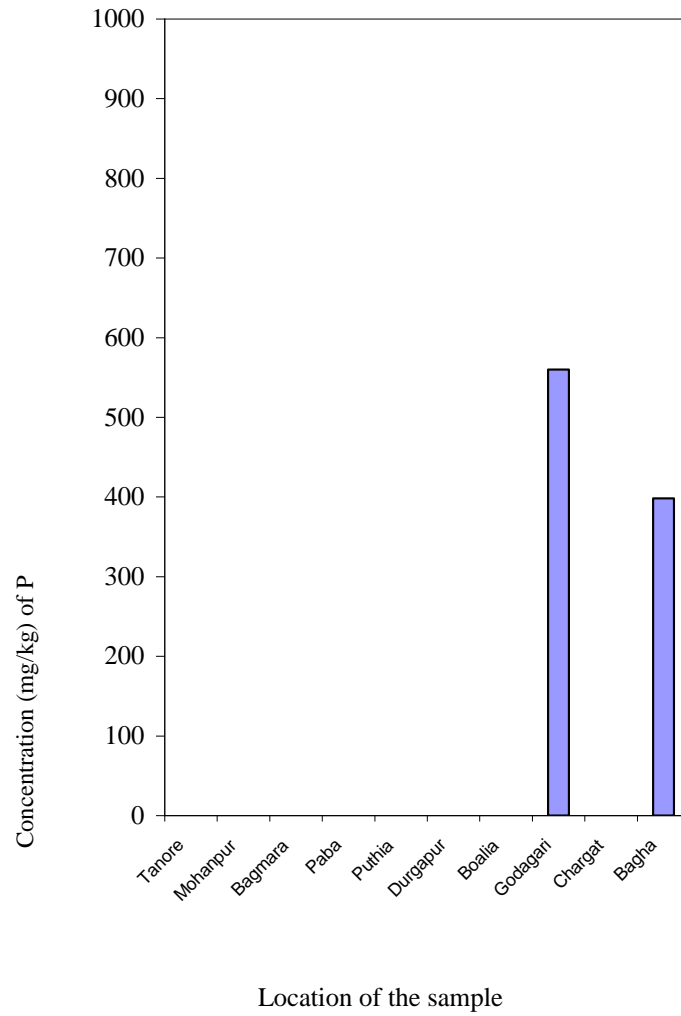
The optimum range of fluorine in soil is 300mg/kg, and in water it is 1 mg/L. By maintaining the concentrations within the optimum range, different fluorine originated diseases can be avoided (Newman, 1984).

### **Phosphorus (P)**

Sample wise concentrations of P are shown in appendix table XV and XVI. The optimum requirement of P in arable soil is 800mg/kg (Underwood, 1997). In the present study P was found in only two samples (567mg/kg and 409mg/kg) out of ten samples in monsoon season and (560 mg/kg & 398 mg/kg) in winter season and which is much lower than the optimum value shown in graph 3.15 & 3.16. So, P enriched fertilizer should be used in the study area and the farmer of this area should be made aware about the P deficiency of their lands.







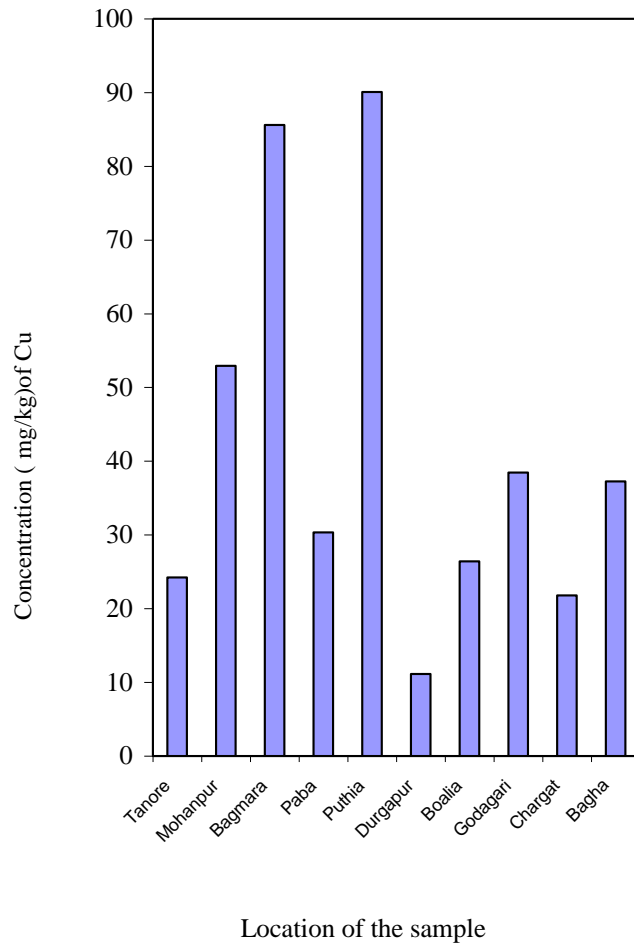
### **Copper (Cu)**

In the present study concentration vary from 11.12 to 90.06 mg/kg shown in appendix table XVII in monsoon season and vary from 20.12 mg/Kg to 80 mg/Kg in winter season shown in appendix table XVIII. Highest concentration was found in Puthia thana of Rajshahi district and lowest concentration was found in Tanore thana of Rajshahi district in monsoon season. Highest concentration was found in Bagmara thana and lowest concentration was found in Tanore thana of Rajshahi thana in winter season shown in graph 3.17 and 3.18 respectively.

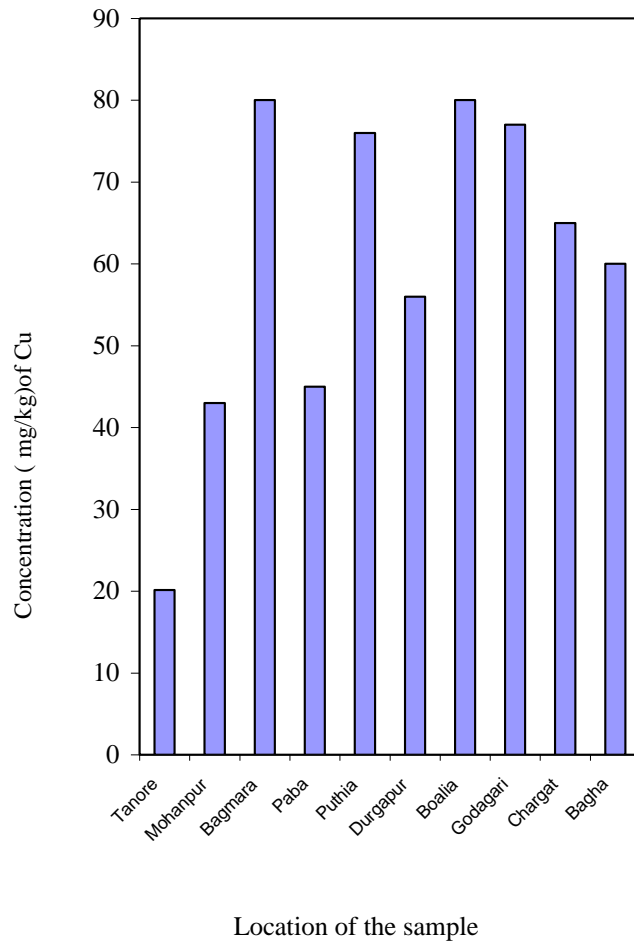
Cu is found in relatively high levels in mafic rocks (60-120 mg/kg) and argillaceous sediments (40-60 mg/kg), and in much lower levels in limestones (2-10 ppm). Values for soil contents vary but generally range

from 1-40mg/kg with an average of 9 mg/kg in the United States (Tisdale *et al.*, 1985). Mean copper contents for uncontaminated soils world wide range from 13-24mg/kg, but the overall range for world soils is higher (1-140mg/kg) depending on the nature of the soil parent materials. (Kabata-Pendias and Pendias, 1992).

So, the investigated soils in the present study lie between the world wide soil ranges. Without adequate Cu, plants will fail to grow properly. Therefore, maintaining fair amounts of Cu for the agriculture is important. Cu is an important component of proteins found in the enzymes that regulate the rate of many biochemical reactions in plants. Plants would not grow without the presence of these specific enzymes.



Graph 3.17 Sample wise distribution of Cu in the arable soil of Rajshahi district in monsoon season.

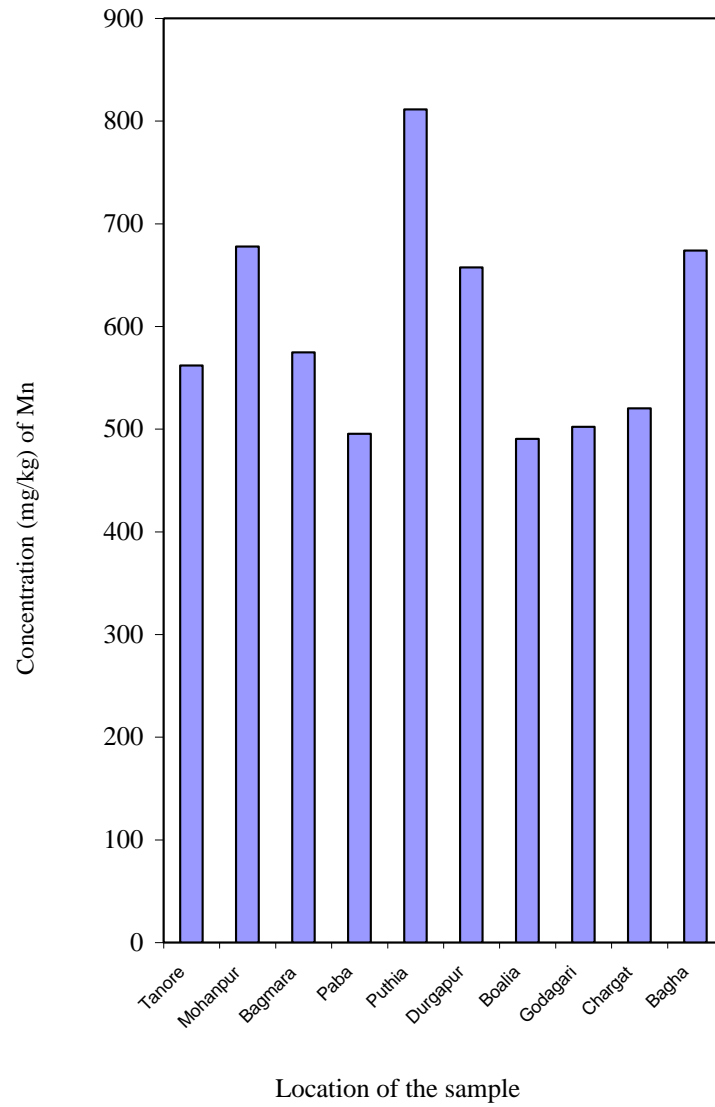


Graph 3.18 Sample wise distribution of Cu in the arable soil of Rajshahi district in winter season.

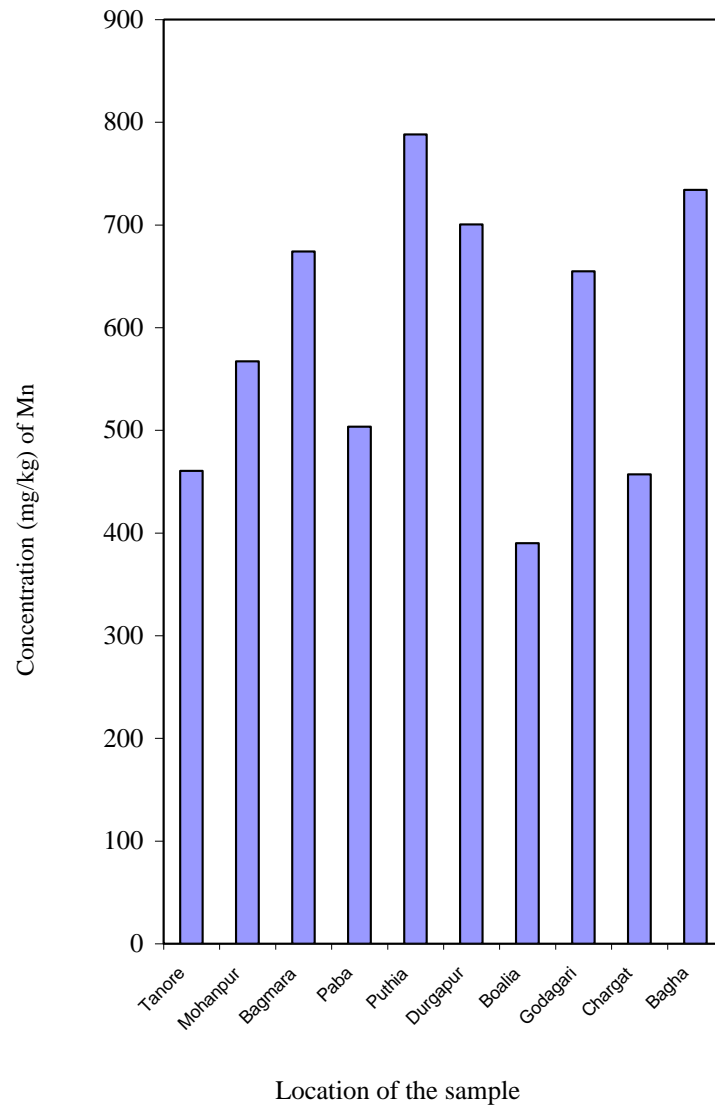
### **Manganese (Mn)**

In the present study concentration vary from 490 to 811mg/kg in monsoon season shown in appendix table XIX and concentration vary from 500 to 711mg/Kg shown in table XX in winter season. Highest concentration was found in Puthia thana of Rajshahi district in monsoon and also in winter season shown in graph 3.19 and 3.20 respectively. Lowest concentration was found in Boalia thana of Rajshahi district in monsoon season and also in winter season.

There is a wide range of Mn in world soils (7-9200mg/kg).The global average for Mn in soil has been estimated as 437mg/kg (Kabata-Pendias and Pendias, 2001). So the investigated soil in the present study higher than the global average range. Soil samples of the study area are found to be rich in Mn.



Graph 3.19 Sample wise distribution of Mn in the arable soil of Rajshahi district in monsoon season.



Graph 3.20 Sample wise distribution of Mn in the arable soil of Rajshahi district in winter season.

### **Magnesium (Mg)**

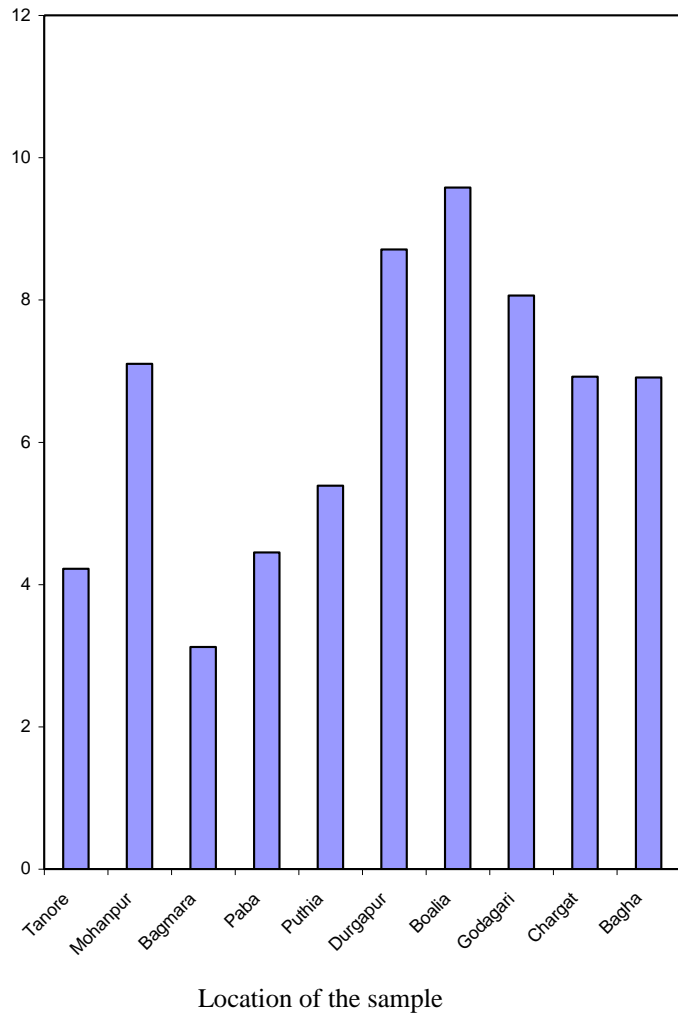
For particular element, PIGE reaction may occur with all its isotopes present in the sample. In the case of Mg, the relative abundances of 79% ( $^{24}\text{Mg}$ ), 10% ( $^{25}\text{Mg}$ ), and 11% ( $^{26}\text{Mg}$ ), Total magnesium is calculated from the relative abundance of  $^{24}\text{Mg}$ . Sample wise distributions of Mg are given in appendix table XXI & XXII and in graph 3.21 & 3.22 respectively.

Mg is present in plant pigment chlorophyll, making plant foods a rich source of this mineral. In the present study concentration of Mg in soil lies in the range of 3.12 g/kg to 9.58 g/kg with a mean of 6.44 g/kg in monsoon and lies in the range 3.22 g/kg to 10.45 g/kg with a mean 6.43 g/kg in winter season. The optimum being 6g/kg (Hoque and Khaliquzzaman, 2002). Mg is found in all the 10 samples analyzed and the soil in the region is slightly Mg rich.

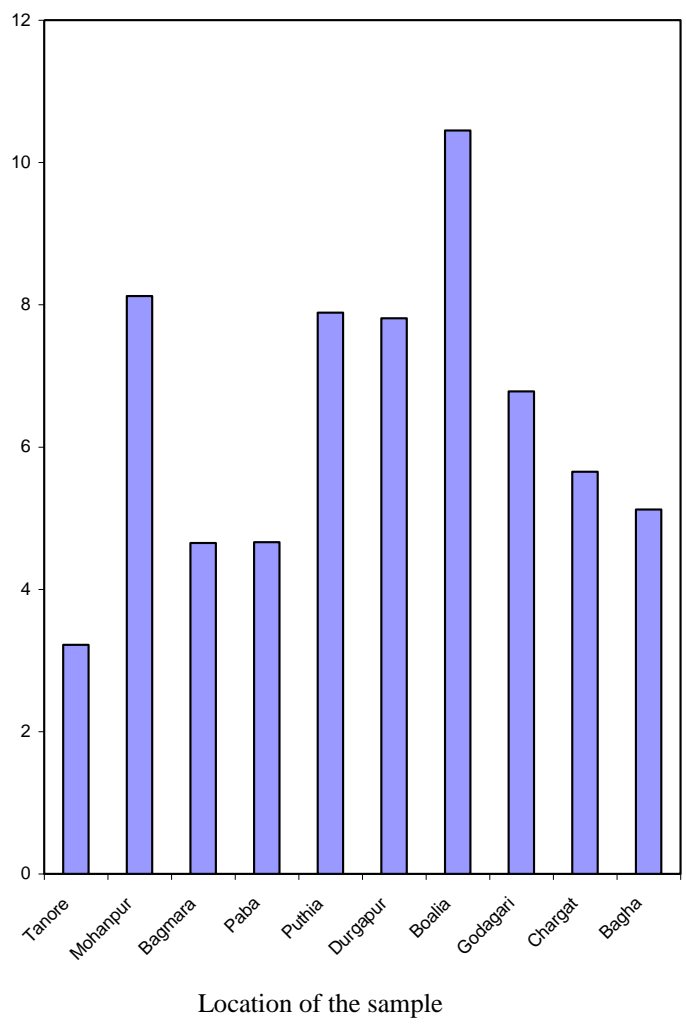
### **Potassium (K)**

In the present study concentration varies from 47 to 245mg/kg in monsoon season and concentration varies from 55 to 180 mg/kg in winter season shown in appendix table XXIII & XXIV and graph 3.23 & 3.24 respectively. Highest concentration was found in Baghat thana of Rajshahi district in monsoon and also in dry season. Lowest concentration was found in Boalia thana of Rajshahi district in monsoon and Durgapur thana in winter season. There is a wide range of K in world soils (78-273 mg/kg) (Tandon and Sekhon, 1988). So the investigated soils in the present study lower than the global range. Potash fertilizer should be used in the study area and the farmer of this area should be made aware about the Potassium deficiency of their lands.

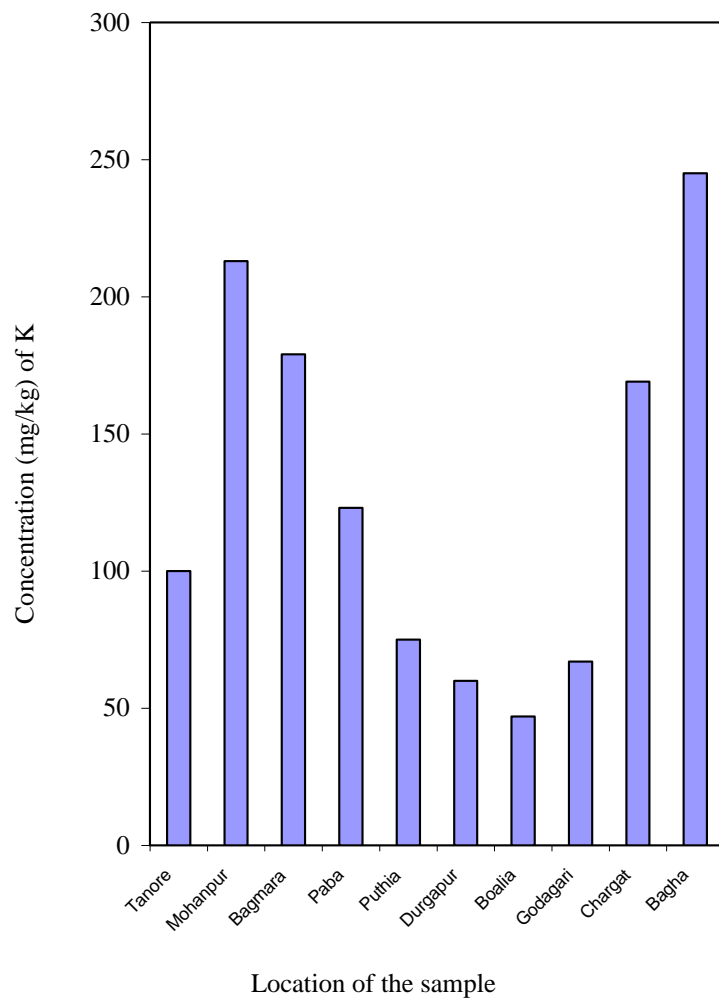




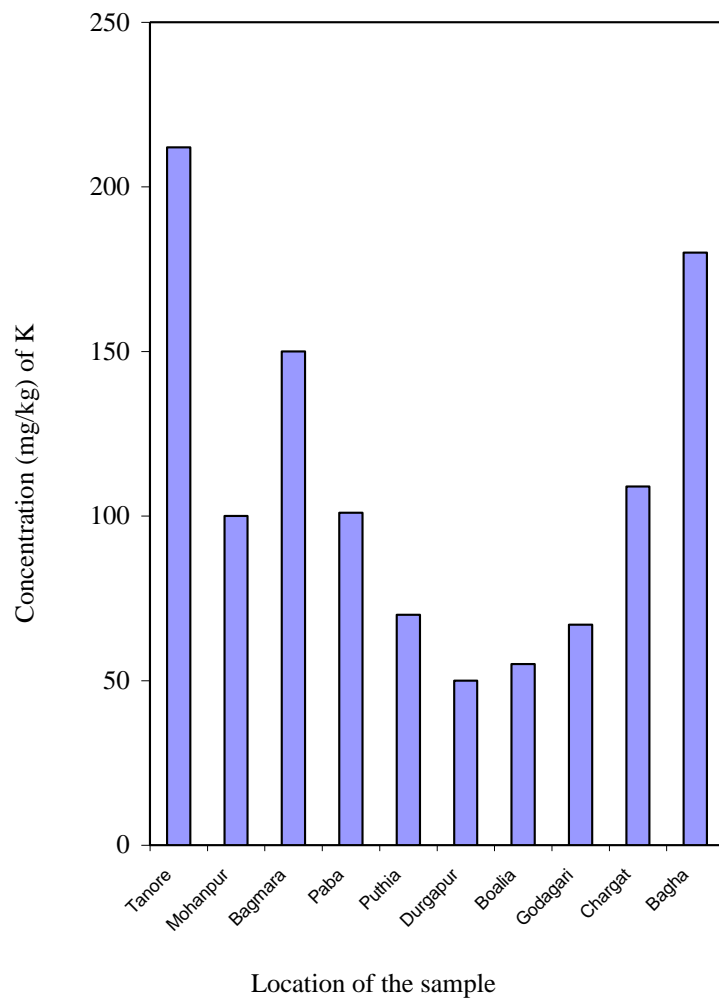
Graph 3.21 Sample wise distribution of Mg in the arable soil of Rajshahi district in monsoon season.



Graph 3.22 Sample wise distribution of Mg in the arable soil of Rajshahi district in winter season.



Graph 3.23 Sample wise distribution of K in the arable soil of Rajshah district in monsoon season.



Graph 3.24 Sample wise distribution of K in the arable soil of Rajshah district in winter season.

### **Calcium (Ca)**

In the present study concentration vary from 145 to 389mg/Kg in monsoon season shown in appendix table XXV and 100 to 364mg/Kg in winter season shown in appendix table XXVI. Highest concentration was found in Bagha thana of Rajshahi district in monsoon season and Charghat thana in winter season. Lowest concentration was found in Paba thana of Rajshahi district in monsoon season and Durgapur thana in winter season. shown in graph 3.25 and 3.26 respectively.

There is a wide range of Ca in world soils (78-273mg/kg) (Tandon and Sekhon, 1988). The investigated soils in the present study higher than the global range. So the soils of this region are slightly Ca rich.

### **Zinc (Zn)**

The concentration of Zn varies from 34.96 to 86.64mg/kg with mean concentration is 60.72 mg/Kg in monsoon season shown in appendix table XXVII and concentration vary from 30.25 to 89.45 mg/Kg with a mean 57.80 in winter season shown in appendix table XXVIII. The highest concentration found in Bagha thana of Rajshahi district in monsoon season and Bhaga thana in winter season. Lowest concentration found in Godagari thana of Rajshahi district in monsoon season and Godagari thana in winter season shown in graph 3.27 and 3.28 respectively.

The mean Zn content in soils of the world ranges from 27 to 235 mg/kg.

with mean zinc concentration of 64 mg/kg (Kabata-Pendias and Pendias,1992) . So the investigated soil in the present study lower than the global average range. Soil samples of the study area are found to be poor in Zn .

Zn-deficient soils can be easily treated with zinc fertilizers to provide an adequate supply of Zn to crops. Several different Zn compounds are utilized as fertilizers but Zn sulphate is by far the most widely used. An adequate supply of Zn is essential for obtaining cost effective yields of crops all over the world. The cost to the farmer of lost production is high but the expense of applying Zn fertilizer when crop symptoms, soil tests or plant analysis show that they are required is relatively low.

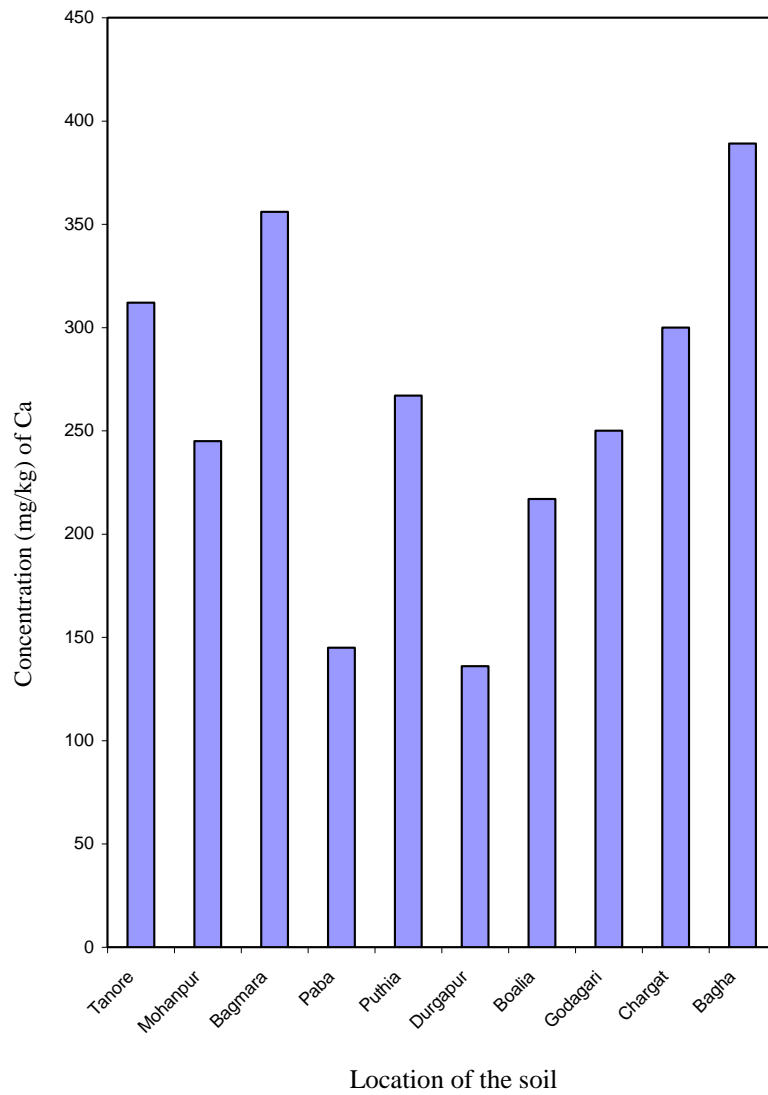
No farmer in areas where soils have been shown to be deficient can afford not to maintain an adequate Zn status in his soils.

However, Zn deficiency is not just a problem in developing countries, it occurs in most of the states in the USA, in Australia, in parts of Europe and many other technologically advanced countries. The main difference

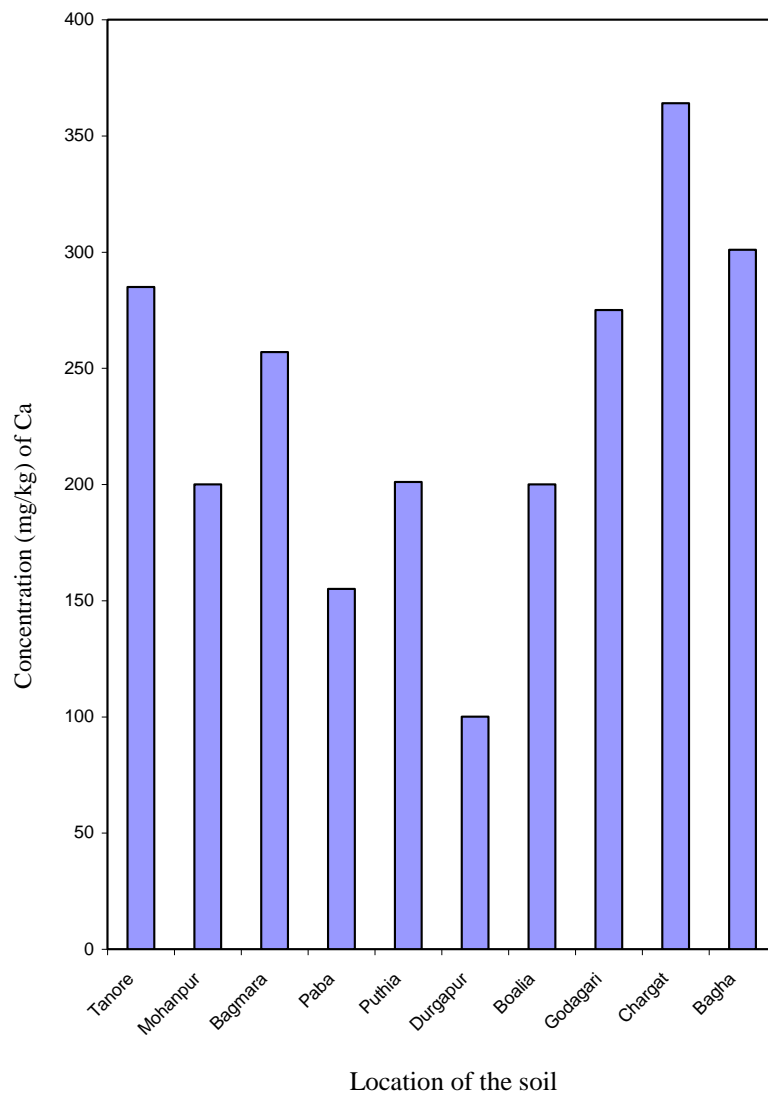
between the situation in these countries and in the developing countries is the existence of an extension agronomy service and rapid access to analytical facilities to minimize time loss before unsuspected conditions can be diagnosed and treated, thus helping to reduce yield losses.

### **Boron (B)**

The concentration of B varies from 1.12 to 2.89mg/kg in monsoon season shown in appendix table XXIX and 1.11 to 2.74mg/Kg in winter season shown in appendix table XXX. The highest concentration found in Durgapur thana of Rajshahi district and lowest concentration found in Bagmara thana of Rajshahi district in monsoon season shown in graph 3.29. The highest concentration found in Durgapur thana of Rajshahi district and lowest concentration found in Godagari thana of Rajshahi district in winter season shown in graph 3.30. Being an important micronutrient, 1mg/kg boron in soil is sufficient for normal growth of plant (Miller and Donahue, 1997). B is rich in all the soil samples of the present studied.

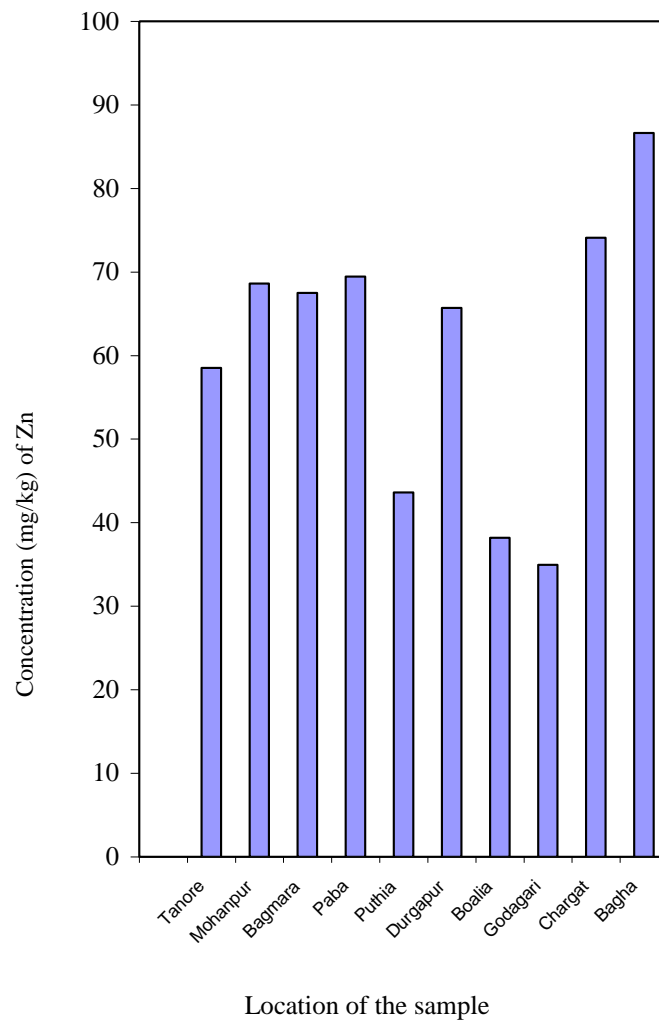


Graph 3.25 Sample wise distribution of Ca in the arable soil of Rajshahi district in monsoon season.

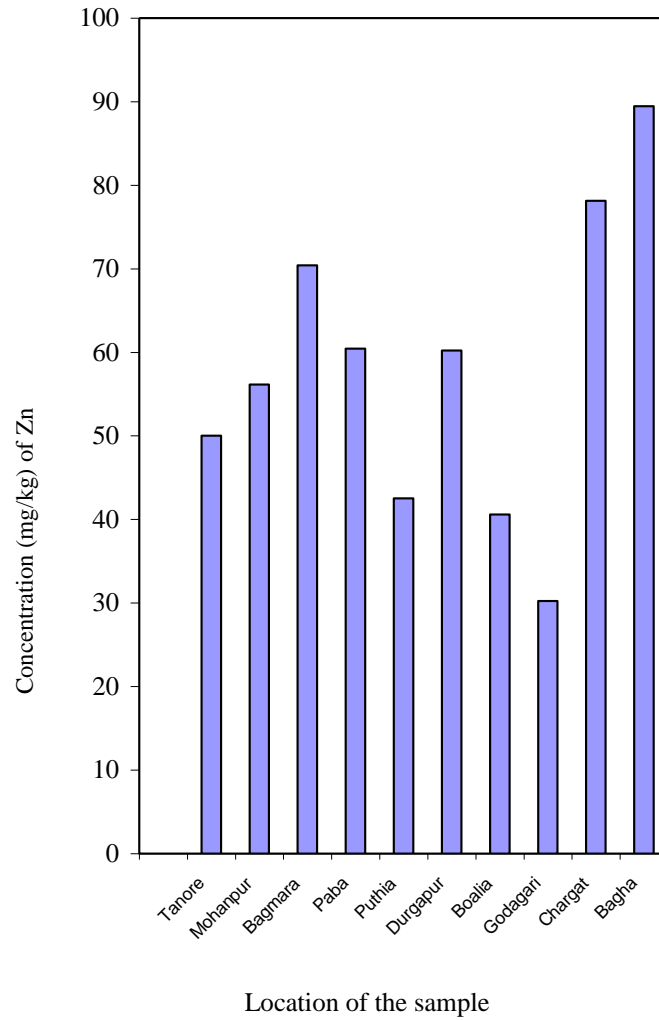


Graph 3.26 Sample wise distribution of Ca in the arable soil of Rajshahi district in winter season.

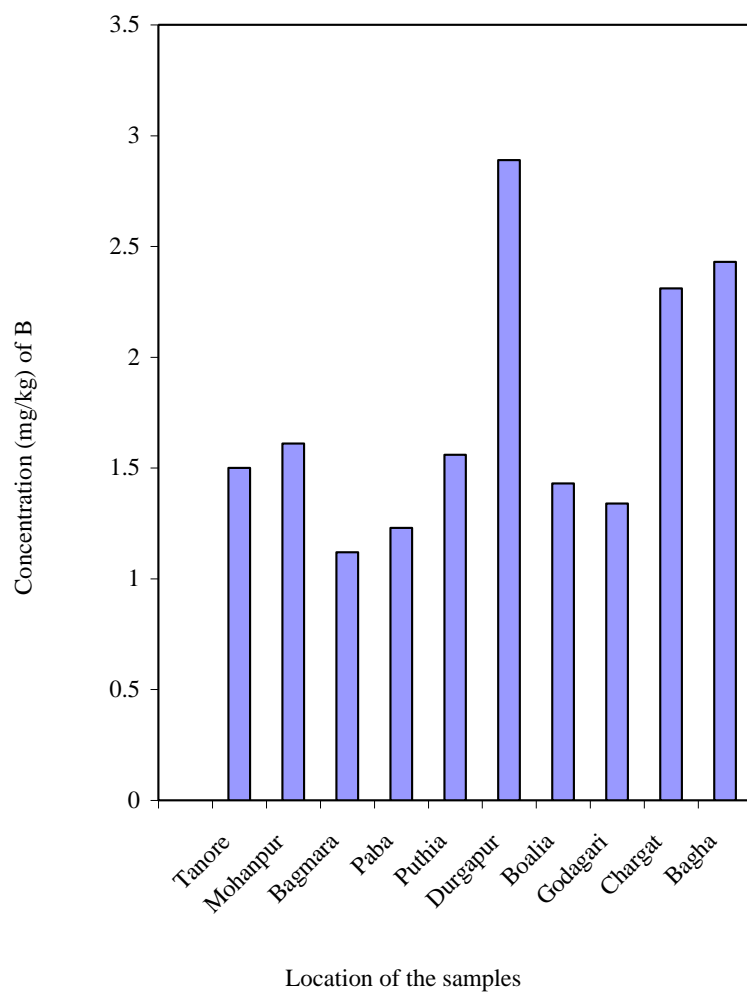




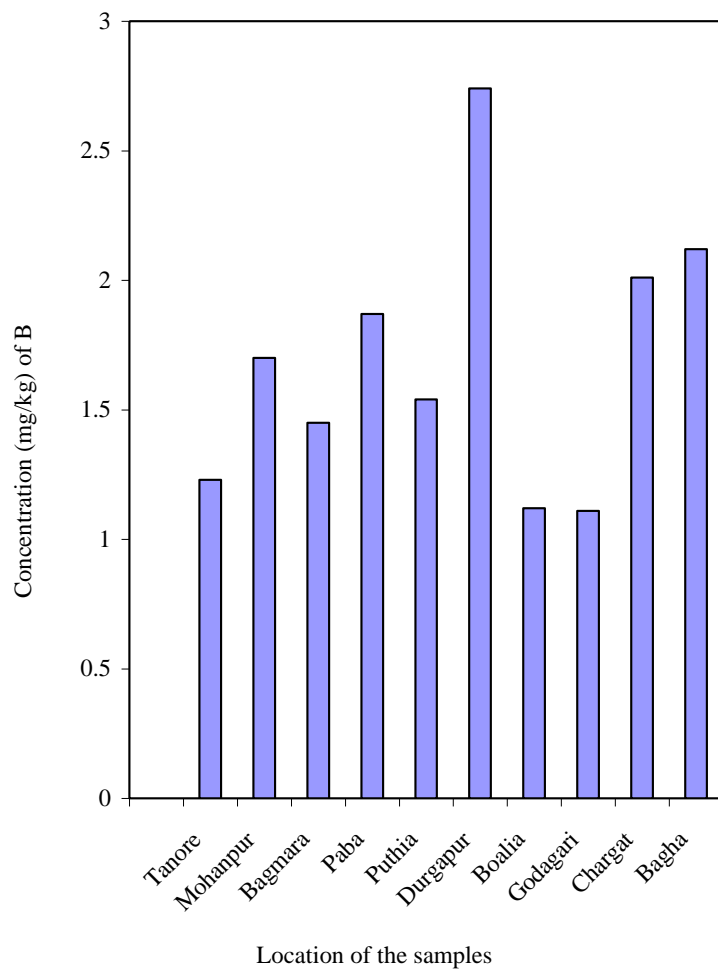
Graph 3.27 Sample wise distribution of Zn in the arable soil of Rajshahi district in monsoon season.



Graph 3.28 Sample wise distribution of Zn in the arable soil of Rajshahi district in winter season.



Graph 3.29 Sample wise distribution of B in the arable soil of Rajshahi district in monsoon season.



Graph 3.30 Sample wise distribution of B in the arable soil of Rajshahi district in winter season.

### 3.4 Fruits and Vegetables Samples Analysis

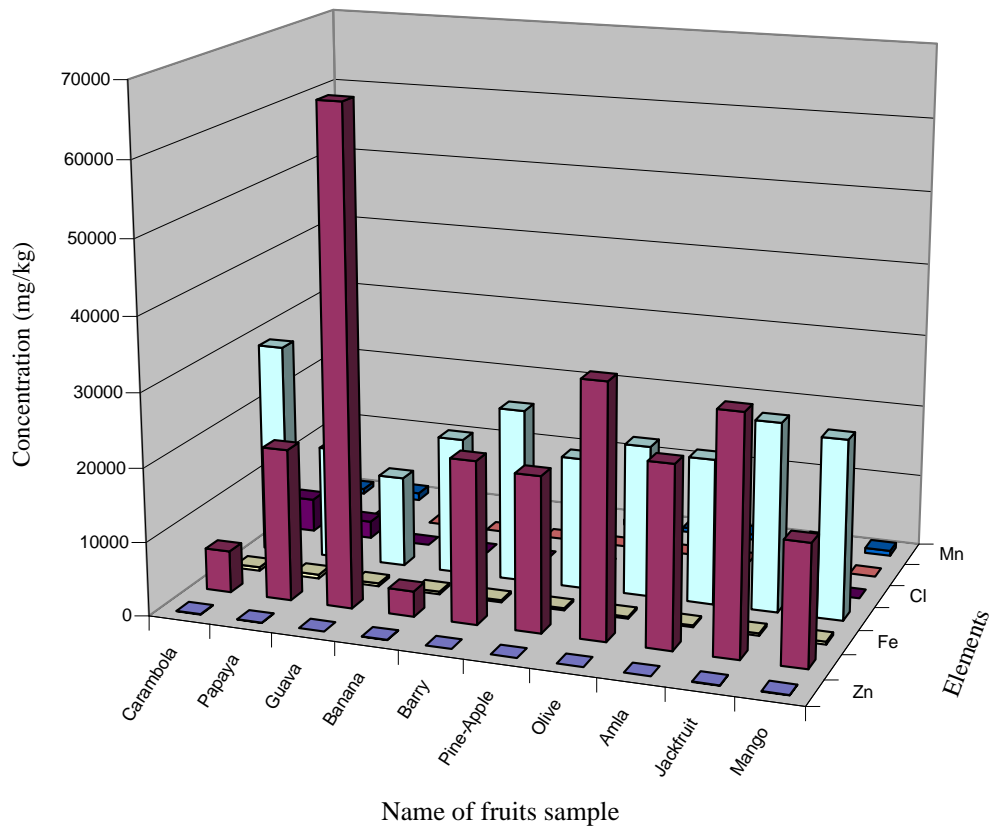
Locally produced 10 fruits and 10 vegetables samples were analyzed and the concentration of the elements are given in appendix table 31 to 32 respectively. Concentration of different elements in fruits and vegetables are also shown in graph 3.31 and 3.32 respectively.

Appendix table XXXI and XXXII summarizes the elemental concentrations of each fruits and vegetables in mg/kg under study. The elements determined in varying concentrations include Zn, Ca, Fe, K, Cl, Cu, Mn and bromine Br. From appendix table XXXI and XXXII, it is seen that the concentrations of the elements vary from one sample to another. The difference in the concentration of these elements within different fruits and vegetables is attributed to factors such as the preferential absorbability of a particular fruit and vegetable for the corresponding elements, the age of the plant, the mineral composition of the soil, in which the plant grows as well as its ambient climatologically conditions (Serfor-Arrnah *et al.*, 2001) . The chlorine ion itself may be important for the blood pressure-raising action of sodium. One research study suggests that a high intake of chloride ions, which tend to remain in the body, may trap positive sodium ions to balance the chemical charge. The minimum chloride requirement for a healthy adult is 700mg/day (Worldlaw, 1999). The deficiency of chlorine causes anorexia, weakness, growth failure, severe convulsions, etc. Highest Cl content is found in vegetables in Potato 7890mg/kg (appendix table XXXII) and Carambola 4567mg/kg (appendix table XXXI) in fruit. It is clear that K and Ca are the most abundant detected elements in the samples (major elements). It is observed from appendix table XXXI to XXXII that in fruits Guava contains the highest amount of Ca 66800mg/kg (appendix table XXXI) and Carambola contain highest of K 28950mg/kg (appendix table XXXI). In vegetables highest amount of Ca and K are exhibited by Turnip 30541mg/kg and Potato 31098mg/kg (appendix table XXXII) respectively. The dietary recommended intakes (DRIs) of K and Ca for adults are 4500 and 1300mg/day respectively (Dietary Reference Intakes, 2004). K has an important in regulation of

the water balance of the body and crucial to heart function (Anderson *et al.*, 2008). Ca is an essential element for maintaining healthy bones and teeth and also of neuromuscular systematic and cardiac functions.

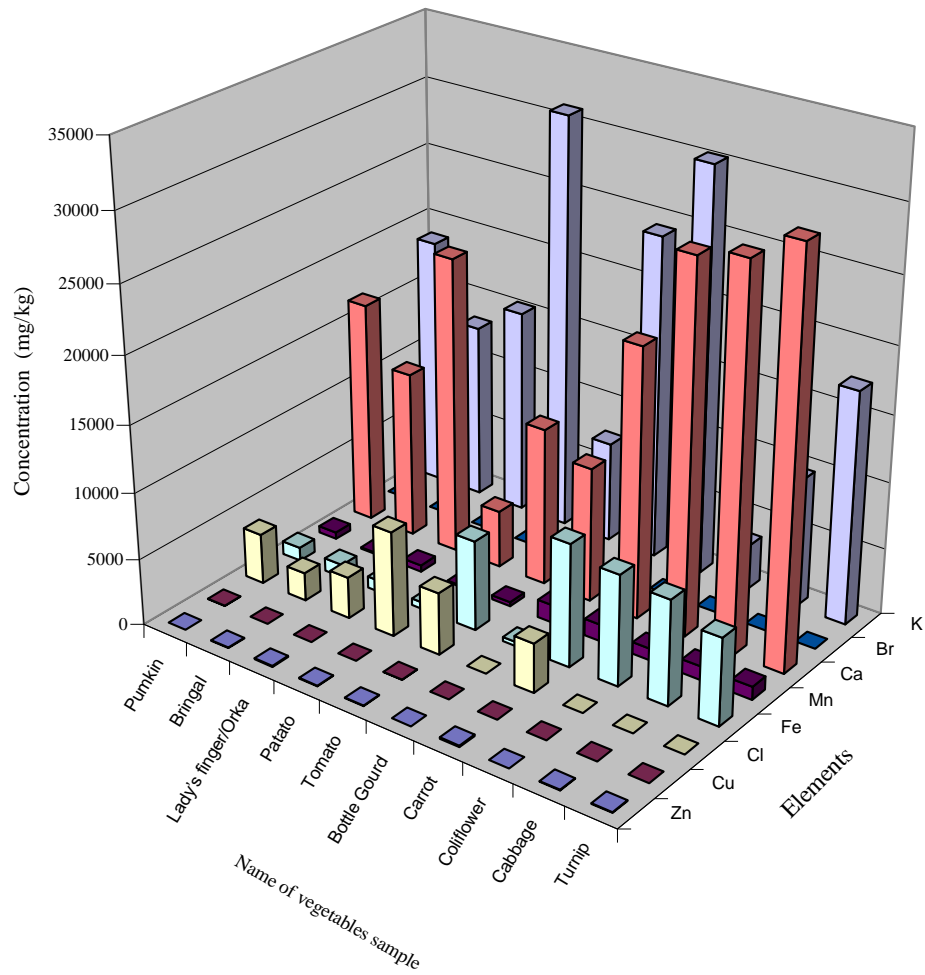
Transition elements Mn, Fe, Cu and Zn are found in varying concentrations in the studied fruits and vegetables. These and other essential elements perform various complementary vital functions in the body to keep the organism healthy. The Fe content fairly good in all fruits samples (varying from 250.30mg/kg in Amla to 545mg/kg in Papaya in appendix table XXXI). The Fe content fairly good in also all vegetables samples (varying from 319mg/kg in Bottle Gourd to 9230mg/kg in Carrot in appendix table XXXII). The daily requirement of Fe is ranged 1.5 - 2.2 mg/day. Worldwide nearly 3.7 million people are iron deficient, the problem being serve enough to cause anemia in two billion people. 35% of the 0 - 5 year old children suffer from Zn or Fe deficiency (Carson *et al.*, 1986). The deficiencies of micronutrients in humans mainly result from their low concentrations and bioavailability in the diet. In this context, the high amount of iron content in these samples can play a significant role in iron deficiency. Zn is an important element responsible for many enzymatic processes and is involved in working of genetic materials, proteins, immune reactions; wound healing, development of the fetus and sperm production. It has been suggested that normal levels of Zn can prevent diarrhea. According to the WHO recommendation fruits and vegetables are poor sources of Zn and ranged up to 1mg/kg and dietary intake for Zn is 10mg/day for an adult (Dietary Reference Intakes, 2004). Zn content in the studied samples is highest in Jackfruit 65mg/kg in fruit (appendix table XXXI) and Carrot 110.05mg/kg in vegetables (appendix table XXXII). Manganese is an antioxidant nutrient and is important in the breakdown of fats and cholesterol and also helps in the nourishment of the nerves and the brain (Hamilton *et al.*, 1988). Mn safe limit for daily intake is 2.0mg/day for an adult (Dietary Reference Intakes, 2004) and its recommended range in fruits and vegetables are 0.42-6.64mg/kg (Chaturvedi *et al.*, 2004). Mn is found in all fruits and vegetables samples. Among them Carrot contains highest amount of Mn 10mg/kg (appendix table XXXII) and in fruit Olive contains highest amount of Mn 9 mg/kg (appendix table XXXI). Cu is an essential

trace element required for proper health in an appropriate limit. Its high uptake in fruits and vegetables can be harmful for human health and in the same way the lower uptake in human consumption can cause a number of symptoms *e.g.* growth retardation, skin ailments, gastrointestinal disorders etc. Copper only found in Carambola 20.56mg/kg (appendix table XXXI) in fruits and Pumpkin 26.87mg/kg (appendix table XXXII) in vegetables respectively. Bromine only found in tomato 118mg/kg in vegetables (appendix table XXXII). Which probably plays a role in prevention of inflammation, given that Cu is a useful anti-inflammatory agent (NRC, 1989). Cu is a factor necessary for the absorption and use of Fe in the formation of hemoglobin, part of many enzymes, and helps in the formation of protective covering around nerves. From appendix table XXXI and XXXII it is found that no samples contain fluorine. So it can be concluded that vegetables and fruits are not a significant source of F uptake of the population of this region.



Graph 3.31 Sample wise distribution of elemental concentration of locally produced fruits.





Graph 3.32 Sample wise distribution of elemental concentration of locally produced vegetables.

### **3.5 Probable effects of the present level of different element on human health**

Effects of Fluorine in human body (Deen, 1942)

#### **Positive effects:**

- Fluorine is one of the 15 essential elements of the human body.
- It is a constituent of bones, teeth, soft tissues and body fluids.
- It confers improved resistance to dental caries.
- Necessary in the maintenance of normal skeleton and in the reduction of osteoporosis in the mature adult population.
- According to WHO survey, fluoridation programs were in progress in 30 countries affecting over 120 million people.
- The water supplies of hundreds of communities have been treated with sodium fluoride or fluorosilicate to maintain the fluorine levels at the range of 0.8 to 1.2 mg/L.

#### **Negative effects**

- Excess fluoride intake causes fluorosis, which is more common than fluorine deficiency.
- The national Institute of Environmental and Health Science in the USA has shown that fluoride causes cancer.
- Fluoride lower the intelligence capacity of humans, especially children are susceptible to early fluoride toxicity.
- Premature aging is the overall effect of fluorosis.
- Molting of teeth is observed from the excessive fluorides (>2mg/L) in drinking water.
- In case of severe fluorosis, dental molting is accompanied by skeletal manifestations including osteosclerosis and calcification of ligaments and tendinous insertions, and leads to deformities such as kyphosis, stiffness of the spine and bony exostoses.
- The Journal of America Medical Association has reported a greater incidence of hip fractures fluoridated areas

Mottling of the teeth enamel is observe among the population who drink

water with fluoride exceeding about 2 mg/L. Fluoride combines to form calcium Fluor apatite in normal hydroxyapatite in the enamel. Damage to ameloblasts to defective matrix formation is seen only when the concentrations of fluorides become exceptionally.

Following prolonged high fluorine intakes the fluorine levels in bones and teeth may become many times higher than normal level. Fluorine being a cumulative bone seeking mineral, the resultant skeleton changes is progressive (Hodge and Smith, 1969). Three stages in this process are teeth/bone fluoridation or chemical fluorosis, teeth/bone mottling, abnormal bone/teeth. The common symptoms of fluorosis are pain on movement, lameness, arthritis of the hip-erosions of teeth enamel and decreased milk yield.

On the other hand, fluorides have been known to influence the prevention of dental caries even though the mechanism of doing so is quite complicated and not yet well understood (Cawson, 1978). The relationship between the prevalence of caries and the fluoride content of drinking water was established on a quantitative basis by the extensive epidemiological surveys carried out by Deen among 7257 children of 12-14 years old, living in 21 cities in the United States. The caries experienced in the permanent teeth was found to decrease as the fluoride concentration in the drinking water was increased above about 1.44 mg/L. Hodge subsequently showed that the decrease in dental caries is a linear function of the fluoride expressed on a logarithmic scale. When considered in relation to the incidence of mottling of the enamel it became evident that the lines intersect at close to fluorine content of 1mg/L in water. This point was therefore considered to be optimal or the point of "Maximum health with maximum safety".

### Health effects of calcium on human body

Calcium is the most abundant metal in the human body: is the main constituent of bones and teeth and it has keys metabolic functions. Calcium is sometimes referred to as lime. It is most commonly found in milk and milk products, but also in vegetables, nuts and beans. It is an essential component for the preservation of the human skeleton and

teeth. It also assists the functions of nerves and muscles. The use of more than 2,5 grams of calcium per day without a medical necessity can lead to the development of kidney stones and sclerosis of kidneys and blood vessels.

A lack of calcium is one of the main causes of osteoporosis. Osteoporosis is a disease in which the bones become extremely porous, are subject to fracture, and heal slowly, occurring especially in women following menopause and often leading to curvature of the spine from vertebral collapse.

Unlike most of the people think, there is an intense biological activity inside our bones. They are being renewed constantly by new tissue replacing the old one. During childhood and adolescence, there's more production of new tissue than destruction of the old one, but at some point, somewhere around the 30 or 35 years of age, the process is inverted and we start to lose more tissue than what we can replace. In women the process is accelerated after the menopause (the period marked by the natural and permanent cessation of menstruation, occurring usually between the ages of 45 and 55); this is because their bodies stop producing the hormone known as estrogen, one of which functions is to preserve the osseous mass.

Evidence suggests that we need a daily intake of 1,000 milligrams of calcium in order to preserve the osseous mass in normal conditions. This is both for man and pre-menopausal women. The recommended daily intake rises to 1,500 for menopausal woman.

Ca works together with magnesium to create new osseous mass. Calcium should be taken together with magnesium in a 2:1 rate, that is to say, if you ingest 1000 mg of calcium, you should also ingest 500 mg of magnesium. Some magnesium sources in the diet are seafood, whole-grains, nuts, beans, wheat oats, seeds and green vegetables. In the present study fruits Guava contains the highest amount of Ca 66800 mg/kg (appendix table XXXI) and in vegetables highest amount of Ca exhibited by Turnip 30541 mg/kg (appendix table XXXII)

#### Health effects of Cl

Cl is a highly reactive gas. It is a naturally occurring element. The largest users of Cl are companies that make ethylene dichloride and other chlorinated solvents, polyvinyl chloride (PVC) resins, chlorofluorocarbons, and propylene oxide. Paper companies use Cl to bleach paper. Water and wastewater treatment plants use Cl to reduce water levels of microorganisms that can spread disease to humans (disinfection).

Exposure to Cl can occur in the workplace or in the environment following releases to air, water, or land. People who use laundry bleach and swimming pool chemicals containing Cl products are usually not exposed to chlorine itself. Cl is generally found only in industrial setting.

Cl enters the body breathed in with contaminated air or when consumed with contaminated food or water. It does not remain in the body, due to its reactivity.

Effects of Cl on human health depend on how the amount of Cl that is present, and the length and frequency of exposure. Effects also depend on the health of a person or condition of the environment when exposure occurs.

Breathing small amounts of Cl for short periods of time adversely affects the human respiratory system. Effects differ from coughing and chest pain, to water retention in the lungs. Cl irritates the skin, the eyes, and the respiratory system. These effects are not likely to occur at levels of chlorine that are normally found in the environment.

Human health effects associated with breathing or otherwise consuming small amounts of Cl over long periods of time are not known. Some studies show that workers develop adverse effects from repeat inhalation exposure to chlorine, but others will not. In the present study highest Cl content is found in vegetables in Potato 7890 mg/kg (table XXXII) and Carambola 4567 mg/kg (table XXXI) in fruit.

#### Health effects of copper (Cu)

Cu can be found in many kinds of food, in drinking water and in air. Because of that we absorb eminent quantities of Cu each day by eating, drinking and breathing. The absorption of Cu is necessary, because Cu is a trace element that is essential for human health. Although humans can handle proportionally large concentrations of Cu, too much Cu can still cause eminent health problems.

Cu concentrations in air are usually quite low, so that exposure to Cu through breathing is negligible. But people that live near smelters that

process copper ore into metal, do experience this kind of exposure.

People that live in houses that still have Cu plumbing are exposed to higher levels of Cu than most people, because Cu is released into their drinking water through corrosion of pipes.

Occupational exposure to Cu often occurs. In the working environment, Cu contagion can lead to a flu-like condition known as metal fever. This condition will pass after two days and is caused by over sensitivity.

### *Effects*

Long-term exposure to Cu can cause irritation of the nose, mouth and eyes and it causes headaches, stomachaches, dizziness, vomiting and diarrhea. Intentionally high uptakes of Cu may cause liver and kidney damage and even death. Whether Cu is carcinogenic has not been determined yet There are scientific articles that indicate a link between long-term exposure to high concentrations of Cu and a decline in intelligence with young adolescents. Whether this should be of concern is a topic for further investigation.

Industrial exposure to Cu fumes, dusts, or mists may result in metal fume fever with atrophic changes in nasal mucous membranes. Chronic copper poisoning results in Wilson's Disease, characterized by a hepatic cirrhosis, brain damage, demyelization, renal disease, and Cu deposition in the cornea. In the present study Copper only found in Carambola 20.56 mg/kg (appendix table XXXI) in fruits and Pumkin

26.87 mg/kg (appendix table XXXII) in vegetables respectively.

#### Health effects of iron (Fe)

Fe can be found in meat, whole meal products, potatoes and vegetables. The human body absorbs iron in animal products faster than Fe in plant products. Iron is an essential part of hemoglobin; the red colouring agent of the blood that transports oxygen through our bodies.

Fe may cause conjunctivitis, choroiditis, and retinitis if it contacts and remains in the tissues. Chronic inhalation of excessive concentrations of iron oxide fumes or dusts may result in development of a benign pneumoconiosis, called siderosis, which is observable as an x-ray change. No physical impairment of lung function has been associated with siderosis. Inhalation of excessive concentrations of Fe oxide may enhance the risk of lung cancer development in workers exposed to pulmonary carcinogens. LD50 (oral, rat) =30 gm/kg. (LD50: Lethal dose 50. Single dose of a substance that causes the death of 50% of an animal population from exposure to the substance by any route other than inhalation. Usually expressed as milligrams or grams of material per kilogram of animal weight (mg/kg or g/kg).)

A more common problem for humans is iron deficiency, which leads to anemia. A man needs an average daily intake 7 mg of iron and a woman 11 mg; a normal diet will generally provided all that is needed.

The Fe content fairly good in all fruits samples (varying from 250.30 mg/kg in Amla to 545mg/kg in Papaya in appendix table XXXI). The Fe content fairly good in also all vegetables samples (varying from 319



mg/kg in Bottle Gourd to 9230mg/kg in Carrot in appendix table XXXII).

### Health effects of Magnesium (Mg)

Humans take in between 250 and 350mg/day of Mg and need at least 200 mg, but the body deals very effectively with this element, taking it from food when it can, and recycling what we already have when it cannot.

There is no evidence that Mg produces systemic poisoning although persistent over-indulgence in taking Mg supplements and medicines can lead to muscular weakness, lethargy and confusion.

Effects of exposure to Mg powder: low toxicity and not considered to be hazardous to health. Inhalation: dust may irritate mucous membranes or upper respiratory tract. Eyes: mechanical injury or particle may embed in eye. Viewing of burning Mg powder without fire glasses may result in "Welder's flash", due to intense white flame. Skin: embedding of particle in skin. Ingestion: unlikely; however, ingestion of large amounts of Mg powder could cause injury. Mg has not been tested, but it's not suspected of being carcinogenic, mutagenic or teratogenic. Exposure to Mg oxide fume subsequent to burning, welding or molten metal work can result in metal fume fever with the following temporary symptoms: fever, chills, nausea, vomiting and muscle pain. These usually occur 4-12 hours after exposure and last up to 48 hours. Mg oxide fume is a by-product of burning Mg.

Physical dangers: Dust explosion possible if in powder or granular form, mixed with air. If dry, it can be charged electrostatically by swirling, pneumatic transport pouring tec.

Chemical dangers: The substance may spontaneously ignite on contact with air or moisture producing irritating or toxic fumes. Reacts violently with strong oxidants. Reacts violently with many substances causing fire and explosion hazard. Reacts with acids and water forming flammable hydrogen gas, causing fire and explosion hazard.

First Aid: Inhalation: remove to fresh air. Eyes: flush eyes with water thoroughly. Consult a physician. Skin: wash with soap and water thoroughly to remove particles. Ingestion: if large amounts of Mg powder are ingested, induce vomiting & consult a physician.

Note to physician: no specific treatment or antidote. Supportive care recommended. Treatment should be based on reactions of the patient.

#### Health effects of manganese (Mn)

Mn is a very common compound that can be found everywhere on earth. Mn is one out of three toxic essential trace elements, which means that it is not only necessary for humans to survive, but it is also toxic when too high concentrations are present in a human body. When people do not live up to the recommended daily allowances their health will decrease. But when the uptake is too high health problems will also occur.

The uptake of Mn by humans mainly takes place through food, such as

spinach, tea and herbs. The foodstuffs that contain the highest concentrations are grains and rice, soya beans, eggs, nuts, olive oil, green beans and oysters. After absorption in the human body manganese will be transported through the blood to the liver, the kidneys, the pancreas and the endocrine glands.

Mn effects occur mainly in the respiratory tract and in the brains. Symptoms of manganese poisoning are hallucinations, forgetfulness and nerve damage. Manganese can also cause Parkinson, lung embolism and bronchitis. When men are exposed to manganese for a longer period of time they may become impotent.

A syndrome that is caused by Mn has symptoms such as schizophrenia, dullness, weak muscles, headaches and insomnia.

Because Mn is an essential element for human health shortages of Mn can also cause health effects. These are the following effects:

- Fatness
- Glucose intolerance
- Blood clotting
- Skin problems
- Lowered cholesterol levels
- Skeleton disorders
- Birth defects
- Changes of hair colour
- Neurological symptoms

Chronic Mn poisoning may result from prolonged inhalation of dust and fume. The central nervous system is the chief site of damage from the disease, which may result in permanent disability. Symptoms include languor, sleepiness, weakness, emotional disturbances, spastic gait, recurring leg cramps, and paralysis. A high incidence of pneumonia and other upper respiratory infections has been found in workers exposed to dust or fume of compounds. Mn compounds are experimental equivocal tumorigenic agents. In present study Mn is found in all fruits and vegetables samples. Among them Carrot contains highest amount of Mn 10mg/kg (appendix table XXXII) and in fruit Olive contains highest amount of Mn 9mg/kg (appendix table XXXI) respectively.

#### Health effects of Potassium (K)

K can be found in vegetables, fruit, potatoes, meat, bread, milk and nuts. It plays an important role in the physical fluid system of humans and it assists nerve functions. K, as the ion  $K^+$ , concentrate inside cells, and 95% of the body's potassium is so located. When our kidneys are somehow malfunctioning an accumulation of potassium will consist. This can lead to disturbing heartbeats.

K can affect you when breathed in. Inhalation of dust or mists can irritate the eyes, nose, throat, lungs with sneezing, coughing and sore throat. Higher exposures may cause a build up of fluid in the lungs, this can cause death. Skin and eye contact can cause severe burns leading to permanent damage. In fruits Carambola contain highest amount of K 28950 mg/kg (appendix table XXXI) and in vegetables highest amount of K are exhibited by Turnip 30541mg/kg and Potato 31098mg/kg

(appendix table XXXII) respectively

### Health effects of Zinc (Zn)

Zn is a trace element that is essential for human health. When people absorb too little zinc they can experience a loss of appetite, decreased sense of taste and smell, slow wound healing and skin sores. Zn-shortages can even cause birth defects.

Although humans can handle proportionally large concentrations of Zn, too much zinc can still cause eminent health problems, such as stomach cramps, skin irritations, vomiting, nausea and anemia. Very high levels of Zn can damage the pancreas and disturb the protein metabolism, and cause arteriosclerosis. Extensive exposure to Zn chloride can cause respiratory disorders. In the work place environment Zn contagion can lead to a flu-like condition known as metal fever. This condition will pass after two days and is caused by over sensitivity.

Zn can be a danger to unborn and newborn children. When their mothers have absorbed large concentrations of Zn the children may be exposed to it through blood or milk of their mothers. In present study Zn content in the studied samples is highest in Jackfruit 65 mg/kg in fruit (appendix table XXXI) and Carrot 110.05mg/kg in vegetables (appendix table XXXII).

Trace elements and their deficiency signs in humans (Chaturvedi *et al.*, 2004).

<u>Element</u>	<u>Deficiency sign</u>
K	Hypertension.
Ca	Hypertension, Osteoporosis, Osteomalacia.
Mn	Decrease in serum cholesterol, triglyceride and phospholipid.

Fe	Anaemia, Hypoferremia.
Cu	Anaemia, hypocupremia, Menke's kinky hair syndrome.
Zn	Hypogonadism, Acrodermatitis enteropathica, diabetes mellitus.
F	Dental caries

### **Importance of elemental analysis at trace level**

Twenty six (26) of the naturally occurring elements are known to be essential for life (Underwood, 1997). Eleven of the major elements, namely, boron, carbon, hydrogen, oxygen, nitrogen, sulfur, calcium, phosphorus, potassium, sodium, chlorine and magnesium are low Z elements. And light elements from the bulk (>90%) of biological materials.

All trace elements are toxic if ingested or inhaled at sufficiently high levels and for long enough periods. The major part of the earth's crust consists of light elements are Si, Al, Na, Mg, Li, Be, K, etc. The most of the industrially and commercially important samples are formed from the light elements. For example, the technical glasses are basically composed of oxygen as anion and eleven cations such as B, Na, Mg, Al, Si, P, K, Ca, Zn, Ba and Pb which mainly determine their properties, most of them  $Z < 21$ . Additional cations are only of minor importance. According materials science, light elements play the important role in semiconductor materials; ceramics, lithium-intercalated compounds for the light elements present in the practical specimen is of much interest.

Physical methods, such as NAA, RBS and PIXE have proved to be of value in elemental analysis of the samples of different origin. PIGE method, is however, emerging as an analytical method of choice in verities of materials for determining light elements (Khan *et al.*, 1979).

Among 109 elements in the periodic table at least 36 are known to be associated in one way or another with the fabric of life. From these are derived all the compound chemical structures in the biological systems. The contamination of water supply and foods with trace elements arising from various sources may have, in the long run, deleterious effects on the health and welfare of human population of a country.

This concern has stimulated increasing interest in the study of the trace elements related to the human health and their movement in the environment and to determine their maximum permissible intakes by human through the food chain and drinking water. Realizing the importance of their roles in human health, elemental analysis of samples in various fields is going on in many laboratories all over the world. In the present study 10 water, 10 soil, 10 vegetables and 10 fruit samples were analyzed by proton induced X-ray emission and proton induced  $\gamma$ -ray technique by tandem accelerator. Fluorine has attracted much attention in recent years because of its apparent role in human health. Fluoride ions are readily absorbed by plants, especially from the more acid types of soils, but it is highly toxic if present at a concentration as low as 10mg/L for several species of plants (Hoque, 2000). That is why special emphasis was given in fluorine measurement in all types of samples. The analytical results are presented in the following section.

CONCLUSION



## CONCLUSION

PIXE and PIGE techniques have been used for the elemental analysis of Water, Soil, Fruits and Vegetables from Rajshahi district of Bangladesh.

- The F of ground water contains higher F than the recommended maximum which indicated the probability of F problem in drinking water for the population of Rajshahi region. 100% of the surveyed samples have Mn concentrations exceeding the Bangladesh drinking water standard but do not exceed the WHO health-based guideline value. The 80% of the studied samples contain higher Iron than the recommended maximum. 100% of the studied samples contain higher Ca, Cu and Zn content of ground water found lower than the recommended maximum.
- In the present study P was found in only two out of ten samples and which is much lower than the optimum value. So, P enriched fertilizer should be used in the study area and the farmer of this area should be made aware about the P deficiency of their lands. The concentration of Cu lie between the world wide soil ranges. Soil samples of the study area are found to be rich in Mn , Mg, Ca and B. The investigated soils in the present study lower than the global range of Potassium so Potash fertilizer should be used in the study area and the farmer of this area should be made aware about the K deficiency of their lands. Soil samples of the study area are found to be poor in Zn. Zn-deficient soils can be easily treated with zinc fertilizers to provide an adequate supply of zinc to crops. Several different zinc compounds are utilized as fertilizers but zinc sulphate is by far the most widely used
- Vegetables and fruits samples were analyzed but no samples contain fluorine. So it can be concludes that vegetables and fruits are not a significance source of F uptake of the population of this region. It is clear that K and Ca are the most abundant detected elements in the samples. It is observed that in fruits Guava contains the highest amount of Ca (66800mg/kg) and Carambola contain highest of K (28950mg/kg). The Fe content fairly good in all fruit samples (varying from 250.30mg/kg in Amla

to 545mg/kg in Papaya) and all vegetables samples (varying from 319mg/kg in Bottle Gourd to 9230mg/kg in Carrot). Zn content in the studied samples is highest in Jackfruit (65mg/kg) in fruit and Carrot 110.05 mg/kg in vegetables. Mn is found in all fruits and vegetables samples. Among them in vegetables Carrot contains highest amount (10mg/kg) of Mn and in fruits Olive contain highest amount (9mg/kg) of Mn. Copper content in the studied samples is found only in Pumpkin (26.87 mg/kg) in vegetables and Carambola (20.56 mg/kg) in fruit which probably plays a role in prevention of inflammation, given that Cu is a useful anti-inflammatory agent. Highest Cl content is found in vegetables in Potato (7890mg/kg) and Carambola (4567mg/kg) in fruit. The minimum chloride requirement for a healthy adult is 700mg/day. The deficiency of chlorine causes anorexia, weakness, growth failure, severe convulsions. Bromine only found in tomato (118mg/kg) in vegetables. No toxic heavy metals were detected.

Fruits and vegetables contain elements of vital importance in man's metabolism and that are needed for growth and developments, prevention and healing of diseases. It is hoped that the data from this study provide a preliminary assessment of the background levels of selected trace and essential elements of Rajshahi district. Therefore such studies will help us to understand the environmental pollution and its effects on human health. In the developed countries such analysis are done routinely to ensure the good health and quality of life of their citizens.

# RECOMENDATION

## **RECOMENDATION**

Like Arsenic (As) problem in Bangladesh, pollution of F in ground water is also a major problem. It is also very important to identify areas unacceptable levels of F and to develop water treatment technologies accordingly.

Detection of high concentrations of Mn in groundwater has added a new dimension to the already difficult safe water supply scenario in Bangladesh. However, Mn issue has attracted relatively less attention so far in the water supply sector. It is important to raise awareness among the stakeholders about the Mn issue. It is also very important to identify areas unacceptable levels of Mn and to develop water treatment technologies accordingly.

This will also help to evaluate and implement the future action plans for health service of the country.

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# APPENDICES

**Appendix Table I:** Sample wise distribution of F in the water of Rajshahi district in monsoon season.

Sample no.	Location	Concentration of F (mg/L)	Average Concentration of the present study (mg/L)	Maximum recommended value for Bangladesh (mg/L)( <b>Bangladesh Gazette, 1998</b> ).
AW-1	Tanore	0.96	1.44	1.00
AW-2	Mohanpur	1.02		
AW-3	Bagmara	1.60		
AW-4	Paba	2.00		
AW-5	Puthia	1.32		
AW-6	Durgapur	1.96		
AW-7	Boalia	1.52		
AW-8	Godagari	2.01		
AW-9	Chargat	0.95		
AW-10	Bagha	1.09		

**Appendix Table II:** Sample wise distribution of F in the water of Rajshahi district in winter season.

Sample no.	Location	Concentration of F (mg/L)	Average Concentration of the present study (mg/L)	Maximum recommended value for Bangladesh (mg/L)( <b>Bangladesh Gazette, 1998</b> ).
AW-1	Tanore	1.05	1.27	1.00
AW-2	Mohanpur	0.87		
AW-3	Bagmara	0.56		
AW-4	Paba	1.21		
AW-5	Puthia	1.58		
AW-6	Durgapur	0.96		
AW-7	Boalia	2.45		
AW-8	Godagari	1.25		
AW-9	Chargat	1.96		
AW-10	Bagha	0.87		

**Appendix Table III:** Sample wise distribution of Mn in the water of Rajshahi district in monsoon season.

Sample no.	Location	Concentration of Mn (mg/L)	Average Concentration of Mn (mg/L)	Bangladesh standard (mg/L) (WHO, 2004)	WHO guideline value (mg/L) (WHO, 2004)
AW-1	Tanore	0.33	0.23	0.1	0.4
AW-2	Mohanpur	0.27			
AW-3	Bagmara	0.24			
AW-4	Paba	0.19			
AW-5	Puthia	0.17			
AW-6	Durgapur	0.11			
AW-7	Boalia	0.20			
AW-8	Godagari	0.25			
AW-9	Chargat	0.27			
AW-10	Bagha	0.26			



**Appendix Table IV:** Sample wise distribution of Mn in the water of Rajshahi district in winter season.

Sample no.	Location	Concentration of Mn (mg/L)	Average Concentration of Mn (mg/L)	Bangladesh standard (mg/L) (WHO, 2004)	WHO guideline value (mg/L) (WHO, 2004)
AW-1	Tanore	0.24	0.33	0.1	0.4
AW-2	Mohanpur	0.23			
AW-3	Bagmara	0.51			
AW-4	Paba	0.18			
AW-5	Puthia	0.17			
AW-6	Durgapur	0.35			
AW-7	Boalia	0.58			
AW-8	Godagari	0.34			
AW-9	Chargat	0.39			
AW-10	Bagha	0.29			

**Appendix Table V:** Sample wise distribution of Fe in the water of Rajshahi district in monsoon season.

Sample no.	Location	Concentration of Fe (mg/L)	Average Concentration of Fe (mg/L)	Bangladesh standard (mg/L) (WHO, 2006)	WHO guideline value (mg/L) (WHO, 2006)
AW-1	Tanore	1.5	1.44	1	0.3
AW-2	Mohanpur	1.33			
AW-3	Bagmara	2.03			
AW-4	Paba	0.95			
AW-5	Puthia	0.97			
AW-6	Durgapur	1.11			
AW-7	Boalia	1.56			
AW-8	Godagari	1.85			
AW-9	Chargat	1.52			
AW-10	Bagha	1.61			

**Appendix Table VI:** Sample wise distribution of Fe in the water of Rajshahi district in winter season.

Sample no.	Location	Concentration of Fe (mg/L)	Average Concentration of Fe (mg/L)	Bangladesh standard (mg/L) (WHO, 2006)	WHO guideline value (mg/L) (WHO, 2006)
AW-1	Tanore	1.18	1.29	1	0.3
AW-2	Mohanpur	1.28			
AW-3	Bagmara	1.85			
AW-4	Paba	1.25			
AW-5	Puthia	0.98			
AW-6	Durgapur	0.99			
AW-7	Boalia	0.86			
AW-8	Godagari	1.52			
AW-9	Chargat	1.25			
AW-10	Bagha	1.74			

**Appendix Table VII:** Sample wise distribution of Ca in the water of Rajshahi district in monsoon season.

Sample no.	Location	Concentration of Ca (mg/L)	Average Concentration of Ca (mg/L)	WHO guideline value (mg/L) (WHO, 2006)
AW-1	Tanore	100	94.6	75
AW-2	Mohanpur	96		
AW-3	Bagmara	89		
AW-4	Paba	110		
AW-5	Puthia	80		
AW-6	Durgapur	90		
AW-7	Boalia	115		
AW-8	Godagari	93		
AW-9	Chargat	85		
AW-10	Bagha	88		

**Appendix Table VIII:** Sample wise distribution of Ca in the water of Rajshahi district in winter season.

Sample no.	Location	Concentration of Ca (mg/L)	Average Concentration of Ca (mg/L)	WHO guideline value (mg/L) (WHO, 2006)
AW-1	Tanore	99	100	75
AW-2	Mohanpur	100		
AW-3	Bagmara	95		
AW-4	Paba	120		
AW-5	Puthia	89		
AW-6	Durgapur	98		
AW-7	Boalia	102		
AW-8	Godagari	105		
AW-9	Chargat	97		
AW-10	Bagha	101		

**Appendix Table IX:** Sample wise distribution of Cu in the water of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/L)of Cu	Average Concentration (mg/L)of Cu	Bangladesh standard (mg/L) (GoB, 1997)	Maximum permissible value (mg/L) (Pravilnik, 1990 )
AS-1	Tanore	0.95	0.72	1.3	1
AS-2	Mohanpur	1.1			
AS-3	Bagmara	0.85			
AS-4	Paba	0.45			
AS-5	Puthia	0.56			
AS-6	Durgapur	0.74			
AS-7	Boalia	0.85			
AS-8	Godagari	0.29			
AS-9	Chargat	0.47			
AS-10	Bagha	0.96			

**Appendix Table X:** Sample wise distribution of Cu in the water of Rajshahi district in winter season

Sample no.	Location	Concentration (mg/L) of Cu	Average Concentration (mg/L) of Cu	Bangladesh standard (mg/L) (GoB, 1997)	Maximum permissible value (mg/L) (Pravilnik, 1990 )
AS-1	Tanore	0.90	0.81	1.3	1
AS-2	Mohanpur	1.01			
AS-3	Bagmara	0.80			
AS-4	Paba	0.65			
AS-5	Puthia	0.70			
AS-6	Durgapur	0.82			
AS-7	Boalia	0.79			
AS-8	Godagari	0.81			
AS-9	Chargat	0.49			
AS-10	Bagha	1.13			

**Appendix Table XI:** Sample wise distribution of Zn in the water of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/L) of Zn	Average Concentration (mg/L) of Zn	Maximum permissible value (mg/L) (Pravilnik, 1990 )
AS-1	Tanore	1.20	3.44	5.0
AS-2	Mohanpur	4.10		
AS-3	Bagmara	3.21		
AS-4	Paba	2.25		
AS-5	Puthia	4.52		
AS-6	Durgapur	4.23		
AS-7	Boalia	4.12		
AS-8	Godagari	3.45		
AS-9	Chargat	3.21		
AS-10	Bagha	4.14		



**Appendix Table XII:** Sample wise distribution of Zn in the water of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/L) of Zn	Average Concentration (mg/L) of Zn	Maximum permissible value (mg/L) (Pravilnik, 1990 )
AS-1	Tanore	2.20	3.41	5.0
AS-2	Mohanpur	3.10		
AS-3	Bagmara	3.12		
AS-4	Paba	2.45		
AS-5	Puthia	3.51		
AS-6	Durgapur	4.12		
AS-7	Boalia	3.45		
AS-8	Godagari	4.15		
AS-9	Chargat	4.25		
AS-10	Bagha	3.75		

**Appendix Table XIII:** Sample wise distribution of F in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/Kg) of F	Average concentration of the present study (mg/kg)	Maximum permissible value (mg/Kg) ( <b>Pravilnik, 990</b> )
AS-1	Tanore	275	312.5	300
AS-2	Mohanpur	306		
AS-3	Bagmara	365		
AS-4	Paba	400		
AS-5	Puthia	300		
AS-6	Durgapur	245		
AS-7	Boalia	267		
AS-8	Godagari	415		
AS-9	Chargat	243		
AS-10	Bagha	309		

**Appendix Table XIV:** Sample wise distribution of F in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/Kg) of F	Average concentration of the present study (mg/kg)	Maximum permissible value (mg/Kg) ( <b>Pravilnik, 990</b> )
AS-1	Tanore	230	306.7	300
AS-2	Mohanpur	356		
AS-3	Bagmara	301		
AS-4	Paba	401		
AS-5	Puthia	315		
AS-6	Durgapur	412		
AS-7	Boalia	203		
AS-8	Godagari	250		
AS-9	Chargat	356		
AS-10	Bagha	243		

**Appendix Table XV:** Sample wise distribution of P in the arable soil of Rajshahi district in monsoon season .

Sample no.	Location	Concentration (mg/kg) of P	Optimum requirement of P (mg/kg) (Underwood, 1997)
AS-1	Tanore	0	800
AS-2	Mohanpur	0	
AS-3	Bagmara	0	
AS-4	Paba	0	
AS-5	Puthia	0	
AS-6	Durgapur	0	
AS-7	Boalia	0	
AS-8	Godagari	567	
AS-9	Chargat	0	
AS-10	Bagha	409	

**Appendix Table XVI:** Sample wise distribution of P in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/kg) of P	Optimum requirement of P (mg/kg) (Underwood, 1997)
AS-1	Tanore	0	800
AS-2	Mohanpur	0	
AS-3	Bagmara	0	
AS-4	Paba	0	
AS-5	Puthia	0	
AS-6	Durgapur	0	
AS-7	Boalia	0	
AS-8	Godagari	560	
AS-9	Chargat	0	
AS-10	Bagha	398	

**Appendix Table XVII:** Sample wise distribution of Cu in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/kg) of Cu	Average Concentration (mg/kg) of Cu	Range for world soils (mg/kg) ( <b>Kabata-pendias and Pendias, 1992</b> ).
AS-1	Tanore	24.22	41.8	1-140
AS-2	Mohanpur	52.94		
AS-3	Bagmara	85.6		
AS-4	Paba	30.34		
AS-5	Puthia	90.06		
AS-6	Durgapur	11.12		
AS-7	Boalia	26.42		
AS-8	Godagari	38.46		
AS-9	Chargat	21.78		
AS-10	Bagha	37.25		

**Appendix Table XVIII:** Sample wise distribution of Cu in the arable soil of Rajshahi district in winter season..

Sample no.	Location	Concentration (mg/kg) of Cu	Average Concentration (mg/kg) of Cu	Range for world soils (mg/kg) ( <b>Kabata-pendias and Pendias, 1992</b> ).
AS-1	Tanore	20.12	60.21	1-140
AS-2	Mohanpur	43		
AS-3	Bagmara	80		
AS-4	Paba	45		
AS-5	Puthia	76		
AS-6	Durgapur	56		
AS-7	Boalia	80		
AS-8	Godagari	77		
AS-9	Chargat	65		
AS-10	Bagha	60		

**Appendix Table XIX:** Sample wise distribution of Mn in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/kg) of Mn	Average concentration of the present study (mg/kg)	Global Average (mg/kg) of Mn (Kabata-Pendias and Pendias, 2001)
AS-1	Tanore	561.83	596.48	437
AS-2	Mohanpur	677.72		
AS-3	Bagmara	574.81		
AS-4	Paba	495.31		
AS-5	Puthia	811.22		
AS-6	Durgapur	657.47		
AS-7	Boalia	490.41		
AS-8	Godagari	502.12		
AS-9	Chargat	520.15		
AS-10	Bagha	673.81		



**Appendix Table XX:** Sample wise distribution of Mn in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/kg) of Mn	Average concentration of the present study (mg/kg)	Global Average (mg/kg) of Mn (Kabata-Pendias and Pendias, 2001)
AS-1	Tanore	460.45	593	437
AS-2	Mohanpur	567.20		
AS-3	Bagmara	674.21		
AS-4	Paba	503.45		
AS-5	Puthia	788.12		
AS-6	Durgapur	700.56		
AS-7	Boalia	390.13		
AS-8	Godagari	654.78		
AS-9	Chargat	456.98		
AS-10	Bagha	734.12		

**Appendix Table XX:** Sample wise distribution of Mg in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration(g /kg) of Mg	Average Concentration of the present study (g/kg)	Optimum concentration (g/kg) (Hoque, and Khaliquzzaman, 2002)
AS-1	Tanore	4.22	6.44	6.00
AS-2	Mohanpur	7.10		
AS-3	Bagmara	3.12		
AS-4	Paba	4.45		
AS-5	Puthia	5.39		
AS-6	Durgapur	8.71		
AS-7	Boalia	9.58		
AS-8	Godagari	8.06		
AS-9	Chargat	6.92		
AS-10	Bagha	6.91		

**Appendix Table XXII:** Sample wise distribution of Mg in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration(g /kg) of Mg	Average Concentration of the present study (g/kg)	Optimum concentration (g/kg) (Hoque and Khaliquzzaman, 2002)
AS-1	Tanore	3.22	6.43	6.00
AS-2	Mohanpur	8.12		
AS-3	Bagmara	4.65		
AS-4	Paba	4.66		
AS-5	Puthia	7.89		
AS-6	Durgapur	7.81		
AS-7	Boalia	10.45		
AS-8	Godagari	6.78		
AS-9	Chargat	5.65		
AS-10	Bagha	5.12		

**Appendix Table XXIII:** Sample wise distribution of K in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/kg) of K	Average Concentration (mg/kg) of K	Permissible Range (mg/kg) (Tandon and Sekhon, 1988)
AS-1	Tanore	100	127.8	78-273
AS-2	Mohanpur	213		
AS-3	Bagmara	179		
AS-4	Paba	123		
AS-5	Puthia	75		
AS-6	Durgapur	60		
AS-7	Boalia	47		
AS-8	Godagari	67		
AS-9	Chargat	169		
AS-10	Bagha	245		

**Appendix Table XXIV:** Sample wise distribution of K in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/kg) of K	Average Concentration (mg/kg) of K	Permissible Range (mg/kg) (Tandon and Sekhon, 1988)
AS-1	Tanore	212	109.4	78-273
AS-2	Mohanpur	100		
AS-3	Bagmara	150		
AS-4	Paba	101		
AS-5	Puthia	70		
AS-6	Durgapur	50		
AS-7	Boalia	55		
AS-8	Godagari	67		
AS-9	Chargat	109		
AS-10	Bagha	180		

**Appendix Table XXV:** Sample wise distribution of Ca in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/kg) of Ca	Average Concentration (mg/kg) of Ca	World Soil range (Tandon and Sekhon, 1988)
AS-1	Tanore	312	261.7	78-273
AS-2	Mohanpur	245		
AS-3	Bagmara	356		
AS-4	Paba	145		
AS-5	Puthia	267		
AS-6	Durgapur	136		
AS-7	Boalia	217		
AS-8	Godagari	250		
AS-9	Chargat	300		
AS-10	Bagha	389		

**Appendix Table XXVI:** Sample wise distribution of Ca in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/kg) of Ca	Average Concentration (mg/kg) of Ca	World Soil range (Tandon and Sekhon, 1988)
AS-1	Tanore	285	233.8	78-273
AS-2	Mohanpur	200		
AS-3	Bagmara	257		
AS-4	Paba	155		
AS-5	Puthia	201		
AS-6	Durgapur	100		
AS-7	Boalia	200		
AS-8	Godagari	275		
AS-9	Chargat	364		
AS-10	Bagha	301		

**Appendix Table XXVII:** Sample wise distribution of Zn in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/kg) of Zn	Average concentration of the present study (mg/kg)	Worldwide mean concentration (mg/kg) of Zn ( <b>Tandon and Sekhon, 1988</b> )
AS-1	Tanore	58.5	60.72	64
AS-2	Mohanpur	68.6		
AS-3	Bagmara	67.5		
AS-4	Paba	69.44		
AS-5	Puthia	43.6		
AS-6	Durgapur	65.7		
AS-7	Boalia	38.18		
AS-8	Godagari	34.96		
AS-9	Chargat	74.09		
AS-10	Bagha	86.64		



**Appendix Table XXVIII:** Sample wise distribution of Zn in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/kg) of Zn	Average concentration of the present study (mg/kg)	Worldwide mean concentration (mg/kg) of Zn
AS-1	Tanore	50.01	57.8	64
AS-2	Mohanpur	56.14		
AS-3	Bagmara	70.42		
AS-4	Paba	60.44		
AS-5	Puthia	42.5		
AS-6	Durgapur	60.21		
AS-7	Boalia	40.58		
AS-8	Godagari	30.25		
AS-9	Chargat	78.12		
AS-10	Bagha	89.45		

**Appendix Table XXIX:** Sample wise distribution of B in the arable soil of Rajshahi district in monsoon season.

Sample no.	Location	Concentration (mg/kg) of B	Average Concentration (mg/kg) of B	Optimum requirement (mg/kg) (Miller and Donahue, 1997).
AS-1	Tanore	1.50	1.74	1.0
AS-2	Mohanpur	1.61		
AS-3	Bagmara	1.12		
AS-4	Paba	1.23		
AS-5	Puthia	1.56		
AS-6	Durgapur	2.89		
AS-7	Boalia	1.43		
AS-8	Godagari	1.34		
AS-9	Chargat	2.31		
AS-10	Bagha	2.43		

**Appendix Table XXX:**Sample wise distribution of B in the arable soil of Rajshahi district in winter season.

Sample no.	Location	Concentration (mg/kg) of B	Average Concentration (mg/kg) of B	Optimum requirement (mg/kg) (Miller and Donahue, 1997).
AS-1	Tanore	1.23	1.68	1.0
AS-2	Mohanpur	1.70		
AS-3	Bagmara	1.45		
AS-4	Paba	1.87		
AS-5	Puthia	1.54		
AS-6	Durgapur	2.74		
AS-7	Boalia	1.12		
AS-8	Godagari	1.11		
AS-9	Chargat	2.01		
AS-10	Bagha	2.12		

**Appendix Table XXXI:** Sample wise distribution of different elements in the locally produced fruits.

Smample No	Bengali name	English name	Scientific name	Concentrations (mg/kg)								
				Zn	Ca	Fe	K	Cl	Cu	Mn	F	Br
AF-1	Kamranga	Carambola	<i>Averrhoa carambola</i>	64.00	5700	511.00	28950	4567	20.56	8	0	0
AF-2	Papay	Papaya	<i>Carica papaya</i>	45.00	20600	545.00	15374	2343	0	5	0	0
AF-3	Peyara	Guava	<i>Psidium guajava</i>	0	66800	404.70	12374	0	0	7	0	0
AF-4	Kola	Banana	<i>Musa sapientum</i>	50.67	3450	350.60	18705	0	0	6	0	0
AF-5	Kalo Jam	Black Berry	<i>Syzygium cumini</i>	0	22003	301.20	23643	0	0	7	0	0
AF-6	Anaras	Pine-Apple	<i>Ananus comosus</i>	0	20987	300.00	17890	0	0	8	0	0
AF-7	Jalpi	Olive	<i>Olea europea</i>	0	34211	290.50	20579	1324	0	9	0	0
AF-8	Aamalaki	Amla	<i>Emblica officinalis</i>	0	24532	250.30	19780	1098	0	4	0	0
AF-9	Kathal	Jackfruit	<i>Artocarpus heterophyllus</i>	65.00	32101	345.70	25673	0	0	8.5	0	0
AF-10	Aam	Mango	<i>Mangifera indica</i>	50.00	16431	377.90	24356	0	0	4	0	0

**Appendix Table XXXII:** Sample wise distribution of different elements in the locally produced vegetables.

Sample No	Bengali name	English name	Scientific name	Concentrations (mg/kg)					
				Zn	Ca	Fe	K	Cl	Cu
AV-1	Misti kumra	Pumkin/ Sweet gourd	<i>Cucurbita moschata</i>	0	16754	1021	18790	3789	26.87
AV-2	Begun	Bringal	<i>Solanum melongena</i>	45.00	12541	983	13211	2113	0
AV-3	Dherosh	Lady's finger/Orka	<i>Abelmoschus esculentus</i>	87.90	22314	879	15432	3123	0
AV-4	Alu	Patato	<i>Solanum tuberosum</i>	50.67	4321	511	31098	7890	0
AV-5	Tomato	Tomato	<i>Lycopersicon esculentum</i>	42.00	11856	6780	7560	4658	0
AV-6	Lau	Bottle Gourd	<i>Lagenaria siceraria</i>	0	10123	319	24288	0	0
AV-7	Gajor	Carrot	<i>Daucus carota</i>	110.05	20264	9230	30342	3706	0
AV-8	Phul kopi	Coliflower	<i>Brassica oleracea var botrytis.</i>	0	27621	8321	3421	0	0
AV-9	Bandhakapi	Cabbage	<i>Brassica oleracea</i>	40.00	28431	7890	9712	0	0
AV-10	Shalgom	Turnip	<i>Brassica rapa</i>	45.23	30541	6530	17456	0	0