Assessment of Geochemical Variabilities in Tea Garden Soils and their Effects on the Quality of Tea in Bangladesh

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Assessment of Geochemical Variabilities in Tea Garden Soils and their Effects on the Quality of Tea in Bangladesh

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Declaration

I hereby declare that this thesis has been composed by myself, and has not been accepted in any previous application for a degree. The work presented in this thesis has been performed by myself. All quotations have been distinguished by quotation marks, and the sources of information presented in this thesis have been specifically acknowledged using references, to the best of my knowledge.

Apu Biswas	Dhaka, June 2023
(Name)	(Place and Date)

To Whom It May Concern

This is to confirm that the research presented in this PhD thesis entitled "Assessment of Geochemical Variabilities in Tea Garden Soils and their Effects on the Quality of Tea in Bangladesh" has been done by Apu Biswas at the Department of Soil, Water and Environment, University of Dhaka, Dhaka-1000, Bangladesh. This PhD thesis has been entirely composed by Apu Biswas. All quotations have been distinguished by quotation marks, and the sources of information are specifically acknowledged. This thesis has not been accepted for any previous degree.

As Apu Biswas's supervisors, we wish to confirm that he is the main contributor to this thesis, and we have supervised his PhD project leading to writing the thesis.

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The Author.
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Dedicated to my family

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Abbreviations

AAS = Atomic Absorption Spectroscopy

ANOVA = Analysis of variance

CD = Churamoni Dust

g = gram

GBOP = Golden Broken Orange Pekoe

GIS = Geographic Information System

GPS = Global Positioning System

Kg = kilogram

mg = milligram

PCA = Principal Component Analysis

PF = Pekoe Fannings

r = Pearson correlation coefficient

R^2 = R-squared value for regression models

s.e. = Standard error of the mean

SRDI = Soil Resource Development Institute

TF = Theaflavin

TF = Transfer factor

TR = Thearubigin

Assessment of Geochemical Variabilities in Tea Garden Soils and their Effects on the Quality of Tea in Bangladesh

Abstract

Tea cultivation in Bangladesh is done mainly in the hilly regions of Moulvibazar, Sylhet and Chattogram. Recently, tea cultivation has also been started in the plain lands of Panchagarh. A large amount of chemical fertilizers and agrochemicals are used in the soils of tea gardens to increase the production of tea. These fertilizers and agrochemicals are sources of heavy metals accumulation in tea garden soils. To date there is very limited information on the concentration of heavy metals in tea garden soils, tea leaves and made tea of Bangladesh. In the present study, 160 tea garden soils (surface soil, n = 112 and sub-surface soil, n = 48) and 32 non-tea garden soils from the geographically different tea gardens of Bangladesh were collected and analyzed for nitrogen, boron, chromium, copper, iron, manganese, nickel and zinc to assess their variabilities among Old Himalayan Piedmont Plain, Northern and Eastern Hills and Northern and Eastern Piedmont Plains based on landforms, soil texture, drainage class, soil series as well as the age of tea plants. Tea leaves (n = 112) and made tea (n = 39) samples were also collected and analyzed to assess the concentrations of heavy metals. A range of biochemical parameters (total polyphenol, theaflavin, thearubigin and protein content) of made tea were also evaluated. The concentrations of chromium, copper, iron, manganese, nickel, and zinc in the surface soils of the different tea gardens were ranged between 2 - 73, 0.86 - 23, 3084 – 47120, 21 – 1274, 3 – 23 and 2 – 465 mg/kg, respectively. In the sub-surface soils of different tea gardens, the concentration of chromium, copper, iron, manganese, nickel and zinc were ranged between 3 - 89, 1 – 22, 2640 – 52540, 49 – 466, 4 – 24 and 2 - 71 mg/kg, respectively. Significant variations ($0.001 > p < 0.05$) in the concentrations of the heavy metals were observed between the surface and sub-surface soils. The mean concentrations of chromium, copper, iron, manganese, nickel, and zinc in the tea leaves were 12.86, 14.67, 185, 641, 4, and 43.79 mg/kg, respectively. The mean concentrations of chromium, copper, iron, manganese, nickel and zinc in the made tea were 21, 17.51, 373, 613, 6.29 and 134 mg/kg, respectively. The concentrations of iron and nickel in the tea leaves and made tea and zinc in the tea leaves were within the permissible limits of heavy metals in plants. Significant variations ($p < 0.001$) in the concentrations of copper, iron, manganese, nickel, and zinc in the tea leaves and made tea were observed. The concentrations of heavy metals in the soils were

found to be varied significantly within the different physiographic regions, tea gardens, landforms, soil texture, and soil series. The tea leaves and made tea of different tea gardens also had variabilities in their heavy metal contents. Zinc had significant positive relationship with caffeine ($p < 0.01$) in the made tea while had the significant negative relationships with theaflavin and thearubigin. Significant positive relationships between caffeine and zinc indicated that with increase zinc concentration caffeine content also increased in the made tea. Excess zinc might decrease the quality of tea through decreasing theaflavin and thearubigin contents. Nickel had significant negative ($p < 0.01$) relationships with polyphenol and manganese had significant negative relationships with thearubigin in the made tea. Excess nickel might decrease the quality of tea through decreasing the total polyphenol content. The made tea of different tea gardens had significant differences ($p < 0.01$) in the concentrations of biochemical parameters such as caffeine, protein, total polyphenol, theaflavin and thearubigin. The transfer factors for chromium, copper, iron, manganese, nickel, and zinc were 0.72, 1.72, 0.009, 3.44, 0.44 and 1.15**,** respectively. The findings of present research can be regarded as the baseline information on heavy metals in soils, tea leaves and made tea of the tea gardens located in different physiographic regions of Bangladesh, which can be useful for further understanding of the origin, extent, and bioavailability of toxic heavy metals in the tea garden soils of Bangladesh. As tea is a popular drink, this study will be supportive for the further investigation to assess the release of toxic heavy metals from made tea into tea brew.

Chapter One

General Introduction

1.1 Tea plant

Tea plant is a perennial and evergreen flowering tree which is botanically known as *Camellia sinensis* (L). Mountain regions of southwestern China and neighboring countries are the native place of tea. This plant belongs to the family Theaceae. If pruning is not done, in natural condition a tea plant can grow up to a height of 10 m. A tea plant can give commercially profitable crop up to 50–60 years in favorable environments. This plant is self-sterile, highly cross-pollinated and heterogeneous crop. There are two main commercial varieties of tea such as *Camellia sinensis* variety *sinensis* and *Camellia sinensis* variety *assamica*. Tea variety *sinensis* has small leaves which grow well in cool weather and higher altitude area while tea variety *assamica* has large leaves which grow well in tropical and subtropical environments. (Chang 1981; Chang and Bartholomew 1984; Ming and Zhang 1996).

1.2 Origin of tea

Tea is one of the most admired and healthy beverages in the world which is manufactured from the leaves of evergreen shrub *Camellia sinensis*. In all levels of society, varied range of age groups peoples drink tea. All over the world, more than three thousand million cups of tea are consumed daily. Coffee, cocoa and tea are the three nonalcoholic stimulating beverages has hitherto discovered by human civilization. In Chinese mythology, tea is deep rooted plant and its use as a drink has been trend long before the Christian era which regarded tea as the most prevalent temperance drink (Choudhury, 2010).

Tea encompasses a long and momentous history. This beverage has been enjoyed through the world for centuries. The discovery of tea by the Chinese Emperor, Shen Nung and its legendary history dates back to 2737 B. C. who said to own invented tea during one amongst of his travels round the Chinese Empire. While taking a brief rest, some leaves fell off a shrub, straight into his cup of boiling water. A beautiful scent came up from the cup and therefore the curious emperor tasted the drink made unintentionally. The beverage did not only have an exquisite taste, but a refreshing, energizing effect additionally. The emperor became so overwhelmed by the new discovery that he began to promulgate the enchanted shrub (Weatherstone, 1992).

1.3 The Scenario of global tea production

Currently, tea has gained admiration as a healthy drink worldwide. More than fifty countries of the world produce tea and the global production of tea in the year 2021 was 6455.19 million kg (ITC, 2022) (Table 1.1). Only twelve countries are the main producers out of the fifty tea-growing countries, specifically, Bangladesh, China, India, Indonesia, Japan, Sri Lanka, Turkey, Russia, Kenya, Malawi, Uganda and Argentina. China secured 1st position by producing 3063.15 million kg tea which is 47.45% of the total world tea production and followed by India stands 2nd position by producing 1343.06 million kg tea (20.81%). The three largest tea producer countries such as China, India and Kenya contributed 76.59% of the total world tea production (ITC, 2022). Global tea production has increased by 2.55% per year in the last two decades. In the last 15 years the total area of 2.34 million ha accounted as stable yielding tea plantations, of which 86.7% in Asia, while only 8.64% covered in Africa (PDU, 2020).

Table 1.1: World tea production (million kg).

Country	2017	2018	2019	2020	2021
China	2496.41	2610.39	2799.38	2986.02	3063.15
India	1321.76	1338.63	1390.08	1257.53	1343.06
Kenya	439.86	493.00	458.85	569.54	537.83
Sri Lanka	307.72	304.01	300.13	278.49	299.34
Vietnam	175.00	185.00	190.00	186.00	180.00
Indonesia	134.00	131.00	128.80	126.00	127.00
Others	843.64	904.18	882.84	875.92	904.81
Total	5718.39	5966.21	6150.0	6279.50	6455.19

Source: ITC, 2022.

1.4 Tea growing areas of Bangladesh

Tea is the cheapest healthy beverage. In Bangladesh it has enormous importance as a perennial crop to meeting the entire internal demand. Up to the independence in 1971 in Bangladesh tea was cultivated in only two districts, one in Sylhet district which was known as 'Surma Valley' and the other in Chattogram district which was known as 'Halda Valley'. Tea cultivation was also introduced in the *Korotoa valley* in Panchagarh district (extreme northwest of Bangladesh) since 2002. Tea areas were also extended at Lalmonirhat, Thakurgaon, Nilphamari, Dinajpur and Bandarban in the Chittagong Hill Tracts recently. District and management-wise number of tea estates in Bangladesh are given in the Table 1.2.

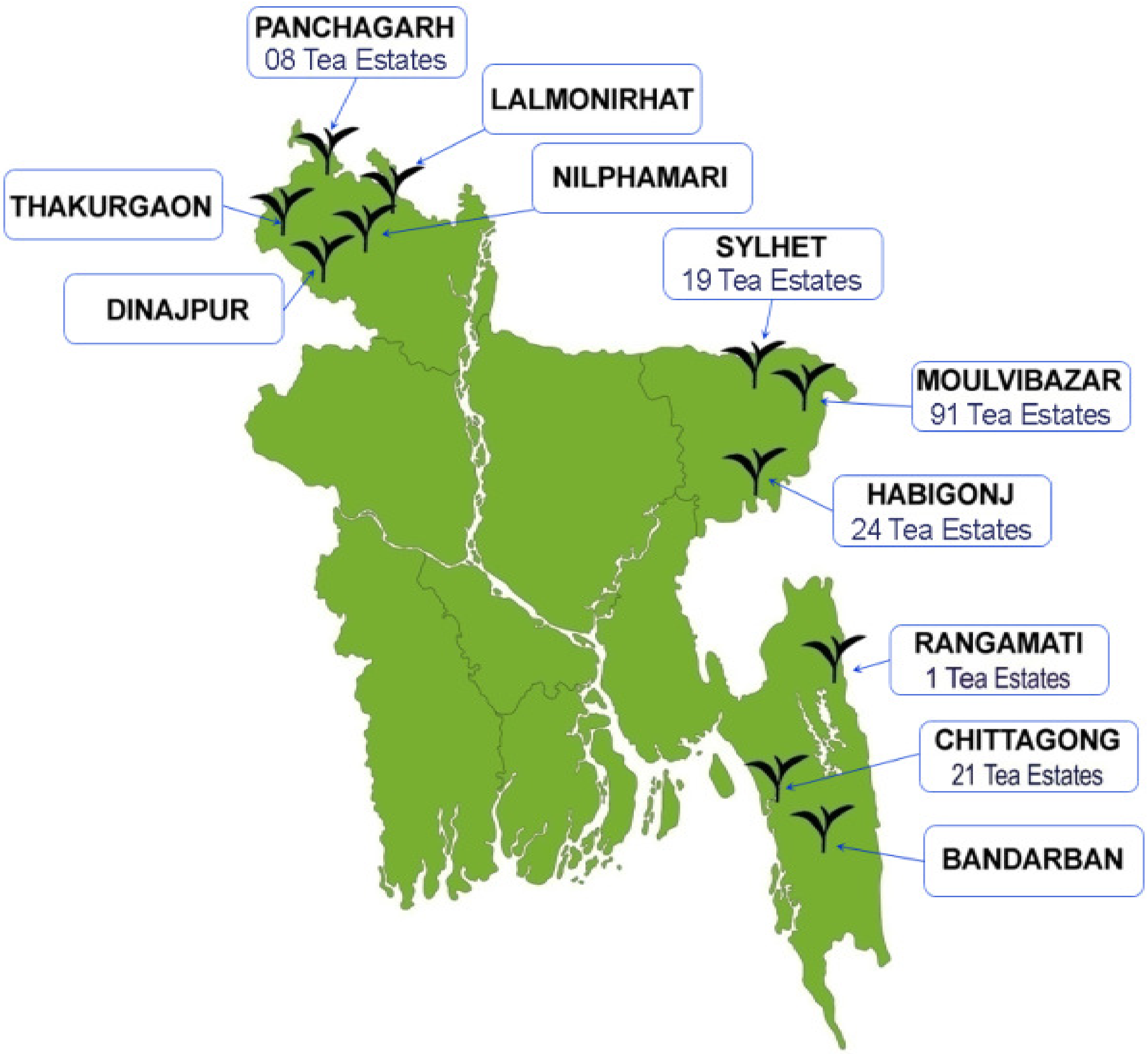


Figure 1.1: Tea growing regions in Bangladesh (Source: Web1).

At the present, Bangladesh is producing about 96.07 million kg of made tea per year with a mean yield of about 1518 kg per hectare in about 63,261 ha plantation area of 167 tea estates (PDU, 2020). The tea industry contributes 0.11% of GDP (PDU, 2020). In respect of area, production and export, Bangladesh tea is now ranked 8th, 10th and 12th positions in the world

(Mamun, 2019). The production, internal consumption and export of our tea are given in the Table 1.3.

Table 1.2: District and management-wise number of tea gardens in Bangladesh.

District	No. of Tea Gardens	Type of Management	No. of Tea Gardens
Moulvibazar	90	Duncan Brothers (BD) Ltd.	16
Habiganj	26	Deundi Tea Co. Ltd.	03
Sylhet	19	Noyapara Tea Co. Ltd.	01
Chattogram	21	The New Sylhet Tea Estate Ltd.	01
Rangamati	02	Bangladesh Tea Board	05
Panchagarh	08	National Tea Co. Ltd.	12
Thakurgaon	01	Finlays	07
Total	**167**	Private Ltd. Co.	68
		Proprietary	54
		Total	**167**

Source: PDU, 2020.

1.5 Key factors influencing tea plantation in Bangladesh

1.5.1 Geographical characteristics of Bangladesh and Tea

Bangladesh is a low-lying country of South Asia situated on the banks of the rivers. It lies in Indian subcontinent within latitudes 20°25´ and 38°40´N and longitudes 88°01´ and 92°40´E. Bangladesh has 1, 43,999 square kilometers or 55,598 square miles total area. The country is formed by a delta plain at the confluence of the Padma, Meghna and Jamuna rivers and their tributaries (Brammer, 2012). In Bangladesh the alluvial soil is prone to flood and drought although it is highly fertile. Chittagong hill tracts in the far South-east and the hills of Sylhet division in the North-east region have only risen above the plain. The hilly area of South-east and North-east region is famous for tea plantation. Among the major tea growing countries in the world Bangladesh is one of them due to its suitable physical factors and cheap labourer.

Tea cultivation is regulated by some abiotic factors such as physiography, topographic features and climatic factors. Tea plantation needs a suitable climatic condition (Sana, 1989). But Bangladesh grows tea under minimal land and environmental requirements such as low height, poor topography, the presence of impermeable subsoil layer and low fertility status.

Table 1.3: Production, internal consumption, and export of Bangladesh tea.

Year	Production (million kg)	Expected Internal Consumption (million kg)	Export (million kg)	Export Value (million taka)
2001	53.15	36.95	12.92	894.99
2002	53.62	41.50	13.65	939.93
2003	58.30	37.44	12.18	915.07
2004	56.00	43.33	13.11	934.04
2005	60.14	43.30	9.01	742.62
2006	53.41	40.51	4.79	469.59
2007	58.19	46.27	10.56	899.01
2008	58.66	52.12	8.39	976.95
2009	59.99	53.74	3.16	433.50
2010	60.04	57.63	0.91	176.68
2011	59.13	58.50	1.48	213.51
2012	62.52	61.19	1.56	222.28
2013	66.26	64.00	0.54	133.04
2014	63.88	67.17	2.66	281.72
2015	67.38	77.57	0.54	105.13
2016	85.05	81.64	0.62	140.56
2017	78.95	85.93	2.56	377.29
2018	82.13	90.45	0.65	203.93
2019	96.07	95.20	0.60	194.26
2020	86.39		2.17	347.14
2021	96.51		0.68	180.57
2022	49.09 (Up to August)		0.59 (Up to September)	

Source: Web 2.

On the other hand, the winter months of Bangladesh very often go without rain and the minimum temperature sometimes goes as low as 7°c. As a result, during the months from

December to March very little crop is harvested. During January to mid-March, almost all the garden stopped its production as no green leaf harvested (Sana, 1989; Ahmed, 2005). These are such conditions, a careful consideration of tea plantation is very much essential to increase production and sustainability of tea garden in Bangladesh.

1.5.2 Physical Factors

There are some physical factors which influences tea plantation. Tea plantation and production of green leaf depends on these factors. The key influencing physical factors of tea plantation in Bangladesh is discussed below.

Physiography

Tea growing areas in Bangladesh are generally located in three physiographic regions such as Northern and Eastern Hills, Northern and Eastern Piedmont Plains and Old Himalayan Piedmont Plain.

Northern and Eastern Hills

The Northern and Eastern Hills physiography covers total area of 18,171 km^2. Khagrachhari, Chittagong Hill Tracts, Bandarban, Cox's Bazar, and small areas along the northern border of the Sherpur, Mymensingh, Sunamganj, and Sylhet districts are belong to this tract. In general, more rolling relief occurs locally with very steep slopes and a few low hills have flat peaks. Topography is complex. Over different rocks, hills have been dismembered to different magnitude. The soils of the hills are mainly excessively well drained. The valley soils are subjected to flash floods due to poorly drained condition. The nature of the soils are very strongly acidic with yellowish brown to vivid brown in colour, porous, fragile, loamy in texture and low in water holding capacity. Brown hill soil is the prime common soil category of the area with very steep slopes. In general, there are low content of organic matter and fertility status (FAO-UNDP, 1988; Huq and Shoaib, 2013).

Northern and Eastern Piedmont Plains

Excluding in the greater Chattogram district and part of the Feni district, Northern and Eastern Piedmont Plains is an uneven region happening as a slender strip of land at the bottom of the northern and eastern hills. The region covers total 4,038 km^2 area. Sherpur, Netrakona, Sunamganj, Sylhet, Moulavi Bazar, Habiganj, Brahmanbaria, and Comilla districts are belong to this physiography. From the bottom of the northern and eastern hills, this region inclusion the alluvial fans that slope mildly outward into smooth low-lying basins. The topography is uneven and close to rivers and streams crossing the region on high land adjacent to the hills. Greater portions are above usual inundation level. In the rainy season lower portions are narrowly to relatively intensely inundate. During successive flash floods the uneven accumulation of alluvium of various textures are responsible for the complex soil patterns in this region. Grey piedmont soils and non-calcareous grey floodplain soils are the main general soil natures of the area. Soils with sandy loam to silty clay textural classes occupied the greater part of this area. The soil reaction is slightly acidic to strongly acidic in nature and the fertility status is low to medium (FAO-UNDP, 1988; Huq and Shoaib, 2013).

Old Himalayan Piedmont Plain

Old Himalayan Piedmont Plain is developed in an old Tista alluvial fan extending from the foot of the Himalayas. This physiographic region covers an area of 4,008 km^2. The northwestern part of the Dinajpur district as well as most of the Panchagarh and Thakurgaon districts are belongs to this zone. This region has a difficult topographic arrangement. Sandy loams and sandy clay loams, rapidly permeable and deep soils are largest in this area. Sands happen locally on the top of ridge, and clays occupy middles of a few basins. Due to differences in the wideness of the dark colored topsoil, texture of the subsoil, depth to a sandy stratum and drainage soil patterns often are complex. They are low in weatherable potassium minerals with topsoil of strong acidity and subsoil of moderate acidity. Non-calcareous brown floodplain soils, black Terai soils, non-calcareous dark grey floodplain soils and non-calcareous grey floodplain soils are predominate in this physiographic region. The organic matter contents are usually greater than in most floodplain soils of Bangladesh. The natural fertility of the soil is modest but well persistent (FAO-UNDP, 1988; Huq and Shoaib, 2013).

Topography

The topography of the tea producing regions is different from other places of the country. The topography of traditional and new plantation area is discussed below.

Topography of traditional tea garden of Sylhet and Chattogram

The topography of traditional garden is divided in three types. There are- tillah, high tillah or low flat.

***Tillah*:** Nearly 32% of tea land is tillah. The flat toped hills with their slopes are included in this category. Tillah or low hills are outliers of the Tripura hill range in India. These tillahs are up to 9 meter in height steeply rounded with unconsidered sandy soils derived from tertiary sandstone rocks and shale's or broken pieces of literate occur below 2 to 5 feet. The south slopes of these tillahs are more susceptible to drought during dry season. The tillah is appropriate for tea but exposed to drought and erosional loss of soil. Seed plant is more suitable for plantation in tillah because of its tolerance capacity (Sana, 1989; Rahman, 2009).

***High Flat*:** High flats are the upper valleys create about 45% of tea area composed of sand, silt and clay. The high tillahs are undulating 20 to 30 feet beyond the plains and separated by slender valleys formed by erosion. Soils are well drained without any rock fragments in the substratum but gentler slopes than tillahs. The soils are suitable for tea plantation (Sana, 1989; Rahman, 2009).

***Low Flat*:** Nearly 23% of tea land is constituted with low flats. Low flats are the valley floors. Low flats are undulating with moderately well drained and subjected to flush flooding up to a day after heavy shower in monsoon. The top soils of low flat is brown, the substratum is grey, may be comparatively opulent in organic matter but put through to water logging. Low flat land has a semi-permanent water Table. Recently most of the gardens use clone plant on low land. In low flat land, clone plants give more production than seed plants (Sana, 1989; Rahman, 2009).

Topography of new tea garden of Panchagarh

Tea plantation was a great challenge in the harsh terrain with little vegetation in Northwestern region. The soil of new plantation is plain but sandy with rocks and rubble underneath. The drainage capacity of this soil is well. Topography of the Panchagarh district is higher, 60 to 85 meters above from the mean sea level, well drained and free from usual inundation risks. But it was a great challenge to plant tea in the harsh terrain with vegetation and rubble underneath (Ahmed and Ahmed, 2015).

1.5.3 Tea soils

Tea requires certain specific soil conditions as well as suitable climatic conditions for good growth and productivity. Understanding the nature and properties of tea soils would therefore be of great importance in order to provide and maintain the necessary soil conditions for maximizing and sustaining the growth and productivity of tea (Natesan, 1999). Optimum physical, chemical and biological properties of the soils are essential for the primary establishment of tea plants which are also necessary for attaining high yield subsequently. Information on the physical and chemical characteristics of tea soils is available from the work of Ranganathan (1977), Othieno (1992) and Barua (1989) whereas only limited information is available on the biological conditions of soils suitable for tea (Balamani Bezbaruah, 1994). Since tea grows well in strongly acidic soils that are porous and well drained, the physico-chemical characteristics of the tea soils are of paramount importance. Though the tea soils from various tea-growing countries are derived from different parent materials, they all have certain common characteristics. Favorable soil pH is the utmost essential circumstance for sufficient growth of tea. In tea-growing countries usually the pH of the soils differs from 3.3 to 6.0. In many countries the ideal pH for optimum development and well use of nutrients, specifically for nitrogen, is 4.5 to 5.0. However, some high pH tolerant varieties of tea can grow at pH levels between 6.0 and 6.5. Tea can accumulate large quantities of aluminium ion (Al^{3+}) due to its more availability in tea soils under acidic conditions (Chenery, 1955; Foy *et al*., 1978; Matsuda *et al*., 1979; Natesan and Ranganathan, 1990).

Tea garden soils of Bangladesh

Soils of varied geological origins are suitable for growing Tea. Quaternary and recent alluvia are most of the soils in North-east India and Bangladesh. From the gneiss and sandstone many soils formed in situ. Sometimes as an indicator of suitability of tea cultivation the hue of soils is considered. The colour of the tea soils of famous tea growing areas such as Sri Lanka, Java, South India, North-East India (Assam) and Bangladesh are red. In North-East India and

Bangladesh many parts of the tea soils are derived from the grasslands and grey in colours (Sana, 1989). Fertility status of tea soils are low, moreover it is highly weathered, extremely acidic in nature. These soils suffer from erosion and do not obtain deposits of productive silt by inundation. Organic matter and nitrogen content of tea soils are low with medium textured. Sana (1989) and Alam (1999) reported that kaolinite, quartz, hematite, goethite and gibbsite type's minerals are found in tea soils of Bangladesh. These soils have strong phosphate fixing capacity. Cation exchange capacity and nutrient releasing capacity of these soils are also low. Moreover, some deficiency symptoms and aluminium toxicity shown due to the content of 85% aluminium in the exchangeable sites of tea soils of Bangladesh and the uptake of indispensible nutrients, such as calcium, phosphorus, magnesium and iron retarded due to high concentration of aluminium. Unless lime is used phosphorus released from the phosphatic fertilizer will readily be immobile by such high concentration of aluminium. An overall content of nitrogen is 0.07 to 0.09 % and organic matter in tea soils of Bangladesh varies from 1.0 to 1.2%. The critical values of organic matter and total nitrogen have been fixed at 1% and 0.1%. The content of base-saturation, available phosphorus and magnesium is low. The critical values of nutrients of tea soil is 10 mg/kg for phosphorus, 80 mg/kg for potassium, 25 mg/kg for magnesium, 90 mg/kg for calcium and carbon-nitrogen ratio is 10 (Alam, 1999).

1.5.4 Environmental factors for tea cultivation

In different parts of the world tea crop is grown in a variability of climates and soils due to its wide adaptability. Commercial tea plantations are wide spread as far North as Georgia (42°N) and as far South as Argentina (27°S), at elevations extending from almost sea level to 2,700 m above the mean sea level. The young tender shoots of tea plant processed the most extensively consumed beverages after water is tea. Geographical, climatic and environmental factors are responsible for variations in yields and excellence of tea (Owuor *et al*., 2011). The most important factors determining tea productivity are light, carbon dioxide (CO_2), temperature and water availability (Hajiboland, 2017).

Climatic Factors

The tea agro-ecosystem consisted by various abiotic elements along with tea plants, shade trees and other ancillary crops. Due to the variation of a country the climatic parameters such as geographical locations, the physiographic regions, land characteristics and circulation of water, elevation, hill obstructions, ocean flows, pressure-belts, wind, storms, etc are vary from location to location as well as season to season (Sana, 1989). Hot period (middle of February to middle of May), monsoon period (middle of May to middle of October) and cold period (middle of October to middle of February) is the three distinctive seasons in the tropical monsoon climatic zone where Bangladesh is located. With dry periods the year begins and ends (Alam, 1999). Precipitation, temperature, moisture-evaporation and sun light are the four important environmental factors of the tea growing areas in Bangladesh which is responsible for effective tea cultivation (Islam *et al*., 2021).

Rainfall

The distribution of precipitations has an important role on the development of the tea plant. It was observed that minimum 1270 mm rainfall per year is required for tea cultivation except there are other alleviating conditions. Over a period of several months if monthly averages rainfall fall below 50 mm the crop production suffers rigorously because without exception the rainfall in the dry season is important (Sana, 1989). In Bangladesh, precipitation is the most significant weather component which differentiates climatic differences. The quality and quantity of rainfall pattern is zonal and the productivity of tea is influenced by the rainfall distribution. In Bangladesh the annual precipitation varies from 1400 to 5000 mm (Alam, 1999). The wet area comprises by the tea cultivation regions of Sylhet, Chattogram and Panchagarh (Islam *et al*., 2021). November to April is the dry season in the tea cultivation regions of the country, while May to October is the wet season and during June to September more than 80% of mean annual rainfall is acquired in different tea valley circles (Alam, 1999). In North Sylhet valley circle, the volume of annual precipitation is greater whereas it is lower in the Luskerpur valley circle (Sana, 1989).

Temperature

For tea cultivation the temperature should be above 13 °C and below 30 °C. The ideal ambient temperature for optimum tea growth is 18 to 25 °C. In Bangladesh generally the temperature raised in April to May, slightly decrease during monsoon season, and steadily reduces after September or October. Sometimes long dry spell arises when rain is meager next to November or December (Islam *et al*., 2021). Temperature increases up to a highest 40°C in the tea areas of Sylhet and Chittagong. In January the extreme minimum in the Sylhet region drops to 1.7°C and rising to 2.8°C in February. In contrast, the extreme minimum temperature in Chattogram region decreases to the extent of 7.8°C to 8.9°C (Alam, 1999). The cropping season of tea is found to vary from 5 to 12 months in the existing tea growing areas of the world. Tea areas between latitudes 18°N and 21°S have 12 months of cropping season, while outside these latitudes it varies from 6 to 9 months. Undoubtedly, temperature change over the year is the primary cause which stops are flushing (Sana, 1989).

Humidity-Evaporation

The development and production of tea has a close relationship with desiccation of the air. The complete relative humidity of tea areas in Sylhet ranges from 56% to 80% whereas in the tea areas of Chattogram range from 68% to 80%, respectively (Islam *et al*., 2021). In Bangladesh, temperature raised over the country is at the peak during April and May and shows a primary maximum evaporation regime. When temperature is lowest in December and January evaporation is also the lowest (Alam, 1999). Tea areas of Sylhet and Chattogram have low vaporation than the Panchagarh district of Bangladesh (Islam *et al*., 2021).

Sunshine

The development of tea plants also depends on the intensity and duration of sunshine. Tea is responsive to the length of the day and it seems that winter dormancy in tea occurs only in areas of higher latitudes. The mean lowermost length of day is 10.6 hours in December and mean longest length of day is 13.7 hours in June. In the rainy season of Bangladesh, the length of shining sunlight is lower due to the overcast clouds (Islam *et al*., 2021; Alam, 1999).

Favourable Condition

Tea plant favors mild jungle condition. 70–90% relative moistness, warm and tropical condition having 26–28°C temperature and more than 2000 mm annual rainfall are appropriate for tea husbandry. About 12 hours day length is required for tea plant. Tea plants can't tolerate water logging condition. For tea excessive as well as insufficient rainfall regime is unfavorable (Islam *et al*., 2021). The elementary environmental necessities for the growth and improvement of tea are listed in the Table 1.4. The elementary factors required for the growth and improvement of tea plants such as pH of soil, moisture content, organic matter, availability of macro and micro nutrients, pest and disease management, and ecological balance of the surrounding tea gardens (Han *et al*., 2018).

Table 1.4: Basic environmental requirements for the growth and development of tea plant.

Climate factors	Extreme lowest	Normal range	Ideal
Temperature (°C)	-20 (*Camellia sinensis*) -8 (*Camellia assamica*)	13-26	18-23
Deposited temperature per annum (≥10°C)	3000	4000-8000	6000-7000
Rainfall per year (mm)	500	800-2500	1500-2000
Relative humidity per annum (%)	60	70-90	80-85
Water holding capacity of soil (%)	50	60-95	70-90
Soil pH (in water suspension)	3.0	3.5-6.5	4.5-5.5

(Source: Han *et al*., 2018).

1.5.5 Fertilizer management practices for tea cultivation in Bangladesh

Recommended doses of fertilizer for young tea

A well-adjusted combination of nitrogen, phosphorus and potassium fertilizers are required for the development of their frame and root system in the primary phase of life of the tea plants. In Bangladesh condition, the blend of nitrogen, phosphorus and potassium in the ratio of 2:1:2 has evidenced to be favorable for young tea. Fertilizer recommendations for the young tea are given in Table 1.5. Due to pruning, plucking, high precipitation, volatilization, evapotranspiration, etc. an enormous amount of nutrients are being lost from the tea field. With the purpose of improve the soil nutrient status and increase the yield and quality of tea through balanced fertilizer application inorganic fertilizers must be added to the soil (Motalib and Dutta, 2011).

Table 1.5: Fertilizer recommendations for young tea.

Age of plant	Fertilizer mixture kg/ha.			Total mixture	Dose per plant	
	Urea	TSP	MOP		Chemical	Cow dung
1st year	176	90	160	426	39 g	1 kg/per plant
2nd year	200	100	180	480	44 g	1 kg/per plant
3rd year	265	90	160	515	47 g	5 tons/ha.
4th year	300	100	180	580	53 g	5 tons/ha.
5th year	330	110	200	640	60 g	5 tons/ha.

In early 1980s dolomite application was started in the tea plantation of Bangladesh which supplies the essential nutrient magnesium as well as raises the soil pH. To replenish the deficiencies of phosphorus in tea triple super phosphate (TSP) was mainly used as phosphatic fertilizer since 1960. On the basis of growth and development of the seedlings NPK fertilizers should be applied per plant. For young tea fertilizer recommendations formulated on the basis of age while for mature tea it was formulated on the basis of yield per hectare in a plantation (Motalib and Dutta, 2011). After 6 months of planting tea (1st year) the application of fertilizer should be done. Usually after a little good shower of rain of 15-20 cm when the soil contains adequate moisture is considered the best time for fertilizer application. For better utilization of fertilizer the mixture should be applied in 3 split doses.

Dose	Time of application
1st dose	April/May
2nd dose	August
3rd dose	October/November

Recommended doses of fertilizer for mature tea

This fertilizer recommendation for mature tea has been formulated on the basis of yield of tea in kg per hectare which is given in the Table 1.6. Minimum 2 split doses should be applied for getting better yield. Up to 2000 kg/ha yield for every 100 kg yield increase amount of N/ha should be increased by 5kg. Above 2000 kg/ha yield for every 100 kg yield increase amount of N/ha should be increased by 6kg and it will be continue up to 3000 kg/ha yield. Moreover, from 1000 to 3000 kg/ha yield for every 100 kg yield increase amount of P_2O_5/ha should be increased by 1 kg. Additionally, from 1000 to 2500 kg/ha yield for every 100 kg yield increase amount of K_2O/ha should be increased by 3 kg and after that the amount will be remain constant (Motalib and Dutta, 2011).

Table 1.6. Fertilizer recommendations for mature tea.

yield kg/ ha	Fertilizer kg/ha in 1st split		
	N	P_2O_5	K_2O
Upto 1000	50	20	30`
	2nd split		
1000-3000	60	-	30

After a few good showers with sufficient moisture in the soil 1st dose of fertilizer application should be done in the month of March/April. By the last week of July or 1st week of August 2nd dose of fertilizer application should be done.

1.6 Pest management in tea plantation

Chen and Chen (1989) reported total 1116 species of various types of pests observed in the tea gardens of different parts of the world. In Bangladesh tea there are hitherto twenty-five insects, four mites and ten nematodes species were observed (Ahmed, 2005). Loss of yield and degradation of the quality of made tea are results from pest infestation in tea plantation. Numerous pests mainly insects, mites and nematodes can damage huge crop. When pest outbreak or disease frequency is very severe then crop loss may be extended to as high as 50%. Muraleedharan and Chen (1997) reported that yield loss can be 100% in some cases. Ahmed

(2005) reported that about 10–15% annual loss of the yield of tea occurred by pest attack in Bangladesh. The main pests of mature tea plantation are red spider mites, tea mosquito bug and termites while the main pests of tea nursery and young tea plantations are flushworms, thrips, aphids, jassids and nematodes. Since 1960 diverse types of pesticides such as organophosphate, carbamates, organochlorine, pyrethroids etc. have been used in the tea plantations to control the invasion of red spider mite (Ahmed, 2005). Gradually the uses of pesticides are rising in the agriculture sector. Pesticides are greatly necessary for raising the production of tea. Uses of pesticides to regulate pest attack in the tea gardens of Bangladesh are an important strategy (Alam, 1999). Diverse types of pesticides such as dimethoate, sulphur, propargite, ethion, quinalphos, abamectin, fenvalerate, bifenthrin, fenpropathrin etc. are being used to regulate the invasion of red spider mite in the tea plantation of Bangladesh (Mamun *et al*., 2014a; Alam *et al*., 2017). Uses of synthetic chemicals for pest management in tea plantation are doing during the last several decades. An integrated pest management programme is a vital segment of viable tea cultivation (Mamun *et al*., 2014b). In the last few years non-chemical methods has been tried to introduce to regulate the infestations of pests, diseases and weeds in the tea gardens. Integrated pest management (IPM) is the wise application of chemicals along with biological and cultural practices to decrease the pest attack in the tea plantation (Mamun *et al*., 2014b).

1.7 Tea disease management

Fungal pathogens and nematodes are mainly responsible for diseases in tea plants. Algae are responsible for a disease in tea plants. In Bangladesh tea, nineteen fungal and one algal disease have been identified. Gall disease on a particular plant has been observed in the year 1998. However, the disease occurred sporadically, but its intensity was severe (Ali *et al*., 2013). Few year observations revealed that 10–15% crop loss can cause by disease in tea (Ali *et al*., 2013). Gall disease, die back, black rot, brown blight, grey blight and blister blight are the diseases of upper canopy of tea. Branch canker, red rust, horse hair blight and thread blight are the mid-canopy diseases of tea. Charcoal stump rot, collar rot, violet root rot, purple root rot and nematode are the lower canopy diseases of tea (Rahman, 2009). To combat the infestation of diseases in tea plantation various types chemicals and fungicides are using at different doses such as Carbendazim, Copper Oxychloride, Copper Hydroxide, Tridemorph, Mancozeb, Bordeaux mixture+Cupraneb, Azoxystrobin+Difenoconazole, Formalin 40% etc (Akonda *et al*., 2017).

1.8 Weed management in tea

In Bangladesh, thirty-six species of monocot and dicot weeds have been observed in tea plantations (Sana, 1989). The widespread monocot weeds found in the tea plantations of Bangladesh are ulu or sun grass, motha, durba, makarjuri, bindi Motha, bahia ghash and enella as well as the dicot weeds found in tea plantations are bagrakote, lojjaboti, mikania, potti, gheto, gandhali bon, katanote, button, boro dudia, bond honey, dudia, neel pathar, ayapan, shoto dudia and shathoddron (Sana, 1989). It is reported that in the world wide about 15-20% crop loss in the tea plantations occur due to weed invasion. The assessed loss of the yield of tea crop by weeds invasion in tea areas of Bangladesh, Northeast India, South India and Sri Lanka is known to be 15–20%, 12%, 6–12% and 9%, respectively (Singh, 1999; Kotoky, 2011).

Weed infestation has many adverse effects on tea plantation. Weeds will contest with tea plants for nutrient elements, moisture, sunlight and space as well as hamper the other agronomic operations of tea crops. Moreover, they will play role as a different hosts for pests, diseases and upsurge the production cost. Weeds will decrease the yield and excellence of tea (Singh, 1999; Kotoky, 2011). Observing and identification of weed species as well as preference of active weeds resistor approaches are very important. Moreover, choice of proper herbicides, identification of right time and place of use as well as safety should be considered with precautions (Singh, 1999; Kotoky, 2011). Traditional, mechanical, biological and chemical weed control methods are using in the tea plantations of Bangladesh. Traditional method includes uses of mulch materials, growing cover crops, planting with close spacing and infilling the empty place in the tea garden. Sickling, cheeling, forking and hand weeding are mechanical methods (Sana, 1989). Living organisms such as insects, pathogens, herbivores are using in biological method of controlling weeds while chemicals or herbicides are using in chemical method. Depends on the chemical structures, process of action and selectivity different types of chemicls and herbicides such as 2,4-D, MCPA, Atrazine, BecAno, Simazine, Gramoxone, Paraxone (paraquat), Roundup (glyphosate), Bimastar, Dalapon etc. are using in the tea plantations of Bangladesh (Akonda *et al*., 2017).

1.9 Biochemistry and quality of tea

Biochemistry of tea is related to the biochemical changes during the growth of the tea plants and manufacturing of tea (Choudhury, 2010). Polyphenolic compounds are mainly responsible for the quality of black tea. Approximate biochemical composition of the young tea shoots are presented in Table 1.7.

Tea quality mainly assessed in two ways, one is biochemical test and another one organoleptic test. In the auction markets, organoleptic tests are accepted traditionally due to low time and cost consuming having a large number of samples available for assessment. Tea quality is usually assessed by the tea taster, who ascertains his judgement on the basis of previous experience of tea from the producing area and knowledge of national and regional condition and feeling of taste of the consuming countries (Dutta, 2009). In our country, taster considers the characteristics of (a) the appearance of tea before preparation of liquor, (b) appearance of the infused leaf, (c) color of the liquor, (d) liquor taste and (e) the creaming down (Dutta, 2009). But the biochemical tests were carried out due to only when the product was suspected for the adulterated or shown abnormal characteristics (ISO 3720, 1977). Bangladesh Government has realized the necessity of a tea standard for internal and external human consumption and trade. Bangladesh Standards and Testing Institution (BSTI) adopted a tea standard on April 17, 1974 on the endorsement of the Agriculture Food Divisional Committee under the caption “Bangladesh Standard Specification for Tea, BDSS 808:1974”.

In the year 2008, BSTI again amended its standard BDSS 808:1974 (amended in 1988 and 1995) specification of black tea and green tea and has cordially accepted the ISO 3720 with a view to accelerate export of Bangladesh tea in the world market. ISO 3720 was applicable only for black tea, whereas BSTI has accepted it for both black tea and green tea (Choudhury, 2010). The required parameters for standard black tea are presented in Table 1.8.

Table 1.7: Approximate biochemical composition of young tea shoots (Choudhury, 2010).

Components	% of dry weight
Soluble in cold water	
Flavanols: Epigallocatechin gallate	9-13
Epigallocatechin	3-6
Epicatechin gallate	3-6
Epicatechin	1-3
Gallocatechin	1-2
Catechin	1-2
Flavanols and their glycosides	3-4
Leucoanthocyanins	2-3
Phenolic acids: Theogallin	2
Others	2
Total phenolics	**30**
Caffeine	3-4
Amino acids: Theanine	2
Others	2
Carbohydrates	4
Organic acids	0.5
Volatile substances	0.01
Partially soluble in hot water	
Polysaccharides: Starch	2-5
Others	12
Protein	15
Ash (inorganic material)	5
Insoluble in water	
Cellulose	7
Lignin	6
Lipids	3

Table 1.8: Requirements for standard black tea.

Characteristics	ISO requirements	Bangladesh
Water extract (%) minimum	32.0	32.0
Total ash (%)	4.0–8.0	4.0–8.0
Water soluble ash as percentage of total ash minimum	45.0	45.0
Alkalinity of water soluble ash (as KOH) %	1.0–3.0	1.0–3.0
Acid insoluble ash % maximum	1.0	1.0
Crude fiber	16.5	16.5

Source: Choudhury, 2010.

Some of the biochemical properties of tea are described briefly in below:

Tea polyphenols**:** Tea comprises small amounts of proteins, carbohydrates, amino acids, lipids, vitamins, minerals and some antioxidant properties. Polyphenols are mainly responsible for the aroma of tea which has some health benefits effects. Polyphenol has antioxidant properties. The concentrations of polyphenols are higher in the green tea than black or oolong tea. The unique polyphenolic compounds such as catechins, (-)-epigallocatechin-3-gallate (EGCG), (-)-epigallocatechin (EGC), (-)-epicatechin-3-gallate (ECG) and (-)-epicatechin (EC) are present in green tea. 50–70% of catechins in tea are (-)-epigallocatechin-3-gallate (EGCG). One cup of green tea contains 200 mg of EGCG which have chemopreventive / chemotherapeutic effects against several types of cancers (Khan and Mukhtar, 2019).

Caffeine**:** Caffeine is one of the most consumed biochemical components which are found in tea and coffee. Quality characteristics of tea such as taste, astringency and bitterness can be affected by caffeine (Tfouni *et al*., 2018).

Protein**:** Water-insoluble compound which comprise 82% glutelin and 13% prolamin is tea protein. Non-animal sources of proteins have more importance in the food industry. Tea protein is very active in decreasing blood lipid and removing peroxide free radicals (Wang *et al*., 2014).

***Theaflavin*: ** Theaflavin is one of the polyphenols which are primary red pigments in the black tea. It has several health benefits including abilities to reduce glucose and fat. Theaflavin has some properties to prevent atherosclerotic, obesity, viral infection, cancer, inflammatory, osteoporotic, dental caries and bacterial infections (Takemoto and Takemoto, 2018).

***Thearubigin*: ** Thearubigins (TRs) are typical dark, brown colour which is a key element of black tea. It has been assessed that 60%-82% of the solids in a usual infusion of black tea is the TRs are the most profuse group of phenolic pigments. In the 1960s the presence of TRs in black tea was first acknowledged and that are ascribed to its health and mouth-feel properties as well as its characteristic colour. One of the leading bioactive components present in black tea is TRs that could be acknowledged to some of these health effects. The inhibition of oxidative stress such as pro-oxidant enzymes and redox-sensitive transcription factors can be protected by TRs. Capability to control gastrointestinal motility and skeletal health, antioxidant, anticancer effects and antimutagenic are the recommended biofunctions of TRs (Bond and Derbyshire, 2020).

1.9.1 Component of Tea Quality

Made tea quality depends on some components of green leaf and maintaining manufacturing process. The quality of tea is a combination of some factors such as color, appearance and flavor which are influenced by chemical components produced during tea processing (Wood and Roberts, 1964). About 200 varieties of volatile and non-volatile bio-chemical components are present in tea such as caffeine, tea polyphenols, proteins, amino acids, soluble sugar, volatiles, lipids and vitamins. Each of these components is responsible for the quality of tea (Ravichandran and Parthiban, 2000). The made tea quality depends on chemical components present in tea leaves, cultivars use, plucking standard, pruning types and fermentation process (Ravichandran and Parthiban, 2000).

Shahiduzzaman and Eunus (2010) conducted a study about the plucking round which had an impact on tea quality. Within the same locality at different times of the year as well as from place to place leaf period varies. They mentioned that different plucking rounds starting from 6 to 10 days are in practice in Bangladesh. They found in their study that shorter round (6-7 day) was more suitable for tea. Plucking round have significant effect on yield of tea. Ravichandran and Parthiban (2000) stated that the quality of manufactured tea depends on the type of bush, plucking standard, handling of leaf, withering, rolling, fermentation, drying and general condition of the factory. Finally, quality of made tea as a finished product is affected by the producing techniques. Fine plucking gives shoots which are physically and chemically most suitable for the production of quality tea with good liquoring character and favorable character.

1.10 Heavy metals

Heavy metals have a density higher than 5 g/cm^3. Non-biodegradable and non-thermodegradable heavy metals are extremely persistent in the environment. They are readily accumulated at toxic levels. Heavy metals are not easily eliminated from our body due to their high molecular weight (Jana *et al*., 2017). Heavy metals are geologically occurring substances and the uptake of the metals by the plants and animals accelerates their entrance into the food chain (Jana *et al*., 2017). Weathering and volcanic eruptions are natural phenomena which introduced heavy metals to the environment aside from normal natural causes (Ntwampe and Moothi, 2018). At very low concentrations some of the heavy metals have toxic effects and classified as "dark side of chemistry" (Koller and Saleh, 2018).

1.10.1 Sources of heavy metals in tea garden soils and tea

Soil contamination by heavy metals and it's entry into the food chain is a threat to the environment and human health. Information about the concentrations of heavy metals in the tea garden soils of Bangladesh and tea (*Camellia sinensis*) plants are very limited. Consequently, an effort has made to create a databank on the important heavy metals such as chromium (Cr), Copper (Cu), Iron (Fe), Manganese (Mn), nickel (Ni) and Zinc (Zn). Among the heavy metals chromium and nickel are soil pollutants while iron, manganese, zinc and copper are recognized as micronutrients or trace elements. Trace elements are essential in minor quantities and produce optimum effects. Shortage or surplus of trace elements is detrimental to the plants. Uses of fertilizers in the tea fields supply the essential nutrients as well as increase the load of heavy metals (Nath, 2013). Soil, its organic matter contents, tea manufacturing process and environmental pollutions are the factors may be contributing to the accumulation of heavy metals in the tea leaves (Marbaniang *et al*., 2011). Acid soils are suitable for the growing of tea plants. Production of acid from the tea plant root secretions and application of acid forming fertilizers are generally could cause soil acidification (Zhang and Fang, 2007). In the low pH of soil, hydrogen ions can displace the heavy metals and tea plants can uptake the metal ions which ultimately enhance the agglomerate of heavy metals in the tea plants (Zhang and Fang, 2007).

Over the past fifty years, the use of agrochemicals such as fertilizers and pesticides has been increased which has been reported as a major source of heavy metals in tea plants (Yaqub *et al*., 2018). Macronutrients and micronutrients are required for the growth and development of tea plants. Synthetic fertilizers are widely applied to tea plantations to accelerate productivity. Although tea is a calcifuge crop, certain amount of calcium is required for the growth and productivity of tea (Seenivasan *et al*., 2016). Magnesium is a secondary nutrient and of vital importance. To correct soil acidity application of dolomitic lime has become a regular practice in Bangladesh which also met up the requirement of Ca and Mg to some extent (Alam, 1999). The practice of chemical fertilizer in tea gardens of Bangladesh is crucial. A huge amount of Urea, Triple superphosphate (TSP), and Muriate of potash (MOP) fertilizers are applying in the tea plantations to ensure the balanced nutrition for the tea plants. Zinc sulphate ($ZnSO_4$) fertilizer is also applied as foliar spray. Mohiuddin *et al*. (2017) reported that chemical fertilizers used in the agricultural field contained chromium, copper and zinc 280 – 1293, 107.4 - 527.8 and 284.9 - 968.3 mg/kg, respectively. Nath (2013) reported that chemical fertilizers contained 7 – 38 mg/kg nickel. These fertilizers are regularly used in the tea gardens which added heavy metals in the soil as well. In Bangladesh tea, chemical pesticides are still the basis of almost all the pest control (Alam, 1999). Foliar and soil application of adulterated fertilizers are the sources of nickel contamination in tea garden soils and tea plants. Franklin *et al*. (2005) reported 2.7 to 16 mg/kg nickel in potassic fertilizers and 19 to 24 mg/kg nickel in commercial phosphatic fertilizers as impurity. The heavy metal concentrations in the agrochemicals used in the tea gardens should be assessed as some of the toxic heavy metals do not have any tolerance limit in tea (Seenivasan *et al*., 2008a). The accumulation of Cu in tea plants might be increased due to the application of copper fungicides in the tea plantation. The main source of Cu contamination is copper oxychloride (Zazouli *et al*., 2010). Black teas prepared by CTC process contain greater concentrations of chromium as a local contaminant because the rollers use in CTC tea manufacturing techniques are made of stainless steel which contain chromium (17% w/w), whereas chromium content is lower in orthodox processed tea (Seenivasan *et al*., 2008a).

1.10.2 Status of heavy metals in tea soils, fresh tea leaves and made tea

Tea is a popular beverage which is economically and socially important. Approximately 18 to 20 billion cups of tea are consumed daily in the world. Drinking of tea can reduce serum cholesterol. It can prevent lipoprotein oxidation, decrease the risk of cardiovascular disease and cancer (Chung *et al*., 2003). Tea contains polyphenols, caffeine, fluoride, and essential elements which has immense importance due their antioxidative, antimutagenic and anticarcinogenic effects (Yao *et al*., 2004, Cabrera *et al*., 2003). Tea plants are grown in highly acidic soils where trace elements are possibly more bioavailable for the uptake by root which is the main reason of the existence of trace elements in tea (Han *et al*., 2006). On human health trace element contents of tea may have both advantageous and adversative effects. The daily dietary requirements of some of these elements can meet up from the regular consumption of tea (Deng *et al*., 1998). Human health may be adversely affected by trace elements content. Practices should be used to ensure food safety from excessive trace element contamination in tea leaves as it is a matter of alarming. Trace element contents of many South Asian teas have been investigated where tea is a traditional drink (Powell *et al*., 1993). There are two important aspects of trace elements determination in tea (a) to evaluate their nutritional value and (b) to protect against any possible ill-effects.

Chromium (Cr)

Although chromium (Cr) is an important trace element for both human being and animals but it can cause antagonistic effects due to the introduction of chromium (Cr) in human beings through food chain (Schwarz, 1972). Chromium is found in all segments of the environment. The concentration of chromium in soil ranges from 10 to 50 mg/kg depending on the parent material (Adriano, 1986). Plants can uptake chromium in two forms such as, Cr (III) and Cr (VI). Chromium (VI) is more soluble in water and highly toxic to biota while chromium (III) is less soluble and less toxic (Adriano, 1986). Chromium causes toxicity and disorders some metabolic processes of plants. Chlorosis, photosynthetic impairing, stunting, reduction of root growth and finally plant death occurs due to chromium toxicity to the plants (Gardea-Torresdey *et al*., 2004). The effects of chromium toxicity on leaf and root growth in plants reduced the yield as well as inhibit enzymatic activities and mutagenesis (Gardea-Torresdey *et al*., 2004).

Chromium in tea garden soils

Chromium was not found in the tea garden soil samples of Moulvibazar and Sylhet of Bangladesh (Rashid *et al*., 2016). Zhou *et al*. (2009) reported that the concentration ranges of chromium in tea plantation of 12 counties in Zhejiang Province of China were 10.0-121 mg/kg. The concentrations of chromium in surface (0-15 cm), subsurface I (15-30 cm) and subsurface II (30-60 cm) soils were 68.73 to 102.02, 56.0 to 94.07 and 46.58 to 88.93 mg/kg, respectively in the tea garden soils of Dibrugarh and Sivasagar districts of Assam, India (Nath, 2013). Sun *et al*. (2017) reported that the concentration range of chromium in soil was 11.80 to 48.80 mg/kg in the Tieguanyin tea Garden of southeastern China. Zhang and Fang (2007) reported that the concentrations of chromium in the surface soils of the tea plantations of different age such as 2, 7, 18, 40 and 70 years at Jinghua, Zhejiang province, Southeast China were 53.1, 49.7, 50.3, 47.7 and 41.3 mg/kg, respectively.

Chromium in tea leaves

Research data of chromium in tea leaves is very limited. Tea plants uptake about 1 g of chromium from soils to produce 1 ton of made tea (Natesan and Ranganathan, 1990). Chen *et al*. (2009) reported that 0.39 and 0.97 mg/kg chromium was found in leaves of eight tea cultivar. Rashid *et al*. (2016) reported that the concentration of chromium in fresh tea leaves of the tea gardens of Moulvibazar and Sylhet of Bangladesh was below the detection limit. The concentration of chromium in Japanese, Chinese, Iranian and Thai green tea leaves were 0.024, 0.14, 0.05 and 0.06 μg/g, respectively (Limmatvapirat *et al*., 2012; Cabrera *et al*., 2003).

Chromium in made tea

The concentration of chromium varied from 2.5 to 11.4 mg/kg in marketable black tea of South India (Seenivasan *et al*., 2008b). However, Cr was below 10 mg/kg in the majority

(90%) of the samples tested. Narin *et al*. (2004) reported 16.9±1.5 mg/kg chromium in the maximum Turkish tea samples. The concentration of chromium in 801 tea samples tested in China was found from the BDL (Below Detection Limit) up to 16.10 mg/kg (Han *et al*., 2005). Yaqub *et al*. (2018) reported that the concentration of chromium in tea samples collected from the tea gardens of Pakistan were 2 ± 0.7 mg/kg. Rashid *et al*. (2016) reported that high concentrations of chromium were detected in the black tea (3.581 ± 3.941 mg/kg) of Bangladesh. Moreover, the concentrations of chromium in black tea from India, China, Sri Lanka and Turkey were 0.371, 0.155, 0.050 and 3.000 mg/kg (Cabrera *et al*., 2003; Soylak and Aydin, 2011).

Copper (Cu)

Copper (Cu) is a micronutrient for plants which is highly phytotoxic at high concentrations (Brun *et al*., 2001). It is detrimental to human health if copper consumption from food and beverages is excess (Kawada *et al*., 2002). Therefore, it is essential to evaluate and regulate the concentration of copper in water, beverages, and food regularly.

Copper in tea garden soils

The concentrations of copper in surface (0-15 cm), subsurface I (15-30 cm) and subsurface II (30-60 cm) soils were 16.73 to 36.33, 15.35 to 31.73 and 13.17 to 29.13 mg/kg, respectively in the tea garden soils of Dibrugarh and Sivasagar districts of Assam, India (Nath, 2013). Sun *et al*. (2017) reported that the concentration range of copper in soil was 7.580 to 26.50 mg/kg in the Tieguanyin tea Garden of southeastern China. Chand *et al*. (2011) reported that the concentrations of copper in the soils of the tea gardens of Palampur, India were ranged from 22 to 51 mg/kg. Zhang and Fang (2007) reported that the concentrations of copper in the surface soils of the tea plantations of different age such as 2, 7, 18, 40 and 70 years at Jinghua, Zhejiang province, Southeast China were 37.3, 35.3, 29.3, 30.6 and 23.6 mg/kg, respectively.

Copper in tea leaves

The concentration of copper in the tea leaves of Yuyao County, China was ranged from 8.05 to 33.50 mg/kg, with an average of 13.26 mg/kg (Jin *et al*., 2008). The Chinese Ministry of Health imposed the permissible limit of copper is 60 mg/kg. Zhang *et al*. (2018) reported that the copper concentrations in young tea leaves are significantly higher than mature tea leaves. The concentration of copper in the tea leaves of Puan tea area of southwestern China ranges from 14.90 to 26.10 mg/kg, with an average concentration of 18.57 mg/kg.

Copper in made tea

The presence of higher concentrations of copper in black tea might be due to the overuse of copper fungicides in the tea plantation (Seenivasan *et al*., 2008a). Lepp and Dickinson (1985) reported that the concentrations of copper in the tea samples collected from nine tea-growing regions of the world exceeded the normal range and the highest concentration of copper 78 mg/kg was found in the samples produced from China. The concentrations of copper in the made tea samples of South India were ranged from 15.9 to 32.2 mg/kg (Seenivasan *et al*., 2008a). According to Food Adulteration Act, 1954 (PFA), India the permissible limit of Cu is 150 mg/kg. The concentrations of copper in the tea samples of main tea producing provinces of China were ranged from 2.04 to 447.50 mg/kg (Han *et al*., 2005). Ramakrishna *et al*. (1986) reported that the concentrations of copper in tea samples of Sri Lanka were ranged from 10.0 to 25.0 mg/kg. The concentration of copper in the tea samples of Sri Lanka were much lower than the concentrations of copper in the tea samples of Australia, the United Kingdom and the United States (150mg/kg; Kumar *et al*., 2005), Turkey (120 mg/kg; Narin *et al*., 2004) and Japan (100 mg/kg; Fuchinokami and Fuchinokami, 1999). The concentrations of copper in the green tea of Japan were ranged from 4.7 to 36.5 mg/kg with a mean value of 11.4 mg/kg (Tsushida and Takeo, 1977). Podwika *et al*. (2018) reported that in southern Poland the mean concentration of copper in black tea was 21.3 ± 6.9 mg/kg and the mean concentration of copper in green tea was 17.5 ± 5.0 mg/kg.

Iron (Fe)

Iron is one of the most important constituents of the lithosphere. About 5% of the earth's crust is made up of iron. In plants, animals, and humans iron plays a distinct character in the activities of several trace elements. For all forms of life iron is an indispensable nutritive element. As a cofactor, iron can act for several enzymes. Plant injury is most likely to occur due to iron toxicity in the strongly acid soils (Ultisols, Oxisols). The necessities of iron for individuals may vary from 8 to 18 mg per day. Iron is essential for oxygen carriage and the electron transmission. Meanwhile iron can be lethal in superfluous concentrations (Kabata-Pendias, 2011; Prkić *et al*., 2018).

Iron in tea garden soils

The concentrations of iron in surface (0-15 cm), subsurface I (15-30 cm) and subsurface II (30-60 cm) soils were 4.933 to 10.766, 4.405 to 9.962 and 3.206 to 8.531 mg/kg, respectively in the tea garden soils of Dibrugarh and Sivasagar districts of Assam, India (Nath, 2013). Zhang and Fang (2007) reported that the concentrations of iron in the surface soils of the tea plantations of different age such as 2, 7, 18, 40 and 70 years at Jinghua, Zhejiang province, Southeast China were 48.9, 46.1, 46.3, 42.4 and 38.3 mg/kg, respectively.

Iron in tea leaves

The levels of iron in fresh tea leaves have not been frequently documented. Yaylalı-Abanuz and Tűysűz (2009) reported that 0.015%, 0.017% and 0.023% iron in tea leaves of eastern Black Sea region, NE Turkey. Kotoky *et al*. (2013) reported that iron concentration of tea leaves collected from Assam, India ranges from 69.20 to 85.60 mg/kg with an average 80 mg/kg. Moseti *et al*. (2013) also reported that iron concentration of 24 tea leaves collected from Kenya ranges from 55 to 203 mg/kg.

Iron in made tea

Iron is the most desirable micronutrient in plants. Polyphenol can influence the bioavailability of this element as well as can markedly inhibit the absorption of iron. The standard of iron for black tea in Sri Lanka is 500 mg/kg (Soliman, 2016). Achudume and Owoeye (2010) reported greatest concentration of iron in made tea of Nigeria ranges from 442 to 1344 mg/kg. The concentrations of iron in 30 made tea samples of different countries ranged from 103 to 523 mg/kg (Street *et al*., 2006). The concentrations of iron in diverse varieties of black tea of Pakistan were ranged from 5.47 to 11.63 mg/kg (Ahmad *et al*., 2012). Moseti *et al*. (2013) reported that the concentrations of iron in the made tea samples of Kenya were ranged from 151 to 369 mg/kg. The average concentrations of iron in the made tea of Uganda, Rwanda and Tanzania were 344, 262 and 195 mg/kg, respectively. The concentrations of iron in commercially available Ethiopian black tea were ranged from 319 to 467 mg/kg (Gebretsadik and Chandravanshi, 2010).

Manganese

For humans and other animals, manganese is an indispensable element and has pervasive significance. However, both deficiency and overexposure of manganese has adverse effects. Particularly, inhalation exposure of manganese is responsible for neurological problems. An epidemiological study reported that prolonged exposure to very high levels of manganese in drinking water have adverse neurological effects (Powell *et al*., 1998).

Manganese in tea garden soils

The concentrations of manganese in surface (0-15 cm), subsurface I (15-30 cm) and subsurface II (30-60 cm) soils were 118.53 to 420.53, 103.73 to 390.33 and 92.07 to 377.50 mg/kg, respectively in the tea garden soils of Dibrugarh and Sivasagar districts of Assam, India (Nath, 2013). Zhang and Fang (2007) reported that the concentrations of manganese in the surface soils of the tea plantations of different age such as 2, 7, 18, 40 and 70 years at Jinghua, Zhejiang province, Southeast China were 257, 263, 223, 183 and 155 mg/kg, respectively.

Manganese in tea leaves

Tea plants can store manganese within the concentration range of 390 to 900 mg/kg in leaves (AL-Oud, 2003). Yemane *et al*. (2008) reported that the concentrations of manganese in five tea clones of Ethiopia were ranged from 501 and 1281 mg/kg. The concentrations of manganese in eight different tea cultivars of China were ranged from 950.1 to 1224.2 mg/kg (Chen *et al*., 2009).

Manganese in made tea

Manganese in tea leaves has biochemical significance. Kumar *et al*. (2005) reported that the concentrations of manganese in black tea of India were ranged from 371 to 758 mg/kg. The concentrations of manganese in the tea leaves of various countries were ranged from 300 to 900 mg/kg while tea leaves of Turkey and Japan were contained manganese from 1100 to

2678 mg/kg (Kumar *et al*., 2005). Fernandez-Caceres *et al*. (2001) reported that the concentrations of manganese in 46 commercial made tea samples in Spain were ranged from 148 to 1595.4 mg/kg with an average 824.8 mg/kg.

Nickel

Nickel (Ni) is an important heavy metal due to its possible toxicity to plants and animals. The soils can be polluted by nickel through agricultural activities and mining (Everhart *et al*., 2006; Peltier *et al*., 2006). A low concentration of nickel is indispensable for plant growth which is a well-known fact (Brady and Weil, 1999; Reeves and Baker, 2000). Nickel becomes lethal for maximum plant species if its concentration increases beyond certain limit (Brady and Weil, 1999; Peralta-Videa *et al*., 2002).

Nickel in tea garden soils

The concentrations of nickel in surface (0-15 cm), subsurface I (15-30 cm) and subsurface II (30-60 cm) soils were 34.40 to 65.37 mg/kg, 30.67 to 60.00 mg/kg and 19.13 to 46.27 mg/kg, respectively in the tea garden soils of Dibrugarh and Sivasagar districts of Assam, India (Nath, 2013). Chand *et al*. (2011) reported that the concentrations of nickel in the tea garden soils of Palampur, India were ranged from 3.1 to 4.0 mg/kg. Zhang and Fang (2007) reported that the concentrations of nickel in the surface soils of the tea plantations of different age such as 2, 7, 18, 40 and 70 years at Jinghua, Zhejiang province, Southeast China were 23.4, 25.4, 20.3, 19.2 and 17.6 mg/kg, respectively.

Nickel in tea leaves

Zhang *et al*. (2018) reported that the concentrations of nickel in young tea leaves of southwestern China were ranged from 6.33 to 14.90 mg/kg while the concentrations of nickel in the mature tea leaves were ranged from 3.43 to 14.20 mg/kg. Wen *et al*. (2018) reported that the concentrations of nickel in tea leaves Nanjing, China were ranged from 4.26 to 24.08 mg/kg. Nickel can accumulate in various parts of the tea plant in a diverse pattern. Its accumulation gradually increases from bud to the feeder roots. The concentration of nickel in mature tea leaf of south India was 2.59 mg/kg (Seenivasan *et al*., 2016).

Nickel in made tea

Nickel has no permissible limit in tea. Due to the toxic nature of nickel, the agrochemicals used in the tea gardens need to be tested for the identification of heavy metals contamination. The concentrations of nickel in black tea samples of South India were ranged from 1.1 to 5.3 mg/kg (Seenivasan *et al*., 2008a). Marcos *et al*. (1998) reported the concentrations of nickel in made tea samples ranged from 2.99 to 22.6 mg/kg. The concentrations of nickel in black tea of Pakistan were varied from 0.07 to 0.64 mg/kg (Ahmad *et al*., 2012). The concentrations of nickel in Chinese tea samples were 4.92 mg/kg while nickel concentrations in Indian and Sri Lankan tea samples were 4.08 mg/kg (Moreda-Piñeiro *et al*., 2003). The mean concentration of nickel in made tea samples of Asia and Africa was found 4.24 and 4.76 mg/kg, respectively (Moreda-Piñeiro *et al*., 2003).

Zinc

The earth's crust and worldwide soils contained an average of 70 mg/kg zinc. During weathering processes zinc is very mobile and it readily formed precipitate by reaction with carbonates while in the presence of sulfur anions it is absorbed by minerals and organic compounds. Zinc is very mobile, and it readily formed precipitate by reaction with carbonates during weathering processes. The zinc fractions which are most available to plants are related to the iron and manganese oxides. Zinc is the constituent of various types of enzymes which are indispensable for metabolic activities in plants. The continued use of zinc fertilizers increased the concentrations of zinc in soils and zinc toxicity. The common symptoms of zinc toxicity are chlorosis in young leaves and stunted growth of plants (Kabata-Pendias, 2011).

Zinc in tea garden soils

The concentrations of zinc in surface (0-15 cm), subsurface I (15-30 cm) and subsurface II (30-60 cm) soils were 21.43 to 65.20, 21.07 to 56.47 and 17.70 to 48.87 mg/kg, respectively in the tea garden soils of Dibrugarh and Sivasagar districts of Assam, India (Nath, 2013). Sun *et al*. (2017) reported that the concentrations of zinc in the tea garden soils of southeastern China were ranged from 54.90 to 639.0 mg/kg. Zhang and Fang (2007) reported that the concentrations of zinc in the surface soils of the tea plantations of different age such as 2, 7, 18, 40 and 70 years at Jinghua, Zhejiang province, Southeast China were 57.6, 52.3, 48.6, 44.4 and 42.3 mg/kg, respectively.

Zinc in tea leaves

Yaylalı-Abanuz and Tűysűz (2009) reported that the concentrations of zinc in tea leaves of eastern Black Sea region, NE Turkey were 22.90, 23.08, 24.30 and 27.52 mg/kg. The concentrations of zinc in tea leaves of Assam, India were ranged from 53.55 to 74.26 mg/kg with an average 69.16 mg/kg (Kotoky *et al*., 2013). Moseti *et al*. (2013) reported that the concentrations of zinc in the tea leaves of Kenya were ranged from 15.4 to 32.6 mg/kg. Zhang *et al*. (2018) reported that the concentrations of zinc in young tea leaves of southwestern China were ranged from 35.8 to 50.3 mg/kg while the concentrations of zinc in the mature tea leaves were ranged from 9.1 to 20.0 mg/kg.

Zinc in made tea

Tamuly *et al*. (2016) reported that the concentration of zinc in made tea of Assam, India was ranged from 20.5 to 39.26 mg/kg. The concentrations of zinc in black tea and green tea of South India were 25.39 ± 0.59 and 26.39 ± 0.92 mg/kg, respectively (Srividhya *et al*., 2011). The mean concentration of zinc in made tea samples of Pakistan were 0.1 ± 0.2 mg/kg (Yaqub *et al*., 2018). The concentrations of zinc in 30 made tea samples of different countries ranged from 21.5 to 82.2 mg/kg (Street *et al*., 2006). The concentrations of zinc in diverse varieties of black tea of Pakistan were ranged from 1.05 to 3.21 mg/kg (Ahmad *et al*., 2012). Moseti *et al* (2013) reported that the concentrations of zinc in made tea of Kenya were ranged from 18.8 to 44.9 mg/kg. The maximum permissible concentration (MPC) of zinc in tea is 50 mg/kg in the Kenya Standard KS 65: 2009 set by the Kenya Bureau of Standards (Moseti *et al*., 2013).

1.11 Tea and health

Tea leaves comprise numerous biochemical components with medicinal values, such as, caffeine, polyphenol and alkaloids etc. (Khormali *et al*., 2007). Polyphenols is responsible for the colour and strength of the liquor while caffeine acts as a mild stimulant. Useful fluorides, volatile oils, vitamin B, theobromine and theophylline are also present in tea (Ahmad *et al*., 2013). To a tea-drinker's self-contentment, a cup of tea may contain as much as 64 mg (1 grain) caffeine, 128 mg (2 grains) polyphenols, 0.299 gm amino acid, 0.09 gm pectin, 0.025 gm potassium, 0-006 gm calcium, and vitamin B equivalent to about 2 per cent of daily requirement of an adult. Polyphenols in tea are components of abundant importance because of their antioxidant capacity (Pereira *et al*., 2014). Tea is considered advantageous to human health due to its high concentrations of phenolic antioxidant components (Pereira *et al*., 2014). Both black and green tea contain very high mounts of antioxidant polyphenols. Therefore, drinking tea could reduce the adverse effects of excessive and prolonged inflammation (Amarakoon and Grimble, 2017).

Green tea has numerous health benefits and it significantly reduces the risk of several types of cancer (Yusuf *et al*., 2007; Schwarz *et al*., 2008; Wu *et al*., 2009). Therefore, tea could be consumed to provide part of the daily requirement of fluids. Extensive research on tea has not found adverse effects of regular tea consumption. It has been established that tea consumption could improve the antioxidant defenses in the human body. Further, research has established that tea consumption could contribute to the lowering of the risk of NCD's, such as heart disease, stroke, cancer and diabetes (Ortmann, 2004). It can improve the oral and digestive tract health and contribute to weight reduction. Due to the action of caffeine and theanine a cup of tea could refresh you and improve your performance, making it an ideal beverage for the workplace. Therefore, tea could be considered as a prudent choice to provide part of the daily requirement of fluids.

Permissible limits of heavy metals

According to Food Safety & Standard Authority in India the permissible limit of copper is 150 mg/kg specific for tea only (Jana *et al*., 2017). There are no permissible limits of other heavy metals for tea. The permissible limits of heavy metals in plants are given in Table 1.9.

Table 1.9: Permissible limits of heavy metals in plants.

Elements	Permissible limits (mg/kg)
Chromium	1.3
Copper	10
Iron	450
Manganese	200
Nickel	10
Zinc	50

Source: Dabanović *et al*., 2016.

1.12 Objectives of the study

Tea is a popular drink in Bangladesh. Tea is cultivated in different areas of the country. While tea has numerous health benefits, it has also been reported to contain toxic heavy metals. The soils of Bangladesh are inherently variable in their geochemical fingerprints. There are geomorphological differences in the different tea growing areas along with topographical variations and differences in the age of tea plantation, which may have potentials to affect the accumulations of different elements particularly heavy metals in the tea. Different tea management practices, such as applications of inorganic and organic fertilizers and pesticides, tea manufacturing processes and others can also affect the elemental concentrations in soil as well as in tea plants. Considering these variabilities, it is important to understand the elemental concentrations, particularly of different heavy metals, in the different the garden soils, tea plants and made tea of Bangladesh and their effects on the quality of tea. There are very limited information on the concentration of heavy metals in tea garden soils, tea leaves and made tea of Bangladesh. There is also limited scope to understand the extent, variabilities and relationships of elemental concentrations in the soils of the geomorphologically different tea growing areas of Bangladesh and their possible effects on the quality of tea produced. The present study involves an extensive tea garden soil survey with an aim to assess the extent and distribution of heavy metals in soils, tea leaves and made tea of the tea gardens located in different physiographic regions of Bangladesh.

The major objectives of the research presented in the thesis were:

1. To assess the extent and variability of a range of geochemical elements including heavy metals in the geographically different tea garden soils of Bangladesh;
2. To understand the geochemical relationships of the elements in the tea garden soils;
3. To assess the variability of the geochemical elements in the soils with respect to different physiographic regions, land topographic features, soil texture, drainage class, soil series and the age of tea plantation.
4. To assess the concentrations of heavy metals in tea leaves and made tea of different tea gardens of Bangladesh and their possible effects on the chemical and biochemical qualities of made tea.
5. To understand the relationships of heavy metals in the tea leaves and made tea; and
6. To estimate the transfer factors of heavy metals from soil to tea plants.

Chapter Two

General Methodology

2.1 Strategy for sample collection

Bangladesh covers a total area of 147,570 km^2 (56,977 square miles), and is divided into 8 administrative divisions: Barisal, Chattogram, Dhaka, Khulna, Mymensingh, Rajshahi, Rangpur, and Sylhet. These divisions are subdivided into districts, popularly known as "zila". There are 64 administrative districts in Bangladesh (Figure 2.1) and each district is subdivided into a number of sub-districts, called "upazila" (or, "thana"). Each upazila (except for those in metropolitan areas) is divided into a number of "unions", and each union comprises several villages, called "gram". Bangladesh has 3 major geomorphological units: Tertiary hills, Pleistocene terraces and Holocene floodplains (Brammer, 1996; Huq and Shoaib, 2013). To understand and characterize the physiography of the geomorphological areas, Bangladesh is divided into 20 main physiographic regions (FAO-UNDP, 1988) (Figure 2.2).

Tea growing areas in Bangladesh are located in 3 different ecological zones, namely Sylhet zone in the Surma valley, Chattogram zone in the Halda valley, and Panchagarh zone in the Korotoa valley. There are 167 tea gardens in Bangladesh. The Sylhet zone has 135 tea gardens, among the 135 tea gardens 91 tea gardens are in Moulvibazar district, 25 tea gardens are in Habiganj district and 19 tea gardens are in Sylhet district. The Chattogram zone has 23 tea gardens of which 21 tea gardens are in Chattogram and 2 tea gardens are in Rangamati districts. In the Panchagarh zone, Panchagarh district includes 8 tea gardens, and 01 tea garden belongs to Thakurgaon district (PDU, 2020). In Bangladesh, total leased area for tea gardens is 1,17,915.14 hectare among which tea are growing in 64,138.21 hectare. Other areas are covered by fallow land, rubber plantation, natural forest, stream/pond, infrastructure etc. (PDU, 2020).

Soil, tea leaf and made tea samples were collected from 13 tea gardens located in 6 sub-districts (Srimangal, Kamalganj, Sylhet Sadar, Fatikchhari, Panchagarh Sadar and Tetulia) of 4 districts (Moulvibazar, Sylhet, Chattogram and Panchagarh) of Bangladesh (Figure 2.1). A set of control soil samples were also collected from the fallow lands adjacent to the tea gardens. The samples were collected through 4 separate fieldwork campaigns conducted during the months of April 2019 (Tea gardens: Allynugger, Patrakhola, Sathgao, Rajghat, BTRI, Jagcherra and Malnicherra; Sub-districts: Kamalganj, Srimangal and Sylhet Sadar; District: Moulvibazar and Sylhet), November 2019 (Tea gardens: Karnafuli, Neptune and Oodaleah;

Sub-districts: Fatikchhari; District: Chattogram), October 2020 (Tea garden: Bilashcherra; Sub-district: Srimangal; District: Moulvibazar) and March 2021 (Tea gardens: Karotoa, Kazi and Kazi; Sub-districts: Panchagarh Sadar and Tetulia; District: Panchagarh).

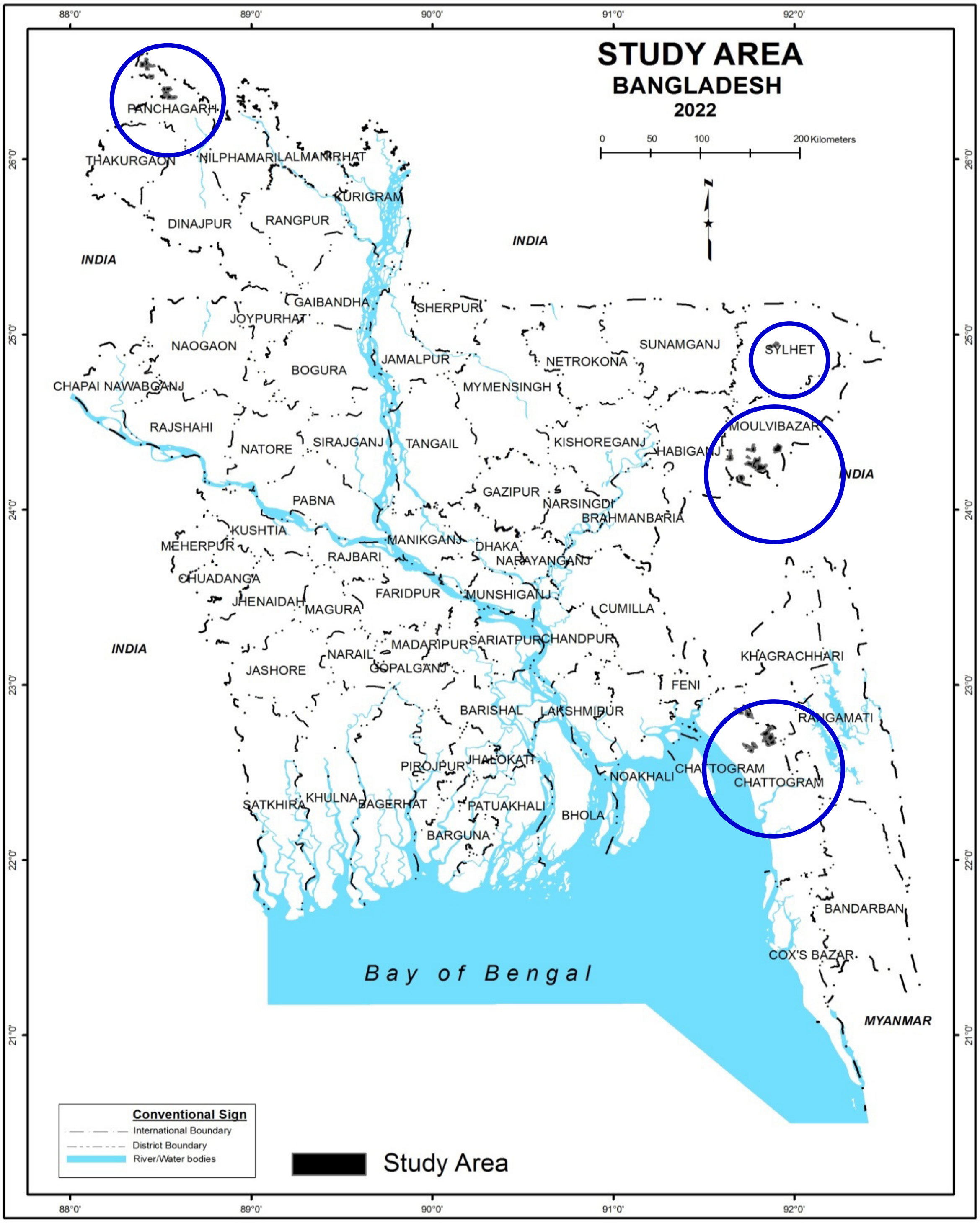


Figure 2.1: Study areas (4 districts Chattogram, Moulvibazar, Sylhet and Panchagarh are shown in blue circles) in the administrative district map of Bangladesh.

Before collection of the soil sample, soil series (the lowest category of soil taxonomy) identification was done. The sampling sites were selected considering different physiography, topographic position, age of tea plants, total area, presence of tea manufacturing unit and also considering different ownership & management of the tea gardens. The sampling locations belonged to 3 different physiographic regions of Bangladesh, namely the Northern and Eastern Hills, Northern and Eastern Piedmont Plains, and Old Himalayan Piedmont Plain (Figure 2.2). Among these Northern and Eastern Hills belongs to hills, whereas Northern and Eastern Piedmont Plains and Old Himalayan Piedmont Plain are floodplains. However, these tea growing areas have topographical variation. The tea growing areas of Sylhet and Moulvibazar are situated in the North-east of the country which is generally low hills, locally known as tillah with heights varying from 30 to 90 meters. Chattogram hills constitute the only significant hill system in the country with an altitude from 600 to 900 meters above from the sea level. Panchagarh, the new tea plantation region is situated on the Piedmont Plain of Himalayas. The old Himalayan Piedmont Plain is an old alluvial fan, apparently deposited by former courses of the Tista, Mahananda and Korotoa rivers.

Tea is a perennial crop which lives more than 100 years. Depending on the age, tea plants can be classified as immature tea, young tea, mature tea, old tea and very old tea. The economic life of tea plant is up to 50 years. After 50 years, with increase the age of tea plants yield might be reduce. Prior to sample collection, the presence of young tea (6-10 years), mature tea (11-40 years) and old tea (41-60 yrs) plantations in each selected tea gardens of Moulvibazar, Sylhet and Chattogram districts was confirmed. Soil and tea leaf samples were collected from young tea, mature tea and old tea sections of the tea gardens. In Panchagarh district tea plantation started in the year 2000, therefore, soil and leaf samples were collected only from young and mature (up to 20 years) tea plantation.

In general, soil samples from tea gardens are collected from three depths (0-23 cm, 23-46 cm and 46-92 cm). Soil samples up to 92 cm depth are tested for the feasibility study of a new area for tea cultivation. For uprooting, rehabilitation and replanting of tea plants, soil samples up to 46 cm depth are tested. Top soil layer (0-23 cm) is mostly considered for fertilizer application. In the present study we initially aimed to collect and analyzed top soil (0-23 cm) as generally recommended and most biologically active layer of the tea garden. Accordingly we collected 61 no of only top (0-23 cm) soil samples from 7 tea gardens. However, according to the expert suggestions given during the research presentation of first year progress we considered to collected soil samples from 0-23 cm and 23-46 cm depth.

It should be mentioned here that although a target was made in the initial sampling plan to collect more samples from different tea gardens of Bangladesh, it was not entirely possible due to the COVID-19 Pandemic situation in Bangladesh, and therefore, the length of the fieldwork in 2020 to 2021 was cut short.

2.2. Sampling sites

The name of the selected seven (7) tea gardens of Moulvibazar district are Sathgao (Proprietary garden), BTRI Farm (Bangladesh Tea Board), Bilascherra Farm (Bangladesh Tea Board), Rajghat (Finlay Tea Co), Jagcherra (Finlay Tea Co; only one green tea producing garden in Moulvibazar), Allynugger (Duncan Brothers) and Patrakhola (National Tea Company Ltd). Malnicherra (Private Ltd. Co.) of Sylhet district is the first commercial tea garden of Bangladesh which was established in the year 1854 were also selected for sample collection. From Chattogram district three (3) tea gardens such as Oodaleah (Private Ltd. Co.), Karnafuli (Private Ltd. Co.) and Neptune (Private Ltd. Co.) were selected for sample collection. From Panchagarh district two (2) tea gardens such as Kazi & Kazi (Proprietary garden) and Karatoa (Proprietary garden) were selected for sample collection. The numbers of soil, green tea leaf and made tea samples were collected from the selected tea gardens of Moulvibazar, Sylhet, Chattogram and Panchagarh districts are given below in Table 2.1.

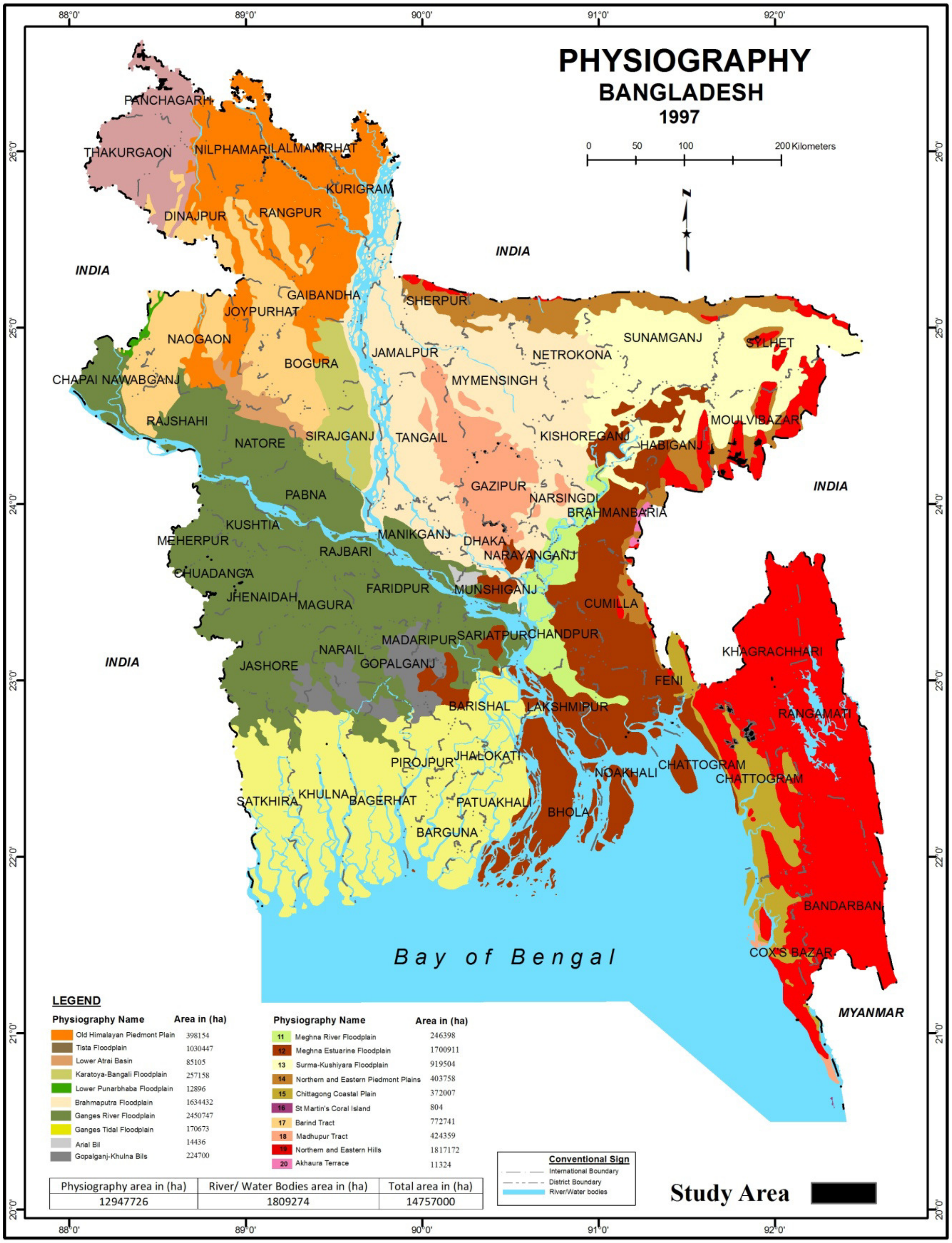


Figure 2.2: Study area in physiographic map sourced from Soil Resource Development Institute (SRDI).

Table 2.1: The districts, sub-districts and tea gardens from where the samples were collected. The numbers of soil, tea leaf and made tea samples from each district, sub-district and tea gardens are given, respectively, within the parentheses.

Sl. No.	Districts	Sub-districts	Tea gardens
1	Chattogram	Fatickcharri (surface soil = 27, sub-surface soil = 16; tea leaves = 23; made tea = 9)	Karnafuli (surface soil = 8, sub-surface soil = 4; tea leaves = 7, made tea = 3); Neptune (surface soil = 10, sub-surface soil = 6; tea leaves = 8; made tea = 3) Oodaleah (surface soil = 9, sub-surface soil = 6; tea leaves = 8; made tea = 3)
2	Moulvibazar	Kamalganj (surface soil = 16; tea leaves = 14; made tea = 6)	Allynugger (surface soil = 11, tea leaves = 9, made tea = 3); Patrakhola (surface soil = 5, tea leaves = 5, made tea = 3)
		Srimangal (surface soil = 56, sub-surface soil = 21; tea leaves = 48; made tea = 13)	BTRI (surface soil = 5; tea leaves = 5, made tea = 4), Bilashcherra (surface soil = 21, sub-surface = 21; tea leaves = 18; made tea = 4); Jagcherra (surface soil = 11; tea leaves = 9, made tea = 3) Rajghat (surface soil = 9; tea leaves = 8, made tea = 3); Sathgao (surface soil = 10; tea leaves = 8, made tea = 3)
3	Panchagarh	Panchagarh Sadar (surface soil = 9, sub-surface soil = 9; tea leaves = 6, made tea = 3)	Karotoa (surface soil = 9, sub-surface soil = 9; tea leaves = 6; made tea = 3)
		Tetulia (surface soil = 14, sub-surface soil = 14, tea leaves = 12; made tea = 5)	Kazi & Kazi (surface soil = 14, sub-surface soil = 14; tea leaves = 12; made tea = 5)
4	Sylhet	Sylhet Sadar (surface soil = 10; tea leaves = 9; made tea = 3)	Malnicherra (surface soil = 10; tea leaves = 9; made tea = 3)

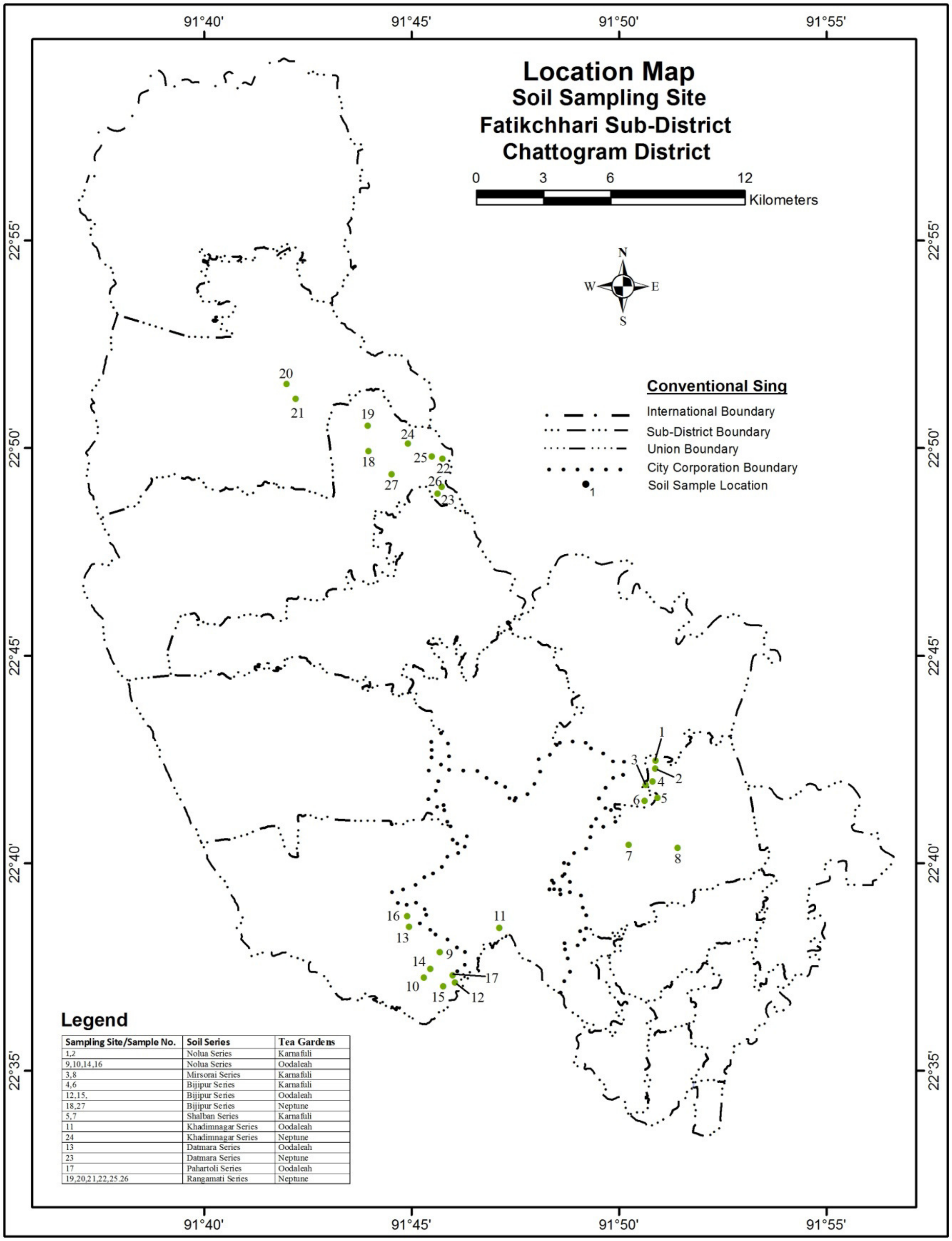


Figure 2.3: Map of Fatikchhari sub-district of Chattogram district with sampling locations indicated by green circles with sample numbers (surface soil = 27, sub-surface soil = 16; tea leaves = 23; made tea = 9).

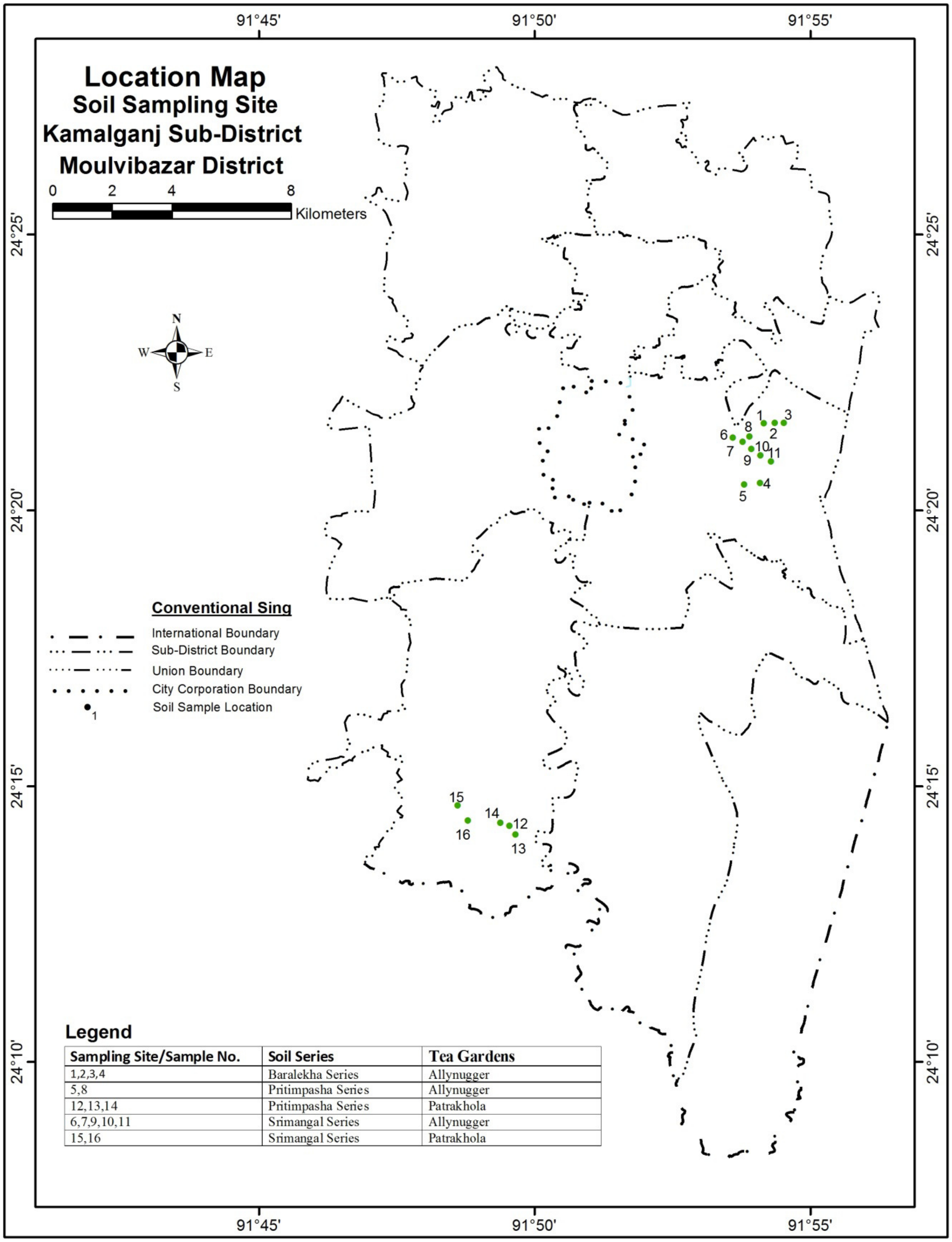


Figure 2.4: Map of Kamalganj sub-district of Moulvibazar district with sampling locations indicated by green circles with sample numbers (surface soil = 16; tea leaves = 14; made tea = 6).

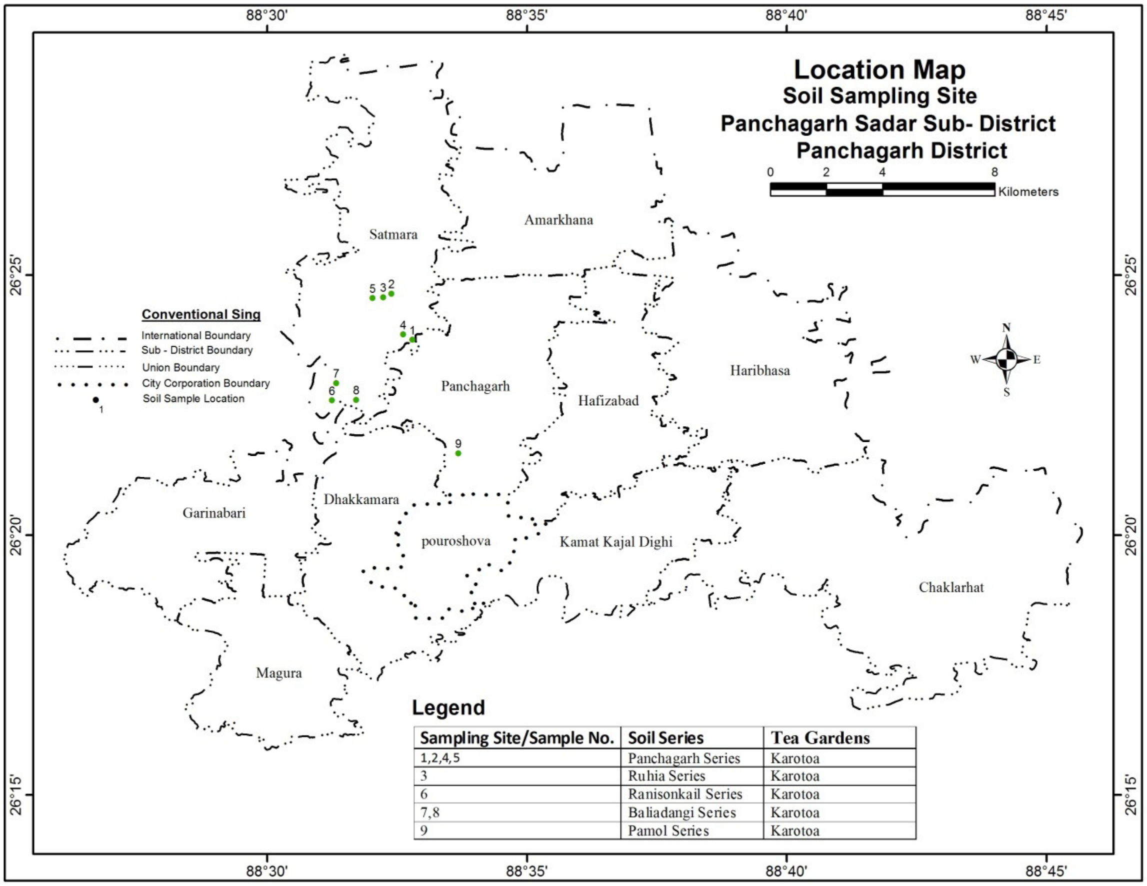


Figure 2.5: Map of Panchagarh Sadar sub-district of Panchagarh district with sampling locations indicated by green circles with sample numbers (surface soil = 9, sub-surface soil = 9; tea leaves = 6, made tea = 3).

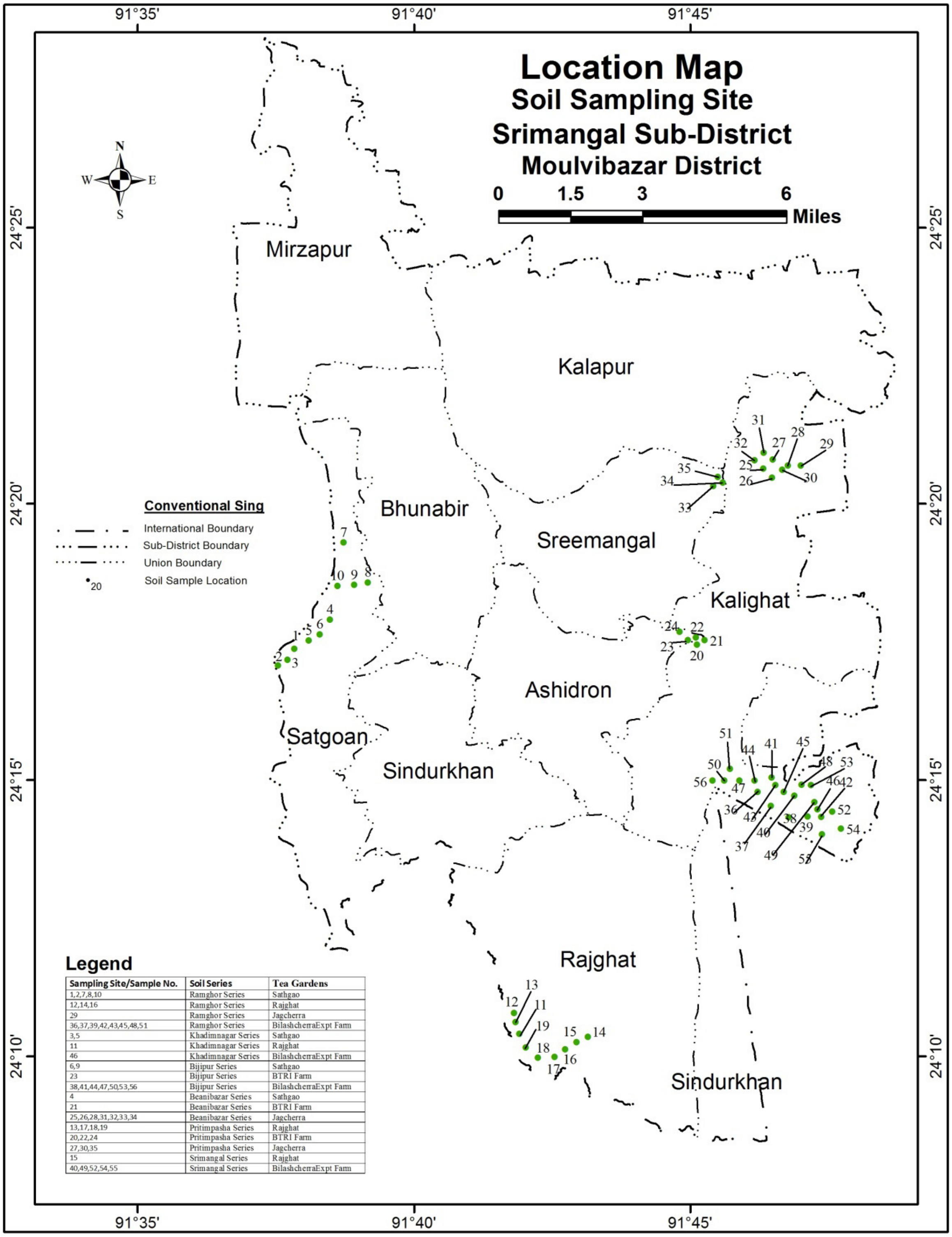


Figure 2.6: Map of Srimangal sub-district of Moulvibazar district with sampling locations indicated by green circles with sample numbers (surface soil = 56, sub-surface soil = 21; tea leaves = 48; made tea = 13).

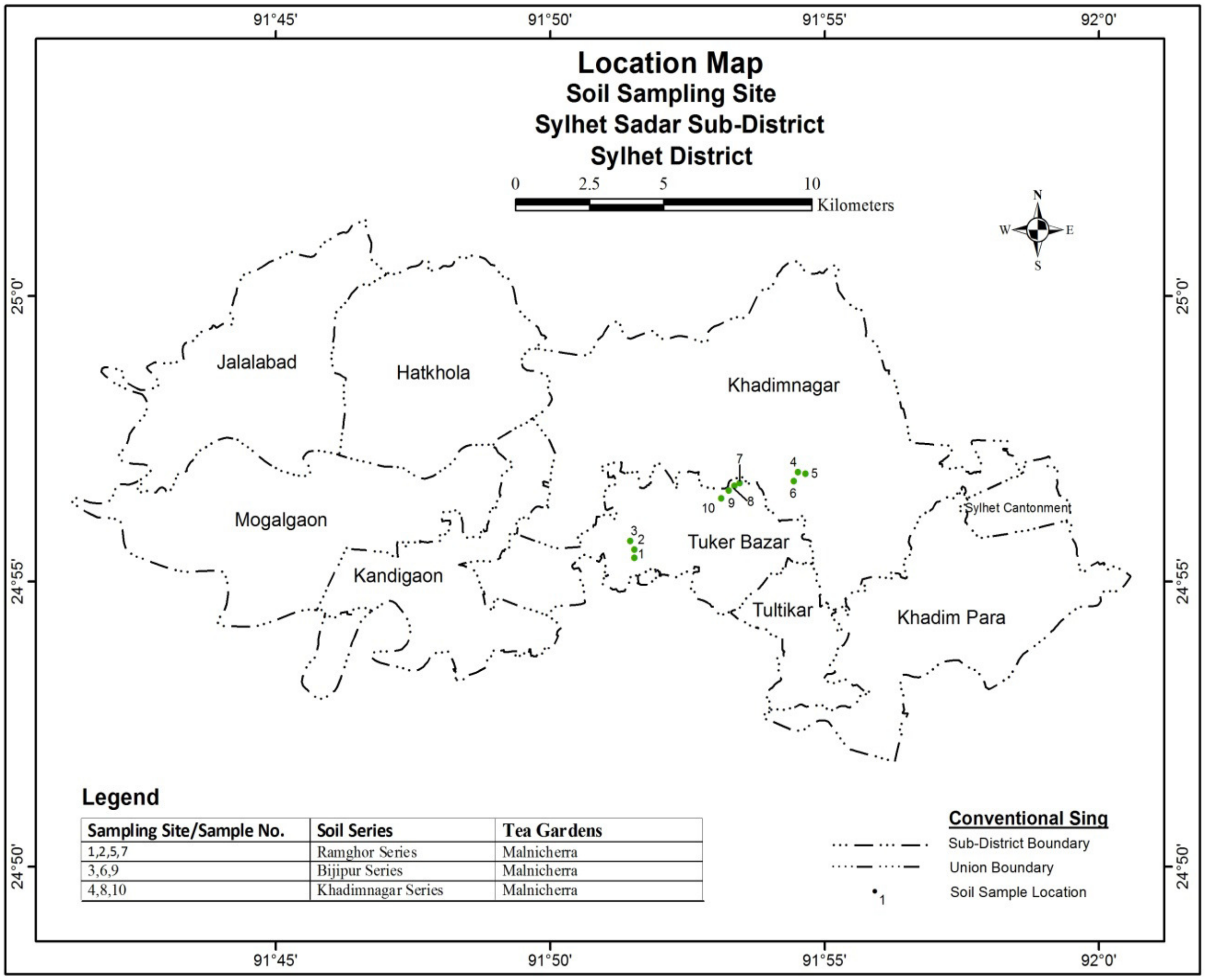
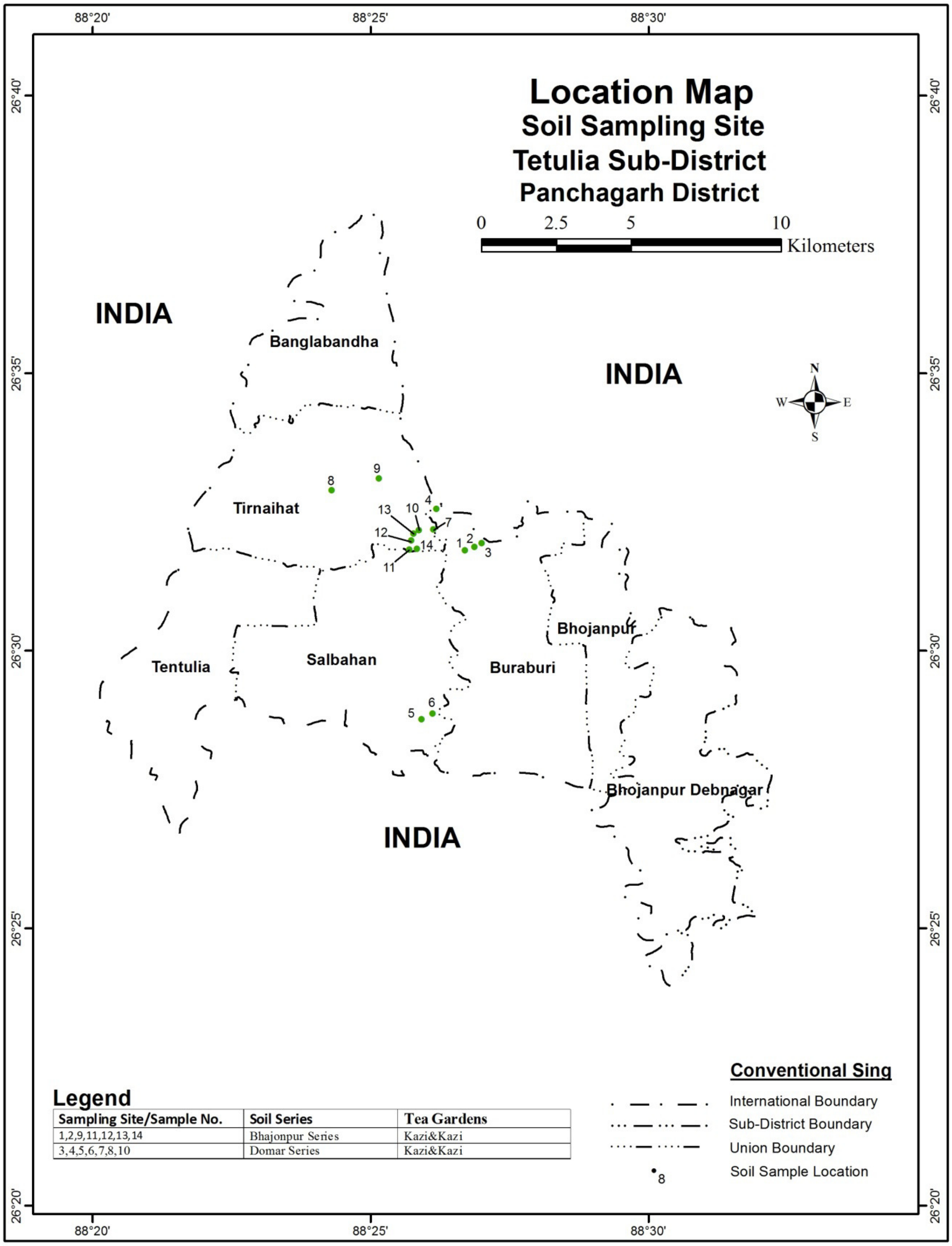


Figure 2.7: Map of Sylhet Sadar sub-district of Sylhet district with sampling locations indicated by green circles with sample numbers (surface soil = 10; tea leaves = 9; made tea = 3).



Sampling Site/Sample No.	Soil Series	Tea Gardens
1,2,9,11,12,13,14	Bhajonpur Series	Kazi&Kazi
3,4,5,6,7,8,10	Domar Series	Kazi&Kazi

Figure 2.8: Map of Tetulia sub-district of Panchagarh district with sampling locations indicated by green circles with sample numbers (surface soil = 14, sub-surface soil = 14, tea leaves = 12; made tea = 5).

2.3 Collection of samples

A total of 192 soil samples (surface soils at 0-23 cm depth = 132; subsurface soils at 23-46 cm depth = 60), 112 tea leaf samples, and 39 made tea samples were collected from 13 tea gardens. Soil (~1 kg) and tea leaf (~0.5 kg) samples were collected from young tea (6-10 yrs), mature tea (11-40 yrs) and old tea (41-60yrs) sections of each of the gardens. From the tea gardens of Allynugger, Patrakhola, Sathgao, Rajghat, Jagcherra, BTRI and Malnicherra, soil samples were collected from only 0-23 cm depth. Soil samples were collected by using the soil sampling tools called 'Nirani' and 'Boring' (Figure 2.9a-c). Soil samples from fallow lands (where tea plantations were never performed) adjacent to tea gardens were also collected, which were considered as the control (non-tea garden soils) soil (Figure 2.9d). Soil samples were collected from the tillah top, slope, and flat area (at the bottom of the tillah) of a tea plantation. The topology of the tea growing areas from where the samples were collected was categorized as level ridge, valley ridge, terrace, low hill and medium high hill. The longitude and latitude of sampling sites were recorded by using a GPS machine (Table A1). Sampling location maps were prepared with these longitude and latitude of sampling sites (Figure 2.3-2.8). The series of each of the soils was identified on the sites according to the information given in the sub-district land and soil resources utilization guide (locally known as Upazila Nirdeshika of SRDI) (Figure 2.9e-f). After collection, soil samples were kept in plastic bags, which were sealed and labeled properly. Soil samples were carried to the laboratory for analysis. Made tea samples (250-300 g) of different grades were collected from the tea factory at each of the gardens. Bilashcherra tea garden has no tea factory. All the tea leaves of Bilashcherra tea garden are processed in the tea factory of BTRI. The black tea grades such as GBOP, PF and CD were collected from the tea manufacturing units of the gardens. These grades are available in the local markets which are consumed by the people regularly. Green tea had no grades available in the tea factory. Made tea samples were collected in plastic bags, which were sealed and labeled properly. Samples of made tea were brought to the laboratory for chemical and biochemical analysis.

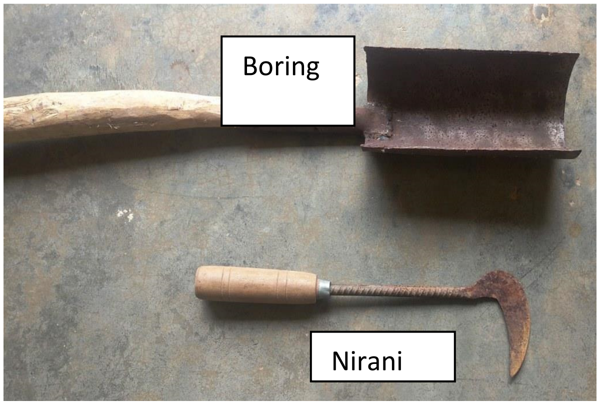


Figure 2.9a. Soil sample collection tool 'Nirani' and 'Boring'.

Figure 2.9b. Soil collection from a tea field using a boring.

Figure 2.9c. Soil collection from a tea field using a nirani.

Figure 2.9d. Soil collection from a fallow land.

Figure 2.9e. Soil sample collected for soil series/texture identification.

Figure 2.9f. Soil series identification using map of Upazila Nirdeshika of SRDI.

2.4 Samples processing and preparation

After collection at various tea gardens, the soil samples were transferred to and processed in the laboratory of Soil Science Division at Bangladesh Tea Research Institute, Srimangal. The soil samples were spread on clean polyethylene bags, and air-dried (Figure 2.10a). The air-dried soils were then ground using a wooden hammer. The big lumps were broken down, and plant roots, pebbles and other undesirable matters were removed. After homogenization, each sample was passed through a 2 mm mesh sieve. The cleaned soils were dried in the oven at 80°C ± 5°C for 48 h. Then, a portion of each of the oven-dried soil samples was preserved in clean sealed polyethylene bags for analysis.

Collected tea leaf samples were rinsed with distilled water and air-dried on sheets of paper (Figure 2.10b). After air dry samples were placed in a paper bag and marked the date, location of sampling and other relevant information on the bag. The paper bags were put in draft oven at 80°C for 48 hours (Huq and Alam, 2005). Then, the tea leaves samples were ground and stored in envelopes. Envelopes were then stored in polythene bag in a cool, dark place for later heavy metals analysis.

Made tea samples means processed tea from green tea leaves. It did not require further processing after collection of made tea samples from the manufacturing unit of a tea garden. The collected samples were stored in airtight condition because of its very hygroscopic nature.

(a) (b)

Figure 2.10: Processing of samples: soils are being air-dried (a), tea leaves are being air-dried (b).

2.5 Samples preparation for analysis

Digestion of the soil samples were performed with nitric acid (70% solution, RCI Labscan) using block digester. The method for the digestion procedure was: 1 g of sample was taken into an acid washed glass digest tube, and 10 ml of concentrated nitric acid was added to the tube and left overnight for predigestion at room temperature. Then, 5 ml of hydrogen peroxide was added to the soil sample just before digesting. The glass tube was heated on the block digester at 80°C for 1 hour, at 100°C for 1 hour, at 120°C for 1 hour, and finally, at 140°C for 3 hours until the solution was clear (Chowdhury, 2016). Once cooled, the digested samples were transferred into 50 ml volumetric flasks while rinsing the glass tubes thoroughly 3-4 times with distilled water. The volumes were made up to 50 ml mark (final volume accurately recorded). The samples were then shaken well to ensure that they were mixed properly. Then the samples transferred to plastic bottle through filter paper (Whatman No. 42). Digestion of tea leaves and made tea made tea samples were performed with nitric-perchloric acids suggested by Huq and Alam (2005).

2.6 Analysis of soil samples

The moisture content (%) of the soil samples were measured by halogen moisture analyzer (Precisa, EM 120-HR) as reported by Rasti *et al*. (2020). The particle size analysis of soil samples and determination of soil texture by hydrometer method was done (Huq and Alam, 2005). The pH of the soil samples was measured using a pH meter (Jenway-3510). Distilled water was used to make the soil suspension at a soil:water ratio of 1:2.5 (Huq and Alam, 2005). The suspension was allowed to stand for 0.5 to < 3 hour before taking the pH reading. The organic carbon contents of the soil samples were measured by Walkley and Black wet oxidation method. The soil samples were analysed for exchangeable potassium (K) using a flame photometer (Jenway PFP7). Exchangeable calcium (Ca) and magnesium (Mg) of the soil samples were measured by using Atomic Absorption Spectrophotometer (AA-7000, Shimadzu, Japan). The available phosphorus (P) was measured by UV-spectrophotometer (Shimadzu UV-1800). The boron (B) content was measured by UV-spectrophotometer (Shimadzu UV-1800). The soil sulfur was measured by turbidimetric method. The total concentrations of chromium, copper, iron, manganese, nickel and zinc was measured by Atomic Absorption Spectroscopy (AAS, Varian AA240, Australia). The tea garden soils are generally rich in iron and manganese. In addition, the fertilizers such as Triple superphosphate (TSP), Diammonium phosphate (DAP), Muriate of potash (MOP) and zinc sulphate, which are regularly used in the tea gardens of Bangladesh, usually contain chromium, copper, zinc and nickel as impurities (Nath, 2013; Mohiuddin *et al*., 2017). Considering these, the six heavy metals such as chromium, copper, iron, manganese, nickel and zinc were selected in the present study.

2.7 Chemical analysis of leaf samples

The total concentrations of chromium, copper, iron, manganese, nickel, and zinc of the leaf samples was measured by the Atomic Absorption Spectroscopy (AAS, Varian AA240, Australia).

2.8 Physical, chemical and biochemical analysis of made tea samples

The moisture content (%) of the made tea samples were measured by halogen moisture analyzer (Precisa, EM 120-HR). After acid digestion, the total concentrations of chromium, copper, iron, manganese, nickel and zinc of the made tea samples was determined by the Atomic Absorption Spectroscopy (AAS, Varian AA240, Australia). According to Belay *et al*. (2008) 50 mg of made tea sample was weighed and dissolved in 25ml distilled water of a 50 ml beaker. The solution was stirred for one hour using magnetic stirrer and heated gently to remove caffeine easily from the solution. In addition, the solution was filtered by a glass filter to get rid of particle from solution. Finally, the digest was cooled, filtered, diluted to 25 ml and transferred into dry plastic bottles. The digest was used for the determination of caffeine content of made tea. The digest was analyzed by a Shimadzu LC-20AT HPLC system with a Shimadzu SPD-M20A detector at 272 nm. The protein contents of tea samples were analyzed by determining the nitrogen content and multiplying the nitrogen with factor 6.25 according to Anon (2000) by using micro Kjeldahl method. Moreover, total polyphenol content was measured by UV-spectrophotometer (T60, PG Instruments) as reported by Yao *et al*. (2006). Additionally, theaflavin and thearubigin content of made tea was measured by UV-spectrophotometer (T60, PG Instruments) as reported by Jiang *et al*. (2018).

2.9 Soil Mapping

ArcCatalog 10.5 and ArcMap 10.5 (ArcGIS software, Esri) were used to create soil maps with sample locations.

2.10 Statistical analysis

Statistical analyses, such as descriptive statistics, correlation coefficient, regression analysis, one-way analysis of variance (ANOVA), Principal Component Analysis (PCA), and Tukey's significance test were performed, and a variety of graphs were plotted using the statistical software Minitab 21 and SigmaPlot 14.5. The ANOVA was performed to determine the variability of the parameters, and correlation and regression techniques were used to determine the relationships between the parameters tested.

Chapter Three

Extent and Distribution of Geochemical Elements in the Tea Garden Soils of Bangladesh

Abstract

Soils from the tea gardens of Bangladesh located at different geographic areas were analyzed to assess the extent and distribution of a range of geochemical elements including heavy metals. The concentration of chromium, copper, iron, manganese, nickel and zinc in the surface soils of the different tea gardens ranged between 2 - 73, 0.86 - 23, 3084 – 47120, 21 – 1274, 3 – 23 and 2 – 465 mg/kg, respectively. In the sub-surface soils of different tea gardens, the concentration of chromium, copper, iron, manganese, nickel and zinc were ranged between 3 - 89, 1 – 22, 2640 – 52540, 49 – 466, 4 – 24 and 2 - 71 mg/kg, respectively. Significant variations ($0.001 > p < 0.05$) in the concentration of the heavy metals were observed between the surface and sub-surface soils of the tea gardens. Chromium in the tea garden soils were observed to be significantly and positively correlated with total nitrogen ($r = 0.43$, $p < 0.001$), boron ($r = 0.37$, $p < 0.001$), copper ($r = 0.39$, $p < 0.001$), iron ($r = 0.69$, $p < 0.001$) and nickel ($r = 0.55$, $p < 0.001$). Significant and positive correlation of copper with nitrogen ($r = 0.21$, $p < 0.05$), iron ($r = 0.40$, $p < 0.001$) and nickel ($r = 0.44$, $p < 0.001$) were observed in the tea garden soils. Iron was found to be significantly and positively correlated with nitrogen ($r = 0.36$, $p < 0.001$), boron ($r = 0.24$, $p < 0.05$) and nickel ($r = 0.65$, $p < 0.001$). A significant and positive correlation of manganese was found with nickel ($r = 0.40$, $p < 0.001$) and zinc ($r = 0.42$, $p < 0.001$). Nickel in the tea garden soils were also observed to be significantly and positively correlated with zinc ($r = 0.21$, $p < 0.05$). The present study warrants further large-scale investigation to assess the origin and bioavailabiliy of different heavy metals in the geographically different tea garden soils across Bangladesh as well as to relate this to the yield and nutritional quality of the produced tea.

Key words: Heavy metals, tea garden soil, non-tea garden soil

3.1. Introduction

Tea plants prefer growing in hilly regions unlike most of the traditional crops (Mondal, 2009). In Bangladesh, tea cultivation is done mainly in the hilly regions of Moulvibazar, Sylhet and Chattogram. Recently, tea cultivation has also been started in the plain lands of Panchagarh, Thakurgaon, Dinajpur and Lalmonirhat. Soils of these areas have differences in their origin, geology and geomorpholy (FAO-UNDP, 1988). Sana (1989) and Alam (1999) reported the presence of kaolinite, quartz, hematite, goethite, and gibbsite minerals in the tea growing soils of Bangladesh. They also reported that the tea soils had strong phosphate fixing capacity and low cation exchange and nutrient releasing capacities. In subtropical and tropical regions like in Bangladesh, tea soils are generally acidic in nature, which are most suitable for growing tea plant. According to Khan *et al*. (2013), the optimum pH of soil for the growth of tea plants in Bangladesh is between 4.5 and 5.8. However, this low pH in the tea soils can have potentials to release higher concentrations of heavy metals in the soil solution posing a risk of plant accumulation (Fung and Wong, 2002).

Soil plays an important role as a source and sink of heavy metals. The total concentrations of metal in soil indicated the geological background and weathering of soil as well as the sources from human activities (Kersten and Förstner, 1989). Vandecasteele *et al.* (2005) and Zhang *et al.* (2020) reported that the higher concentrations of heavy metals were found in the soils originated from the sedimentary rocks, shales or carbonates. Anda (2012) reported that due to the varied dissemination of the components in the same type of rocks, concentrations of heavy metals in soils could be changed within the similar parent rock layer which could ultimately affect the translocation of the heavy metals from soil to plant. Uses of fertilizers, animal manures, sewage sludge, compost and pesticides may be polluted agricultural soil by the accumulation of heavy metals. Besides, depositions from atmosphere and industrial activities playing main roles in the pollution of environment through release heavy metals (Mahanta and Bhattacharyya, 2011; Huq and Shoaib, 2013; Khan and Kathi, 2014). A large amount of fertilizers and agrochemicals are applied in the soils of tea gardens to increase the production of tea. Fertilizers and agrochemicals are important origins of heavy metals in such soils as they contain heavy metals as contaminants (Nath, 2013; Huq and Shoaib, 2013). Therefore, concentrations of heavy metals in the tea garden soils can increase gradually. However, there is very limited scope to understand the extent and variability of heavy metals in the geographically different tea garden soils of Bangladesh. Moreover, while the mobilization of heavy metals in soils are largely regulated by various properties of soils such as pH, texture, organic matter content as well as other geochemical constituents, it is highly important to understand the geochemical relationships of heavy metals in the different tea garden soils of the country.

In the present study, surface (0 – 23 cm depth, n = 132) and sub-surface (23 – 46 cm depth, n = 60) soils from the geographically different tea gardens of Bangladesh were collected and analyzed for a range of geochemical elements such as nitrogen, boron, chromium, copper, iron, manganese, nickel and zinc. The present study is so far the first of a large-scale tea garden soil geochemical survey in Bangladesh to assess the extent as well as local and regional distribution of the different elements in Bangladeshi tea garden soils. The data were also interpreted to understand the geochemical relationships of the elements in the different tea garden soils.

3.2. Objectives

The objectives of the research reported in this chapter are:

1. to determine the heavy metals and other geochemical elements in the geographically different tea garden soils of Bangladesh;
2. to assess the variability of the elements in the different tea garden soils of Bangladesh; and
3. to understand the geochemical relationships of the geochemical elements in the tea garden soils of Bangladesh.

3.3. Materials and Methods

A detail of the *Materials and Methods* for this chapter was described in chapter two (General Methodology).

3.4 Results and Discussion

3.4.1 Geochemical elements in the tea garden soils of Bangladesh

The concentration of chromium in the surface (0-23 cm) and sub-surface (23-46 cm) of the tea garden soils ranged from 2.00 to 73.45 mg/kg (mean, 23.83 mg/kg) and 2.55 to 88.65 mg/kg (mean, 36.39 mg/kg), respectively (Table 3.1), whereas chromium in the surface and sub-surface of the adjacent non-tea garden soils varied from 5.65 to 63.15 mg/kg (mean, 26.09 mg/kg) and 15.10 to 66.60 mg/kg (mean, 37.44 mg/kg), respectively. According to Kabata-Pendias (2011), the world-soil average concentration of chromium is 60 mg/kg. This indicated that average concentration of chromium in the tea garden soils and adjacent non-tea garden soils were below the world-soil average concentrations of chromium. Domingo and Kyuma (1983), Moslehuddin *et al.* (1999), and Chowdhury *et al.* (2021) reported 89 to 196 mg/kg, 24 to 86 mg/kg and 17.79 to 727 mg/kg chromium, respectively, in the agricultural soils of Bangladesh. Chowdhury *et al.* (2021) also reported the chromium concentration in the non-agricultural soils of Bangladesh ranged from 21.40 to 313 mg/kg with an average concentration of chromium was 66.42 mg/kg. In the present study, the concentrations of chromium found in tea garden soils and the adjacent non-tea garden soils were generally within the ranges reported in the previous survey on agricultural and non-agricultural soils of Bangladesh.

The concentration of copper in the surface and sub-surface of the tea garden soils ranged from 0.86 to 23.30 mg/kg (mean, 9.23 mg/kg) and 1.45 to 22.45 mg/kg (mean, 9.20 mg/kg), respectively (Table 3.1), whereas copper in the surface and sub-surface of the adjacent non-tea garden soils varied from 2.53 to 16.10 mg/kg (mean, 7.92 mg/kg) and 2.40 to 15.95 mg/kg (mean, 8.16 mg/kg), respectively. According to Kabata-Pendias (2011), the world-soil average concentration of copper in soil is 39 mg/kg. This indicated that the average concentration of copper in the tea garden soils and adjacent non-tea garden soils were below the world-soil average concentrations of copper. Domingo and Kyuma (1983), Moslehuddin *et al.* (1999), Jahiruddin *et al.* (2000) and Chowdhury *et al.* (2021) reported 6 to 48 mg/kg, 8.5 to 43.3 mg/kg, 9.40 to 43 mg/kg and 5.46 to 68.56 mg/kg copper, respectively in the agricultural soils of Bangladesh. Chowdhury *et al.* (2021) also reported the copper concentration in the non-agricultural soils of Bangladesh ranged from 6.50 to 64 mg/kg with an average concentration was 27.69 mg/kg. The concentrations of copper in the tea garden soils and the adjacent non-tea garden soils, as observed in the present study, were generally within the ranges reported in the previous survey on agricultural and non-agricultural soils of Bangladesh.

The concentration of iron in the surface and sub-surface of the tea garden soils ranged from 3084 to 47120 mg/kg (mean, 16847 mg/kg) and 2640 to 52540 mg/kg (mean, 24415 mg/kg), respectively (Table 3.1), whereas iron in the surface and sub-surface of the adjacent non-tea garden soils varied from 8080 to 39080 mg/kg (mean, 16742 mg/kg) and 2480 to 44020 mg/kg (mean, 23165 mg/kg), respectively. According to Kabata-Pendias (2011), the world-soil average concentration of iron in soil is 35000 mg/kg. This indicated that the average concentration of iron in the tea garden soils and adjacent non-tea garden soils were below the world-soil average concentrations of iron. Moslehuddin *et al.* (1999) and Chowdhury *et al.* (2021) reported 9200 to 47600 mg/kg and 793 to 50802 mg/kg iron, respectively in the agricultural soils of Bangladesh. Chowdhury *et al.* (2021) also reported the concentration of iron in the non-agricultural soils of Bangladesh ranged from 8072 to 50396 mg/kg with an average concentration was 25384 mg/kg. In the present study, the concentrations of iron found in the tea garden soils and the adjacent non-tea garden soils were generally within the ranges reported in the previous survey on agricultural and non-agricultural soils of Bangladesh.

The concentration of manganese in the surface and sub-surface of the tea garden soils ranged from 21.00 to 1274 mg/kg (mean, 219 mg/kg) and 49.00 to 466 mg/kg (mean, 178 mg/kg), respectively (Table 3.1), whereas manganese in the surface and sub-surface of the adjacent non-tea garden soils varied from 19.00 to 483 mg/kg (mean, 152 mg/kg) and 14.00 to 231 mg/kg (mean, 118 mg/kg), respectively. According to Kabata-Pendias (2011), the world-soil average concentration of manganese in soil is 488 mg/kg. This indicated that average concentration of manganese in the tea garden soils and adjacent non-tea garden soils were below the world-soil average concentrations of manganese. Moslehuddin *et al.* (1999), Jahiruddin *et al.* (2000) and Chowdhury *et al.* (2021) reported 122 to 590 mg/kg, 346 to 618 mg/kg and 43.04 to 1198 mg/kg manganese, respectively in the agricultural soils of Bangladesh.

Table 3.1: Elemental composition of the tea garden soils and adjacent non-tea soils of Bangladesh.

Tea garden soils:

	Soil depth (cm)	Total						pH	OM	OC	Total N	B	Available		Exchangeable		
		Cr	Cu	Fe	Mn	Ni	Zn						P	S	K	Ca	Mg
		(mg/kg)							(%)			(mg/kg)	(mg/kg)		(meq/100 g soil)		
Mean	0-23	23.83	9.23	16847	219	9.31	48.14	4.89	2.21	1.29	0.11	0.48	22.75	15.56	0.25	1.59	1.85
s.e.		1.41	0.38	675	19.7	0.41	5.42	0.05	0.11	0.07	0.01	0.05	2.39	1.23	0.04	0.11	0.15
Min.		2.00	0.86	3084	21	3	2.2	3.4	0.50	0.29	0.03	0.01	0.47	0.12	0.05	0.02	0.12
Median		20.13	8.30	14698	153	8.4	36.45	4.90	1.85	1.08	0.09	0.25	10.52	12.05	0.18	1.17	1.44
Max.		73.45	23.3	47120	1274	23.15	465	6.20	6.0	3.49	0.28	2.30	114	69.13	3.85	5.49	7.13
Mean	23-46	36.39	9.20	24415	178	11.52	34.18	5.16	2.49	1.45	0.13	0.79	13.43	27.73	0.16	0.71	0.56
s.e.		2.39	0.63	1412	12.5	0.75	1.93	0.08	0.16	0.09	0.01	0.08	2.66	1.85	0.01	0.08	0.1
Min.		2.55	1.45	2640	49	4.15	1.73	3.80	0.5	0.29	0.03	0.02	0.65	1.89	0.03	0.1	0.13
Median		34.20	9.18	23370	154	10.20	34.26	5.25	2.7	1.57	0.14	0.75	7.00	27.0	0.14	0.62	0.33
Max.		88.65	22.45	52540	466	23.70	71.05	6.20	4.45	2.59	0.23	1.92	117	61.0	0.54	3.64	3.60

s.e. = Standard error of the mean; OM = Organic matter; OC = Organic carbon

Table 3.1: Continued.

Non-tea garden soils:

	Soil depth (cm)	Total						pH				B	Available		Exchangeable		
		Cr	Cu	Fe	Mn	Ni	Zn		OM	OC	Total N		P	S	K	Ca	Mg
		(mg/kg)							(%)			(mg/kg)	(mg/kg)		(meq/100gm soil)		
Mean	0-23	26.09	7.92	16742	152	8.92	36.94	5.08	2.37	1.38	0.12	0.53	12.97	15.85	0.12	0.95	1.39
s.e.		3.57	0.94	1694	24.2	0.93	4.77	0.11	0.22	0.13	0.01	0.14	7.07	2.35	0.01	0.16	0.3
Min.		5.65	2.53	8080	19	2.7	11.88	4.2	0.9	0.52	0.05	0.03	0.37	0.67	0.06	0.07	0.15
Median		23.08	6.85	14770	120	8.1	30.55	4.95	2.3	1.34	0.12	0.27	3.52	17.5	0.1	0.75	0.69
Max.		63.15	16.1	39080	483	16	91.3	6	4.68	2.72	0.23	2.12	142	33.66	0.25	2.24	3.79
Mean	23-46	37.44	8.16	23165	118	10.36	34.99	5.41	1.95	1.13	0.1	0.64	14.89	19.72	0.11	0.37	0.32
s.e.		5.04	1.06	3344	19.8	1.32	4.25	0.15	0.25	0.14	0.01	0.12	9.87	2.45	0.02	0.07	0.04
Min.		15.10	2.4	2480	14	2.5	11.72	4.6	0.5	0.29	0.03	0.21	0.4	8.15	0.05	0.1	0.13
Median		37.35	7.7	19860	116	10.5	31.9	5.5	2.18	1.26	0.11	0.61	4.26	18.54	0.08	0.33	0.33
Max.		66.60	15.95	44020	231	18.25	61.85	6.2	3.03	1.76	0.15	1.52	122	35	0.25	0.84	0.53

s.e. = Standard error of the mean; OM = Organic matter; OC = Organic carbon

Chowdhury *et al.* (2021) also reported the manganese concentration in the non-agricultural soils of Bangladesh ranged from 92.22 to 778 mg/kg with an average concentration was 437 mg/kg. In the present study, the concentrations of manganese found in the tea garden soils and the adjacent non-tea garden soils were generally within the ranges reported in the previous survey on agricultural and non-agricultural soils of Bangladesh.

The concentration of nickel in the surface and sub-surface of the tea garden soils ranged from 3.00 to 23.15 mg/kg (mean, 9.31 mg/kg) and 4.15 to 23.70 mg/kg (mean, 11.52 mg/kg), respectively (Table 3.1), whereas nickel in the surface and sub-surface of the adjacent non-tea garden soils varied from 2.70 to 16.00 mg/kg (mean, 8.92 mg/kg) and 2.50 to 18.25 mg/kg (mean, 10.36 mg/kg), respectively. According to Kabata-Pendias (2011), the world-soil average concentration of nickel in soil is 29 mg/kg. This indicated that average concentration of nickel in the tea garden soils and adjacent non-tea garden soils were below the world-soil average concentrations of nickel. Domingo and Kyuma (1983), Moslehuddin *et al.* (1999) and Chowdhury *et al.* (2021) reported up to 63 mg/kg, 8 to 92 mg/kg and 7.43 to 92.57 mg/kg nickel, respectively in the agricultural soils of Bangladesh. Chowdhury *et al.* (2021) also reported the nickel concentration in the non-agricultural soils of Bangladesh ranged from 8.97 to 86.48 mg/kg with an average concentration was 34.92 mg/kg. In the present study, the concentrations of nickel found in the tea garden soils and the adjacent non-tea garden soils were generally within the ranges reported in the previous survey on agricultural and non-agricultural soils of Bangladesh.

The concentration of zinc in the surface and sub-surface of the tea garden soils ranged from 2.20 to 465 mg/kg (mean, 48.14 mg/kg) and 1.73 to 71.05 mg/kg (mean, 34.18 mg/kg), respectively (Table 3.1), whereas zinc concentration in the surface and sub-surface of the adjacent non-tea garden soils varied from 11.88 to 91.30 mg/kg (mean, 36.94 mg/kg) and 11.72 to 61.85 mg/kg (mean, 34.99 mg/kg), respectively. According to Kabata-Pendias (2011), the world-soil average concentration of zinc in soil is 70 mg/kg. This indicated that average concentration of zinc in the tea garden soils and adjacent non-tea garden soils were below the world-soil average concentrations of zinc. Domingo and Kyuma (1983), Moslehuddin *et al.* (1999), Jahiruddin *et al.* (2000) and Chowdhury *et al.* (2021) reported 10 to 110 mg/kg, 18.9 to 92.3 mg/kg, 45.2 to 111 mg/kg and 11.13 to 181 mg/kg zinc, respectively in the agricultural soils of Bangladesh. Chowdhury *et al.* (2021) also reported the zinc concentration in the non-agricultural soils of Bangladesh ranged from 9.63 to 127 mg/kg with an average concentration was 64.10 mg/kg. In the present study, the concentrations of zinc found in the surface soil of the tea garden soils were higher than the ranges reported in the previous survey while the zinc concentration in the sub-surface of tea garden soils as well as surface and sub-surface of the adjacent non-tea garden soils were generally within the ranges reported in the previous survey on agricultural and non-agricultural soils of Bangladesh.

The pH of the surface and sub-surface of the tea garden soils ranged from 3.40 to 6.20 (mean, 4.89) and 3.80 to 6.20 (mean, 5.16), respectively (Table 3.1), whereas pH in the surface and sub-surface of the adjacent non-tea garden soils varied from 4.20 to 6.00 (mean, 5.08) and 4.60 to 6.20 (mean, 5.41), respectively. The soil pH 4.5 to 5.8 is considered critical limit for growing tea (Khan *et al*., 2013). This indicated that average pH in the tea garden soils and adjacent non-tea garden soils were within the critical limit.

The organic carbon content in the surface and sub-surface of the tea garden soils ranged from 0.29 to 3.49% (mean, 1.29%) and 0.29 to 2.59% (mean, 1.45%), respectively, whereas the organic carbon content in the surface and sub-surface of the adjacent non-tea garden soils varied from 0.52 to 2.72% (mean, 1.38%) and 0.29 to 1.76% (mean, 1.13%), respectively. The critical value of organic carbon in tea garden soil is 1% (Khan *et al*., 2013). This indicated that average organic carbon content in the tea garden soils and adjacent non-tea garden soils were above the critical limit.

The total nitrogen content in the surface and sub-surface of the tea garden soils ranged from 0.03 to 0.28% (mean, 0.11%) and 0.03 to 0.23% (mean, 0.13%), respectively, whereas the total nitrogen content in the surface and sub-surface soil of the adjacent non-tea garden soils varied from 0.05 to 0.23% (mean, 0.12%) and 0.03 to 0.15% (mean, 0.10%), respectively. The critical value of total nitrogen in tea garden soil is 0.1% (Khan *et al*., 2013). This indicated that average total nitrogen content in the tea garden soils and adjacent non-tea garden soils were above the critical limit.

The boron content in the surface and sub-surface soil of the tea garden soils ranged from 0.01 to 2.30 mg/kg (mean, 0.48 mg/kg) and 0.02 to 1.92 mg/kg (mean, 0.79 mg/kg), respectively, whereas the boron content in the surface and sub-surface soil of the adjacent non-tea garden soils varied from 0.03 to 2.12 mg/kg (mean, 0.53 mg/kg) and 0.21 to 1.52 mg/kg (mean, 0.64 mg/kg), respectively. The critical value of boron is 0.5 mg/kg (Ma. *et al.,* 2003).

3.4.2 Variability of geochemical elements in the different tea garden soils of Bangladesh

The distribution of heavy metals and other elements in the tea garden soils are presented in the tables 3.2 and 3.3. The highest concentration of chromium (73.45 mg/kg) in the surface soil was found in the Karotoa tea garden while the lowest concentration of chromium (2.00 mg/kg) was found in the Jagcherra tea garden. The highest mean concentration of chromium (51.42 mg/kg) was found in the surface soils of Karotoa tea garden and the lowest mean concentration of chromium (9.56 mg/kg) was found in the BTRI tea garden (Table 3.2). The highest concentration of chromium (88.65 mg/kg) of the sub-surface soil was found in the Karotoa tea garden while the lowest concentration of chromium (2.55 mg/kg) was found in the Neptune tea garden. The highest mean concentration of chromium (56.79 mg/kg) was found in the sub-surface soils of Karotoa tea garden and the lowest mean concentration of chromium (25.59 mg/kg) was found in the Neptune tea garden (Table 3.2). Significant variations ($^{ANOVA}F$ = 11.92, $p < 0.001$) were observed in the concentrations of chromium in surface soils among the different tea gardens (Figure 3.1). The concentrations of chromium were also varied significantly ($^{ANOVA}F = 6.85$, $p < 0.001$) in sub-surface soils (Figure A1). The concentration of chromium in the surface soil of Oodaleah and Karotoa tea garden had the wider variability (Figure 3.1). The mean concentration of chromium in the surface soils of Karotoa and Jagcherra tea garden was found significantly different from other tea gardens. There was no significant difference found in the mean concentration of chromium in the surface soils of Kazi and Kazi and Oodaleah tea garden (Figure 3.1). Huq and Alam (2005) reported the typical content of chromium in soil range from 5 to 1500 mg/kg (mean, 65 mg/kg). Kabata-Pendias (2011) reported that the concentration of chromium in agricultural soils of Japan varied from 56 to 70 mg/kg. The concentration of chromium in the surface and sub-surface soil of the tea gardens of Assam, India was found 68.73 to 102.02 mg/kg and 56.0 to 94.07 mg/kg, respectively (Nath, 2013). The concentrations of chromium in the tea producing regions of China were found to be in the range of 20.3-133 mg/kg (Huang *et al*., 2016), while in our present study the concentration of chromium in the surface soil of the tea garden was found up to 73.45 mg/kg.

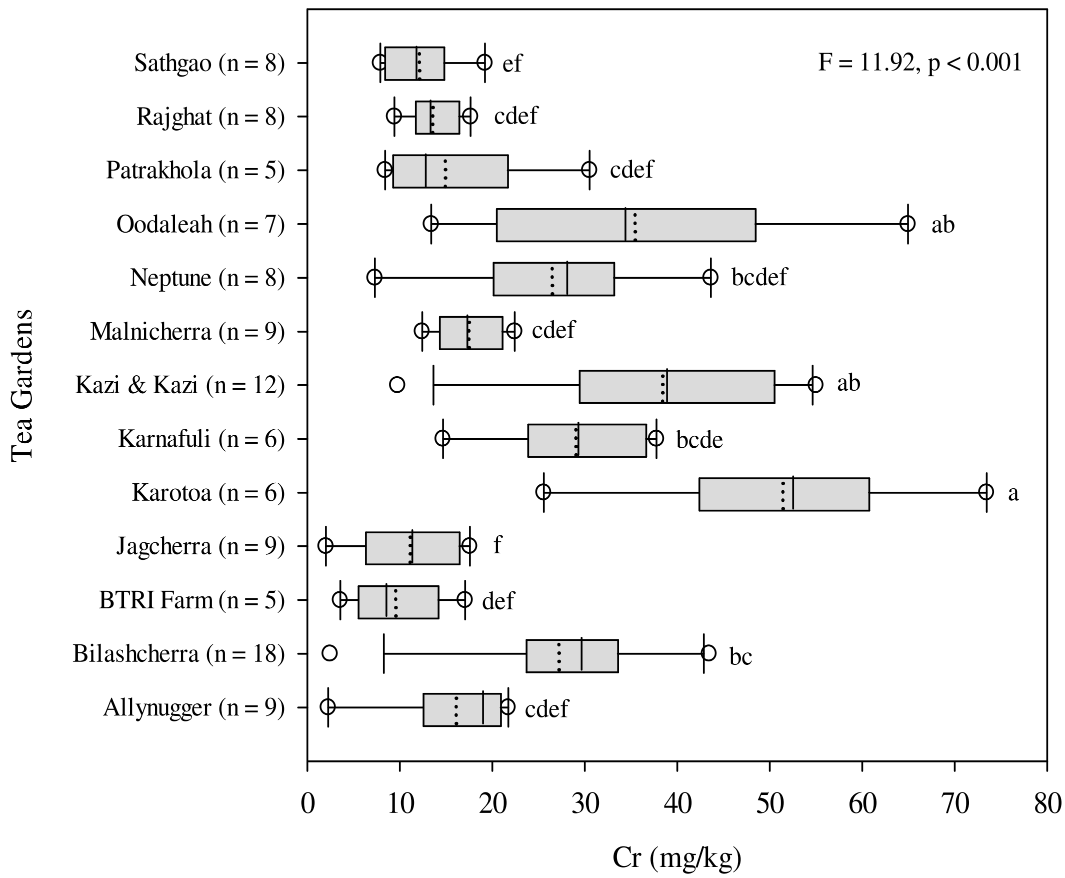


Figure 3.1: Box and whisker plots showing variability of chromium concentration (mg/kg) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, c, d, e, f designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The highest concentration of copper (23.30 mg/kg) in the surface soil was found in the Karotoa tea garden while the lowest concentration of copper (0.86 mg/kg) was found in the Allynugger tea garden. The highest mean concentration of copper (15.88 mg/kg) was found in the surface soils of Karotoa tea garden and the lowest mean concentration of copper (6.01 mg/kg) was found in the surface soils of Patrakhola tea garden (Table 3.2). The highest concentration of copper (22.45 mg/kg) in the sub-surface soil was found in the Karotoa tea garden while the lowest concentration of copper (1.45 mg/kg) was found in the Bilashcherra tea garden. The highest mean concentration of copper (16.28 mg/kg) was found in the sub-surface soils of Karotoa tea garden and the lowest mean concentration of copper (5.87 mg/kg) in the sub-surface soil was found in the Bilashcherra tea garden (Table 3.2). Significant variations ($^{ANOVA}F = 3.99$, $p < 0.001$) were observed in the concentration of copper in surface soils among the different tea gardens (Figure 3.2). The concentrations of copper were also varied significantly ($^{ANOVA}F = 14.68$, $p < 0.001$) in sub-surface soils (Figure A2). The surface soils of Allynugger, Rajghat, BTRI, Karotoa and Oodaleah tea gardens had wider variability in their copper concentrations (Figure 3.2).

The mean concentration of copper in the surface soils of Karotoa tea garden was found significantly different from other tea gardens. There was no significant difference was found in the mean concentration of copper in the surface soils of Rajghat, Kazi and Kazi and BTRI tea garden (Figure 3.2). Huq and Alam (2005) reported the typical concentration of copper in soil range from 2.5 to 60 mg/kg (mean, 26 mg/kg). Kabata-Pendias (2011) reported that, the general values for the average total concentration of copper in soils of different groups all over the world range between 14 and 109 mg/kg. The concentration of copper in the surface and sub-surface soil of the tea estates of Assam, India was found 16.73 to 36.33 mg/kg and 15.35 to 31.73 mg/kg, respectively (Nath, 2013). The concentration ranges of copper were found 4.3-53.8 mg/kg in Zhejiang Province of China (Zhou *et al*., 2009). Khushi *et al*. (2017) reported that the concentration of copper in the soils of tea growing areas of Bangladesh was ranged from 6.00 to 16.50 mg/kg, while in our present study the concentration of copper in the surface soil of the tea garden was found up to 23.30 mg/kg.

The highest concentration of iron (47,120 mg/kg, 4.7%) in the surface soil was found in the Karotoa tea garden while the lowest concentration of iron (3,084 mg/kg, 0.31%) was found in the Kazi and Kazi tea garden. The highest mean concentration of iron (31,840 mg/kg, 3.18%) was found in the surface soils of Karotoa tea garden and the lowest mean concentration of iron (11,363 mg/kg, 1.14%) was found in the surface soils of BTRI tea garden (Table 3.2).

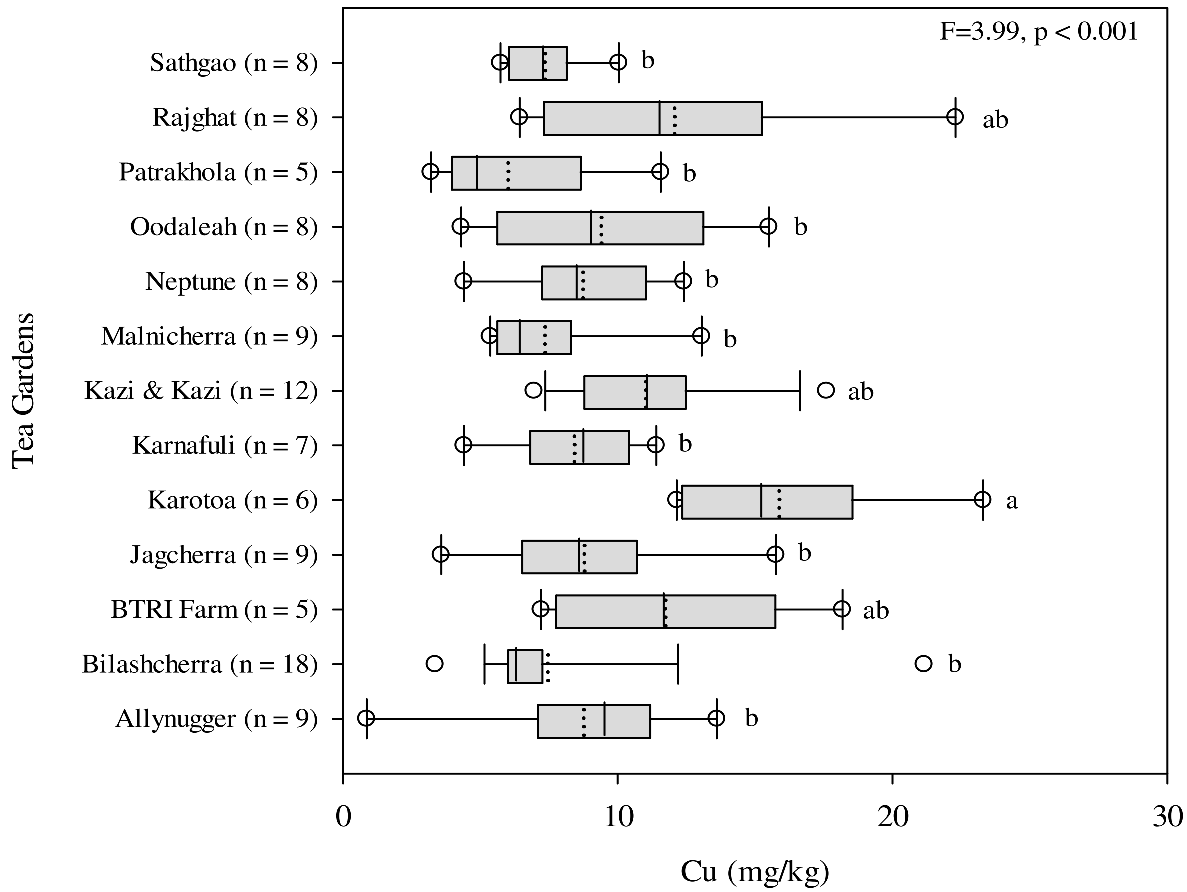


Figure 3.2: Box and whisker plots showing variability of copper concentration (mg/kg) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The highest concentration of iron (52,540 mg/kg, 5.25%) in the sub-surface soil was found in the Oodaleah tea garden while the lowest concentration of iron (2,640 mg/kg, 0.26%) was found in the Bilashcherra tea garden. The highest mean concentration of iron (35,320 mg/kg, 3.53%) was found in the sub-surface soils of Karotoa tea garden and the lowest mean concentration of iron (19,120 mg/kg, 1.91%) was found in the sub-surface soils of Karnafuli tea garden (Table 3.2). Significant variations ($^{ANOVA}F = 7.11$, $p < 0.001$) were observed in the concentration of iron in surface soils among the different tea gardens (Figure 3.3).

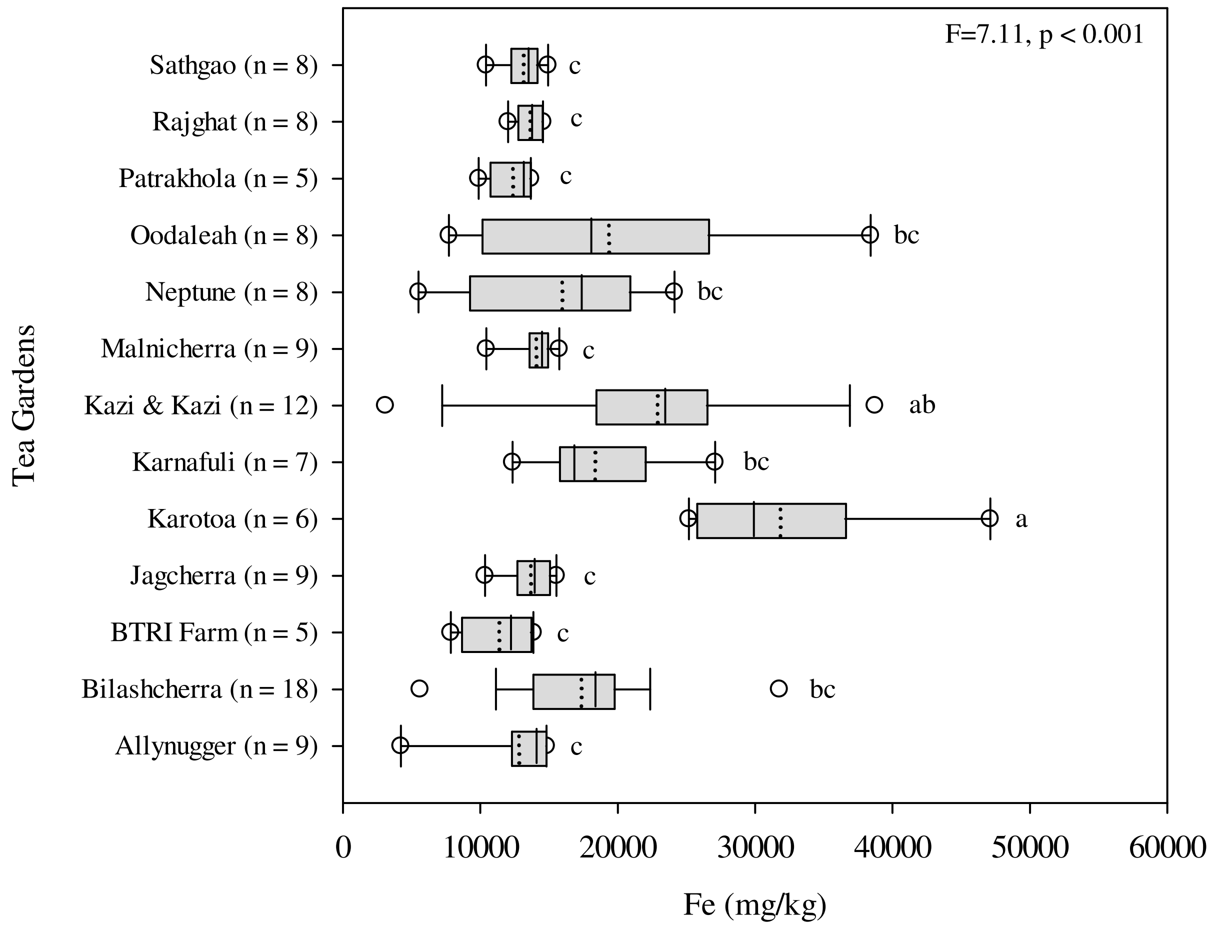


Figure 3.3: Box and whisker plots showing variability of iron concentration (mg/kg) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, c designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

Table 3.2: Concentration of heavy metals in the tea garden soils of Bangladesh.

Tea Estate	Depth (cm)		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Allynugger	0-23 (n=9)	*Min – Max*	2.25 – 21.70	0.85 – 13.61	4206 – 14800	38.40 – 220.30	6.05 – 10.85	8.00 – 66.00
		Mean±s.e.	16.07±2.10	8.76±1.23	12795±1134	114.00±16.80	8.53±0.60	35.03±6.63
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Bilashcherra	0-23 (n=18)	*Min – Max*	2.45 – 43.45	3.35 – 21.15	5600 – 31760	67.00 – 592.00	3.10 – 23.15	14.90 – 38.89
		Mean±s.e.	27.17±2.58	7.46±0.89	17360±1268	268.6±27.60	8.17±1.05	25.64±1.56
	23-46 (n=18)	*Min – Max*	13.30 – 46.75	1.45 – 12.65	2640 – 34400	79.00 – 378.00	4.15 – 21.25	1.73 – 37.69
		Mean±s.e.	27.24±2.14	5.87±0.62	19771±1753	214.20±19.60	8.70±0.93	23.28±2.18
BTRI Farm	0-23 (n=5)	*Min – Max*	3.55 – 17.05	7.21 – 18.18	7845 – 13847	47.90 – 125.60	3.00 – 11.45	35.60 – 69.80
		Mean±s.e.	9.56±2.24	11.72±1.95	11363.±1175	87.70±16.40	5.80±1.46	47.76±6.20
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Jagcherra	0-23 (n=9)	*Min – Max*	2.00 – 17.55	3.57 – 15.76	10340 – 15539	51.50 – 424.20	3.10 – 11.55	10.10 – 465.20
		Mean±s.e.	11.10±1.83	8.78±1.14	13682±554	269.00±41.40	6.68±0.90	102±47.20
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Karnafuli	0-23 (n=7)	*Min – Max*	14.65 – 37.75	4.40 – 11.40	12340 – 27080	29.00 – 140.00	5.35 – 8.80	20.04 – 37.73
		Mean±s.e.	29.02±3.39	8.42±0.88	18354±1833	75.10±12.70	7.10±0.54	30.01±2.71
	23-46 (n=3)	*Min – Max*	28.95 – 42.45	5.70 – 9.95	15020 – 23040	66.00 – 120.00	5.20 – 9.35	20.92 – 27.11
		Mean±s.e.	36.57 ± 3.99	7.77±1.23	19120±2317	87.00±16.70	7.92±1.36	24.48±1.85
Karotoa	0-23 (n=6)	*Min – Max*	25.55 – 73.45	12.15 – 23.30	25180 – 47120	92.00 – 234.00	13.20 – 19.35	16.23 – 51.77
		Mean±s.e.	51.42±6.35	15.88±1.68	31840±3285	146.50±22.90	15.86±0.88	41.37±5.25
	23-46 (n=6)	*Min – Max*	31.50 – 88.65	13.10 – 22.45	27420 – 50080	116.00 – 269.00	15.30 – 20.35	29.03 – 71.05
		Mean±s.e.	56.79±8.70	16.28±1.46	35320±3514	163.80±23.80	17.56±0.87	46.52±5.56
Kazi & Kazi	0-23 (n=12)	*Min – Max*	9.75 – 55.00	6.95 – 17.60	3084 – 38720	101.00 – 281.00	8.20 – 15.15	34.85 – 50.25
		Mean±s.e.	38.41±3.88	11.02±0.84	22904±2510	160.80±15.30	11.65±0.62	42.42±1.68
	23-46 (n=12)	*Min – Max*	34.20 – 62.35	8.95 – 14.40	19060 – 37400	116.00 – 191.00	10.20 – 16.55	22.10 – 50.09
		Mean±s.e.	46.07±2.68	11.00±0.39	27870±1998	149.50±6.45	12.92±0.64	41.59±2.17

Table 3.2: Continued.

Tea Estate	Depth (cm)		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Malnicherra	0-23 (n=9)	*Min – Max*	12.40 – 22.40	5.36 – 13.05	10424 – 15741	81.00 – 1274	7.90 – 14.85	68.40 – 145.10
		Mean±s.e.	17.47±1.17	7.34±0.81	14067±522	555±130	11.44±0.83	87.51±8.25
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Neptune	0-23 (n=8)	*Min – Max*	7.30 – 43.65	4.40 – 12.40	5500 – 24120	53.00 – 268.00	3.95 – 13.50	20.47 – 48.34
		Mean±s.e.	26.47±3.81	8.74±0.90	15955±2318	123.80±24.50	6.84±1.10	29.18±3.34
	23-46 (n=4)	*Min – Max*	2.55 – 41.85	5.55 – 9.15	13800 – 25700	49.00 – 134.00	5.20 – 8.90	29.44 – 39.72
		Mean±s.e.	25.59±8.27	7.36±0.86	19745±2616	81.30±19.90	6.75±0.88	34.51±2.50
Oodaleah	0-23 (n=8)	*Min – Max*	13.40 – 64.95	4.30 – 15.50	7720 – 38420	21.00 – 955	4.15 – 21.60	18.00 – 394.60
		Mean±s.e.	35.41±6.88	9.41±1.46	19360±3678	285±104	13.42±2.34	77.70±45.50
	23-46 (n=5)	*Min – Max*	14.50 – 60.50	5.05 – 17.70	11400 – 52540	121.00 – 466.00	6.20 – 23.70	34.05 – 63.61
		Mean±s.e.	32.02±8.13	10.70±2.29	26668±7423	263.60±60.10	16.46±3.61	46.38±5.19
Patrakhola	0-23 (n=5)	*Min – Max*	8.40 – 30.50	3.19 – 11.56	9881 – 13683	85.40 – 209.60	5.25 – 7.80	6.70 – 53.70
		Mean±s.e.	14.92±3.98	6.01±1.45	12361±733	161.30±22.20	6.59±0.48	28.62±8.87
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Rajghat	0-23 (n=8)	*Min – Max*	9.40 – 17.65	6.43 – 22.30	12017 – 14544	32.00 – 957	4.55 – 18.20	40.20 – 92.20
		Mean±s.e.	13.55±0.98	12.06±1.86	13616±334	323±112	10.78±1.91	61.80±7.26
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Sathgao	0-23 (n=8)	*Min – Max*	7.90 – 19.20	5.73 – 10.04	10417 – 14924	47.90 – 175.80	3.25 – 13.05	2.20 – 51.00
		Mean±s.e.	12.12±1.45	7.34±0.49	13131±501	99.40±17.80	7.61±1.18	26.50±6.40
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-

The concentrations of iron were also varied significantly ($^{ANOVA}F = 3.97$, $p < 0.01$) in sub-surface soils (Figure A3). The concentration of iron in the surface soil of Oodaleah, Kazi and Kazi, Karotoa and Neptune tea gardens had the wider variability (Figure 3.3). The mean concentration of iron in the surface soils of Karotoa tea garden was found significantly different from other tea gardens. There was no significant difference was found in the mean concentration of iron in the surface soils of Oodaleah, Neptune, Karnafuli and Bilashcherra tea garden (Figure 3.3). Huq and Alam (2005) reported that the typical concentration of iron in soil ranged from 0.01 to 21% (mean, 3.2%). Kabata-Pendias (2011) reported that the abundance of iron in soils average 3.5%. The concentration of iron in the surface and sub-surface soil of the tea estates of Assam, India was found 4.933 to 10.766 mg/kg and 4.405 to 9.962 mg/kg, respectively (Nath, 2013). The concentration of iron in soil of the tea garden of Palampur, India was ranged from 5,494 to 5,627 mg/kg (Chand *et al*., 2011). Khushi *et al*. (2017) reported that the iron concentration in soils of the tea growing areas of Bangladesh was ranged from 0.54 to 2.67%, while in our present study the concentration of iron in the surface soil of the tea garden was found up to 4.7%.

The highest concentration of manganese (1274 mg/kg) in the surface soil was found in the Malnicherra tea garden while the lowest concentration of manganese (21.00 mg/kg) in the surface soil was found in the Oodaleah tea garden. The highest mean concentration of manganese (555 mg/kg) was found in the surface soils of Malnicherra tea garden and the lowest mean concentration of manganese (75.10 mg/kg) was found in the surface soils of Karnafuli tea garden (Table 3.2). The highest concentration of manganese (466 mg/kg) in the sub-surface soil was found in the Oodaleah tea garden while the lowest concentration of manganese (49.00 mg/kg) in the sub-surface soil was found in the Neptune tea garden. The highest mean concentration of manganese (263.60 mg/kg) was found in the sub-surface soils of Oodaleah tea garden and the lowest mean concentration of manganese (81.30 mg/kg) was found in the sub-surface soils of Neptune tea garden (Table 3.2). Significant variations ($^{ANOVA}F = 4.94$, $p < 0.001$) were observed in the concentration of manganese in surface soils among the different tea gardens (Figure 3.4). The concentrations of manganese were also varied significantly ($^{ANOVA}F = 5.14$, $p < 0.001$) in sub-surface soils (Figure A4). The concentration of manganese in the surface soil of Malnicherra, Rajghat and Oodaleah tea gardens had the wider variability (Figure 3.4).

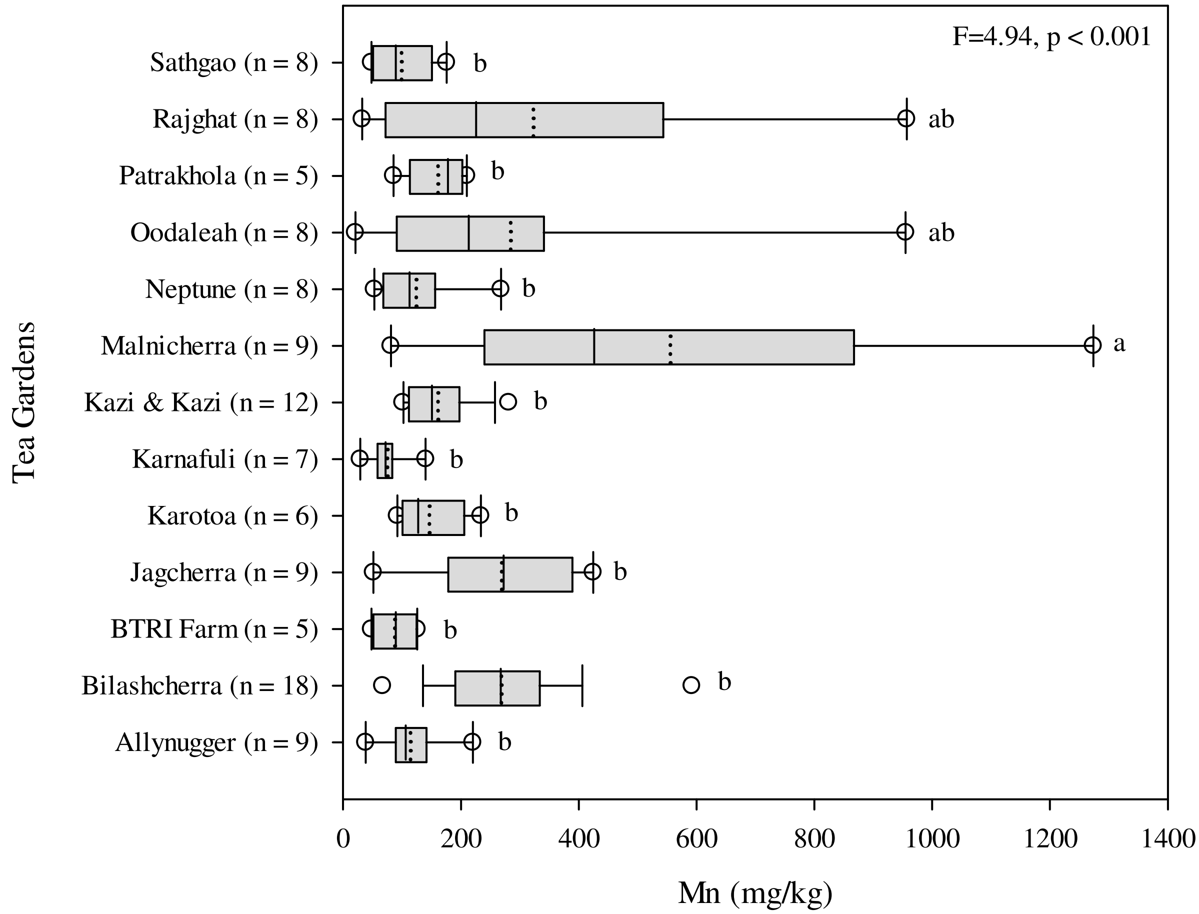


Figure 3.4: Box and whisker plots showing variability of manganese concentration (mg/kg) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10th and 90th percentiles (whiskers) and the 5th and 95th percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The mean concentration of manganese in the surface soils of Malnicherra tea garden was found significantly different from other tea gardens. There was no significant difference was found in the mean concentration of manganese in the surface soils of Rajghat and Oodaleah tea garden (Figure 3.4). Huq and Alam (2005) reported that the typical concentration of manganese in soil ranged from 7 to 8423 mg/kg (mean, 437 mg/kg). The manganese concentrations of worldwide soils varied from 411 to 550 mg/kg (Kabata-Pendias, 2011). The concentration of manganese in the surface and sub-surface soils of the tea gardens of Assam, India was found 118.53 to 420.53 mg/kg and 103.73 to 390.33 mg/kg, respectively (Nath, 2013). The concentration range of manganese in soils of the tea gardens of Palampur, India was ranged from 245 to 435 mg/kg (Chand *et al*., 2011), while in our present study the concentration of manganese in the surface soil of the tea garden was found up to 1274 mg/kg.

The highest concentration of nickel (23.15 mg/kg) in the surface soil was found in the Bilashcherra tea garden while the lowest concentration of nickel (3.00 mg/kg) in the surface soil was found in the BTRI tea garden. The highest mean concentration of nickel (15.86 mg/kg) was found in the surface soils of Karotoa tea garden and the lowest mean concentration of nickel (5.80 mg/kg) was found in the surface soils of BTRI tea garden (Table 3.2). The highest concentration of nickel (23.70 mg/kg) in the sub-surface soil was found in the Oodaleah tea garden while the lowest concentration of nickel (4.15 mg/kg) in the sub-surface soil was found in the Bilashcherra tea garden. The highest mean concentration of nickel (17.56 mg/kg) was found in the sub-surface soils of Karotoa tea garden and the lowest mean concentration of nickel (6.75 mg/kg) was found in the sub-surface soils of Neptune tea garden (Table 3.2). Significant variations ($^{ANOVA}F = 5.32$, $p < 0.001$) were observed in the concentration of nickel in surface soils among the different tea gardens (Figure 3.5).

The concentrations of nickel were also varied significantly ($^{ANOVA}F = 8.73$, $p < 0.001$) in sub-surface soils (Figure A5). The concentration of nickel in the surface soil of Oodaleah and Rajghat tea gardens had the wider variability (Figure 3.5). The mean concentration of nickel in the surface soils of Karotoa tea garden was found significantly different from other tea gardens. There was no significant difference was found in the mean concentration of nickel in the surface soils of Rajghat, Malnicherra and Kazi and Kazi tea garden. The mean concentration of nickel in the surface soils of Sathgao, Patrakhola and Allynugger was also found insignificant differences (Figure 3.5). Zhou *et al*. (2009) reported that the concentration of nickel in the soils of tea gardens in Zhejiang Province of China ranged from 4.2 to 56.9 mg/kg. The concentration of nickel in soil of the Tieguanyin tea garden of southeastern China was ranged from 4.280 to 18.30 mg/kg (Sun *et al*., 2017). Kabata-Pendias (2011) reported that the mean concentrations of nickel in soil throughout the world varied within the range from 13 to 37 mg/kg.

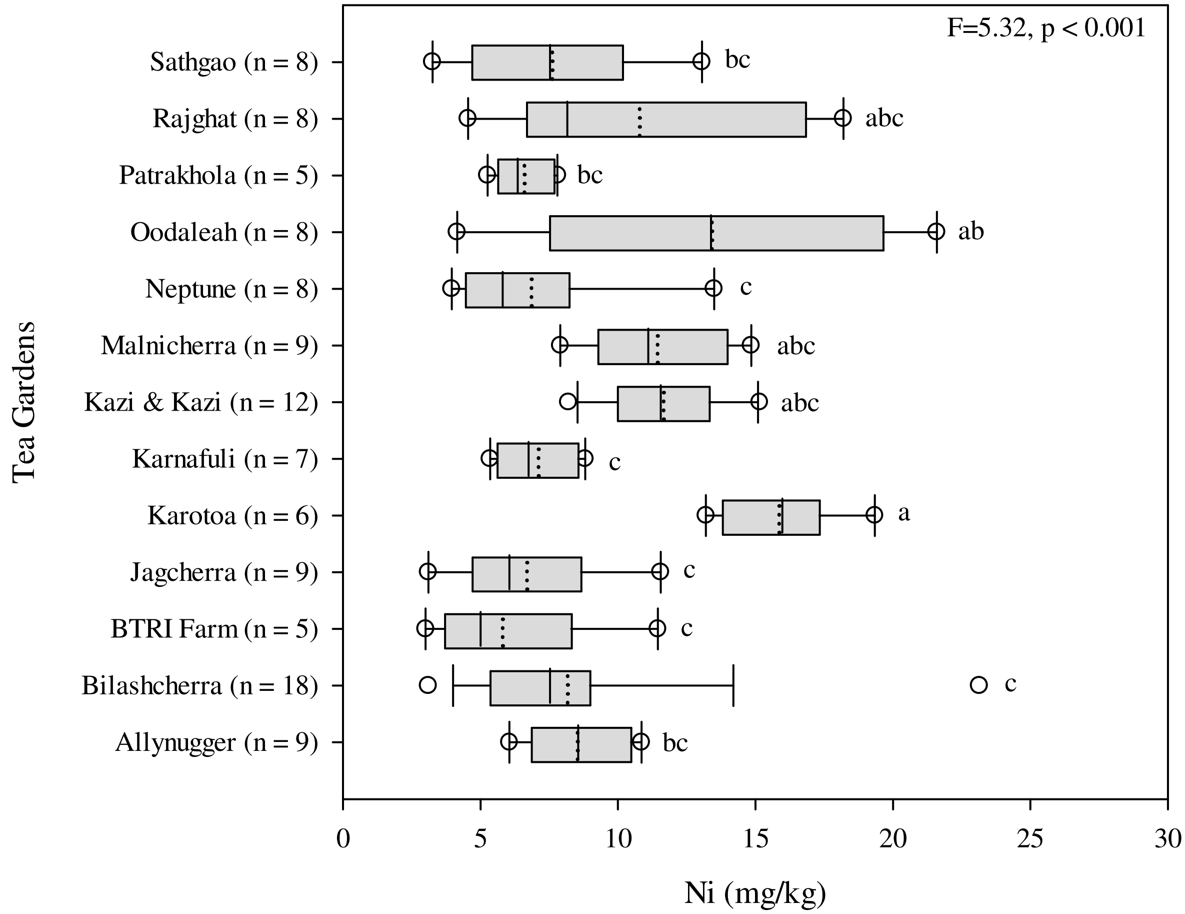


Figure 3.5: Box and whisker plots showing variability of nickel concentration (mg/kg) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, c designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The maximum permissible level of nickel (50 mg/kg) in agricultural soils was established in 1997 (Kabata-Pendias, 2011). The nickel concentration in the surface and sub-surface soil of the tea gardens of Assam, India was found 34.40 to 65.37 mg/kg and 30.67 to 60.00 mg/kg, respectively (Nath, 2013). Khushi *et al*. (2017) reported that the nickel concentration in soils of tea growing areas of Bangladesh was ranged from 3.80 to 15.87 mg/kg, while in our present study the concentration of nickel in the surface soil of the tea garden was found up to 23.15 mg/kg.

The highest concentration of zinc (465 mg/kg) in the surface soil was found in the Jagcherra tea garden while the lowest concentration of zinc (2.20 mg/kg) was found in the surface soils of Sathgao tea garden. The highest mean concentration of zinc (102 mg/kg) was found in the surface soils of Jagcherra tea garden and the lowest mean concentration of zinc (25.64 mg/kg) was found in the surface soils of Bilashcherra tea garden (Table 3.2). The highest concentration of zinc (71.05 mg/kg) in the sub-surface soil was found in the Karotoa tea garden while the lowest concentration of zinc (1.73 mg/kg) was found in the Bilashcherra tea garden. The highest mean concentration of zinc (46.52 mg/kg) was found in the sub-surface soils of Karotoa tea garden and the lowest mean concentration of zinc (23.28 mg/kg) was found in the sub-surface soils of Bilashcherra tea garden (Table 3.2). Significant variations ($^{ANOVA}F = 1.97$, $p < 0.05$) were observed in the concentrations of zinc in surface soils among the different tea gardens (Figure 3.6).

The concentrations of zinc were also varied significantly ($^{ANOVA}F = 10.94$, $p < 0.001$) in sub-surface soils (Figure A6). The concentration of zinc in the surface soil of Jagcherra tea garden had the wider variability. The mean concentration of zinc in the surface soils of Jagcherra and Bilashcherra tea garden was found significantly different from other tea gardens (Figure 3.6). Huq and Alam (2005) reported that the typical concentration of zinc in soil was ranged from 3 to 762 mg/kg (mean, 64 mg/kg). The average total concentrations of zinc in soils of all over the world were ranged between 60 and 89 mg/kg (Kabata-Pendias, 2011). Zhou *et al*. (2009) reported that the concentrations of zinc in tea garden soils in Zhejiang Province of China were varied from 28 to 516 mg/kg. The concentration of zinc in the surface and sub-surface soil of the tea gardens of Assam, India was ranged from 21.43 to 65.20 mg/kg and 21.07 to 56.47 mg/kg, respectively (Nath, 2013). Chand *et al*. (2011) reported that the concentration of zinc in soil of the tea garden of Palampur, India was varied from 30 to 111 mg/kg. The concentration of zinc in the soils of tea growing areas of Bangladesh was ranged from 2.92 to 41.00 mg/kg (Khushi *et al.,* 2017), while in our present study the concentration of zinc in the surface soil of the tea garden was found up to 465 mg/kg.

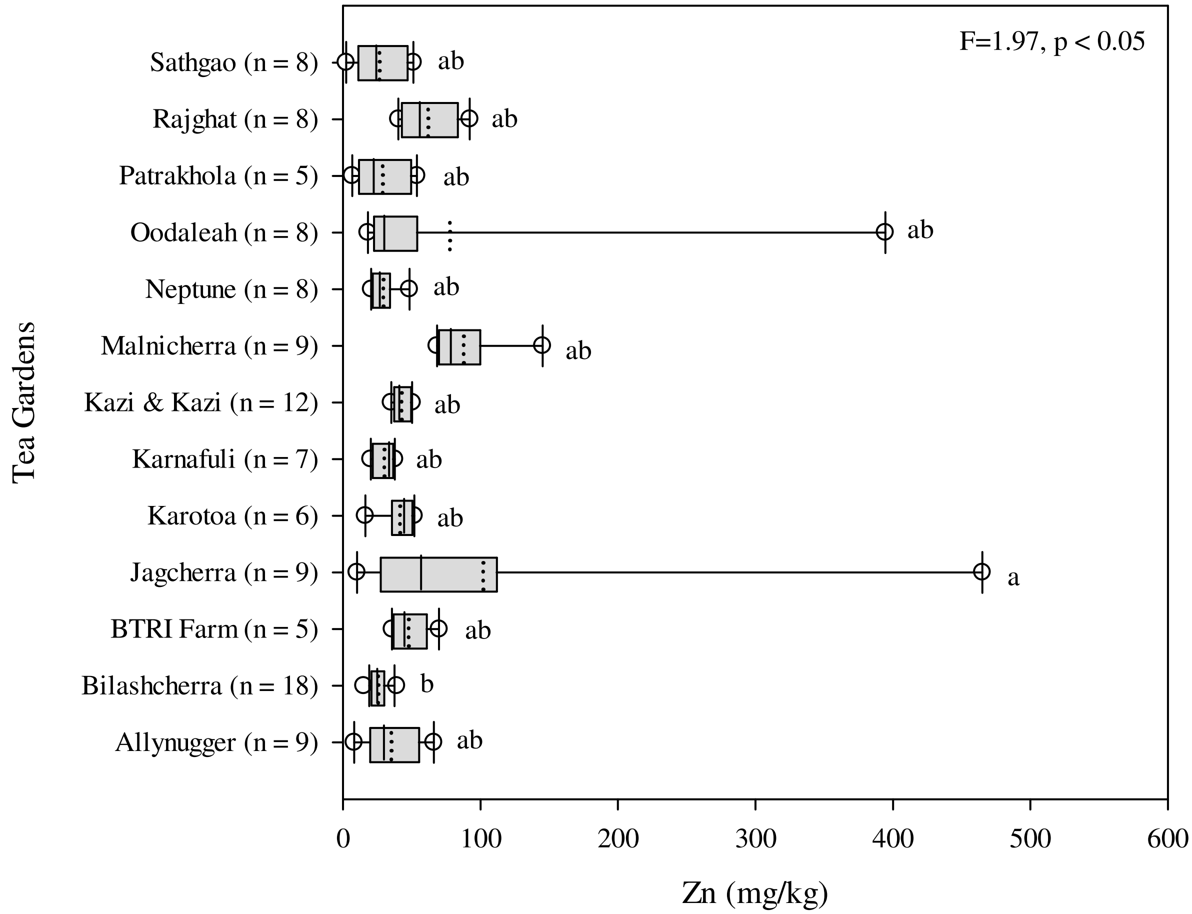


Figure 3.6: Box and whisker plots showing variability of zinc concentration (mg/kg) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The highest pH (6.20) of the surface soil was found in the Bilashcherra tea garden while the lowest pH (3.40) was found in the Neptune tea garden. The highest mean pH (5.57) was found in the surface soil of Kazi & Kazi tea garden and the lowest mean pH (4.44) was found in the surface soil of Patrakhola tea garden (Table 3.3). The highest pH (6.20) of the sub-surface soil was found in the Kazi and kazi tea garden while the lowest pH (3.80) was found in the sub-surface soil of Oodaleah tea garden. The highest mean pH (5.63) was found in the sub-surface soil of Kazi & Kazi tea garden and the lowest mean pH (4.45) was found in the sub-surface soil of Neptune tea garden (Table 3.3). Significant variations ($^{ANOVA}F = 6.36$, $p < 0.001$) were observed in soil pH among the different tea gardens (Figure 3.7).

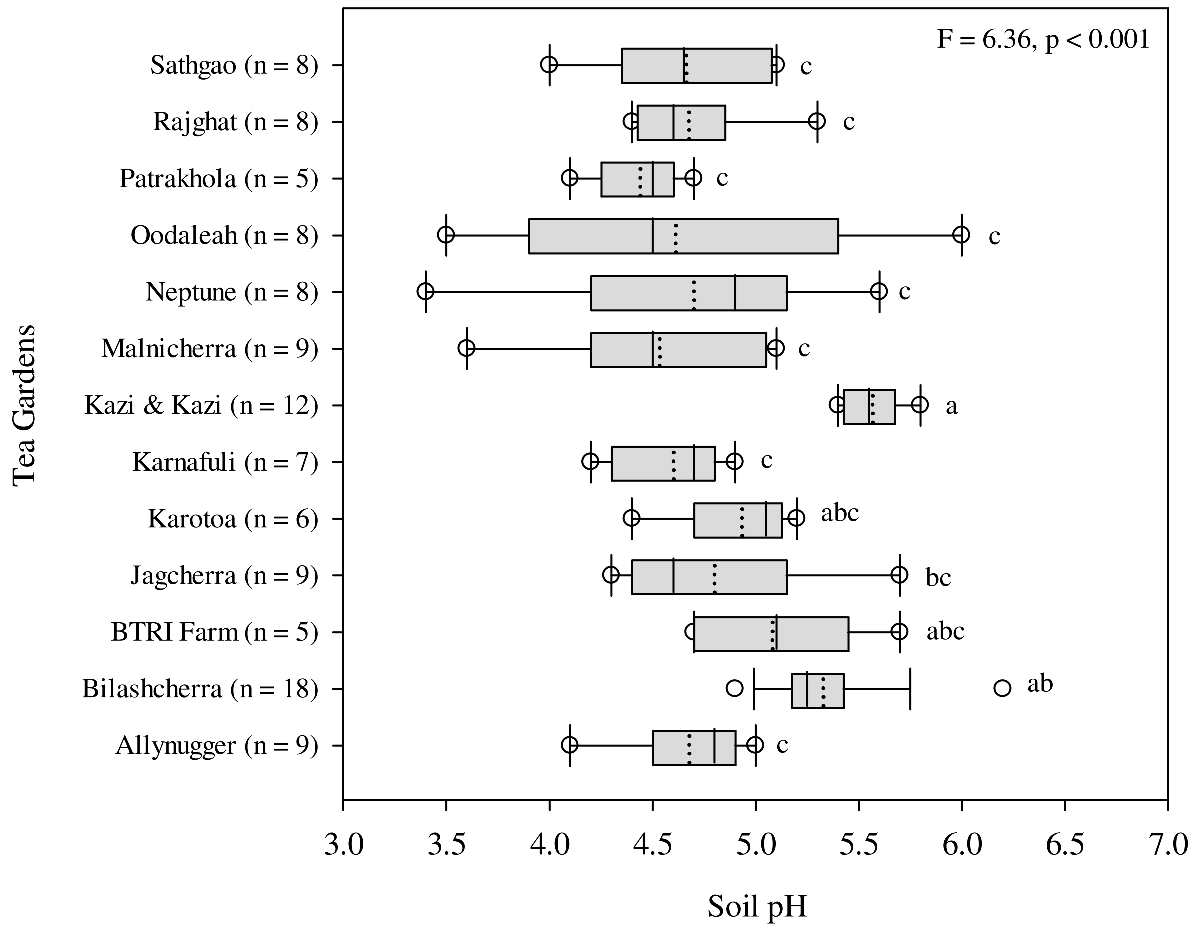


Figure 3.7: Box and whisker plots showing variability of pH in the surface soil (0-23cm depth) in different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, c designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The pH were also varied significantly ($^{ANOVA}F = 8.58$, $p < 0.001$) in sub-surface soils (Figure A7). The surface soil pH in Oodaleah tea garden had a wider variability (Figure 3.7). The mean pH of the surface soils of Kazi and Kazi tea garden was found to be significantly different from other tea gardens. Adhikary *et al*. (2019) reported that surface soil pH of Malnicherra tea garden and Kazi and Kazi tea garden was 4.50 and 5.70, respectively, and sub-surface soil pH of Kazi and Kazi tea garden was 5.55. Sultana *et al*. (2014) reported that the soil pH of Karotoa tea garden was 4.20, while in our present study the pH in the surface soil of the tea garden was found up to 6.20.

The highest organic carbon content (3.49%) of the surface soil was found in the Bilashcherra tea garden while the lowest organic carbon content (0.29%) of the surface soil was found in the Malnicherra tea garden. The highest mean content of organic carbon (2.12%) was found in the surface soils of Bilashcherra tea garden and the lowest mean content of organic carbon (0.53%) was found in the surface soils of Malnicherra tea garden (Table 3.3). The highest organic carbon content (2.59%) of the sub-surface soil was found in the Bilashcherra tea garden while the lowest organic carbon content (0.29%) of the sub-surface soil was found in the Oodaleah tea garden. The highest mean content of organic carbon (1.84%) was found in the sub-surface soils of Karotoa tea garden and the lowest mean content of organic carbon (0.49%) was found in the Karnafuli tea garden (Table 3.3). Significant variations ($^{ANOVA}F = 18.25$, $p < 0.001$) were observed in the contents of organic carbon in surface soils among the different tea gardens (Figure 3.8). The organic carbon contents were also varied significantly ($^{ANOVA}F = 15.83$, $p < 0.001$) in sub-surface soils (Figure A8). The organic carbon content in the surface soil of Bilashcherra, Kazi and Kazi and Rajghat tea garden had the wider variability (Figure 3.8).

The mean organic carbon content in the surface soils of Bilashcherra and Malnicherra tea garden was found significantly different from other tea gardens (Figure 3.8). Ahmed *et al*. (2005) reported that the organic carbon content in some tea garden soils of Bangladesh was varied from 0.79 to 1.24%. The organic carbon content in the surface soil of Malnicherra and Kazi and Kazi tea garden was observed 0.924% and 1.752%, respectively and the organic carbon content in the sub-surface soil of Kazi and Kazi tea garden was observed 1.768% (Adhikary *et al*., 2019), while in our present study the organic carbon content in the surface soil of the tea garden was found up to 3.49%.

Table 3.3: Status of pH, organic matter, organic carbon, total nitrogen and boron in the tea garden soils of Bangladesh.

Tea Estate	Depth (cm)		pH	OM (%)	OC (%)	Total N (%)	B (mg/kg)
Allynugger	0-23 (n=9)	*Min – Max*	4.10 – 5.00	1.60 – 2.70	0.93 – 1.57	0.08 – 0.14	0.12 –0.32
		Mean±s.e.	4.68±0.09	2.09±0.13	1.21±0.07	0.10±0.01	0.23±0.02
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Bilashcherra	0-23 (n=18)	*Min – Max*	4.90 – 6.20	1.95 – 6.00	1.13 – 3.49	0.11 – 0.28	0.74 – 2.30
		Mean±s.e.	5.33±0.07	3.64±0.25	2.12±0.15	0.19±0.01	1.30±0.09
	23-46 (n=18)	*Min – Max*	4.00 – 5.80	1.35 – 4.45	0.79 – 2.59	0.08 – 0.23	0.90 – 1.92
		Mean±s.e.	5.19±0.10	3.07±0.18	1.79±0.10	0.16±0.01	1.37±0.06
BTRI Farm	0-23 (n=5)	*Min – Max*	4.70 – 5.70	1.10 – 2.10	0.64 – 1.22	0.06 – 0.11	0.02 – 0.76
		Mean±s.e.	5.08±0.19	1.82±0.20	1.06±0.11	0.09±0.01	0.23±0.13
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Jagcherra	0-23 (n=9)	*Min – Max*	4.30 – 5.70	0.70 – 1.50	0.41 – 0.87	0.04 – 0.08	0.01 – 0.18
		Mean±s.e.	4.80±0.16	1.14±0.09	0.67±0.05	0.06±0.004	0.06±0.02
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Karnafuli	0-23 (n=7)	*Min – Max*	4.20 – 4.90	1.10 – 1.70	0.64 – 0.99	0.06 – 0.09	0.22 – 0.53
		Mean±s.e.	4.60±0.10	1.33±0.08	0.77±0.05	0.07±0.004	0.35±0.04
	23-46 (n=3)	*Min – Max*	4.30 – 4.60	0.60 – 1.00	0.35 – 0.58	0.03 – 0.05	0.29 – 0.36
		Mean±s.e.	4.47±0.09	0.83±0.12	0.49±0.07	0.04±0.01	0.31±0.02
Karotoa	0-23 (n=6)	*Min – Max*	4.40 – 5.20	2.15 – 4.33	1.25 – 2.52	0.11 – 0.22	0.09 – 1.71
		Mean±s.e.	4.93±0.12	3.57±0.34	2.08±0.20	0.18±0.02	0.47±0.26
	23-46 (n=6)	*Min – Max*	4.70 – 5.50	2.16 – 3.85	1.26 – 2.24	0.11 – 0.19	0.10 – 0.72
		Mean±s.e.	5.30±0.13	3.17±0.23	1.84±0.13	0.16±0.01	0.39±0.11
Kazi & Kazi	0-23 (n=12)	*Min – Max*	5.40 – 5.80	1.38 – 4.82	0.80 – 2.80	0.07 – 0.24	0.18 – 2.27
		Mean±s.e.	5.57±0.04	3.37±0.25	1.96±0.15	0.17±0.01	0.99±0.15
	23-46 (n=12)	*Min – Max*	5.30 – 6.20	1.39 – 4.20	0.81 – 2.44	0.07 – 0.21	0.26 – 1.28
		Mean±s.e.	5.63±0.07	2.77±0.22	1.61±0.13	0.14±0.01	0.69±0.08

Table 3.3: Continued.

Tea Estate	Depth (cm)		pH	OM (%)	OC (%)	Total N (%)	B (mg/kg)
Malnicherra	0-23 (n=9)	*Min – Max*	3.60 – 5.10	0.50 – 1.80	0.29 – 1.05	0.03 – 0.09	0.01 – 0.19
		Mean±s.e.	4.53±0.17	0.91±0.14	0.53±0.08	0.05±0.01	0.09±0.03
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Neptune	0-23 (n=8)	*Min – Max*	3.40 – 5.60	0.70 – 1.90	0.41 – 1.11	0.04 – 0.10	0.13 – 0.63
		Mean±s.e.	4.70±0.24	1.26±0.14	0.73±0.08	0.06±0.01	0.26±0.06
	23-46 (n=4)	*Min – Max*	4.00 – 4.70	0.60 – 1.30	0.35 – 0.76	0.03 – 0.07	0.12 – 0.30
		Mean±s.e.	4.45±0.16	0.95±0.14	0.55±0.08	0.05±0.01	0.21±0.04
Oodaleah	0-23 (n=8)	*Min – Max*	3.50 – 6.00	1.10 – 2.80	0.64 – 1.63	0.06 – 0.14	0.05 – 0.89
		Mean±s.e.	4.61±0.30	1.71±0.22	1.00±0.13	0.09±0.01	0.41±0.09
	23-46 (n=5)	*Min – Max*	3.80 – 6.00	0.50 – 2.20	0.29 – 1.28	0.03 – 0.11	0.02 – 0.38
		Mean±s.e.	4.72±0.36	1.16±0.28	0.68±0.16	0.06±0.01	0.20±0.06
Patrakhola	0-23 (n=5)	*Min – Max*	4.10 – 4.70	1.30 – 2.70	0.76 – 1.57	0.07 – 0.14	0.06 – 0.25
		Mean±s.e.	4.44±0.10	1.90±0.25	1.11±0.14	0.10±0.01	0.13±0.03
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Rajghat	0-23 (n=8)	*Min – Max*	4.40 – 5.30	0.60 – 3.90	0.35 – 2.27	0.03 – 0.20	0.01 – 0.37
		Mean±s.e.	4.68±0.11	2.39±0.34	1.39±0.20	0.12±0.02	0.18±0.04
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Sathgao	0-23 (n=8)	*Min – Max*	4.00 – 5.10	0.90 – 3.00	0.52 – 1.74	0.05 – 0.15	0.03 – 0.44
		Mean±s.e.	4.66±0.14	1.53±0.23	0.89±0.13	0.08±0.01	0.20±0.04
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-

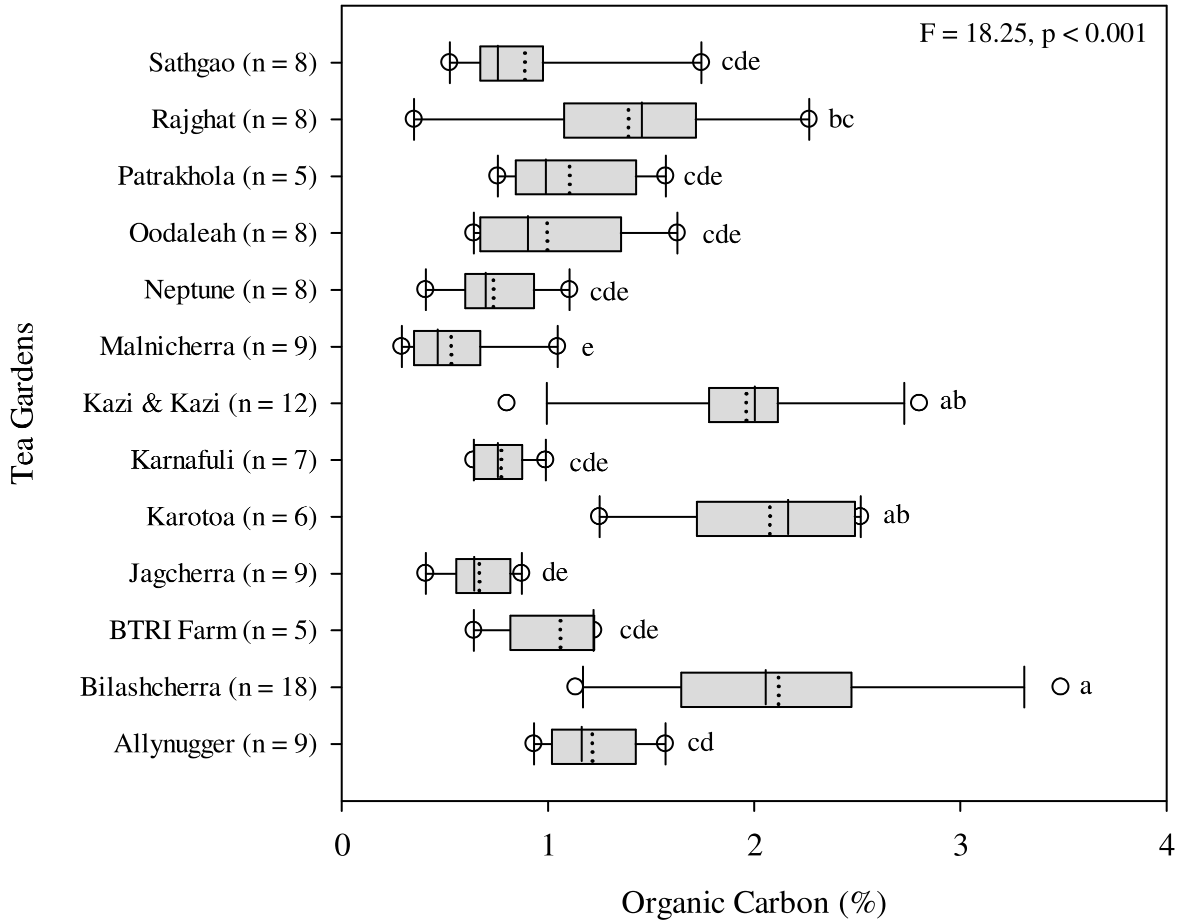


Figure 3.8: Box and whisker plots showing variability of organic carbon (%) content in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, c, d, e designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The highest total nitrogen content (0.28%) of the surface soil was found in the Bilashcherra tea garden while the lowest total nitrogen content (0.03%) of the surface soil was found in the Malnicherra and Rajghat tea gardens. The highest mean content of total nitrogen (0.19%) was found in the surface soils of Bilashcherra tea garden and the lowest mean content of total nitrogen (0.05%) was found in the surface soils of Malnicherra tea garden (Table 3.3). The highest total nitrogen content (0.23%) of the sub-surface soil was found in the Bilashcherra tea garden while the lowest total nitrogen content (0.03%) of the sub-surface soil was found in the Oodaleah tea garden. The highest mean content of total nitrogen (0.16%) was found in the sub-surface soils of Bilashcherra tea garden and the lowest mean content of total nitrogen (0.04%) was found in the Karnafuli tea garden (Table 3.3). Significant variations ($^{ANOVA}F = 20.98$, $p < 0.001$) were observed in the contents of total nitrogen in surface soils among the different tea gardens (Figure 3.9). The total nitrogen contents were also varied significantly ($^{ANOVA}F = 17.89$, $p < 0.001$) in sub-surface soils (Figure A9). The total nitrogen content in the surface soil of Bilashcherra, Kazi and Kazi and Rajghat tea garden had the wider variability (Figure 3.9). The mean total nitrogen content in the surface soils of Bilashcherra and Malnicherra tea garden was found significantly different from other tea gardens (Figure 3.9). Sultana *et al.* (2014) reported that the total nitrogen content of Karotoa and Kazi and kazi tea garden was 0.08% and 0.16%, respectively. The total nitrogen content in the surface soil of Malnicherra and Kazi and Kazi tea garden was observed 0.078% and 0.110%, respectively and the total nitrogen content in the sub-surface soil of Kazi and Kazi tea garden was observed 0.120% (Adhikary *et al*., 2019), while in our present study the total nitrogen content in the surface soil of the tea garden was found up to 0.28%.

The highest concentration of boron (2.30 mg/kg) of the surface soil was found in the Bilashcherra tea garden while the lowest concentration of boron (0.01 mg/kg) was found in the surface soil of Malnicherra, Rajghat and Jagcherra tea gardens. The highest mean concentration of boron (1.30 mg/kg) was found in the surface soils of Bilashcherra tea garden and the lowest mean concentration of boron (0.06 mg/kg) was found in the surface soil of Jagcherra tea garden (Table 3.3). The highest concentration of boron (1.92 mg/kg) was found in the sub-surface soil of Bilashcherra tea garden while the lowest concentration of boron (0.02 mg/kg) was found in the sub-surface soil of Oodaleah tea garden. The highest mean concentration of boron (1.37 mg/kg) was found in the sub-surface soils of Bilashcherra tea garden and the lowest mean concentration of boron (0.20 mg/kg) was found in the sub-surface soils of Oodaleah tea garden (Table 3.3). Significant variations ($^{ANOVA}F = 19.68$, $p < 0.001$) were observed in the concentrations of boron in surface soils among the different tea gardens (Figure 3.10). The concentrations of boron were also varied significantly ($^{ANOVA}F = 38.50$, $p < 0.001$) in sub-surface soils (Figure A10).

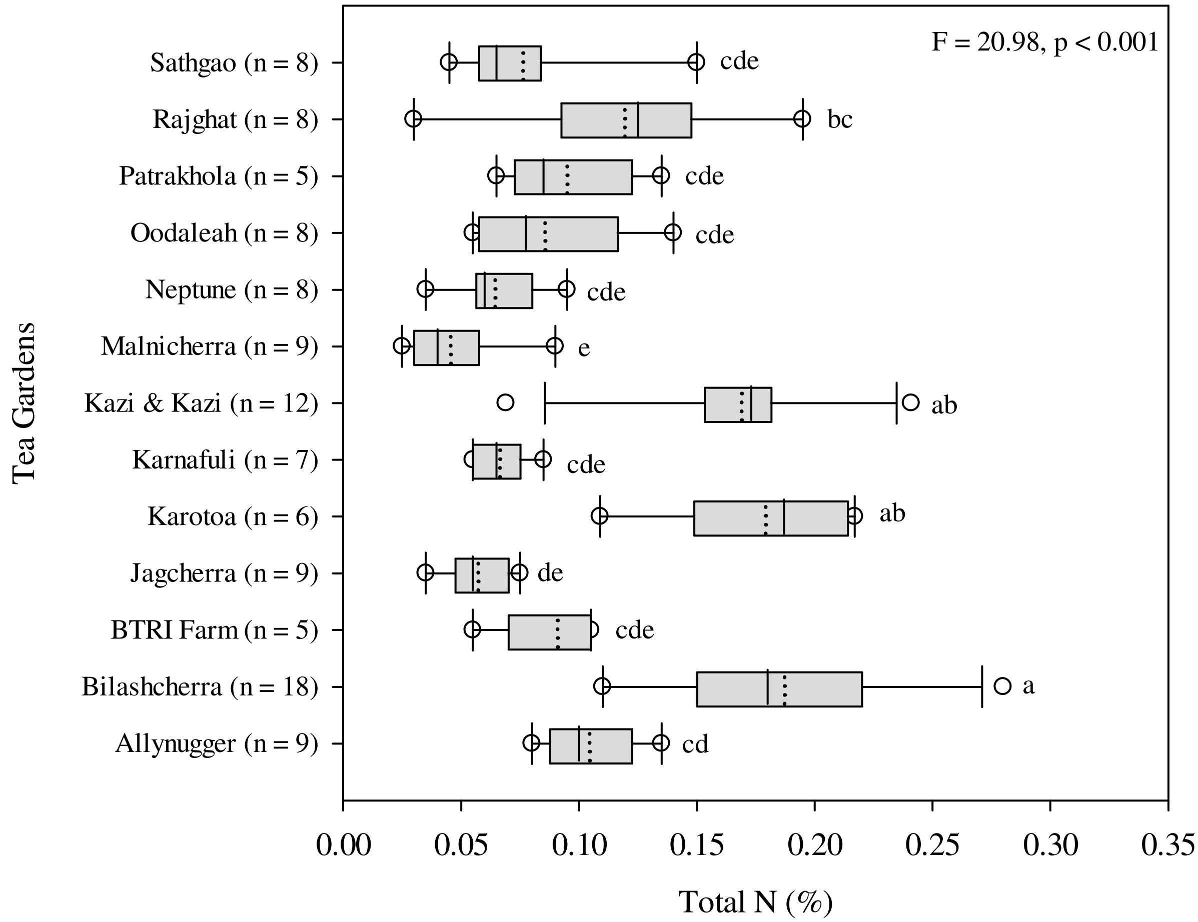


Figure 3.9: Box and whisker plots showing variability of total nitrogen content (%) in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b, c, d, e designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

The concentration of boron in the surface soil of Kazi and Kazi, Karotoa, Bilashcherra, Oodaleah and BTRI tea garden had the wider variability (Figure 3.10). The mean concentration of boron in the surface soils of Kazi and Kazi, and Bilashcherra tea garden was found significantly different from other tea gardens (Figure 3.10). Kabata-Pendias (2011) reported that the worldwide average concentration of boron for soils was estimated as 42 mg/kg. The critical limit of boron is 0.5 mg/kg (Ma. *et al*., 2003). The concentration of boron in the tea garden soils of Assam, India was ranged from 0.039 to 1.45 mg/kg (Baruah *et al*., 2011). In the tea garden soil the average concentration of boron was varied from 0.16 to 0.36 mg/kg (Phukan *et al*., 2014), while in our present study the concentration of boron in the surface soil of the tea garden was found up to 2.30 mg/kg.

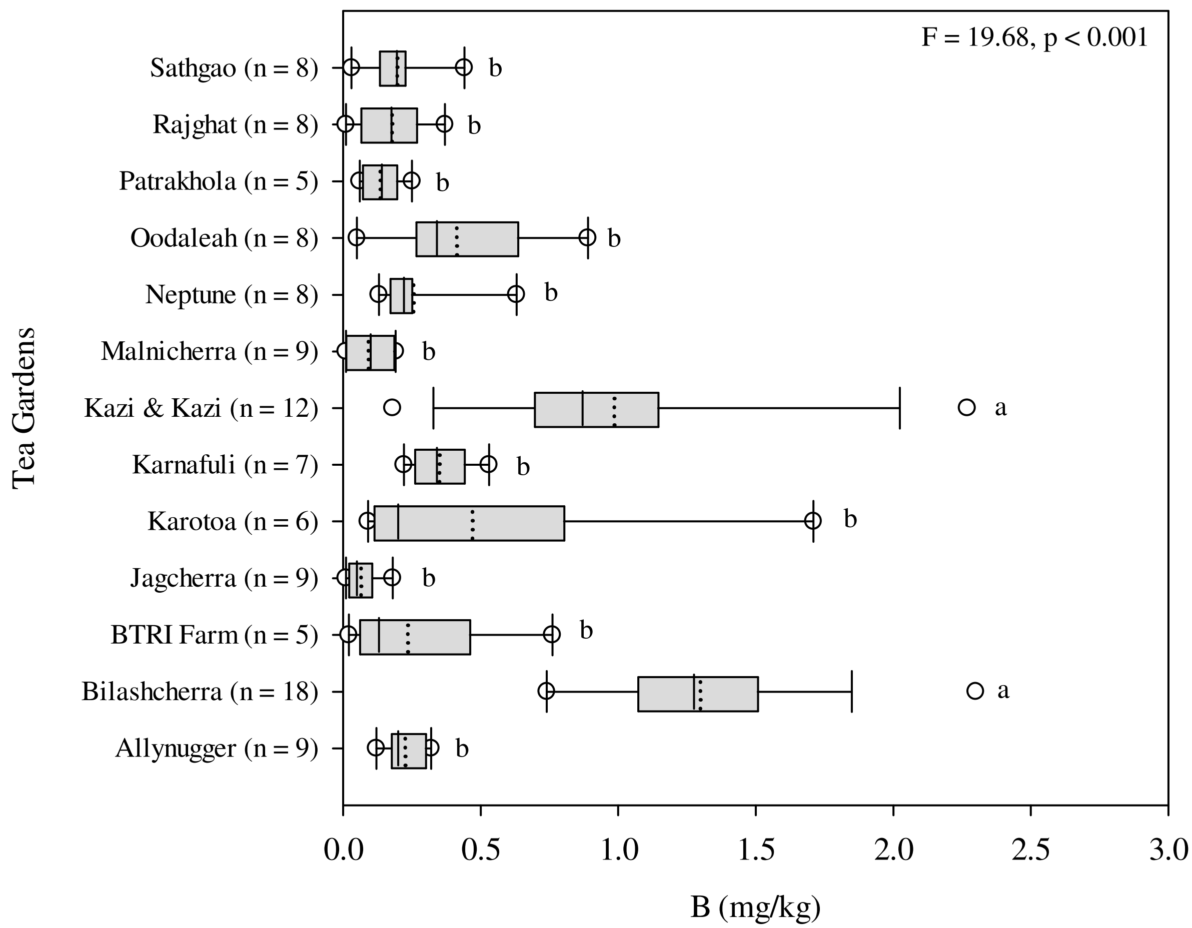


Figure 3.10: Box and whisker plots showing variability of Boron (mg/kg) concentrations in the surface soils (0-23 cm depth) of different tea gardens of Bangladesh. Figures in parentheses are the number of samples. The box plots indicate the lower and upper quartile (box), the median (solid line), the 10th and 90th percentiles (whiskers) and the 5th and 95th percentiles (circles). The F-value and P-value were obtained from one-way analysis of variance (ANOVA) test. The F-value and P-value represent the F-ratio that indicates the degree of variation among the tea gardens and the level of significance of this variation, respectively. The Tukey letters a, b designate the grouping for the tea gardens acquired from one-way ANOVA to differentiate pair-wise the mean of each of the tea gardens. The tea gardens that share same letters are not significantly different.

3.4.3. Geographical distribution and variability of the elements in the tea garden soils of Bangladesh

The distribution of heavy metals and other elements in the tea garden soils located at different sub-districts are presented in the table 3.4 and 3.5.

The mean concentrations of chromium in the surface soils (0-23 cm) of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 51.42 mg/kg > Tetulia, 38.41 mg/kg > Fatikchhari, 30.18 mg/kg > Srimangal, 17.54 mg/kg > Sylhet Sadar 17.47 mg/kg > Kamalganj, 15.66 mg/kg (Table 3.4). The concentrations of chromium in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 18.32$, $p < 0.001$) among the sub-districts (Figure 3.11). The mean concentrations of chromium in the sub-surface soils (23-46 cm) of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 56.79 mg/kg > Tetulia, 46.07 mg/kg > Fatikchhari, 31.01 mg/kg > Srimangal, 27.24 mg/kg (Table 3.4). The concentrations of chromium in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 11.15$, $p < 0.001$) among the sub-districts (Figure 3.11).

The mean concentrations of copper in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 15.88 mg/kg > Tetulia, 11.02 mg/kg > Srimangal, 8.90 mg/kg > Fatikchhari, 8.88 mg/kg > Kamalganj, 7.78 mg/kg > Sylhet Sadar 7.34 mg/kg (Table 3.4). The concentrations of copper in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 5.65$, $p < 0.001$) among the sub-districts (Figure 3.12). The mean concentrations of copper in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 16.28 mg/kg > Tetulia, 10.99 mg/kg > Fatikchhari, 8.85 mg/kg > Srimangal, 5.87 mg/kg (Table 3.4). The concentrations of copper in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 22.28$, $p < 0.001$) among the sub-districts (Figure 3.12).

Table 3.4: Concentrations of heavy metals in the tea garden soils located at different sub-districts of Bangladesh.

Upzila	Depth (cm)		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Fatikchhari	0-23 (n=23)	*Min – Max*	7.30 – 64.95	4.30 – 15.50	5500 – 38420	21.00 – 955.00	3.95 – 21.60	18.00 – 394.60
		Mean±s.e.	30.18±2.87	8.88±0.63	17870±1571	165±40.50	9.21±1.09	46.30±16.00
	23-46 (n=12)	*Min – Max*	2.55 – 60.50	5.05 – 17.70	11400 – 52540	49.00 – 466.00	5.20 – 23.70	20.92 – 63.61
		Mean±s.e.	31.01±4.31	8.85±1.07	22473±3219	159±36.20	11.09±2.00	36.95±3.45
Kamalganj	0-23 (n=14)	*Min – Max*	2.25 – 30.50	0.86 – 13.61	4206 – 14800	38.40 – 220	5.25 – 10.85	6.70 – 66.00
		Mean±s.e.	15.66±1.87	7.78±0.98	12640±756	131±14.30	7.84±0.48	32.74±5.17
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Panchagarh Sadar	0-23 (n=6)	*Min – Max*	25.55 – 73.45	12.15 – 23.30	25180 – 47120	92.00 – 234.00	13.20 –19.35	16.23 – 51.77
		Mean±s.e.	51.42±6.35	15.88±1.68	31840±3285	147±22.90	15.86±0.88	41.37±5.25
	23-46 (n=6)	*Min – Max*	31.50 – 88.65	13.10 – 22.45	27420 – 50080	116.00 – 269.00	15.30 – 20.35	29.03 – 71.05
		Mean±s.e.	56.79±8.70	16.28±1.46	35320±3514	164±23.80	17.56±0.87	46.52±5.56
Srimangal	0-23 (n=48)	*Min – Max*	2.00 – 43.45	3.35 – 22.30	5600 – 31760	32.40 – 957.00	3.00 – 23.15	2.20 – 465.20
		Mean±s.e.	17.54±1.53	8.90±0.60	14717±589	231±25.20	7.98±0.60	48.41±9.60
	23-46 (n=18)	*Min – Max*	13.30 – 46.75	1.45 – 12.65	2640 – 34400	79.00 – 378.00	4.15 – 21.25	1.73 – 37.69
		Mean±s.e.	27.24±2.14	5.87±0.62	19771±1753	214±19.60	8.70±0.93	23.28±2.18
Sylhet Sadar	0-23 (n=9)	*Min – Max*	12.40 – 22.40	5.36 – 13.05	10424 – 15741	81.00 – 1274	7.90 – 14.85	68.40 – 145.10
		Mean±s.e.	17.47±1.17	7.34±0.81	14067±522	555±130	11.44±0.83	87.51±8.25
	23-46 (n=0)	*Min – Max*	-	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-	-
Tetulia	0-23 (n=12)	*Min – Max*	9.75 – 55.00	6.95 – 17.60	3084 – 38720	101.00 – 281.00	8.20 – 15.15	34.85 – 50.25
		Mean±s.e.	38.41±3.88	11.02±0.84	22904±2510	161±15.30	11.65±0.62	42.42±1.68
	23-46 (n=12)	*Min – Max*	34.20 – 62.35	8.95 – 14.40	19060 – 37400	116.00 – 191.00	10.20 – 16.55	22.10 – 50.09
		Mean±s.e.	46.07±2.68	11.00±0.39	27870±1998	150±6.45	12.92±0.64	41.59±2.17

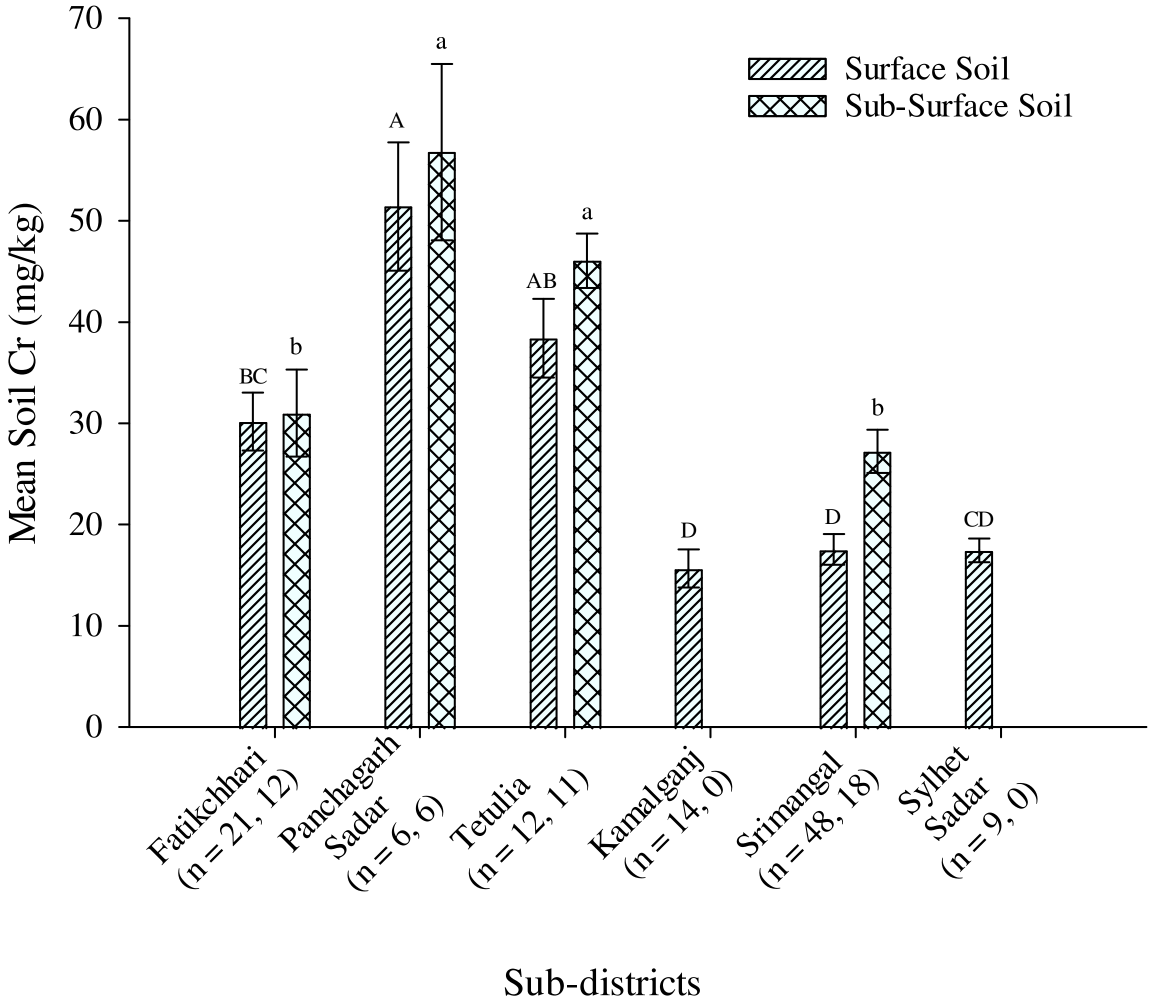


Figure 3.11: The concentrations of chromium (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of chromium in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – D, a – b) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentrations of chromium in the surface and sub-surface soils. Bars with different capital letters indicate that concentrations of chromium in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that concentrations of chromium in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

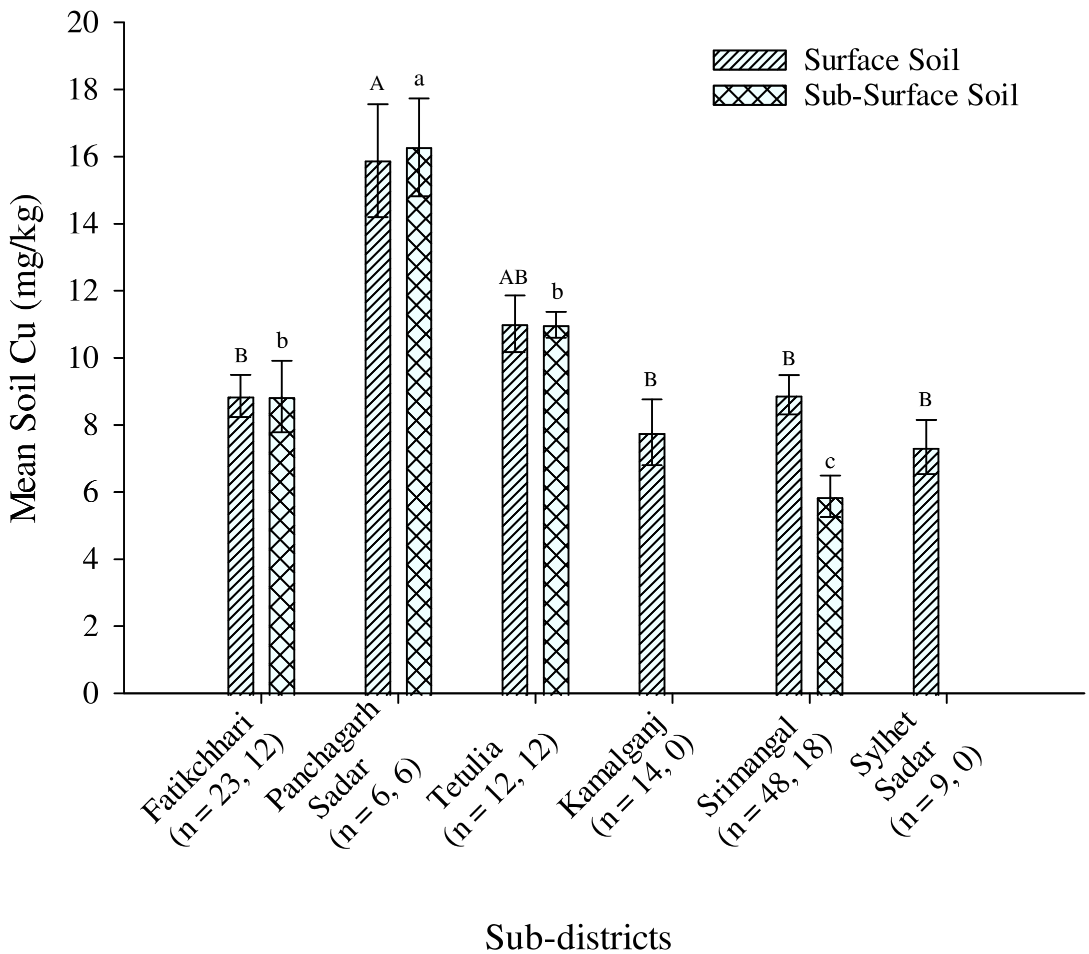


Figure 3.12: The concentrations of copper (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of copper in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – B, a – c) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentrations of copper in the surface and sub-surface soils. Bars with different capital letters indicate that concentrations of copper in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that concentrations of copper in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

The mean concentrations of iron in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 31840 mg/kg > Tetulia, 22904 mg/kg > Fatikchhari, 17870 mg/kg > Srimangal, 14717 mg/kg > Sylhet Sadar 14067 mg/kg > Kamalganj, 12640 mg/kg (Table 3.4). The concentrations of iron in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 15.03$, $p < 0.001$) among the sub-districts (Figure 3.13). The mean concentrations of iron in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 35320 mg/kg > Tetulia, 27870 mg/kg > Fatikchhari, 22473 mg/kg > Srimangal, 19771 mg/kg (Table 3.4). The concentrations of iron in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 5.90$, $p < 0.01$) among the sub-districts (Figure 3.13).

The mean concentrations of manganese in the surface soils of the tea gardens located at different sub-districts were found to in the following order: Sylhet Sadar, 555 mg/kg > Srimangal, 231 mg/kg > Fatikchhari, 165 mg/kg > Tetulia, 161 mg/kg > Panchagarh Sadar, 147 mg/kg > Kamalganj, 131 mg/kg (Table 3.4). The concentrations of manganese in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 7.57$, $p < 0.001$) among the sub-districts (Figure 3.14). The mean concentrations of manganese in the sub-surface soils of the tea gardens located at different sub-districts were found to in the following order: Srimangal, 214 mg/kg > Panchagarh Sadar, 164 mg/kg > Fatikchhari, 159 mg/kg > Tetulia, 150 mg/kg (Table 3.4). The concentrations of manganese in the sub-surface soils of the tea gardens were found insignificantly different among the sub-districts (Figure 3.14).

The mean concentrations of nickel in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 15.86 mg/kg > Tetulia, 11.65 mg/kg > Sylhet Sadar, 11.44 mg/kg > Fatikchhari, 9.21 mg/kg > Srimangal, 7.98 mg/kg > Kamalganj, 7.84 mg/kg (Table 3.4). The concentrations of nickel in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 6.38$, $p < 0.001$) among the sub-districts (Figure 3.15). The mean concentrations of nickel in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 17.56 mg/kg > Tetulia, 12.92 mg/kg > Fatikchhari, 11.09 mg/kg > Srimangal, 8.70 mg/kg (Table 3.4). The concentrations of nickel in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 6.47$, $p < 0.01$) among the sub-districts (Figure 3.15).

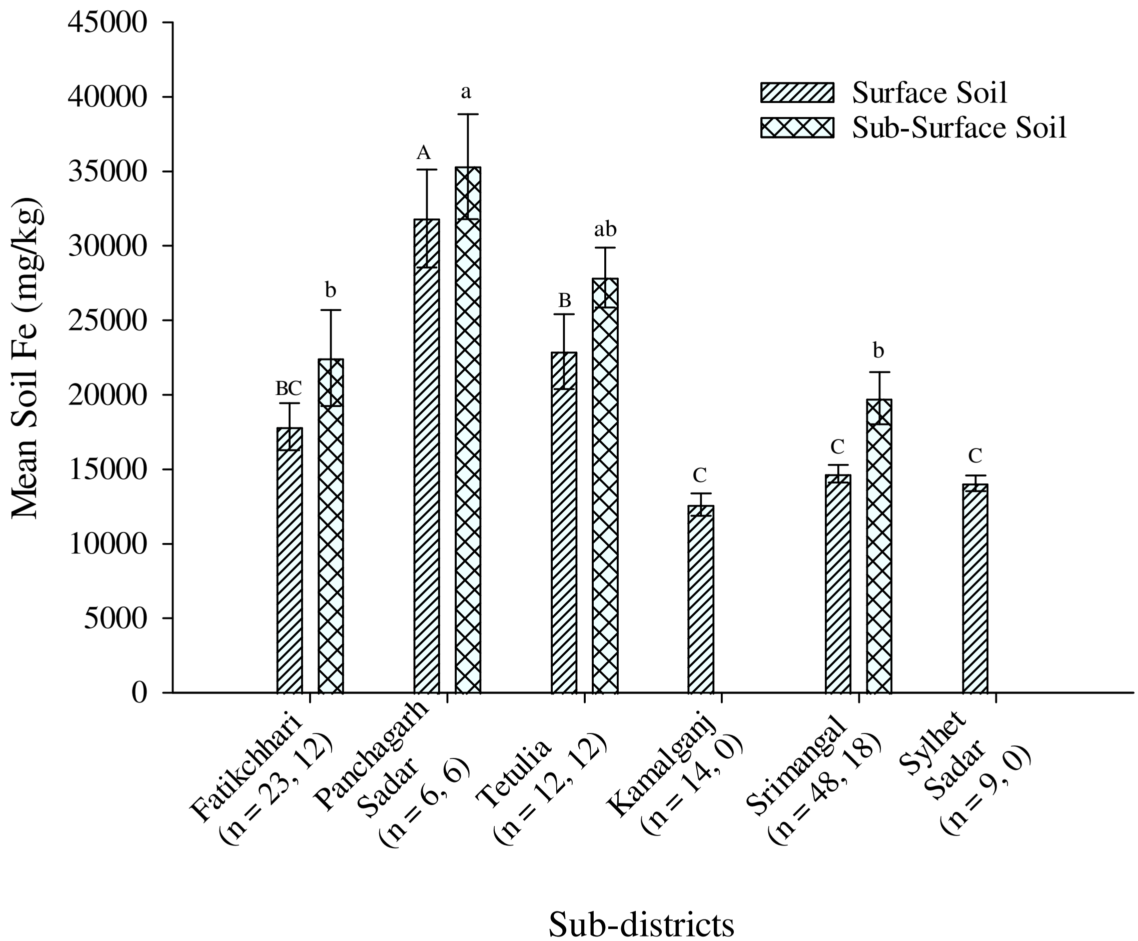


Figure 3.13: The concentrations of iron (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of iron in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – C, a – b) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentration of iron in the surface and sub-surface soils. Bars with different capital letters indicate that concentrations of iron in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that concentrations of iron in the sub-surface soils are significantly different among the sub-districts ($p < 0.01$). The bars are mean±standard error of the mean.

The mean concentrations of zinc in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Sylhet Sadar, 87.51 mg/kg > Srimangal, 48.41 mg/kg > Fatikchhari, 46.30 mg/kg > Tetulia, 42.42 mg/kg > Panchagarh Sadar, 41.37

mg/kg > Kamalganj, 32.74 mg/kg (Table 3.4). The concentrations of zinc in the surface soils of the tea gardens were found insignificantly different among the sub-districts (Figure 3.16).

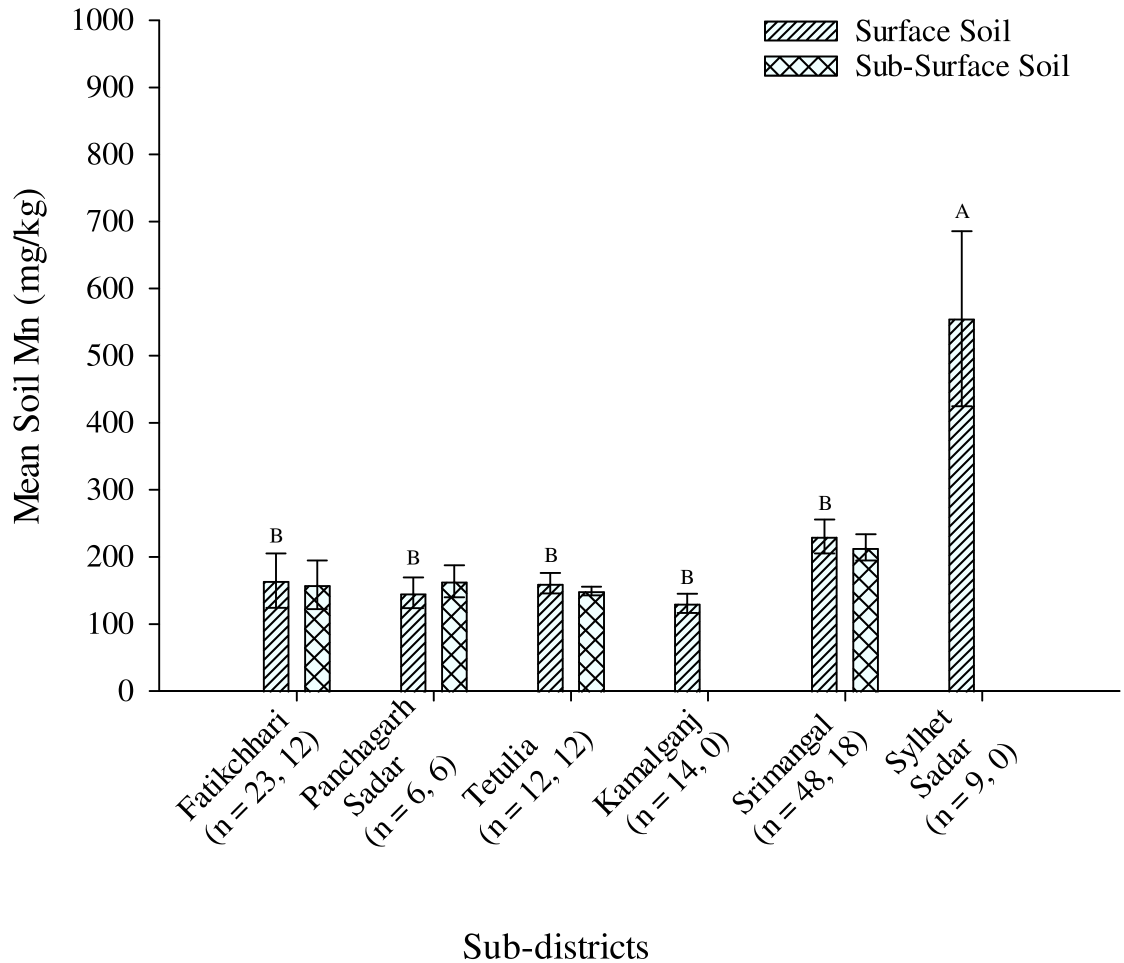


Figure 3.14: The concentrations of manganese (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of manganese in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – B) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentrations of manganese in the surface and sub-surface soils. Bars with different capital letters indicate that concentrations of manganese in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars without letters indicate that concentrations of manganese in the sub-surface soils are insignificantly different among the sub-districts. The bars are mean±standard error of the mean.

The mean concentrations of zinc in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 46.52 mg/kg > Tetulia, 41.59 mg/kg > Fatikchhari, 36.95 mg/kg > Srimangal, 23.28 mg/kg (Table 3.4). The concentrations of zinc in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 12.20$, $p < 0.001$) among the sub-districts (Figure 3.16).

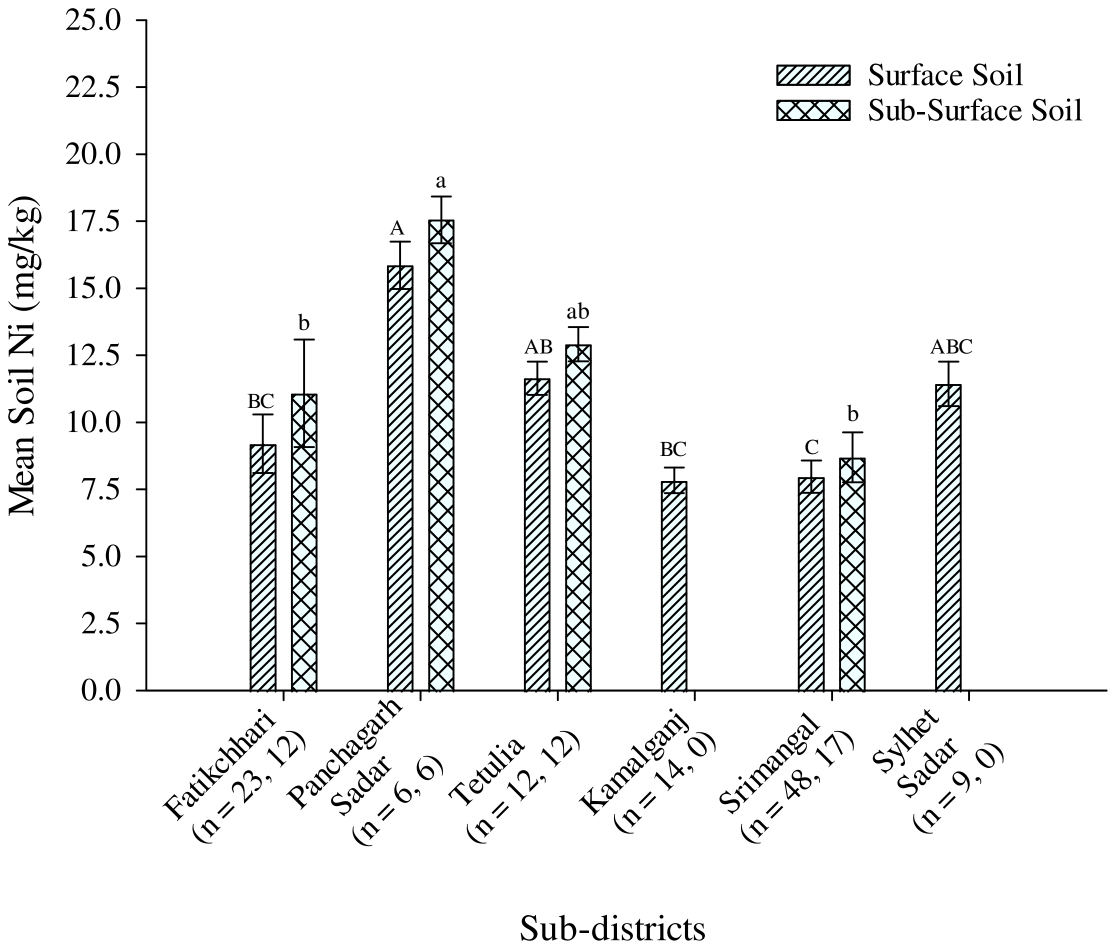


Figure 3.15: The concentrations of nickel (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of nickel in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – C, a – b) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentrations of nickel in the surface and sub-surface soils. Bars with different capital letters indicate that concentrations of nickel in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that concentrations of nickel in the sub-surface soils are significantly different among the sub-districts ($p < 0.01$). The bars are mean±standard error of the mean.

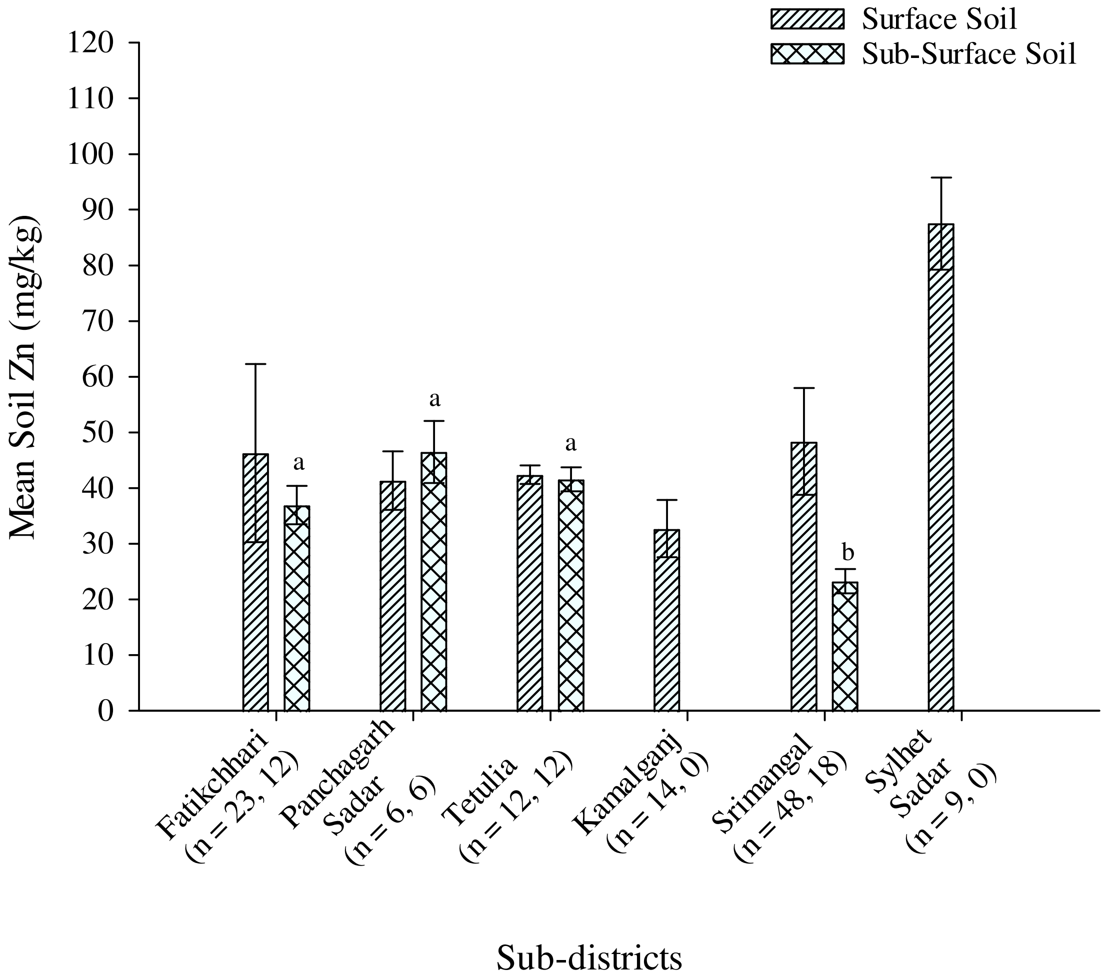


Figure 3.16: The concentrations of zinc (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of zinc in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (a – b) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentrations of zinc in the surface and sub-surface soils. Bars with no letters indicate that concentrations of zinc in the surface soils are insignificantly different among the sub-districts, while bars followed by different lower-case letters indicate that concentrations of zinc in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

The mean pH of the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Tetulia, 5.57 > Srimangal, 4.98 > Panchagarh Sadar, 4.93 > Fatikchhari, 4.64 > Kamalganj, 4.59 > Sylhet Sadar 4.53 (Table 3.5). The pH in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 9.18$, $p < 0.001$)

among the sub-districts (Figure 3.17). The mean pH of the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Tetulia, 5.63 > Panchagarh Sadar, 5.30 > Srimangal, 5.19 > Fatikchhari, 4.57 (Table 3.5). The pH in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 14.15$, $p < 0.001$) among the sub-districts (Figure 3.17). The pH of the tea garden soils of Moulvibazar district was found to be in the range between 4.13 and 5.82 (mean, 4.86) as reported by Sanaullah *et al*. (2016).

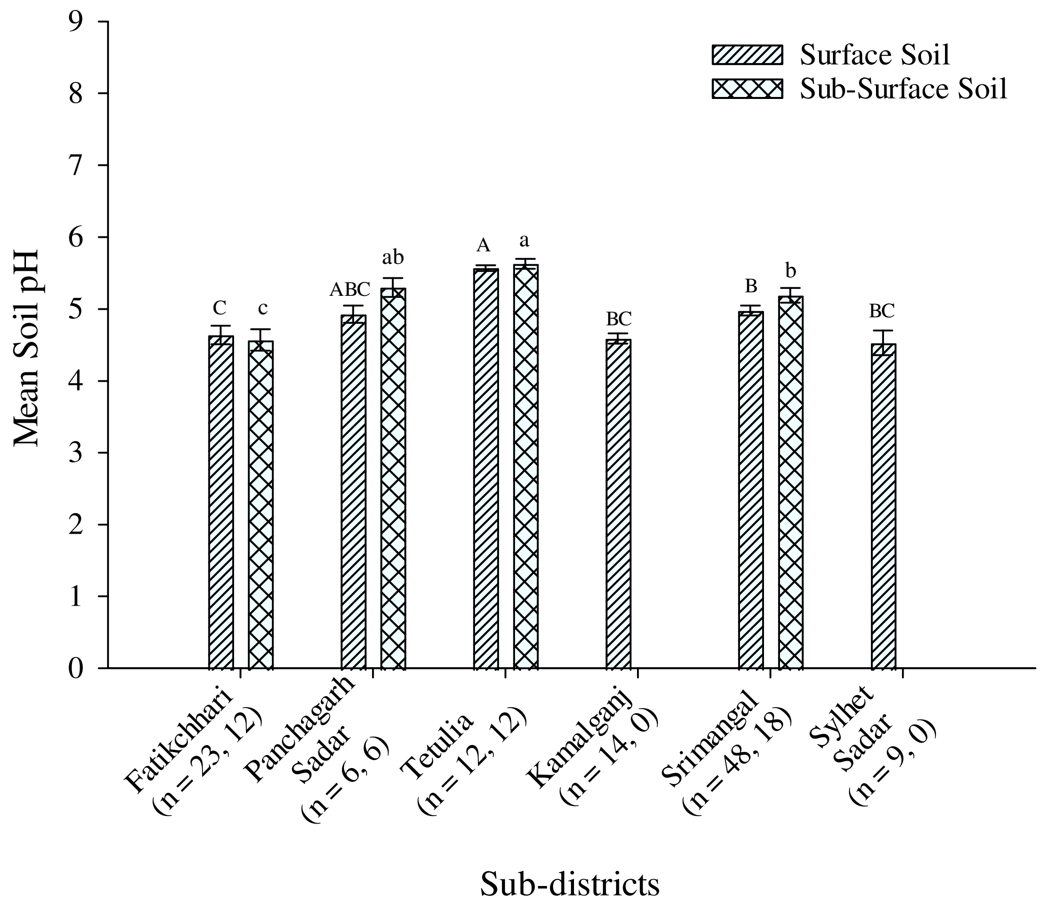


Figure 3.17: The pH in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of pH in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – C, a – c) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean pH in the surface and sub-surface soils. Bars with different capital letters indicate that pH in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that pH in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

Table 3.5: Status of pH, organic matter, organic carbon, total nitrogen and boron in the tea garden soils located at different sub-districts of Bangladesh.

Upzila	Depth (cm)		pH	OM (%)	OC (%)	Total N (%)	B (mg/kg)
Fatikchhari	0-23 (n=23)	*Min – Max*	3.40 – 6.00	0.70 – 2.80	0.41 – 1.63	0.04 – 0.14	0.05 – 0.89
		Mean±s.e.	4.64±0.13	1.44±0.10	0.84±0.06	0.07±0.01	0.34±0.04
	23-46 (n=12)	*Min – Max*	3.80 – 6.00	0.50 – 2.20	0.29 – 1.28	0.03 – 0.11	0.02 – 0.38
		Mean±s.e.	4.57±0.15	1.01±0.13	0.59±0.07	0.05±0.01	0.23±0.03
Kamalganj	0-23 (n=14)	*Min – Max*	4.10 – 5.00	1.30 – 2.70	0.76 – 1.57	0.07 – 0.14	0.06 – 0.32
		Mean±s.e.	4.59±0.07	2.02±0.12	1.18±0.07	0.10±0.01	0.19±0.02
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Panchagarh Sadar	0-23 (n=6)	*Min – Max*	4.40 – 5.20	2.15 – 4.33	1.25 – 2.52	0.11 – 0.22	0.09 – 1.71
		Mean±s.e.	4.93±0.12	3.57±0.34	2.08±0.20	0.18±0.02	0.47±0.26
	23-46 (n=6)	*Min – Max*	4.70 – 5.50	2.16 – 3.85	1.26 – 2.24	0.11 – 0.19	0.10 – 0.72
		Mean±s.e.	5.30±0.13	3.17±0.23	1.84±0.13	0.16±0.01	0.39±0.11
Srimangal	0-23 (n=48)	*Min – Max*	4.00 – 6.20	0.60 – 6.00	0.35 –3.49	0.03 – 0.28	0.01 – 2.30
		Mean±s.e.	4.98±0.07	2.42±0.19	1.41±0.11	0.12±0.01	0.60±0.09
	23-46 (n=18)	*Min – Max*	4.00 – 5.80	1.35 – 4.45	0.79 – 2.59	0.08 – 0.23	0.90 – 1.92
		Mean±s.e.	5.19±0.10	3.07±0.18	1.79±0.10	0.16±0.01	1.37±0.06
Sylhet Sadar	0-23 (n=9)	*Min – Max*	3.60 – 5.10	0.50 – 1.80	0.29 – 1.05	0.03 – 0.09	0.01 – 0.19
		Mean±s.e.	4.53±0.17	0.91±0.14	0.53±0.08	0.05±0.01	0.09±0.03
	23-46 (n=0)	*Min – Max*	-	-	-	-	-
		Mean±s.e.	-	-	-	-	-
Tetulia	0-23 (n=12)	*Min – Max*	5.40 – 5.80	1.38 – 4.82	0.80 – 2.80	0.07 – 0.24	0.18 – 2.27
		Mean±s.e.	5.57±0.04	3.37±0.25	1.96±0.15	0.17±0.01	0.99±0.15
	23-46 (n=12)	*Min – Max*	5.30 – 6.20	1.39 – 4.20	0.81 – 2.44	0.07 – 0.21	0.26 –1.28
		Mean±s.e.	5.63±0.07	2.77±0.22	1.61±0.13	0.14±0.01	0.69±0.08

In the present study, the surface soils of the tea gardens of Srimangal and Kamalganj sub-districts of Moulvibazar district were observed to have pH in the range between 4.00 and 6.20, and 4.10 and 5.00, respectively. Islam and Sanaullah (2011) found the pH of the tea garden soils of Chattogram district in the range between 3.85 and 5.13 (mean, 4.37), while in the present study, the pH of 3.40 - 6.00 was found in the surface soils of the tea gardens of Fatikchhari sub-district of Chattogram district. Zaman *et al*. (2019) reported observed pH from 3.96 to 4.80 (mean, 4.22) in some tea garden soils of Tetulia sub-district of Panchagarh district, whereas in the present study, pH from 5.40 to 5.80 and 4.40 to 5.20 were observed in the surface soils of the tea gardens of Tetulia and Panchagarh Sadar sub-districts of Panchagarh district, respectively.

The mean contents of organic carbon in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 2.08% > Tetulia, 1.96% > Srimangal, 1.41% > Kamalganj, 1.18% > Fatikchhari, 0.84% > Sylhet Sadar 0.53% (Table 3.5). The contents of organic carbon in the surface soils of the tea gardens were found to be significantly different (${}^{ANOVA}F = 12.43$, $p < 0.001$) among the sub-districts (Figure 3.18). The mean contents of organic carbon in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 1.84% > Srimangal, 1.79% > Tetulia, 1.61% > Fatikchhari, 0.59% (Table 3.5). The contents of organic carbon in the sub-surface soils of the tea gardens were found to be significantly different (${}^{ANOVA}F = 27.16$, $p < 0.001$) among the sub-districts (Figure 3.18). Gafur and Sultana (2013) reported that the organic carbon contents in the soils of different tea valleys of Bangladesh was ranged from 0.18 to 2.12%, whereas in the present study, the contents of organic carbon were ranged from 0.29 to 3.49 % in the surface soils of the tea gardens of Bangladesh.

The mean contents of total nitrogen in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Panchagarh Sadar, 0.18% > Tetulia, 0.17% > Srimangal, 0.12% > Kamalganj, 0.10% > Fatikchhari, 0.07% > Sylhet Sadar 0.05% (Table 3.5). The contents of total nitrogen in the surface soils of the tea gardens were found to be significantly different (${}^{ANOVA}F = 12.59$, $p < 0.001$) among the sub-districts (Figure 3.19). The mean contents of total nitrogen in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Srimangal, 0.16% > Panchagarh Sadar, 0.16% > Tetulia, 0.14% > Fatikchhari, 0.05% (Table 3.5).

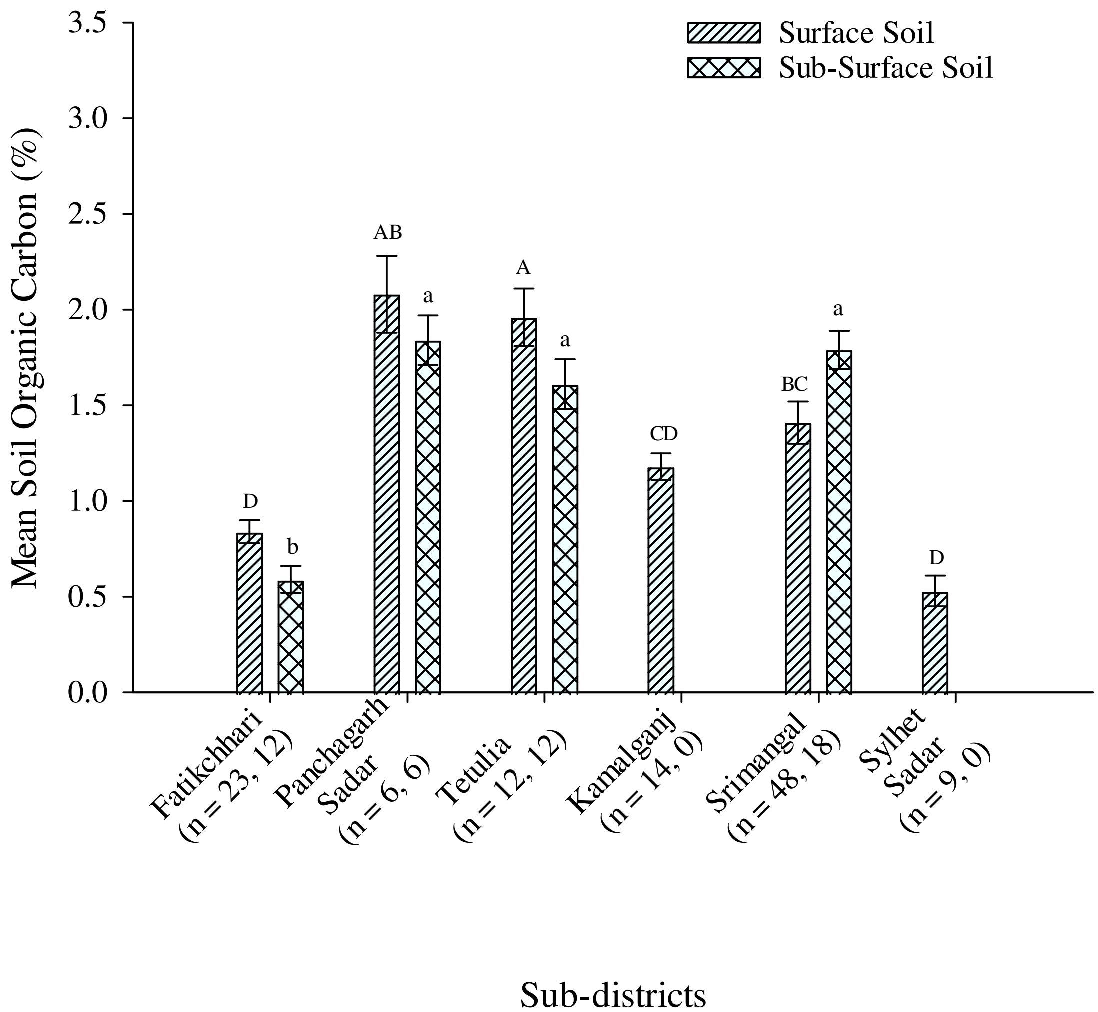


Figure 3.18: The contents of organic carbon (%) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the contents of organic carbon in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – D, a – b) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean contents of organic carbon in the surface and sub-surface soils. Bars with different capital letters indicate that organic carbon contents in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that organic carbon contents in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

The contents of total nitrogen in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 30.71$, $p < 0.001$) among the sub-districts (Figure 3.19).

Zaman *et al*. (2019) reported observed total nitrogen contents from 0.064% to 0.101% (mean, 0.82%) in some tea garden soils of Tetulia sub-district, whereas in the present study, total nitrogen contents from 0.07 to 0.24% and 0.11 to 0.22% were observed in the surface soils of the tea gardens of Tetulia and Panchagarh Sadar sub-districts, respectively.

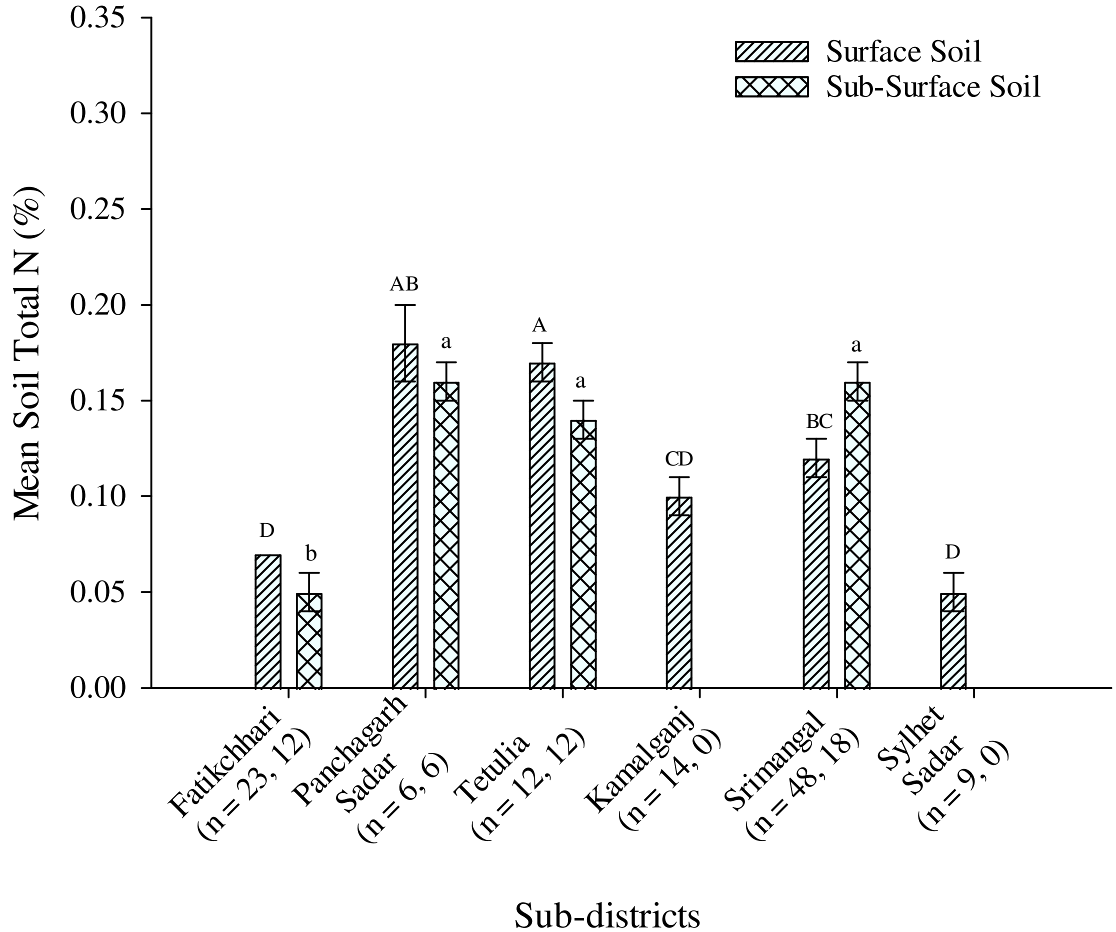


Figure 3.19: The contents of total nitrogen (%) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the contents of total nitrogen in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – D, a – b) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean contents of total nitrogen in the surface and sub-surface soils. Bars with different capital letters indicate that total nitrogen contents in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that total nitrogen contents in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

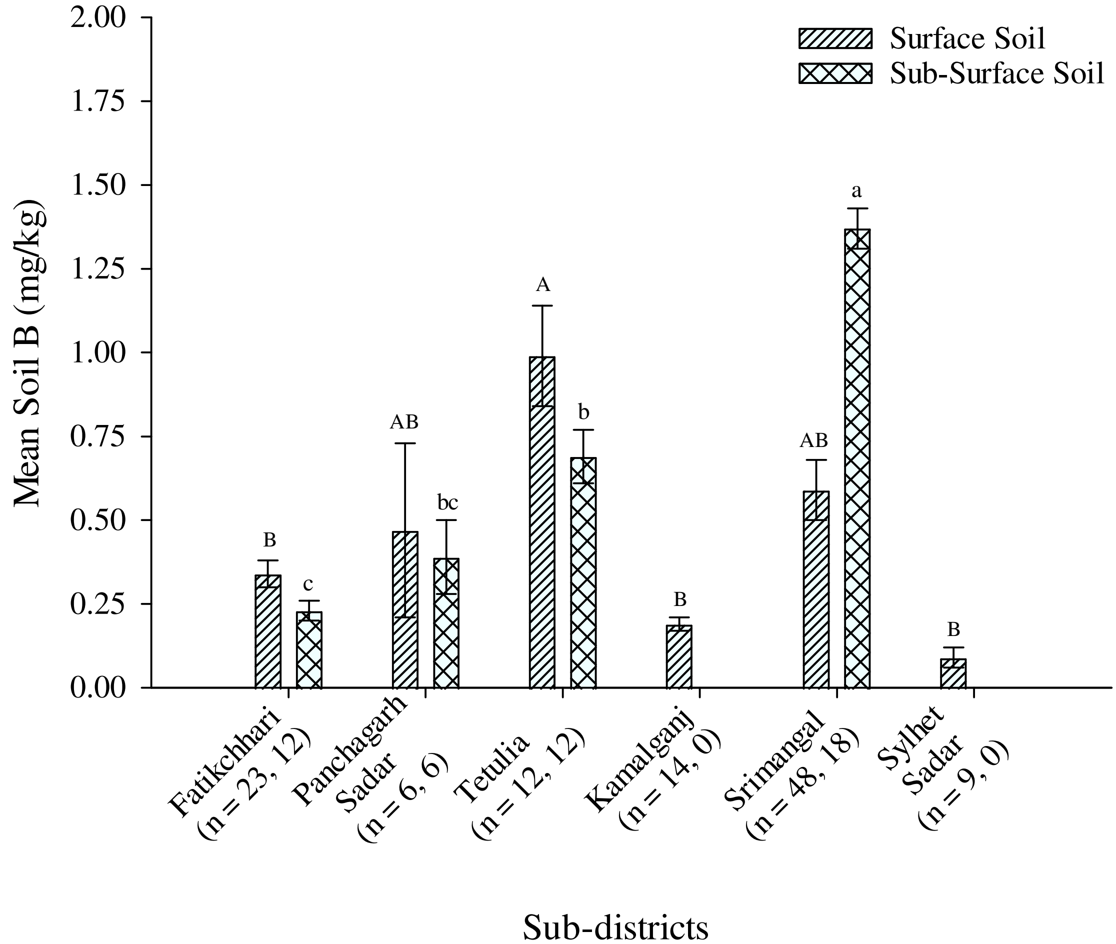


Figure 3.20: The concentrations of boron (mg/kg) in the surface and sub-surface soils of the tea gardens located at different sub-districts of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the mean of the concentrations of boron in the surface and sub-surface soils of different sub-districts. Sub-districts that share the same letter (A – B, a – c) are not significantly different. The letters indicate Tukey groupings for the sub-districts with respect to their mean concentrations of boron in the surface and sub-surface soils. Bars with different capital letters indicate that concentrations of boron in the surface soils are significantly different among the sub-districts ($p < 0.001$), while bars followed by different lower-case letters indicate that concentrations of boron in the sub-surface soils are significantly different among the sub-districts ($p < 0.001$). The bars are mean±standard error of the mean.

The mean concentrations of boron in the surface soils of the tea gardens located at different sub-districts were found to be in the following order: Tetulia, 0.99 mg/kg > Srimangal, 0.59 mg/kg > Panchagarh Sadar, 0.47 mg/kg > Fatikchhari, 0.34 mg/kg > Kamalganj, 0.19 mg/kg > Sylhet Sadar 0.09 mg/kg (Table 3.5). The concentrations of boron in the surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 5.84$, $p < 0.001$) among the sub-districts (Figure 3.20). The mean concentrations of boron in the sub-surface soils of the tea gardens located at different sub-districts were found to be in the following order: Srimangal, 1.37 mg/kg > Tetulia, 0.69 mg/kg > Panchagarh Sadar, 0.39 mg/kg > Fatikchhari, 0.23 mg/kg (Table 3.5). The concentrations of boron in the sub-surface soils of the tea gardens were found to be significantly different ($^{ANOVA}F = 66.30$, $p < 0.001$) among the sub-districts (Figure 3.20).

3.4.4. Geochemical Relationships of the elements in the tea garden soils

A significant positive relationship of chromium in the tea garden soils was found with pH ($R^2 = 0.05$, $p < 0.05$), organic carbon ($R^2 = 0.18$, $p < 0.001$), total nitrogen ($R^2 = 0.18$, $p < 0.001$), boron ($R^2 = 0.13$, $p < 0.001$), copper ($R^2 = 0.16$, $p < 0.001$), iron ($R^2 = 0.48$, $p < 0.001$) and nickel ($R^2 = 0.30$, $p < 0.001$) (Figure 3.21). Khan *et al.* (2016) reported a negative correlation of chromium with soil pH and positive correlation of chromium with organic matter. Soil pH regulates the bioavailability of heavy metals through its influence on the surface charges of clay minerals, metal (iron, manganese, and aluminium) oxides/ hydroxides and organic matter (Adriano, 2001; Bradl, 2004; Du Laing *et al*., 2009). Generally, mobility and availability of heavy metals increases in soil at low pH, whereas retention of the heavy metals increases with increasing soil pH (Adriano, 2001; Bradl, 2004; Sherene, 2010; Rinklebe *et al*., 2016). With increasing soil pH, adsorption of the metal ions increases as the number of negatively charged surface sites increased. But, at high soil pH the mobility and availability of chromium increases, whereas adsorption of chromium increases at low pH (Adriano, 2001; Bradl, 2004; Sherene, 2010). This is due to the formation of hydroxy-complexes of chromium and its oxyanion nature under reducing and oxidizing environments (Fox and Doner, 2003; Bradl, 2004; Sherene, 2010).

Copper in the tea garden soils was found to be significantly and positively related to organic carbon ($R^2 = 0.050$, $p < 0.05$), total nitrogen ($R^2 = 0.050$, $p < 0.05$), iron ($R^2 = 0.16$, $p < 0.001$) and nickel ($R^2 = 0.19$, $p < 0.001$) (Figure 3.22). Soil parameters, likes organic carbon, pH, iron and manganese oxide and total metal content which might affect the distribution of copper (Mohd-Aizat *et al*., 2014).

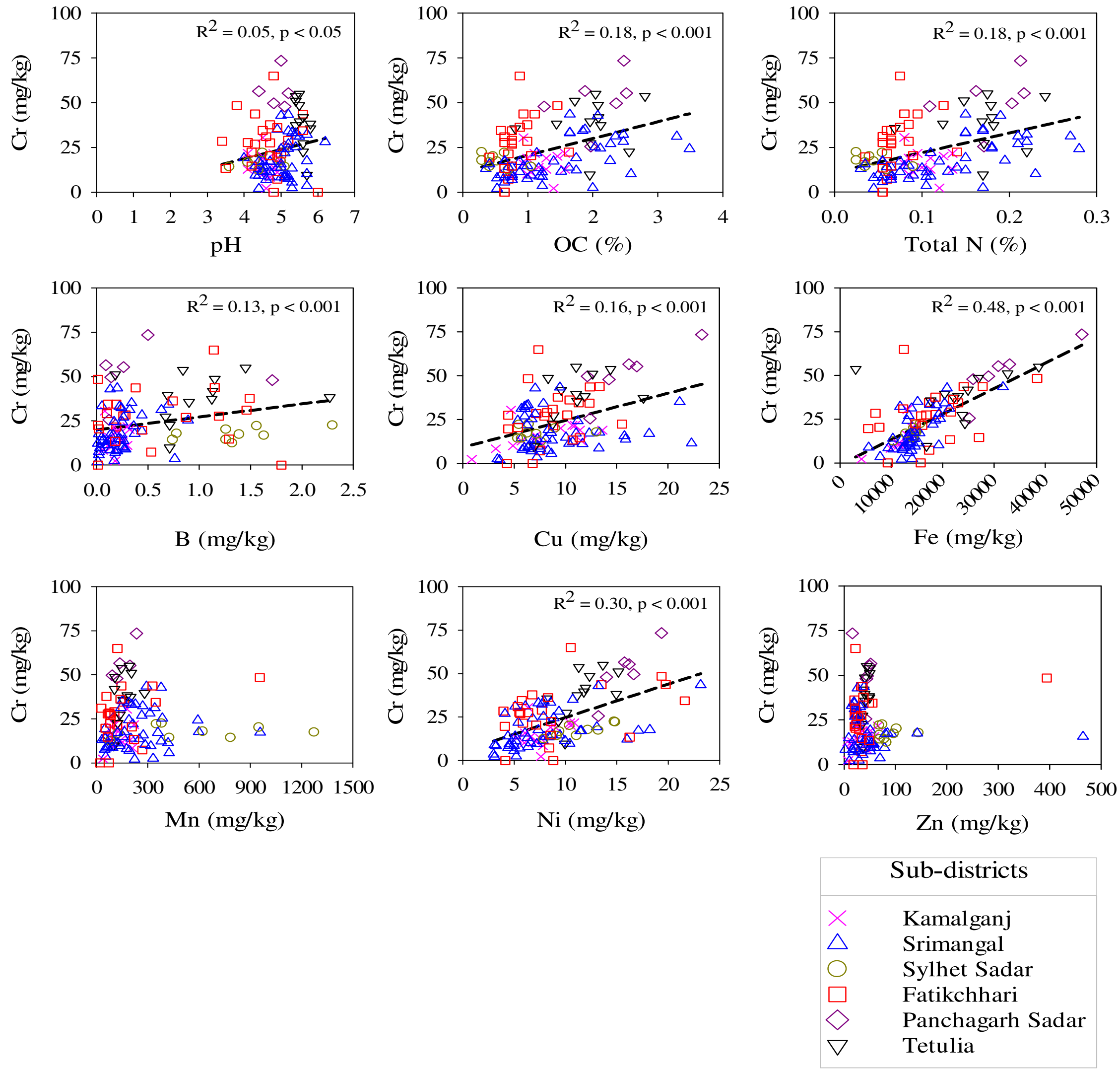


Figure 3.21: Relationships of chromium with other elements in the tea garden soils.

Li *et al.* (2018) reported a significant negative correlation of copper with soil pH and nickel. Sarker *et al.* (2020) observed a non-significant positive interaction of copper with zinc. A significant and positive relationship of iron in the tea garden soils was found with organic carbon ($R^2 = 0.13$, $p < 0.001$), total nitrogen ($R^2 = 0.13$, $p < 0.001$), boron ($R^2 = 0.06$, $p < 0.05$) and nickel ($R^2 = 0.42$, $p < 0.001$) (Figure 3.23). Sarker *et al.* (2020) reported a significant and positive correlation of iron with organic matter, total nitrogen, zinc, copper and manganese as well as a significant negative correlation of iron with soil pH.

A significant and positive relationship of manganese in the tea garden soils was found with nickel ($R^2 = 0.16$, $p < 0.001$) and zinc ($R^2 = 0.17$, $p < 0.001$) (Figure 3.24).

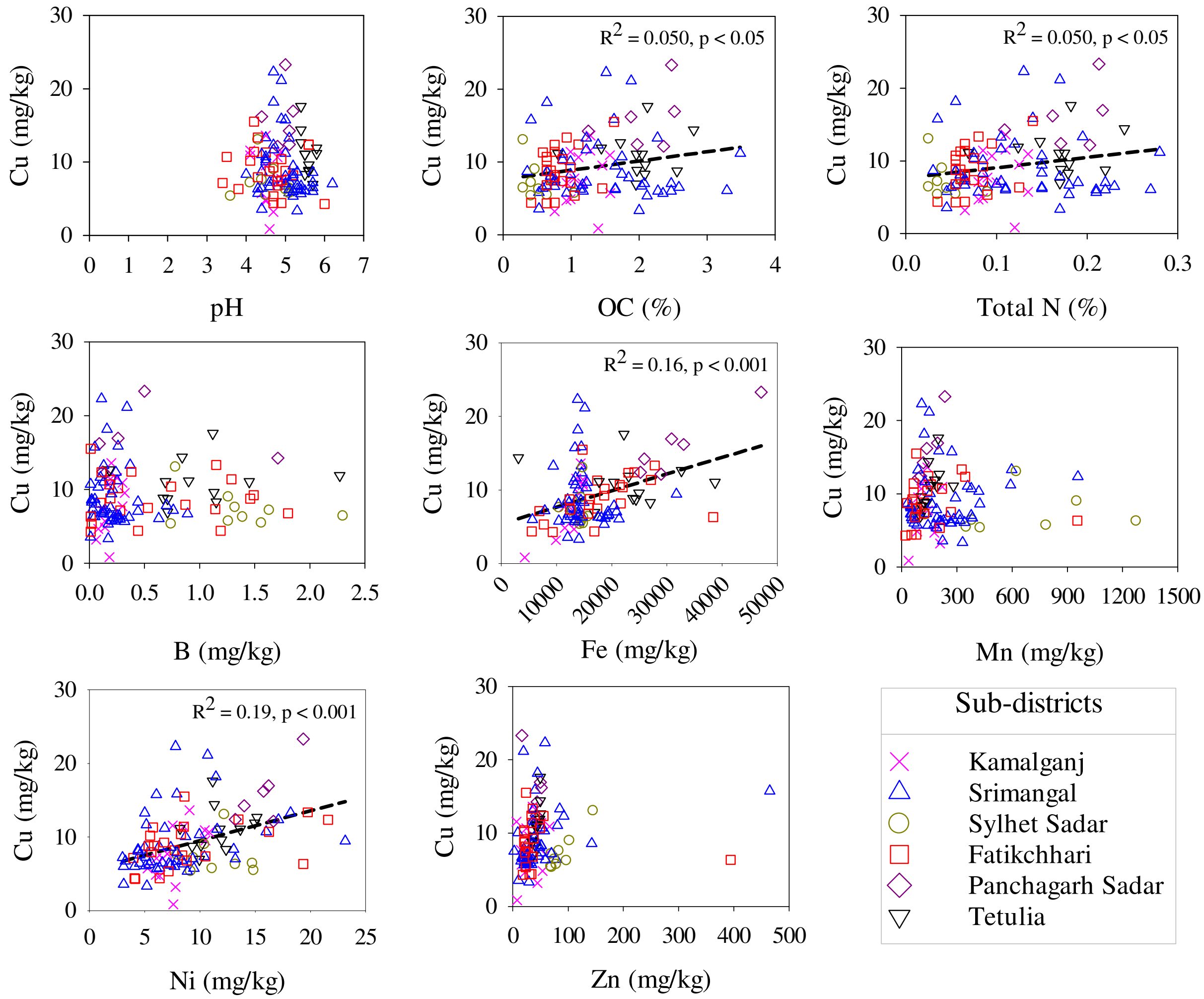


Figure 3.22: Relationships of copper with other elements in the tea garden soils.

Sarker *et al.* (2020) reported a significant positive correlation of manganese with organic matter and total nitrogen. The redox-active iron and manganese in soil are greatly regulated by pH and, therefore, the availability of these elements in soil is largely controlled by pH (Adriano, 2001; Bradl, 2004; Rinklebe *et al*., 2016). In acidic - reducing soil condition (low pH and low E_h), Fe^{3+} is reduced to Fe^{2+} and Mn^{4+} is reduced to Mn^{2+}, thus increasing their concentrations in the soil solution. In contrast, precipitation of iron and manganese as their oxides/ hydroxides occurs at oxidizing - alkaline soil conditions (high pH and high E_h), thus decreasing their bioavailability in the soil. Soil redox potential, thus, strongly influences these reactions (Kabata-Pendias, 2011; Rinklebe *et al*., 2016).

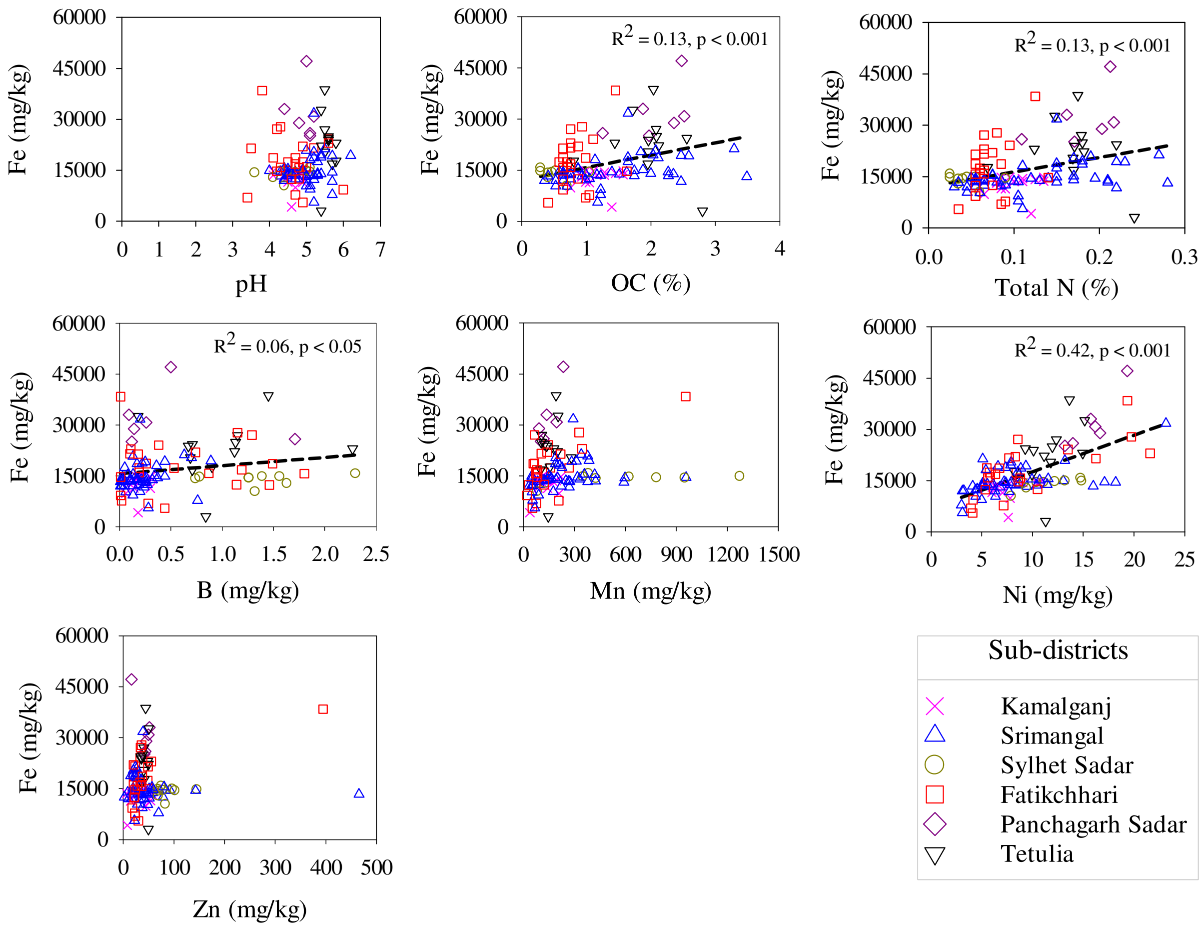


Figure 3.23: Relationships of iron with other elements in the tea garden soils.

Thus, the effects of pH on the retention and mobilization of a particular element is specific to that element and depends on the adsorption constituents of the soil (Barrow and Whelan, 1998).

A significant positive relationship of nickel in the tea garden soils was found with organic carbon (R^2 = 0.052, $p < 0.05$), total nitrogen (R^2 = 0.052, $p < 0.05$) and zinc (R^2 = 0.045, $p < 0.05$) (Figure 3.25). Li *et al.* (2018) reported a significant positive correlation of nickel with soil organic matter as well as a significant negative correlation of nickel with soil pH. No significant relationships of zinc were found with pH, organic carbon, total nitrogen and boron (Figure 3.26).

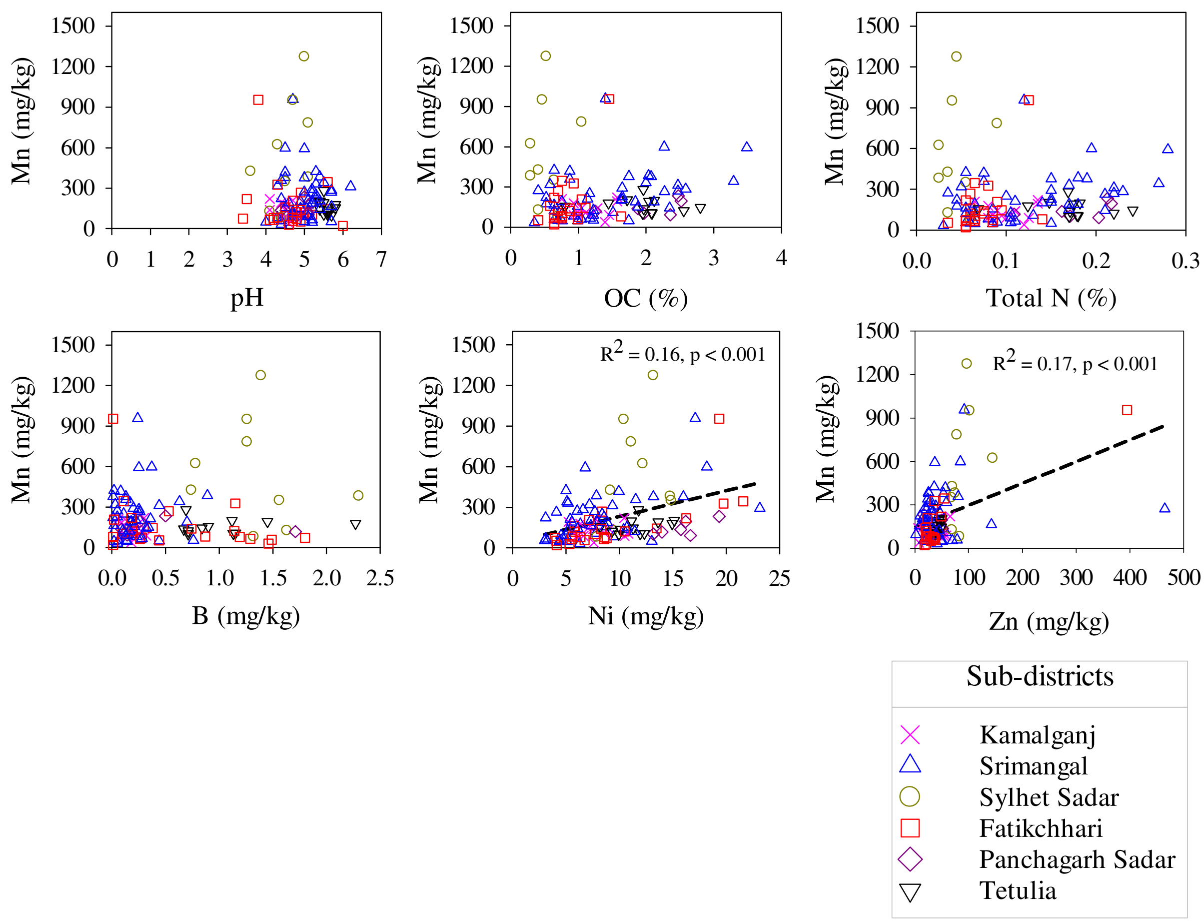


Figure 3.24: Relationships of manganese with other elements in the tea garden soils.

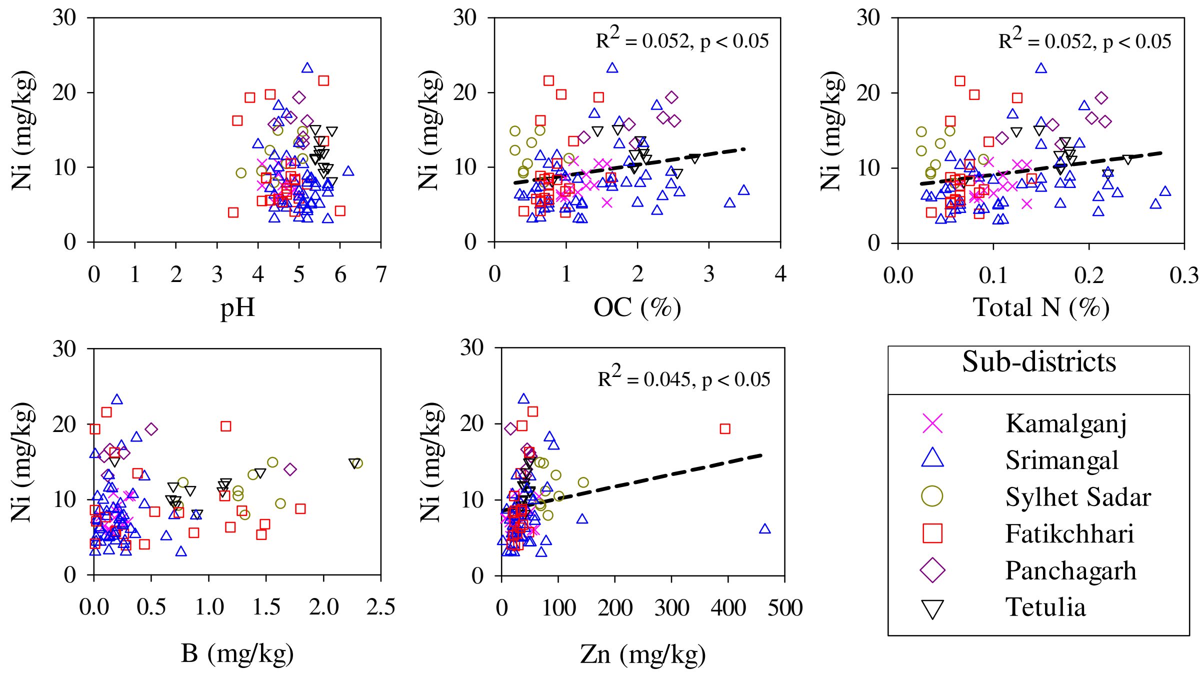


Figure 3.25: Relationships of nickel with other elements in the tea garden soils.

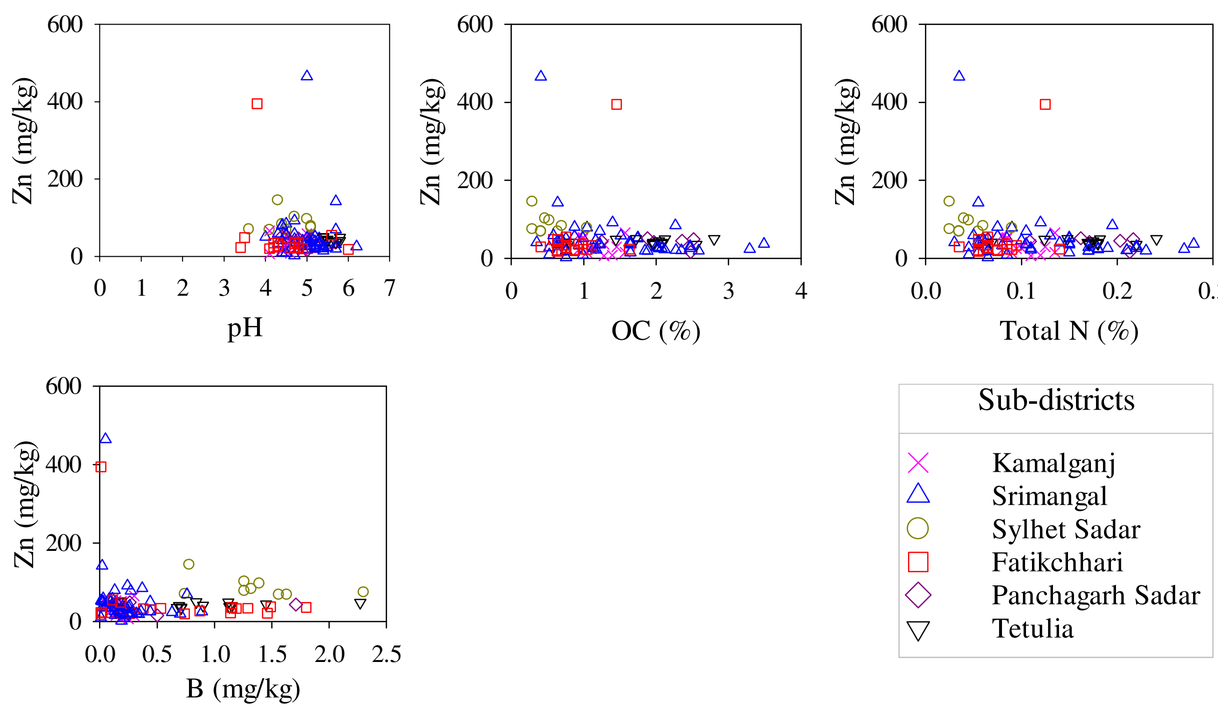


Figure 3.26: Relationships of zinc with other elements in the tea garden soils.

When Principal Component Analysis (PCA) was done to look at the interrelationships between the heavy metals, nitrogen and boron in the soils from the 6 sub-districts together, some differences were observed among the soils of the sub-districts (Figure 3.27). Here, the soils were clustered into individual sub-districts using the first, second, and third components. The eigenvalues of the first three principal components were greater than 1. These three components explained 73% of the variation in the data (Table A2). The PCA indicated that tea garden soils of Tetulia, Panchagarh Sadar and Fatikchhari sub-districts were different from the tea garden soils of the other sub-districts. The first component separated tea garden soils of Fatikchhari sub-district from Tetulia and Panchagarh Sadar. The first component also separated some soils of Srimangal and Kamalganj sub-districts. The second component separated the soils of Tetulia and Panchagarh Sadar from other sub-districts. The second component also separated some soils of Srimangal, Kamalganj and Sylhet Sadar sub-district. The first, second, and third components contributed 37.6, 21.4, and 14.0 percent, respectively, to the variations in the tea garden soils (Table A2). The loading plot visually shows that the second component has large association with manganese, nitrogen and boron. The direction of the loadings for the components indicated that copper was geochemically associated with iron. The chromium, boron, nitrogen, manganese and zinc also followed the similar geochemical trends. It was only nickel that differed from the general trends with other elements in the soils. This is expected, as the tea garden soils of the different physiographic regions also have differences in their inherent soil geochemical characteristics. Bangladeshi soils are highly and inherently variable in their concentrations of geochemical elements (Chowdhury *et al*., 2021). Spatial occurrence and accumulation of elements as well as their mobility and availability in soils are largely regulated by the topography of the landscape (Chowdhury *et al*., 2021).

When PCA was done to look at the interrelationships between heavy metals, nitrogen and boron in the soils of the 13 tea gardens together, some differences were observed among the soils of the tea gardens (Figure 3.28). Here, the soils were clustered into individual gardens using the first, second, and third components. The first component separated the soils of Neptune, Oodaleah, Karnafuli, Patrakhola and some soils of Bilashcherra and Jagcherra tea garden. The second component separated the soils of Kazi & Kazi and Karotoa tea gardens from other tea gardens. The first, second, and third components contributed 37.6, 21.4, and 14.0 percent, respectively, to the variations in the tea garden soils (Table A2). The direction of the loadings for the components indicated that copper was geochemically associated with iron. The chromium, boron, nitrogen, manganese and zinc were also followed the similar geochemical trends. It was only nickel that differed from the general trends with other elements in the soils. The management practices of tea cultivation in different tea gardens such as application of organic and inorganic fertilizers, pesticides and other agronomic practices might also affect the geochemical variability.

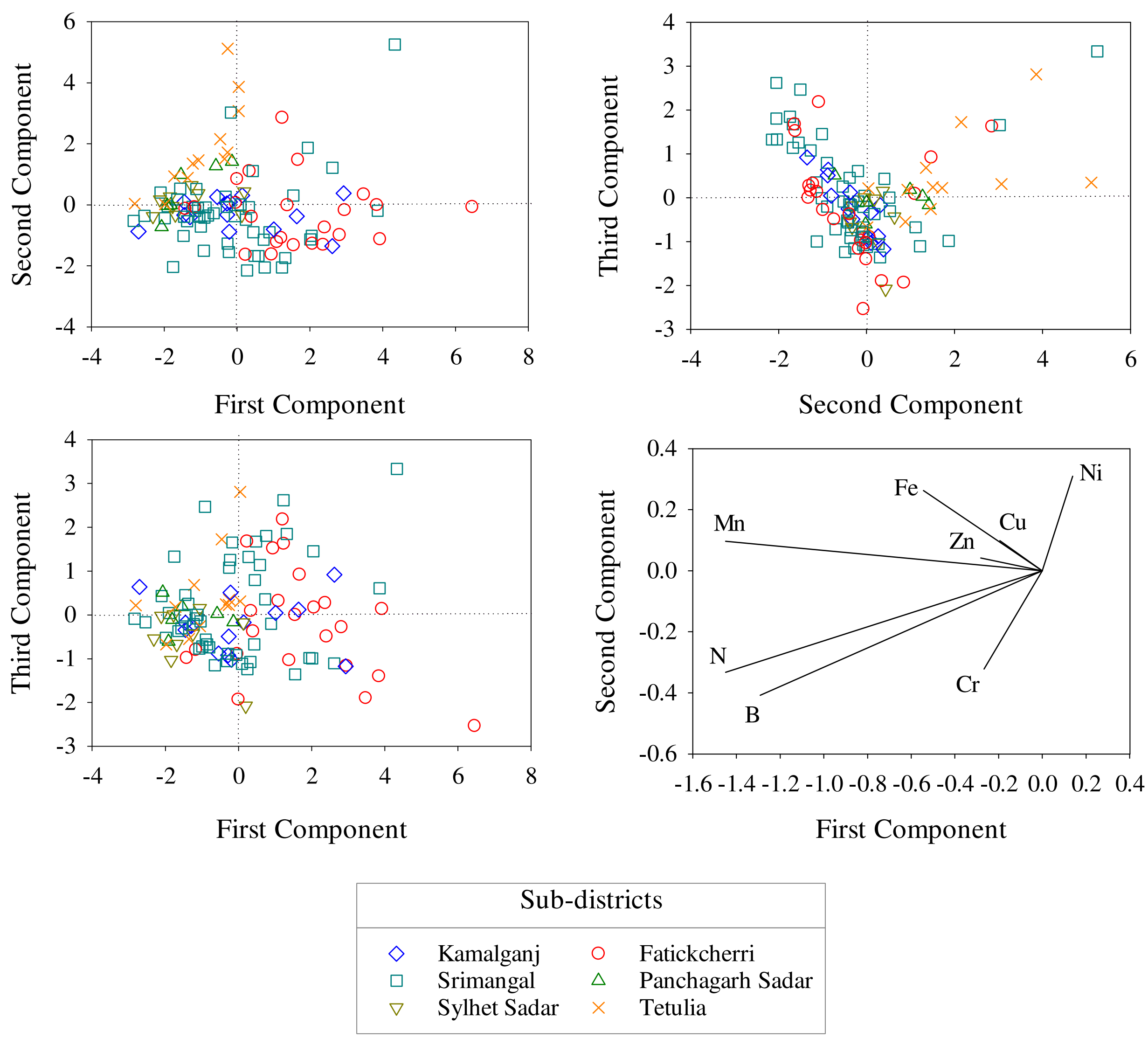


Figure 3.27: PCA of the concentration of the elements in tea garden soils located into different sub-districts of Bangladesh along with loading plots.

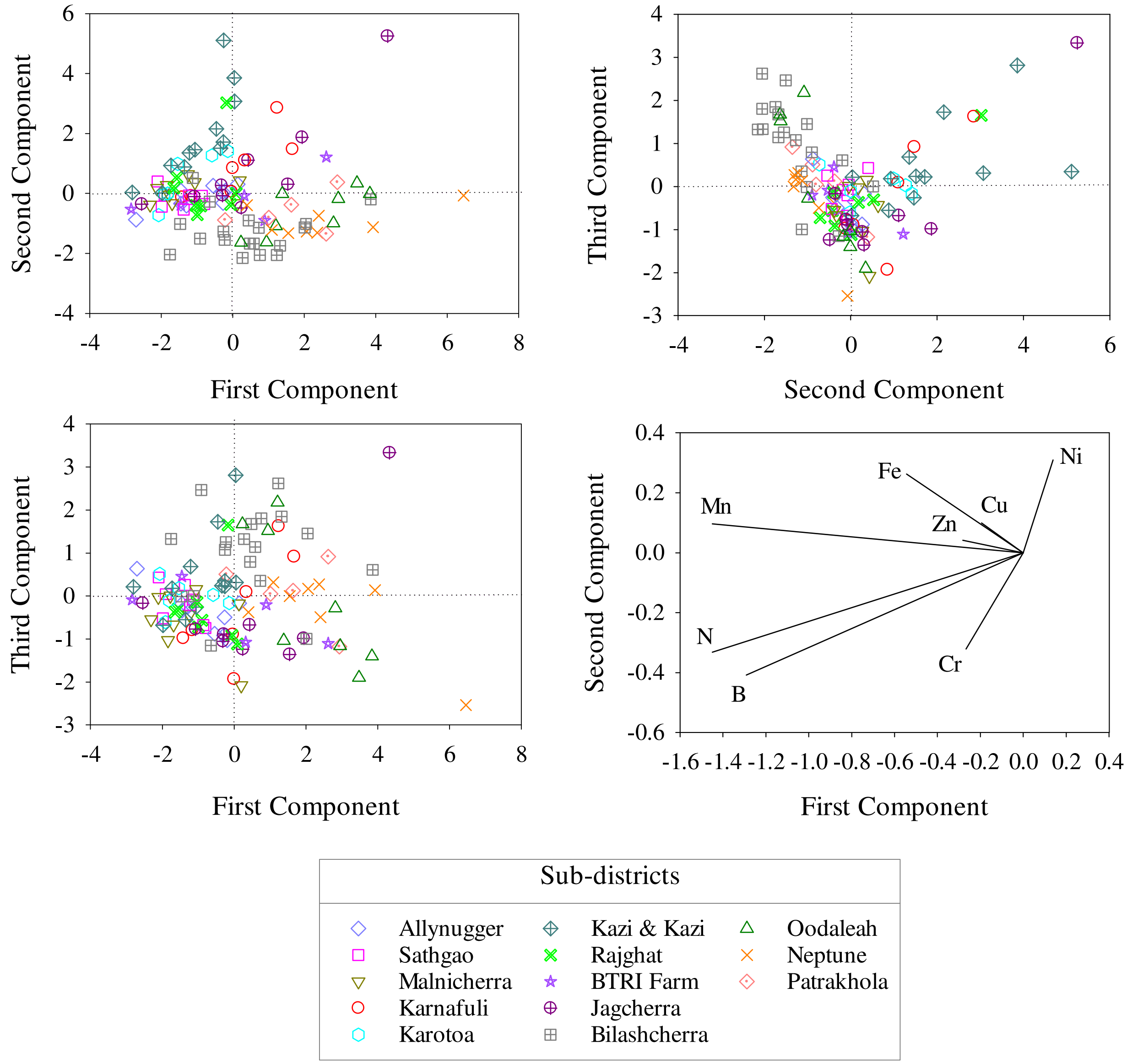


Figure 3.28: PCA of the concentration of the elements in soils of different tea gardens of Bangladesh along with loading plots.

3.5 Conclusion

The mean concentrations of heavy metals in the tea garden soils were found to be, in general, higher than the concentrations in the adjacent non-tea garden soils. The mean concentrations of the heavy metals in the tea garden soils were observed to be in the following order: Fe > Mn > Zn > Cr > Ni > Cu. The highest mean concentrations of chromium, copper, iron and nickel were found in the surface and sub-surface soils of the Karotoa tea garden. The highest mean concentrations of manganese and zinc in the surface soils were observed in the Malnicherra and Jagcherra tea gardens while the highest mean concentrations of manganese and zinc in the sub-surface soils were observed in the Oodaleah and Karotoa tea gardens. Chromium, copper, iron and nickel concentrations were found to be highest in the surface and sub-surface soils of the tea garden located at Panchagarh Sadar sub-district. The highest mean concentrations of manganese and zinc in the surface soils were observed in the tea garden located at Sylhet Sadar sub-district while the highest mean concentrations of manganese and zinc in the sub-surface soils were observed in the tea gardens located at Srimangal and Panchagarh Sadar sub-districts. The mean concentrations of the heavy metals were found within the world-soil average concentrations of the heavy metals in soil. However, the long-term use of large amount of inorganic and organic fertilizers, pesticides and other agrochemicals can potentially increase the concentration of toxic heavy metals in the tea garden soils as well as enhance the mobilization of toxic metals within the soil-plant-environment system. Therefore, further studies and periodic monitoring are required to investigate the origin, extent and bioavailability of toxic heavy metals in the tea garden soils of Bangladesh as well as to assess its possible effects on the soil-plant-environment systems.

Chapter Four

Geomorphological Variabilities in Geochemical Elements in the Tea Garden Soils of Bangladesh

Abstract

While the geomorphologically different soils of Bangladesh are inherently variable in their geochemical fingerprints, it is important to understand the geochemical differences in the geomorphologically different tea garden soils of Bangladesh to better appreciate the underlying geochemistry of elements particularly heavy metals as they are potentially toxic and persistent in the natural environments. Addressing these important issues, A total of 132 surface and 60 sub-surface soil samples were collected from different tea gardens across Bangladesh and were analysed for a range of soil properties including heavy metals (chromium, copper, iron, manganese, nickel and zinc) to assess their variabilities with respect to physiographic regions, land topographic features such as landforms, soil texture, drainage class, soil series as well as the age of tea plants. The concentration of chromium, copper, iron and nickel in the surface soils (0-23 cm) of the tea gardens were found to be significantly different (p < 0.001) among the physiographic regions (Old Himalayan Piedmont Plain, Northern and Eastern Hills and Northern and Eastern Piedmont Plains). The concentration of chromium in the surface soil varied significantly between the different age groups of the tea plantation (${}^{ANOVA}F = 3.27$, $p < 0.05$). The concentration of chromium, copper, iron, nickel and zinc in the surface soils of the tea gardens were found to be significantly different among the soil series ($p < 0.001$ for chromium, iron and nickel, $p < 0.01$ for copper and zinc). In the surface soils of the tea gardens, the concentration of chromium, copper, iron and nickel were found to be significantly different ($p < 0.001$) among the landforms. Chromium, iron and manganese in the surface soils varied significantly among the soils with different drainage classes ($p < 0.001$ for chromium and iron, $p < 0.05$ for manganese). The concentration of iron, manganese, nickel and zinc in the surface soils of the tea gardens were observed to be significantly different among the soil textural classes ($p < 0.001$ for zinc in surface soil, $p < 0.01$ for manganese, $p < 0.05$ for iron and nickel). pH, organic matter, organic carbon, total nitrogen and boron in the tea garden soils also varied significantly among the physiography, soil series, landforms and drainage classes. The present study reveals that the tea garden soils of Bangladesh with different geomorphological characteristics have differences in their geochemical constituents.

Key words: Geomorphology, physiographic regions, heavy metals, topography, soil series.

4.1 Introduction

Bangladesh is a small, flat country, which has a complex geology and geomorphology (Brammer, 2012). It is located at the head of the Bay of Bengal, and occupies most of the Bengal Basin, one of the largest sedimentary basins in the world. The basin is surrounded by the Himalayas and Shillong plateau (uplifted block of Precambrian shield) to the north, the Indo-Burma ranges to the east, and the Indian platform (Indian shield and Himalayan foredeep) to the west (Brammer, 2012). The huge amount of sediments that fluxes from the Himalayan and Indo-Burman mountain ranges are transported by the Ganges-Brahmaputra-Meghna (GBM) river systems (Ahmed *et al*., 2004; Shamsudduha *et al*., 2011), and converge at the lower reaches to form the great delta complex, the GBM Delta that covers almost the entire Bangladesh.

An understanding of the complex geological and geomorphological structure of the landscape in Bangladesh is important to understand the nature and properties of its soils/ sediments (Brammer, 2012). There are three major geological formations in Bangladesh: (i) Tertiary hill sediments in the Northern and Eastern Hills, (ii) Madhupur clay of the Madhupur Tract and the Barind Tract in the central and west, and (iii) recent alluvium in the floodplain and estuarine areas occupying the remainder of the country (Brammer, 1996; Huq and Shoaib, 2013).

Bangladesh has 3 major geomorphological units which are related to the parent geological formations (Brammer, 1996; Huq and Shoaib, 2013). The geomorphological units are: *hills* that occupy 12 percent and include the Northern and Eastern Hills, *terraces* that occupy 8 percent and include mainly the Madhupur Tract and the Barind Tract, and *floodplains* that occupy 80 percent of the country's land area and include piedmont plains (Old Himalayan Piedmont Plain, and Northern and Eastern Piedmont Plains), river floodplains (meander floodplains, active floodplains, old floodplains, and basins), tidal floodplain, and estuarine floodplain (Brammer, 1996; Brammer, 2012; Huq and Shoaib, 2013). Apart from the eastern hills and adjoining piedmont plains, Bangladesh slopes gently south-eastward from near the foot of the Himalayas in the north-west towards the Meghna estuary in the south-centre (Brammer, 2012). The Madhupur Tract and the Barind Tract, the uplifted terraces, are of Pleistocene age, and are therefore known as Pleistocene terraces. The floodplains of Bangladesh are of Holocene age, and are therefore known as Holocene floodplains. These geomorphological units are characterized by land topography, soil formation through sediment deposition over time and also related to the parent geological formations (Brammer, 1996).

Based on the parent material in which individual soil types were formed, and the landscape on which the soils were developed, Bangladesh has been divided into 20 main physiographic regions, some of which have a number of sub-regions bringing the total to 34 (FAO-UNDP, 1988). The physiography of the individual landscapes (that is, the physiographic regions) is particularly important to understand and characterize the landforms and landscapes of the geomorphological areas of Bangladesh. Among the 20 main physiographic regions, the 3 physiographic regions from where the soil samples were collected are Northern and Eastern Hills, Northern and Eastern Piedmont Plains and Old Himalayan Piedmont Plain. A brief description of these three physiographic regions were presented in chapter one (Literature Review). This physiographic classification was based on the parent material and the landscape on which individual soils were developed and soil types were formed (FAO-UNDP, 1988). Therefore, geology, topography, drainage, age of land formation and pattern of sedimentary deposition of the physiographic regions has differences which ultimately influence the nature and properties of the soils in the different physiographic regions.

In Bangladesh, the land levels in relation to the normal depth and duration of seasonal flooding (known as the inundation/ depth-of-flooding land types) have been classified into highland (above flood level), medium highland-1 (normal flooding depth is up to 30 cm), medium highland-2 (normal flooding depth is 30 - 90 cm), medium lowland (normal flooding depth is 90 - 180 cm), lowland (normal flooding depth is 180 - 300 cm), very lowland (normal flooding depth is over 300 cm), and bottomland (depression sites which remain wet throughout the year) (FAO-UNDP, 1988; Brammer, 2012). The lands under tea plantations in Bangladesh are categorized as high land. However, the topographic features of the tea growing lands can also be characterized by different landforms such as level ridge, valley ridge, terrace, low hill and medium high hill (SRDI, 2013). Due to the gradients at the hills, the soils have different drainage conditions such as imperfectly drained, moderately well drained, well drained and excessively drained (SRDI, 2013). The soil series is the lowest category of the USDA Soil Taxonomy. A soil series is a group of soils that have formed in the same way from the same kind of parent material under similar conditions of climate, vegetation, drainage, and time. Bangladesh has 465 soil series, which are inherently different in their nature.

Spatial occurrence and accumulation of the elements as well as their mobility and availability in soils are largely regulated by the topography of the landscape (Du Laing *et al.*, 2009). Besides the inputs from anthropogenic activities, total metal concentrations in agricultural soils also reflect the soil's geological origin and mineral weathering. The soils of Bangladesh are naturally variable in their geochemical fingerprints (Chowdhury *et al.*, 2017). In the geomorphologically different soils of Bangladesh enrichment and variabilities of a range of geochemical elements have been found across the landscape of the country which may perhaps be explained by the differences in sedimentary depositional environments (Chowdhury *et al.*, 2017). It also reformed the regional topological features, drainage conditions as well as soil types (Brammer, 2012). In the tea garden soil environment in Bangladesh, the complex interactions between soil properties, climate, agricultural management practices and land topographic variations can enhance variability in the inherent soil geochemical relationships. While these geomorphological attributes of soils regulate the geochemical cycling of elements in the soils and thus the accumulations of the elements into cultivated crops, it is important to better understand the geochemical variability in the different tea garden soils of Bangladesh.

In the present study, surface (0 – 23 cm depth, n = 132) and sub-surface (23 – 46 cm depth, n = 60) soils from different tea gardens of Bangladesh were collected and analysed for a range of geochemical elements such as nitrogen, boron, chromium, copper, iron, manganese, nickel and zinc to assess their variabilities with respect to physiographic regions; land topographic features such as landforms; soil texture; drainage class; soil series as well as the age of tea plants. The present study is so far the first to assess the geomorphological variability of heavy metals in the tea garden soils of Bangladesh by understanding their geochemical constituents within the different geomorphological settings.

4.2 Objectives

The objective of the research reported in this chapter is:

1. To assess the variability of the geochemical elements in the geomorphologically different tea garden soils of Bangladesh with respect to physiographic regions, land topographic features such as landforms, soil texture, drainage class, soil series as well as the age of tea plants.

4.3 Materials and Methods

A detail of the *Materials and Methods* for this chapter was described in chapter two (General Methodology).

4.4 Results and Discussion

4.4.1 Variabilities in the soil properties in the tea garden soils of different Physiographic regions of Bangladesh

The pH of the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 10.00$, $p < 0.001$) with the maximum pH in Old Himalayan Piedmont Plain (5.36), followed by Northern and Eastern Piedmont Plains (4.88) and Northern and Eastern Hills (4.76) (Figure 4.1). The pH of the sub-surface soils was also observed to be significantly higher in the soils of Old Himalayan Piedmont Plain (5.52) compared to the other physiographic regions (Figure A11). Akhtaruzzaman *et al.* (2014) reported that pH in the surface soils of Northern and Eastern Hill and Northern and Eastern Piedmont Plain ranged from 5.10 to 5.16 and 5.17 to 5.52, respectively, whereas in the present study, lower pH values were observed in the surface soils of the physiographic regions. As soil pH depends on the nature of parent materials lower soil pH might be as a result of the acidic nature of the parent rock (Zhang *et al.*, 2019). Rainfall is also a great factor to increase soil acidity. The use of nitrogenous fertilizers in the soils could also lower the pH of the soils as the released NH_4^+ ions might undergo oxidation and release protons contributing to declined soil pH (Thangarajan *et al.*, 2015).

Organic carbon contents of the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 13.88$, $p < 0.001$) with the maximum organic carbon content in Old Himalayan Piedmont Plain (2%), followed by Northern and Eastern Hills (1.16%) and Northern and Eastern Piedmont Plains (1.13%) (Figure 4.1). The organic carbon contents in the sub-surface soils were observed to be insignificantly higher in the soils of Old Himalayan Piedmont Plain (1.687%) compared to other physiographic regions. Akhtaruzzaman *et al.* (2014) reported that organic carbon contents in the surface soils of Northern and Eastern Hill and Northern and Eastern Piedmont Plain ranged from 1.13% to 1.54% and 1.61% to 1.73%, respectively, whereas in the present study, the organic carbon contents were observed to be higher in the tea garden soils located at Old Himalayan Piedmont Plain physiography. It might be due to the accumulation of a considerable portion of organic matter on piedmont soils through moving down from the nearby hilly areas (Akhtaruzzaman *et al.*, 2014).

The total nitrogen contents of the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 13.48$, $p < 0.001$) with the maximum total nitrogen content in Old Himalayan Piedmont Plain (0.172%), followed by Northern and Eastern Hills (0.101%) and Northern and Eastern Piedmont Plains (0.098%) (Figure 4.1). The total nitrogen contents in the sub-surface soils were observed to be insignificantly higher in the soils of Old Himalayan Piedmont Plain (0.146%) compared to other physiographic regions. Shil *et al.* (2016) reported that the total nitrogen contents in the soils of Old Himalayan Piedmont Plain and Northern and Eastern Piedmont Plain were ranged from 0.078% to 0.103% and 0.068% to 0.117%, respectively, whereas in the present study, the total nitrogen contents were observed to be comparatively higher in the tea garden soils located at Old Himalayan Piedmont Plain physiography. The total nitrogen content in soil is mostly dependent on the organic matter content of the soil (Venkatesan and Murugesan, 2006). The elevated nitrogen in the tea garden soils could be due to the accumulations of organic matter under tea cultivation practices such as pruning, leaves from shade trees and application of nitrogenous fertilizers (Jahan *et al*., 2022).

The concentrations of boron in the surface soils of the tea garden soils at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 4.77$, $p < 0.01$) with the maximum concentration of boron in Old Himalayan Piedmont Plain (0.81 mg/kg), followed by Northern and Eastern Hills (0.44 mg/kg) and Northern and Eastern Piedmont Plains (0.37 mg/kg) (Figure 4.1). The concentrations of boron in the sub-surface soils were observed to be insignificantly higher in the soils of Northern and Eastern Hills (0.95 mg/kg) compared to other physiographic regions. Hasan *et al.* (2020) classified the boron status in agricultural soils of Bangladesh as very low (0.15 μg/gm), low (0.151 – 0.30 μg/gm), medium (0.31 – 0.45 μg/gm), optimum (0.451 – 0.6 μg/gm), high (0.61 – 0.75 μg/gm) and very high (>0.75 μg/gm). In the present study, the average boron content in the surface soils of Old Himalayan Piedmont Plain can be classified as very high and Northern and Eastern Hill and Northern and Eastern Piedmont Plain can be classified as medium. Water evapotranspiration is one of the main reasons for boron accumulation in the top soil (Das and Purkait, 2020).

The concentrations of chromium in the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 27.23$, $p < 0.001$) with the maximum concentration of chromium in Old Himalayan Piedmont Plain (42.74 mg/kg), followed by Northern and Eastern Hills (21.54 mg/kg) and Northern and Eastern Piedmont Plains (17.5 mg/kg) (Figure 4.1). The concentrations of chromium in the sub-surface soils were also observed to be significantly higher in the soils of Old Himalayan Piedmont Plain (49.86 mg/kg) compared to other physiographic regions (Figure A12). Ali *et al*. (2003) reported that the concentrations of chromium in soils ranged from 61.0 to 176.0 mg/kg (mean, 95.9 mg/kg) at different physiography of Bangladesh, where the soils of the Old Himalayan Piedmont Plain and Northern and Eastern Piedmont Plain had 66.4 to 70.2 mg/kg (mean, 68.3 mg/kg) and 80.6 to 113.8 mg/kg (mean, 97.0 mg/kg) chromium, respectively. Concentrations of chromium in the agricultural soils collected from 10 different physiographic regions of Bangladesh were ranged from 17.79 to 727 mg/kg (mean, 72.36 mg/kg) were observed by Chowdhury *et al.* (2017) and Chowdhury *et al.* (2021), whereas in the present study, the mean concentration of chromium found in tea garden soils located at three different physiographic regions were lower than the concentration reported in the previous survey on agricultural soils of Bangladesh.

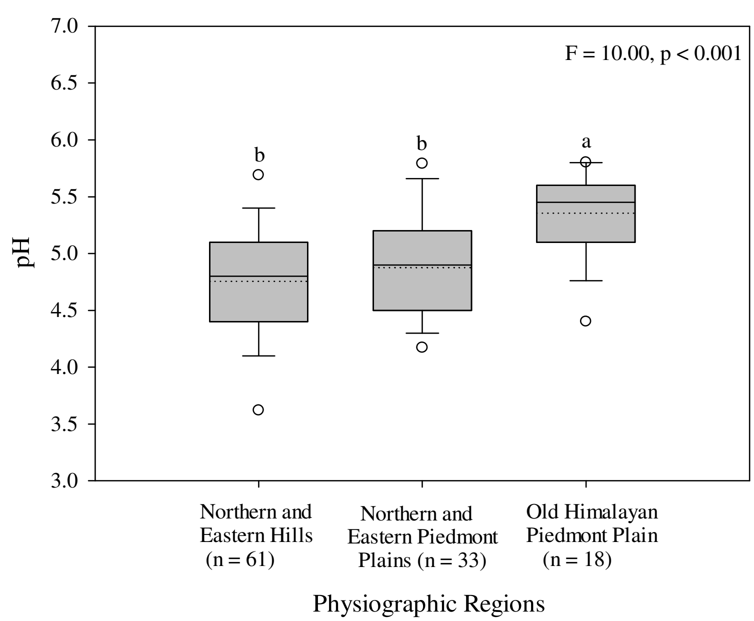
The concentrations of copper in the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 8.93$, $p < 0.001$) with the maximum concentration of copper in Old Himalayan Piedmont Plain (12.64 mg/kg), followed by Northern and Eastern Hills (8.62 mg/kg) and Northern and Eastern Piedmont Plains (8.49 mg/kg) (Figure 4.1). The concentrations of copper in the sub-surface soils were also observed to be significantly higher in the soils of Old Himalayan Piedmont Plain (12.75 mg/kg) compared to other physiographic regions (Figure A13). Jahiruddin *et al*. (2000) reported that the concentrations of copper ranged from 9.40 to 42.9 mg/kg (mean, 22.4 mg/kg) in agricultural soils of Bangladesh. Ali *et al*. (2003) reported that the concentrations of copper ranged from 8.7 to 65.7 mg/kg (mean, 31.6 mg/kg) in the soils of Bangladesh. Ali *et al*. (2003) also observed that the concentration of copper in the soils of Old Himalayan Piedmont Plain and Northern and Eastern Piedmont Plain ranged from 8.7 to 17.5 mg/kg (mean, 13.1 mg/kg) and 9.0 to 30.3 mg/kg (mean, 17.3 mg/kg), respectively. Concentrations of copper in the agricultural soils collected from 10 different physiographic regions of Bangladesh were ranged from 5.46 to 68.56 mg/kg (mean, 28.97 mg/kg) were observed by Chowdhury *et al.* (2017) and Chowdhury *et al.* (2021), whereas in the present study, the mean concentration of copper found in tea garden soils located at three different physiographic regions were lower than the concentration reported in the previous survey on agricultural soils of Bangladesh. It might be due to uptake and accumulation of copper at different concentration in tea plant (Dey *et al*., 2014).

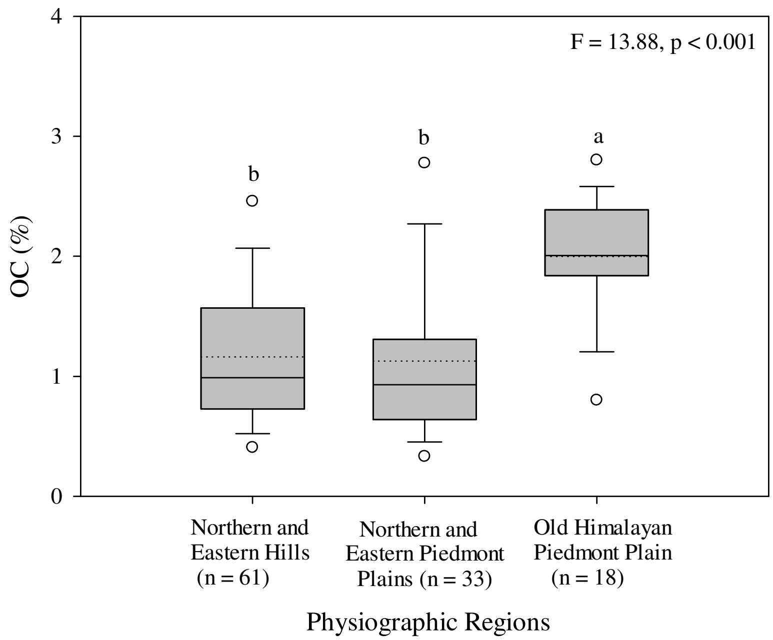
The concentrations of iron in the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 27.49$, $p < 0.001$) with the maximum concentration of iron in Old Himalayan Piedmont Plain (25882 mg/kg), followed by Northern and Eastern Hills (16036 mg/kg) and Northern and Eastern Piedmont Plains (13418 mg/kg) (Figure 4.1). The concentrations of iron in the sub-surface soils were also observed to be significantly higher in the soils of Old Himalayan Piedmont Plain (30353 mg/kg) compared to other physiographic regions (Figure A14). Sanaullah and Akhtaruzzaman (2020) reported that the concentrations of iron in the surface and sub-surface of agricultural soils were 9.83% i.e 98300 mg/kg and 10.50% i.e 105000 mg/kg, respectively. Concentrations of iron in the agricultural soils collected from 10 different physiographic regions of Bangladesh were ranged from 793 to 50,802 mg/kg (mean, 26145 mg/kg) were observed by Chowdhury *et al.* (2017) and Chowdhury *et al.* (2021), whereas in the present study, the mean concentration of iron found in tea garden soils located at three different physiographic regions were lower than the concentration reported in the previous survey on agricultural soils of Bangladesh.

The concentrations of manganese in the surface soils of the tea gardens at different physiographic regions were found to be varied insignificantly with the maximum concentration of manganese in Northern and Eastern Hills (233 mg/kg), followed by Northern and Eastern Piedmont Plains (227 mg/kg) and Old Himalayan Piedmont Plain (156 mg/kg) (Figure 4.1). The concentrations of manganese in the sub-surface soils were also observed to be insignificantly higher in the soils of Northern and Eastern Piedmont Plains (216 mg/kg) compared to other physiographic regions. Jahiruddin *et al.* (2000) reported that the concentrations of manganese in agricultural soils of Bangladesh ranged from 346 to 618 mg/kg (mean, 444 mg/kg). Sanaullah and Akhtaruzzaman (2020) reported that the concentrations of manganese in the surface and sub-surface agricultural soils were 781 and 797 mg/kg, respectively. Concentrations of manganese in the agricultural soils collected from 10 different physiographic regions of Bangladesh were ranged from 43.04 to 1198 mg/kg (mean, 370 mg/kg) were observed by Chowdhury *et al.* (2017) and Chowdhury *et al.* (2021), whereas in the present study, the mean concentration of manganese found in tea garden soils located at three different physiographic regions were lower than the concentration reported in the previous survey on agricultural soils of Bangladesh.

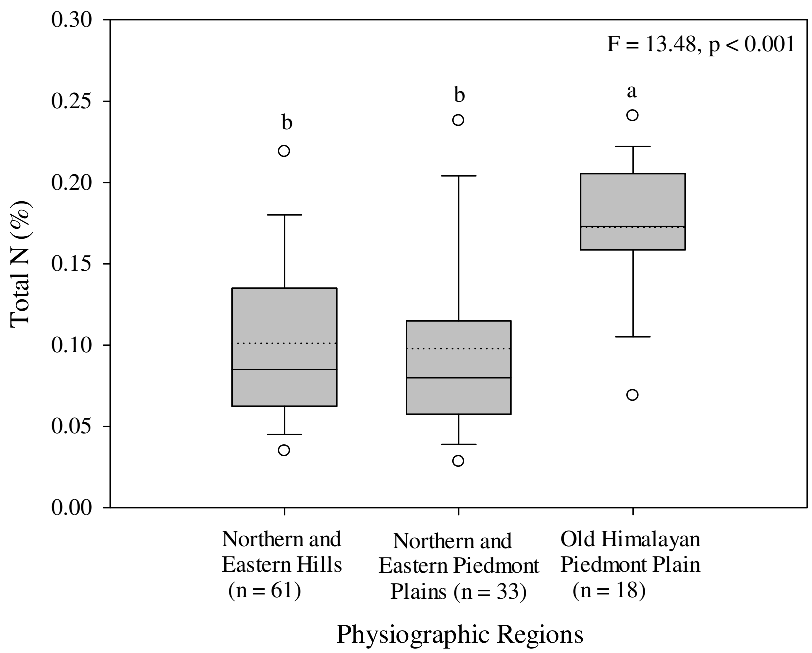
The concentrations of nickel in the surface soils of the tea gardens at different physiographic regions were found to be varied significantly ($^{ANOVA}F = 10.66$, $p < 0.001$) with the maximum concentration of nickel in Old Himalayan Piedmont Plain (13.06 mg/kg), followed by Northern and Eastern Hills (9.06 mg/kg) and Northern and Eastern Piedmont Plains (7.73 mg/kg) (Figure 4.1). The concentrations of nickel in the sub-surface soils were also observed to be significantly higher in the soils of Old Himalayan Piedmont Plain (14.47 mg/kg) compared to other physiographic regions (Figure A15). Ali *et al.* (2003) reported that the concentration of nickel in the soils of Bangladesh ranged from 14.0 to 134.0 mg/kg (mean, 51.0 mg/kg). Ali *et al*. (2003) observed that the concentrations of nickel in the soils of Old Himalayan Piedmont Plain and Northern and Eastern Piedmont Plain ranged from 14.0 to 28.9 mg/kg (mean, 21.5 mg/kg) and 16.8 to 69.6 mg/kg (mean, 42.7 mg/kg), respectively. Concentrations of nickel in the agricultural soils collected from 10 different physiographic regions of Bangladesh were ranged from 7.43 to 92.57 mg/kg (mean, 37.06 mg/kg) were observed by Chowdhury *et al.* (2017) and Chowdhury *et al.* (2021), whereas in the present study, the mean concentration of nickel found in tea garden soils located at three different physiographic regions were lower than the concentration reported in the previous survey on agricultural soils of Bangladesh.

The concentrations of zinc in the surface soils of the tea gardens at different physiographic regions were found to be varied insignificantly with the maximum concentration of zinc in Northern and Eastern Hills (50.74 mg/kg), followed by Northern and Eastern Piedmont Plains (46.65 mg/kg) and Old Himalayan Piedmont Plain (42.07 mg/kg) (Figure 4.1). The concentrations of zinc in the sub-surface soils were observed to be significantly higher in the soils of Old Himalayan Piedmont Plain (43.23 mg/kg) compared to other physiographic regions (Figure A16). Jahiruddin *et al.* (2000) reported that the concentrations of zinc in agricultural soils of Bangladesh ranged from 45.2 to 93.8 mg/kg (mean, 66.4 mg/kg). Ali *et al.* (2003) reported that the concentrations of zinc ranged from 27.8 to 138.0 mg/kg (mean, 75.6 mg/kg) in the soils of Bangladesh. Ali *et al*. (2003) observed that the concentrations of zinc in the soils of Old Himalayan Piedmont Plain and Northern and Eastern Piedmont Plain ranged from 30.6 to 67.4 mg/kg (mean, 49.0 mg/kg) and 32.6 to 89.9 mg/kg (mean, 55.0 mg/kg), respectively. The concentrations of zinc in the surface and sub-surface agricultural soils were observed 117.50 and 119 mg/kg, respectively (Sanaullah and Akhtaruzzaman, 2020). Concentrations of zinc in the agricultural soils collected from 10 different physiographic regions of Bangladesh were ranged from 11.13 to 181 mg/kg (mean, 64.09 mg/kg) as observed by Chowdhury *et al.* (2017) and Chowdhury *et al.* (2021), whereas in the present study, the mean concentration of zinc found in tea garden soils located at three different physiographic regions were lower than the concentration reported in the previous survey on agricultural soils of Bangladesh. The concentrations of chromium, copper, iron, and nickel in the tea garden soils of the Old Himalayan Piedmont Plain were found to be higher than that in the other two physiographic regions. The soils of the physiographic regions of Bangladesh are generally variable in their inherent geochemical fingerprints (Chowdhury *et al.*, 2017). The parent material as well as the topography of a landscape also regulates the spatial occurrence and accumulation of heavy metals in soils (Du Laing *et al.*, 2009). The topography of the tea gardens at the Old Himalayan Piedmont Plain was level ridge, which was comparatively lower than the topography of the landscapes in the other two physiographic regions. Moreover, among the two tea gardens at the Old Himalayan Piedmont Plain, the Kazi & Kazi tea garden practiced a complete organic farming approach (Sultana *et al.*, 2014), which was perhaps the reason behind the elevated amounts of the heavy metals. Different types of organic fertilizers have been reported to increase the inputs of toxic heavy metals in agricultural soils (Yari *et al.*, 2017)

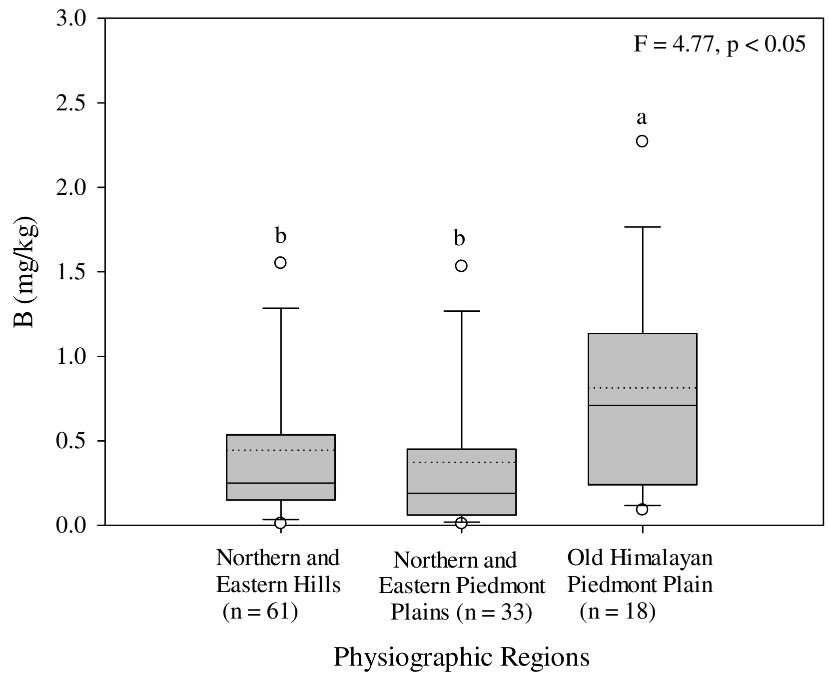




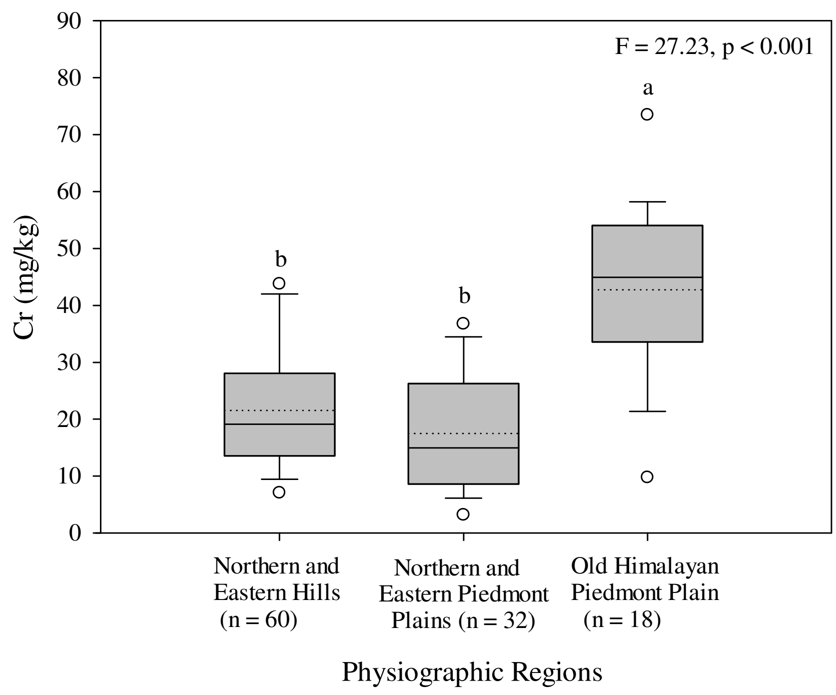
F = 13.88, p < 0.001
OC (%)
0
1
2
3
4
b
b
a
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions



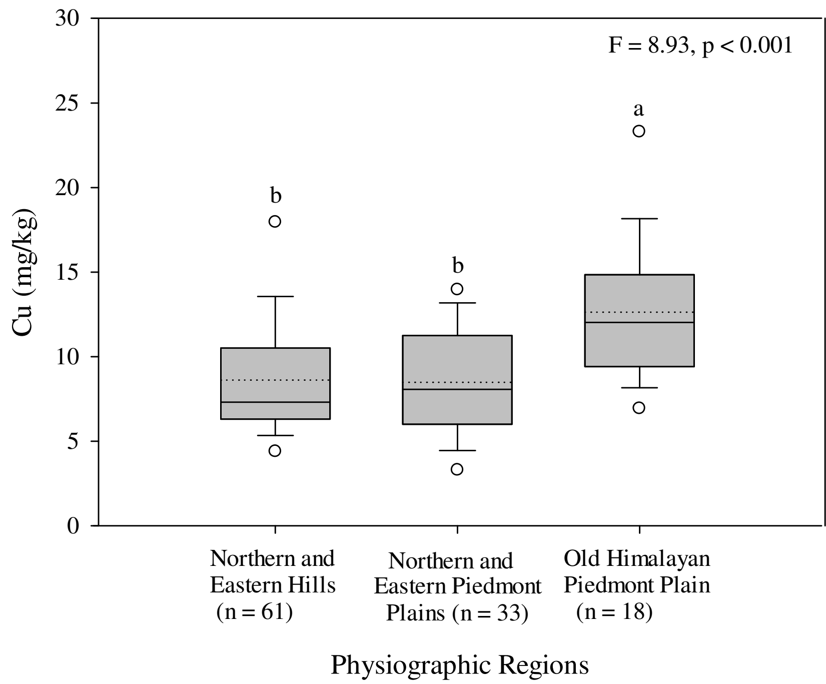
F = 13.48, p < 0.001
Total N (%)
0.00
0.05
0.10
0.15
0.20
0.25
0.30
b
b
a
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions

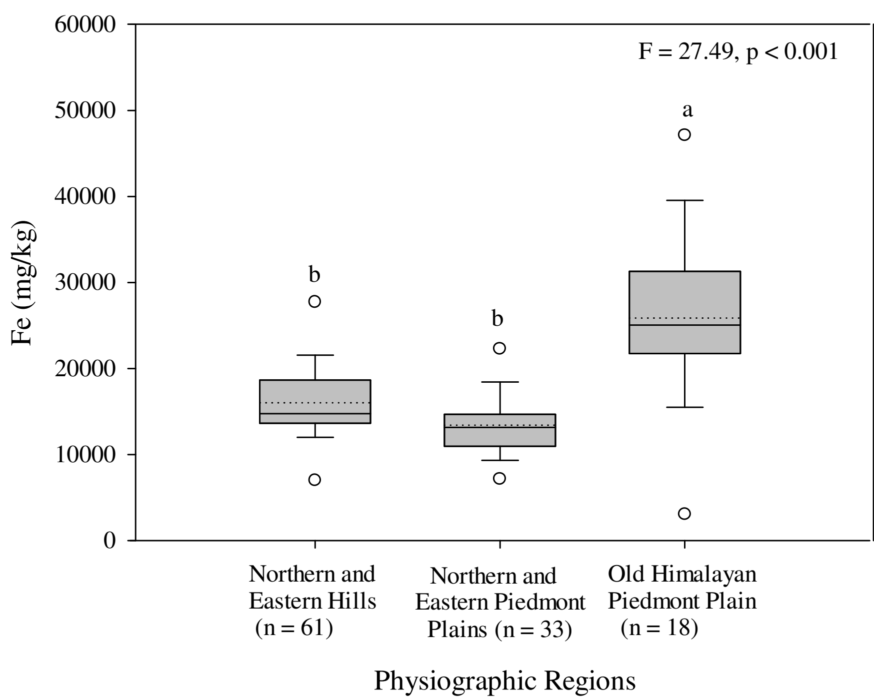


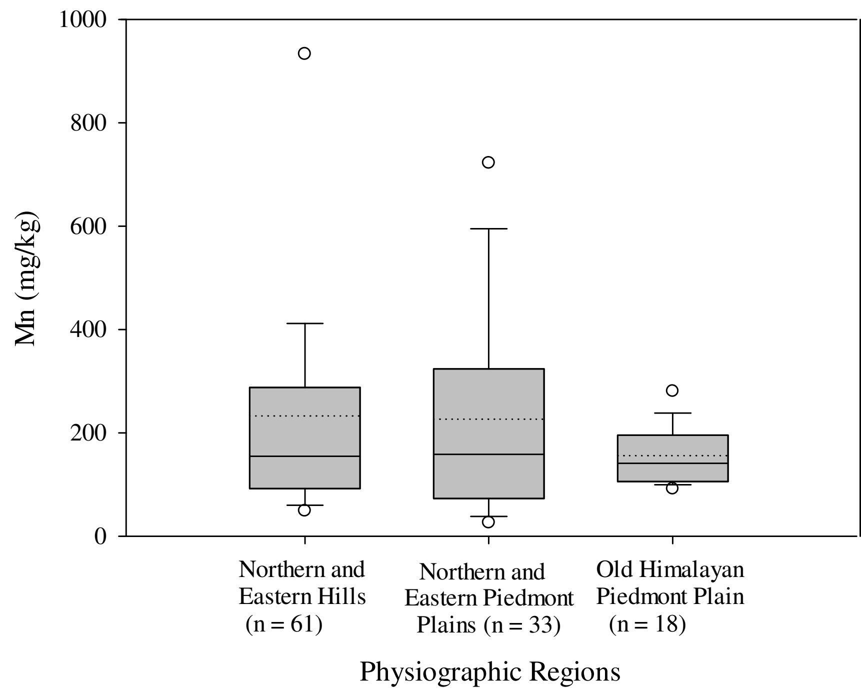
F = 4.77, p < 0.05
B (mg/kg)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
a
b
b
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions



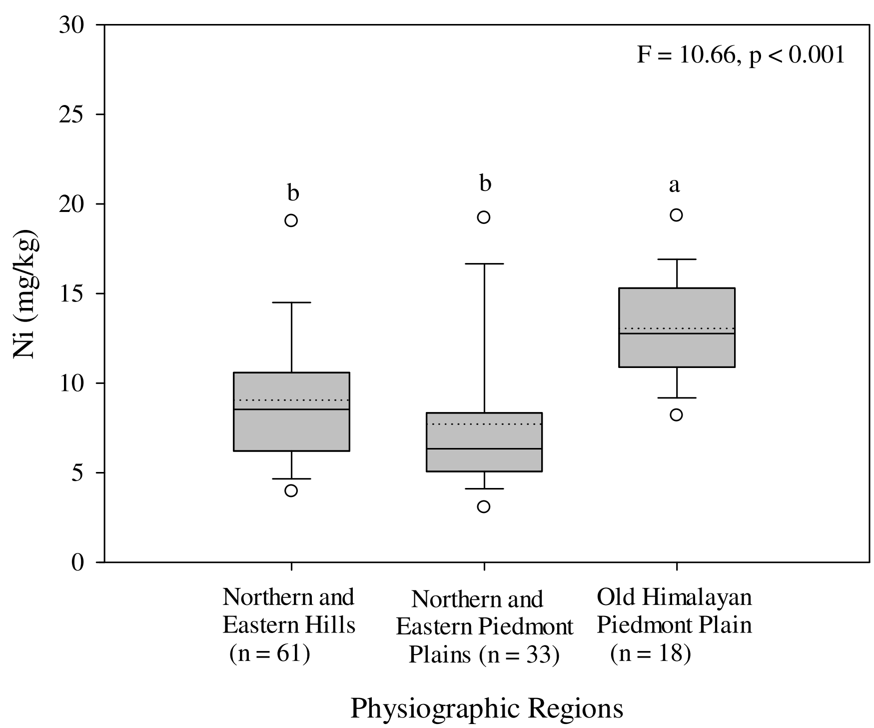
F = 27.23, p < 0.001
Cr (mg/kg)
90
80
70
60
50
40
30
20
10
0
a
b
b
Northern and Eastern Hills (n = 60)
Northern and Eastern Piedmont Plains (n = 32)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions


F = 8.93, p < 0.001
Cu (mg/kg)
30
25
20
15
10
5
0
a
b
b
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions


F = 27.49, p < 0.001
Fe (mg/kg)
60000
50000
40000
30000
20000
10000
0
a
b
b
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions



1000
800
600
400
200
0
Mn (mg/kg)
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions



30
25
20
15
10
5
0
Ni (mg/kg)
F = 10.66, p < 0.001
b
b
a
Northern and Eastern Hills (n = 61)
Northern and Eastern Piedmont Plains (n = 33)
Old Himalayan Piedmont Plain (n = 18)
Physiographic Regions

2016).

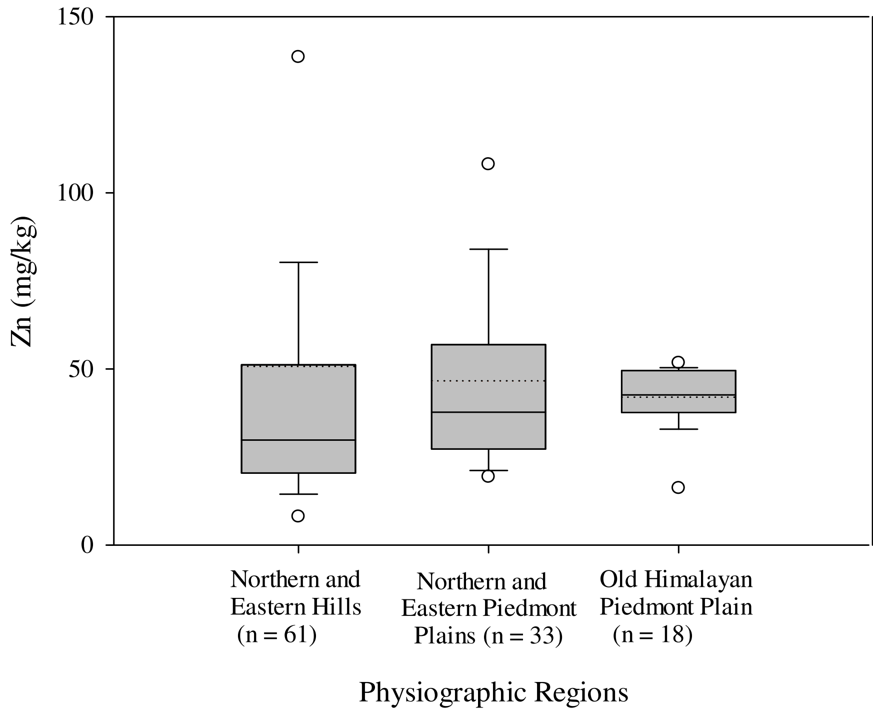


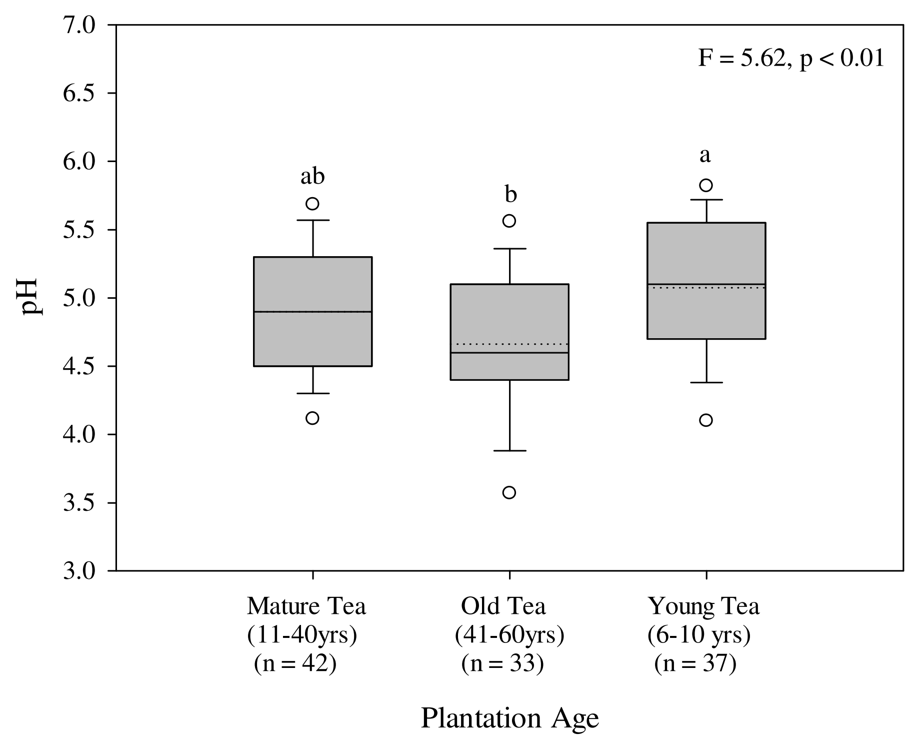
Figure 4.1: Box and whisker plots showing concentrations of different elements in tea garden soils located at different physiographic regions. The boxplots indicate the lower and upper quartile (box), the median (solid line), the mean (dotted line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and *p*-value represent the F-ratio that indicates the degree of variation among the physiographic regions and the level of significance of this variation, respectively, from one-way analysis of variance (ANOVA) test.

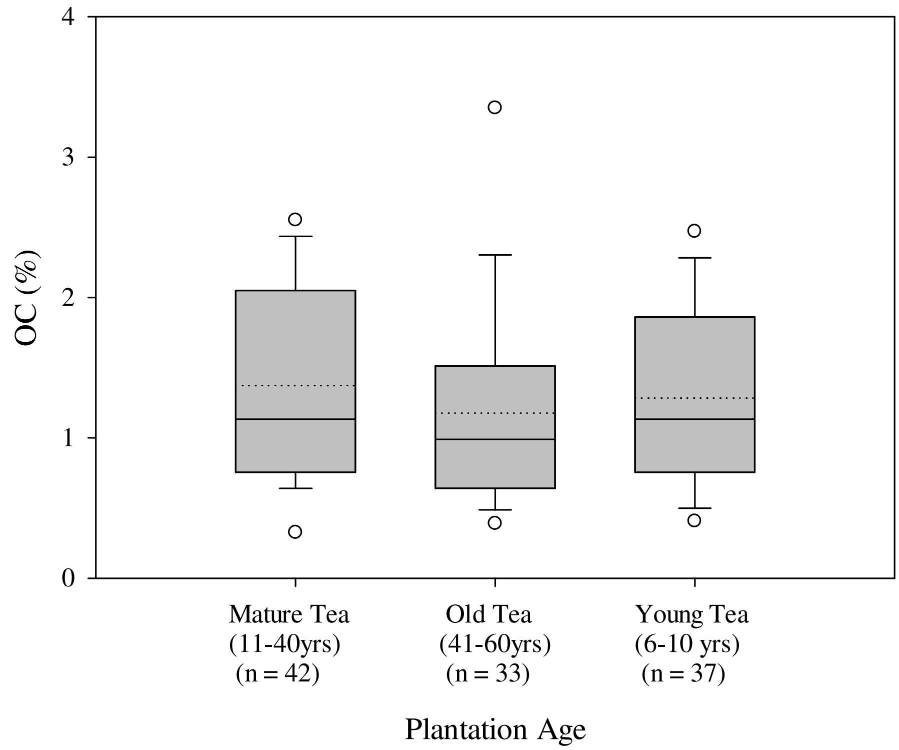
4.4.2 Variabilities in soil properties under different ages of tea plantation

The soils of the tea gardens were grouped based on the age of the tea plantation such as young tea, mature tea and old tea. The pH in the surface soils under tea plantations of different ages were found to be varied significantly ($^{ANOVA}F = 5.62$, $p < 0.01$) with the maximum pH in young tea plantations (5.08), followed by mature tea (4.90) and old tea plantations (4.66) (Figure 4.2). The pH in the sub-surface soils were also observed to be significantly higher in the soils of young tea (5.39) compared to other tea plantations (Figure A17). In the present study, soil pH was gradually decreased with the age of tea plantations. It might be due to the acidification of soil by tea plant itself and also by the application of urea fertilizer (Li *et al*., 2016).

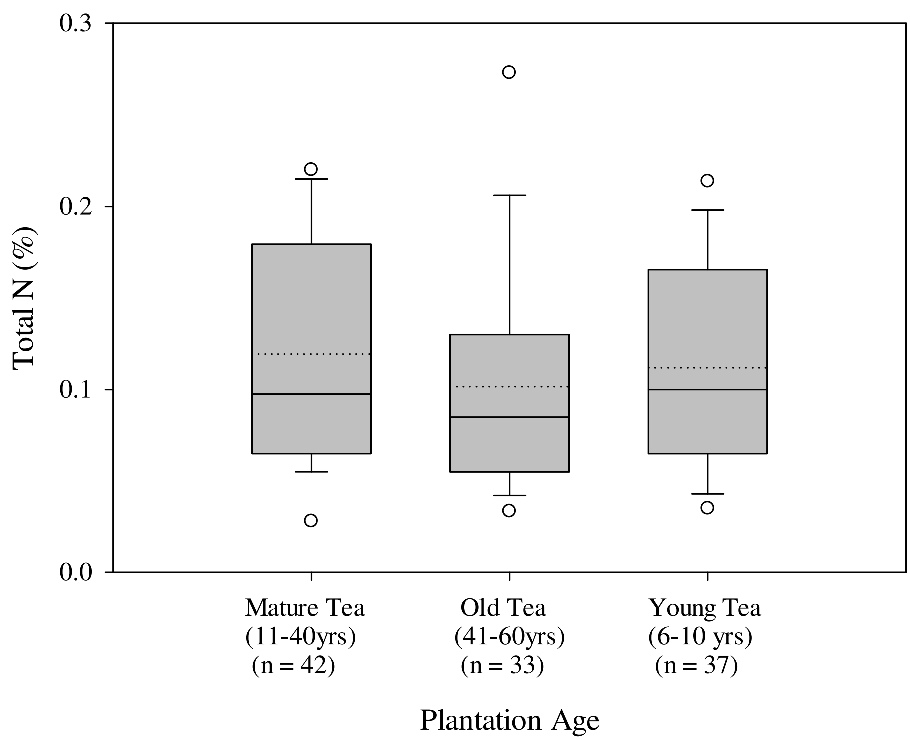
The organic carbon, total nitrogen, boron, copper, iron, manganese, nickel and zinc concentrations in the surface and sub-surface soils of tea gardens were found to be varied insignificantly among the different ages of tea plants (Figure 4.2).

The concentrations of chromium in the surface soils under tea plantations of different ages were found to be varied significantly ($^{ANOVA}F$ = 3.27, $p < 0.05$) with the maximum concentration of chromium in young tea plantations (27.29 mg/kg), followed by mature tea (24.90 mg/kg) and old tea plantations (18.55 mg/kg) (Figure 4.2). Spatial occurrence and accumulation of elements as well as their mobility and availability in soils are largely regulated by the topography of the landscape (Du Laing *et al.,* 2009). Chromium is highly mobile in soil and soil properties such as pH, organic matter and clay minerals can influence the geochemical behaviors of chromium (Sun *et al*., 2022). The lower concentrations of chromium in the soils under old tea plantations could be limited by the lower pH of the soils, which perhaps rendered chromium to be more labile in the soil solution and ultimately to be lost to the environment through leaching, and/or with downslope movement with fine-grain sediments and organic matter (Du Laing *et al*., 2009).

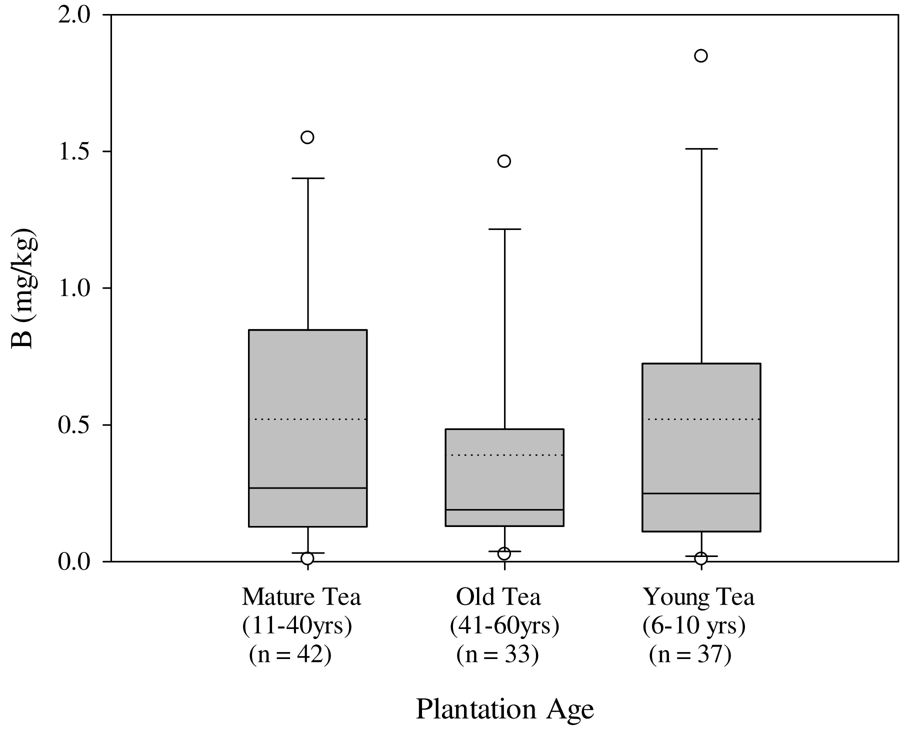


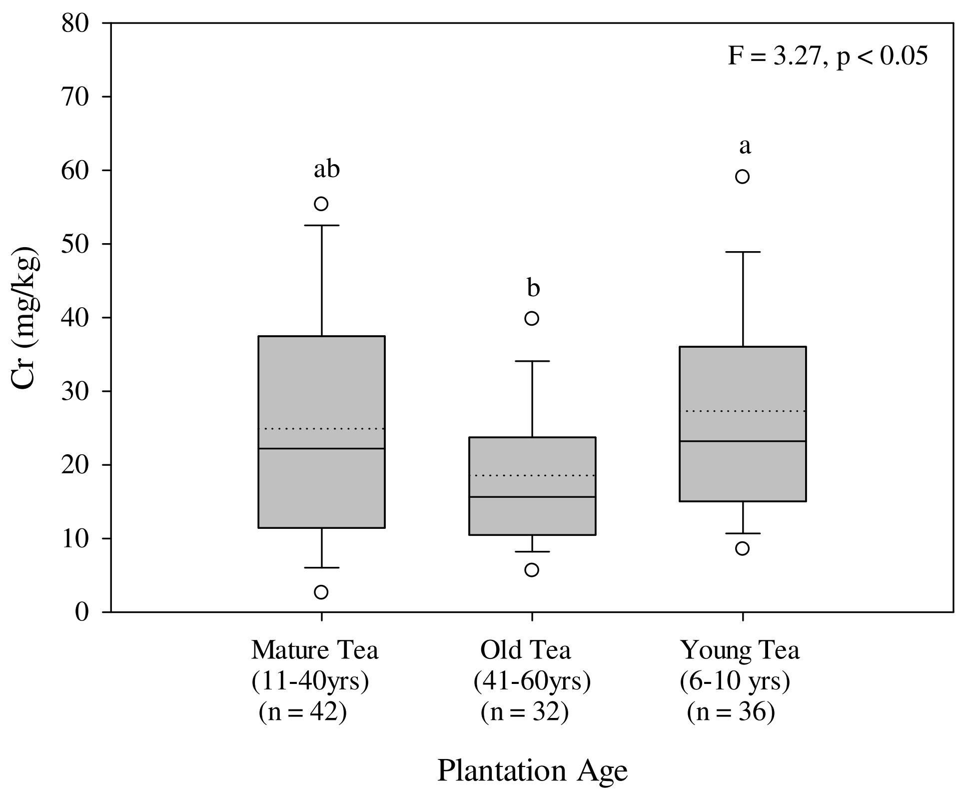


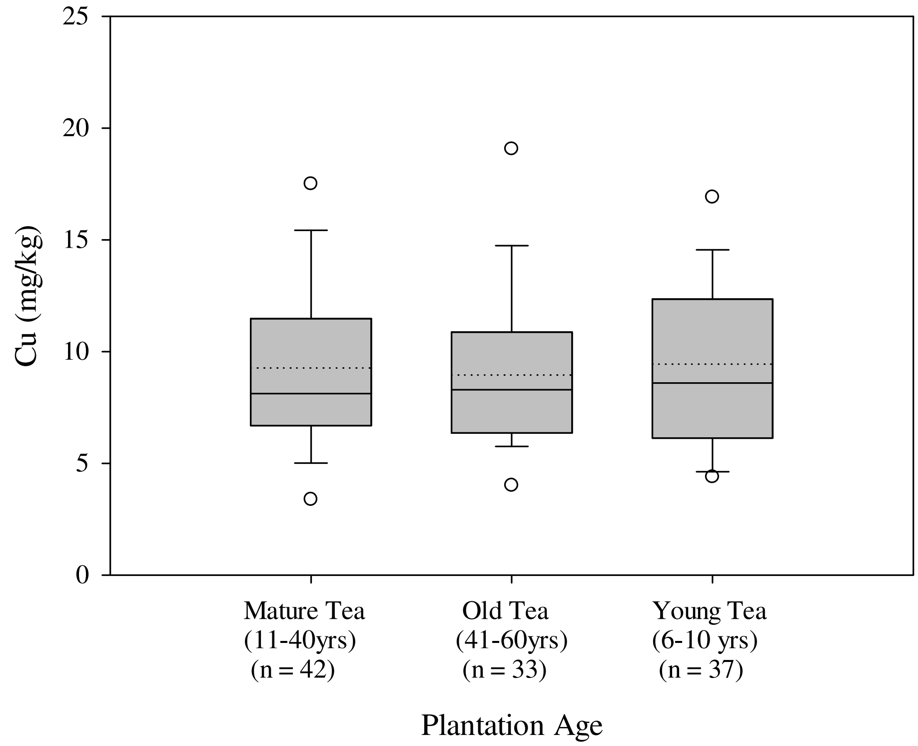
4
3
2
1
0
OC (%)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age

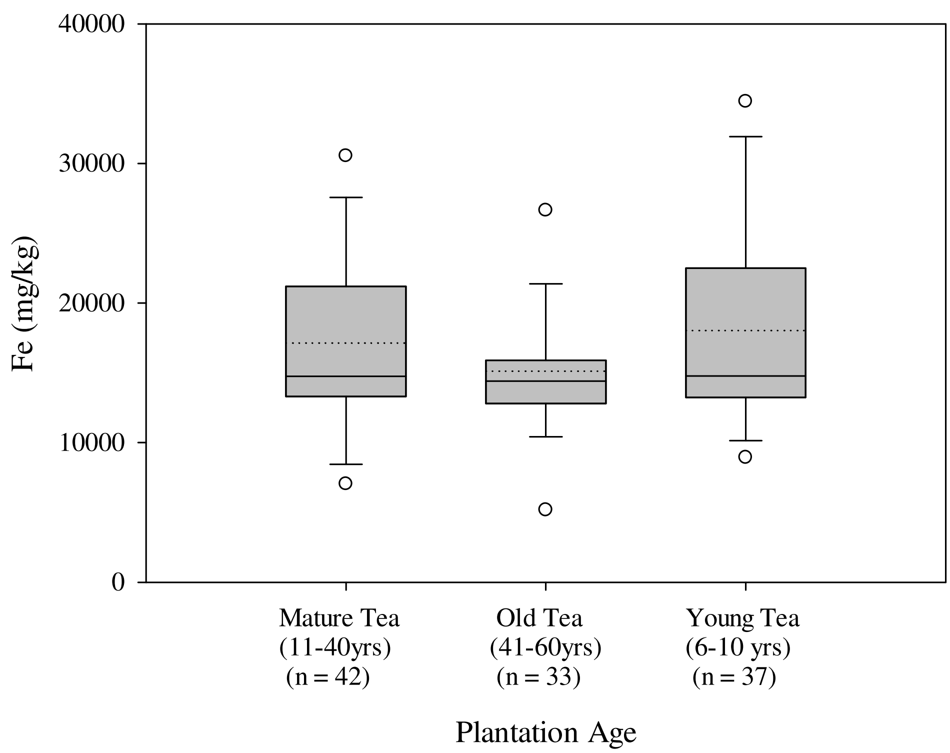


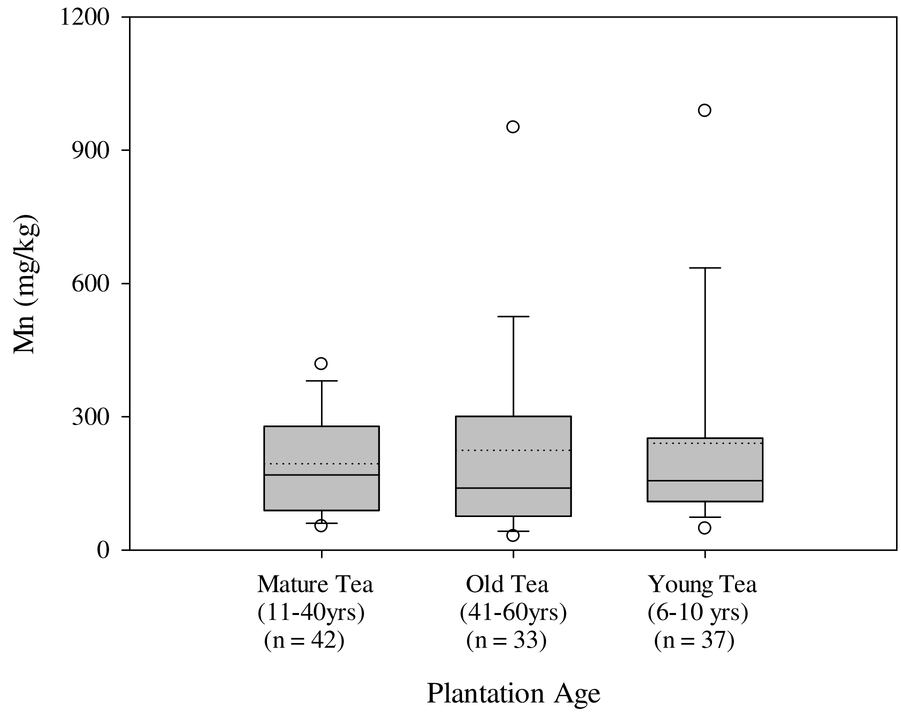
0.3
0.2
0.1
0.0
Total N (%)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age


2.0
1.5
1.0
0.5
0.0
B (mg/kg)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age

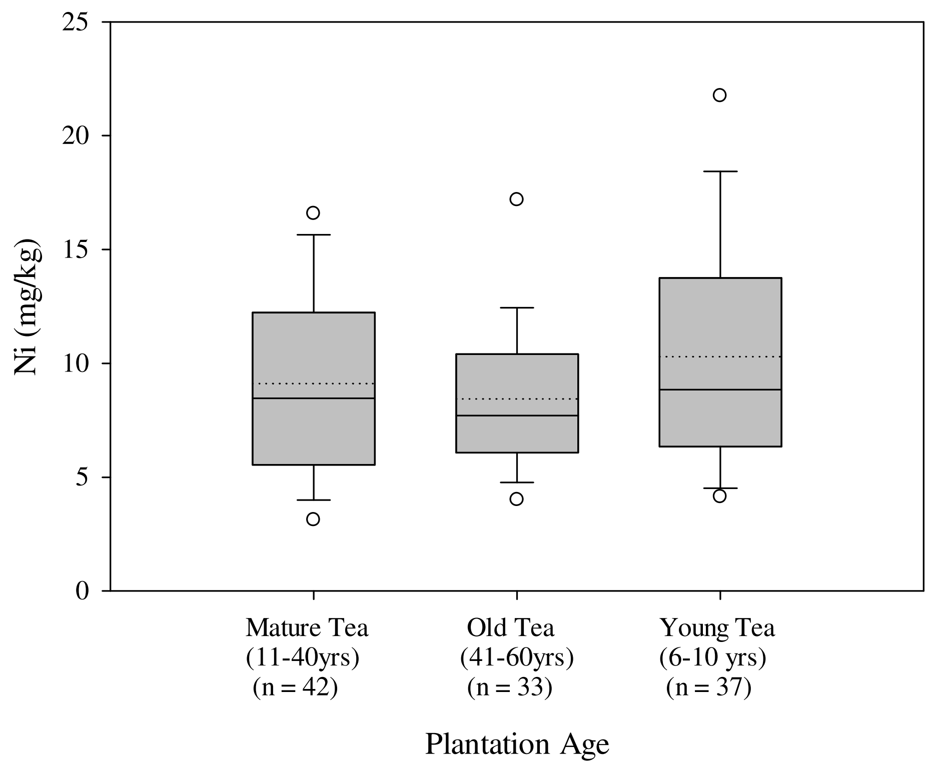

80
70
60
50
40
30
20
10
0
Cr (mg/kg)
F = 3.27, p < 0.05
ab
b
a
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 32)
Young Tea
(6-10 yrs)
(n = 36)
Plantation Age


25
20
15
10
5
0
Cu (mg/kg)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age


40000
30000
20000
10000
0
Fe (mg/kg)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age



1200
900
600
300
0
Mn (mg/kg)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age



25
20
15
10
5
0
Ni (mg/kg)
Mature Tea
(11-40yrs)
(n = 42)
Old Tea
(41-60yrs)
(n = 33)
Young Tea
(6-10 yrs)
(n = 37)
Plantation Age

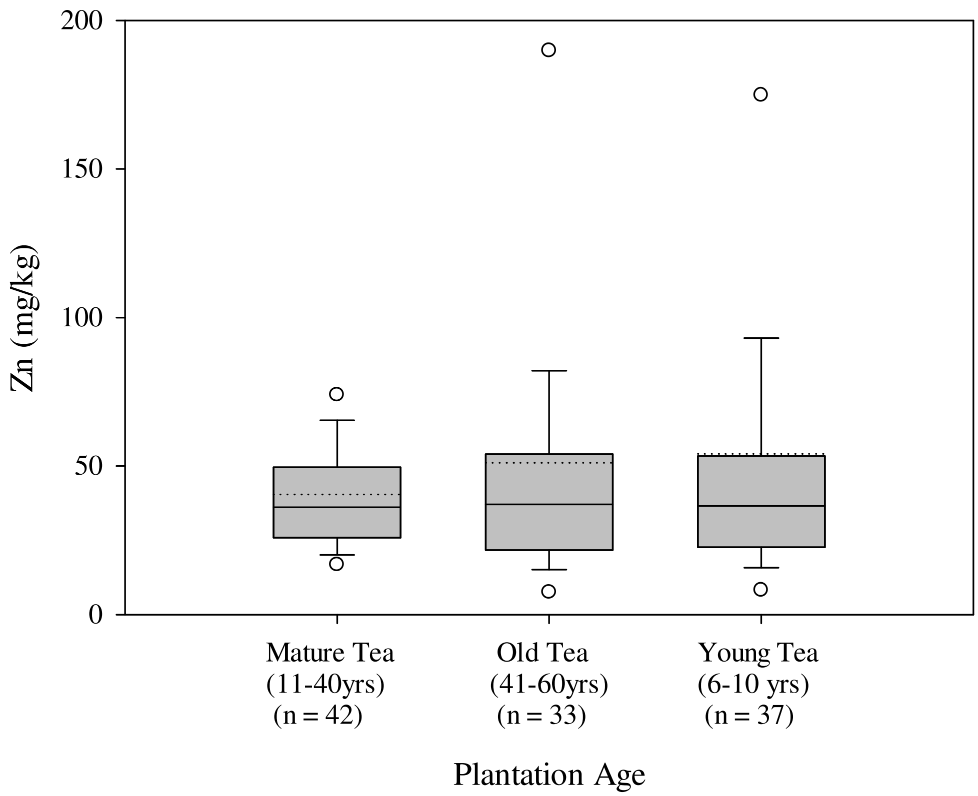


Figure 4.2: Box and whisker plots showing concentrations of different elements in tea garden soils under different ages of tea plants. The boxplots indicate the lower and upper quartile (box), the median (solid line), the mean (dotted line), the 10th and 90th percentiles (whiskers) and the 5th and 95th percentiles (circles). The F-value and *p*-value represent the F-ratio that indicates the degree of variation among the age group tea plantations and the level of significance of this variation, respectively, from one-way analysis of variance (ANOVA) test.

4.4.3 Variabilities in soil properties within different soil series

In the present study 19 soil series have been found in the areas of sampling (Table 4.1).

Table 4.1: Soil Series of the tea gardens

District	Sub-district	Tea gardens	No. of soil series	Soil Series
Moulvibazar	Kamalganj	Allynugger	03	Baralekha Pritimpasha Srimangal
		Patrakhola	02	Pritimpasha Srimangal
	Srimangal	Sathgao	04	Ramghor Khadimnagar Beanibazar Bijipur
		Rajghat	04	Khadimnagar Ramghor Pritimpasha Srimangal
		BTRI Farm	03	Pritimpasha Beanibazar Bijipur
		Jagcherra	03	Beanibazar Pritimpasha Ramghor
		Bilashcherra	04	Ramghor Bijipur Srimangal Khadimnagar
Sylhet	Sylhet Sadar	Malnicherra	03	Ramghor Bijipur Khadimnagar
Chattogram	Fatickcherri	Karnafuli	04	Nolua Mirsorai Bijipur Shalban
		Oodaleah	05	Nolua Khadimnagar Bijipur Datmara Pahartoli

Table 4.1: Continued.

District	Sub-district	Tea gardens	No. of soil series	Soil Series
Chattogram		Neptune	04	Bijipur Rangamati Datmara Khadimnagar
Panchagarh	Panchagarh Sadar	Karotoa	05	Panchagarh Ruhia Ranisonkail Baliadangi Pamol
	Tetulia	Kazi and Kazi	03	Bhajonpur Series Domar Series Baliadangi Series

The pH in the surface soils of different soil series in the tea gardens were found to be varied significantly ($^{ANOVA}F = 2.36$, $p < 0.01$) with the maximum pH in Domar series (5.63), followed by Pahartoli (5.60), Bhajonpur (5.52), Ruhia (5.10), Ranisonkail (5.10), Panchagarh (5.00), Bijipur (4.96), Baliadangi (4.93), Srimangal (4.93), Ramghor (4.89), Mirsorai (4.80), Khadimnagar (4.80), Shalban (4.75), Pritimpasha (4.74), Beanibazar (4.70), Datmara (4.70), Baralekha (4.60), Rangamati (4.43), and Nolua series (4.22) (Figure 4.3). The pH in the sub-surface soils was also observed to be significantly higher in the soils of Pahartoli series (6.00) compared to other soil series (Figure A18).

The organic carbon contents in the surface soil of different soil series in the tea gardens was found to be varied significantly ($^{ANOVA}F = 3.40$, $p < 0.001$) with the maximum organic carbon content in Panchagarh series (2.44%), followed by Bhajonpur (2.16%), Baliadangi (2.03%), Ruhia (1.97%), Domar (1.84%), Srimangal (1.74%), Datmara (1.28%), Ramghor (1.27%), Baralekha (1.26%), Ranisonkail (1.25%), Bijipur (1.24%), Pritimpasha (1.05%), Mirsorai (0.93%), Beanibazar (0.85%), Nolua (0.79%), Rangamati (0.77%), Pahartoli (0.76%), Shalban (0.70%) and Khadimnagar (0.68%) (Figure 4.3). The organic carbon contents in the sub-surface soils were observed to be significantly higher in the soils of Srimangal series (2.08%) compared to other soil series (Figure A19).

The total nitrogen contents in the surface soils of different soil series in the tea gardens were found to be varied significantly ($^{ANOVA}F = 3.44$, $p < 0.001$) with the maximum total nitrogen contents in Panchagarh series (0.21%), followed by Bhajonpur (0.19%), Baliadangi (0.17%), Ruhia (0.17%), Domar (0.16%), Srimangal (0.15%), Ramghor (0.11%), Datmara (0.11%), Ranisonkail (0.11%), Bijipur (0.11%), Baralekha (0.11%), Pritimpasha (0.09%), Mirsorai (0.08%), Beanibazar (0.07%), Nolua (0.07%), Rangamati (0.07%), Pahartoli (0.07%), Shalban (0.06%) and Khadimnagar series (0.06%) (Figure 4.3). The total nitrogen contents in the sub-surface soils were observed to be significantly higher in the soils of Srimangal series (0.19%) compared to other series (Figure A20).

The concentrations of boron in the surface soils of different soil series in the tea gardens were found to be varied significantly (${}^{ANOVA}F = 2.92$, $p < 0.001$) with the maximum concentration in Ranisonkail series (1.71 mg/kg), followed by Domar (1.27 mg/kg), Bhajonpur (0.81 mg/kg), Bijipur (0.65 mg/kg), Ramghor (0.61 mg/kg), Srimangal (0.60 mg/kg), Nolua (0.46 mg/kg), Datmara (0.45 mg/kg), Rangamati (0.31 mg/kg), Shalban (0.30 mg/kg), Baralekha (0.28 mg/kg), Baliadangi (0.26 mg/kg), Mirsorai (0.25 mg/kg), Khadimnagar (0.21 mg/kg), Panchagarh (0.20 mg/kg), Beanibazar (0.15 mg/kg), Ruhia (0.12 mg/kg), Pritimpasha (0.10 mg/kg) and Pahartoli (0.05 mg/kg) (Figure 4.3). The concentrations of boron in the sub-surface soils were observed to be significantly higher in the soils of Ramghor series (1.45 mg/kg) compared to other soil series (Figure A21).

The concentrations of chromium in the surface soils of different soil series in the tea gardens were found to be varied significantly (${}^{ANOVA}F = 7.10$, $p < 0.001$) with the maximum concentration in Baliadangi series (60.35 mg/kg), followed by Panchagarh (52.53 mg/kg), Ranisonkail (47.95 mg/kg), Datmara (46.05 mg/kg), Domar (40.34 mg/kg), Mirsorai (36.98 mg/kg), Pahartoli (34.40 mg/kg), Bhajonpur (33.55 mg/kg), Nolua (32.73 mg/kg), Shalban (27.55 mg/kg), Rangamati (26.66 mg/kg), Ruhia (25.55 mg/kg), Srimangal (21.15 mg/kg), Ramghor (20.30 mg/kg), Bijipur (18.02 mg/kg), Khadimnagar (15.74 mg/kg), Baralekha (15.57 mg/kg), Beanibazar (15.00 mg/kg) and Pritimpasha series (12.96 mg/kg) (Figure 4.3). The concentrations of chromium in the sub-surface soils were observed to be significantly higher in the soils of Baliadangi series (70.10 mg/kg) compared to other soil series (Figure A22).

The concentrations of copper in the surface soils of different soil series in the tea gardens were found to be varied significantly (${}^{ANOVA}F = 2.16$, $p < 0.01$) with the maximum concentration in Baliadangi series (17.38 mg/kg), followed by Panchagarh (14.55 mg/kg), Ranisonkail (14.25

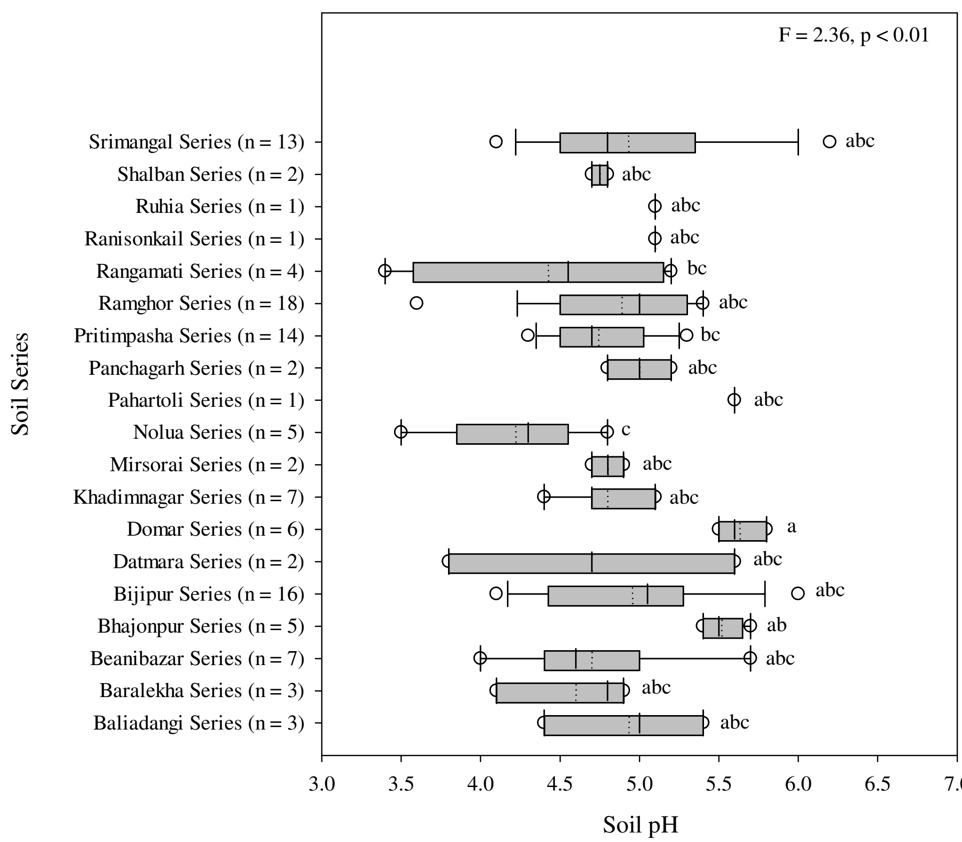
mg/kg), Ruhia (12.40 mg/kg), Pahartoli (12.35 mg/kg), Bhajonpur (11.77 mg/kg), Beanibazar (11.27 mg/kg), Nolua (10.15 mg/kg), Domar (10.13 mg/kg), Mirsorai (9.83 mg/kg), Datmara (9.38 mg/kg), Srimangal (9.37 mg/kg), Pritimpasha (8.66 mg/kg), Rangamati (8.58 mg/kg), Baralekha (8.57 mg/kg), Bijipur (7.94 mg/kg), Ramghor (7.67 mg/kg), Khadimnagar (6.64 mg/kg) and Shalban series (5.60 mg/kg) (Figure 4.3). The concentrations of copper in the sub-surface soils were observed to be significantly higher in the soils of Datmara series (17.70 mg/kg) compared to other soil series (Figure A23).

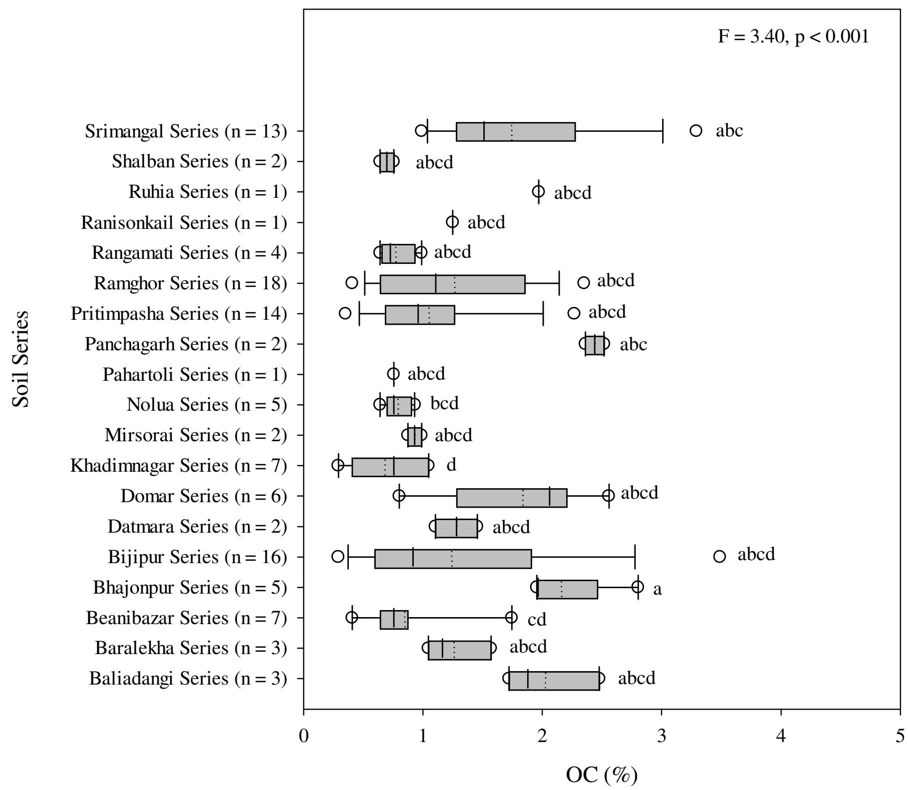
The concentrations of iron in the surface soils of different soils series in the tea gardens were found to be varied significantly ($^{ANOVA}F = 9.64$, $p < 0.001$) with the maximum concentration in Baliadangi series (37607 mg/kg), followed by Datmara (31270 mg/kg), Panchagarh (29890 mg/kg), Domar (25960 mg/kg), Ranisonkail (25920 mg/kg), Ruhia (25180 mg/kg), Pahartoli (23000 mg/kg), Nolua (20924 mg/kg), Mirsorai (20320 mg/kg), Bhajonpur (17285 mg/kg), Srimangal (16604 mg/kg), Shalban (16290 mg/kg), Rangamati (15815 mg/kg), Ramghor (15340 mg/kg), Beanibazar (14484 mg/kg), Baralekha (13259 mg/kg), Bijipur (12713 mg/kg), Pritimpasha (12554 mg/kg) and Khadimnagar (11720 mg/kg) (Figure 4.3). The concentrations of iron in the sub-surface soils were observed to be significantly higher in the soils of Datmara series (52540 mg/kg) compared to other soil series (Figure A24).

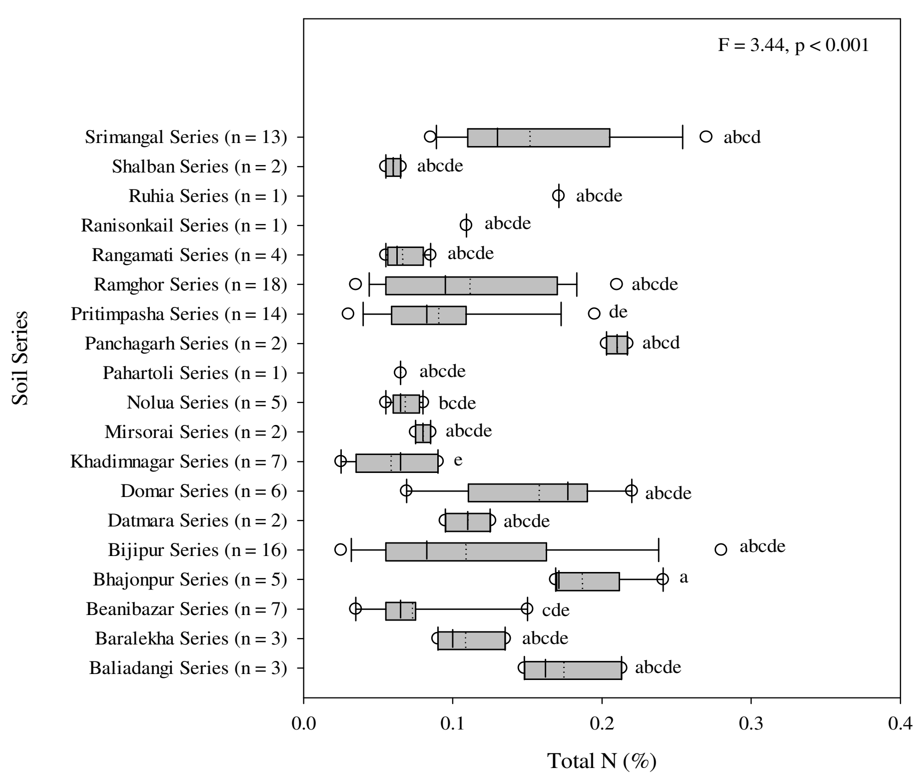
The concentrations of manganese in the surface soils of different soil series in the tea gardens were found to be varied insignificantly with the maximum concentration in Datmara series (550 mg/kg) compared to other soil series (Figure 4.3). The concentrations of manganese in the sub-surface soils were observed to be significantly higher in the soils of Datmara series (466 mg/kg) compared to other soil series (Figure A25).

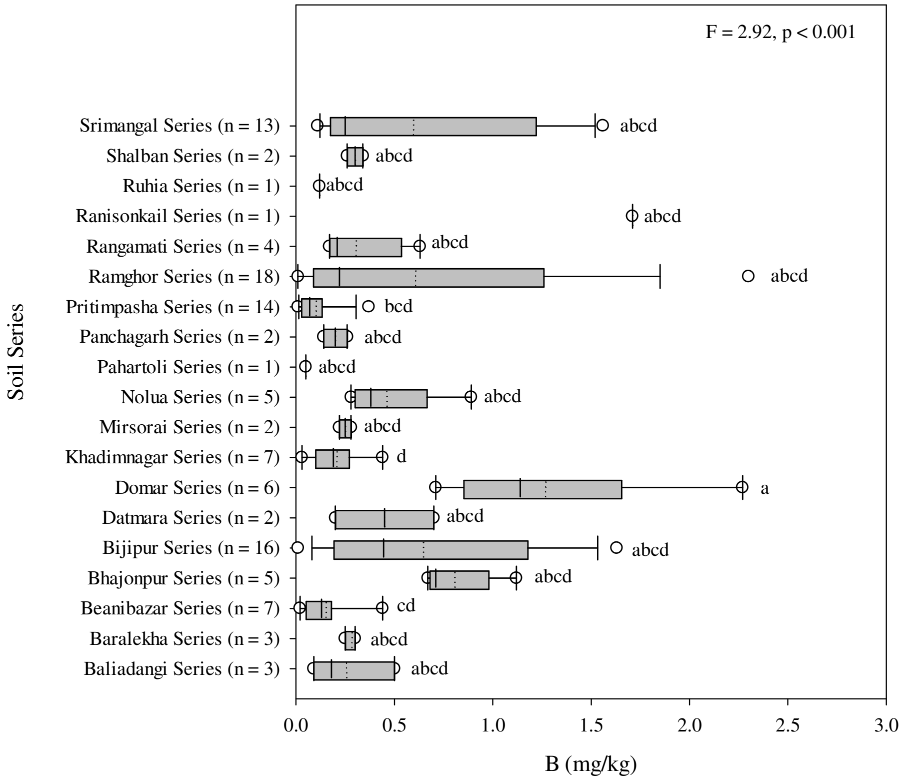
The concentrations of nickel in the surface soils of different soil series in the tea gardens were found to be varied significantly ($^{ANOVA}F = 3.86$, $p < 0.001$) with the maximum concentration in Pahartoli series (21.60 mg/kg), followed by Baliadangi (16.75 mg/kg), Panchagarh (16.43 mg/kg), Datmara (16.43 mg/kg), Ranisonkail (14.00 mg/kg), Ruhia (13.20 mg/kg), Nolua (12.13 mg/kg), Domar (11.73 mg/kg), Bhajonpur (10.86 mg/kg), Srimangal (9.58 mg/kg), Beanibazar (9.28 mg/kg), Ramghor (8.42 mg/kg), Pritimpasha (8.21 mg/kg), Baralekha (8.05 mg/kg), Khadimnagar (7.99 mg/kg), Shalban (7.57 mg/kg), Mirsorai (7.53 mg/kg), Bijipur (6.46 mg/kg) and Rangamati (5.75 mg/kg) (Figure 4.3). The concentrations of nickel in the sub-surface soils were observed to be significantly higher in the soils of Pahartoli series (23.70 mg/kg) compared to other soil series (Figure A26).

The concentrations of zinc in the surface soils of different soil series in the tea gardens were found to be varied significantly ($^{ANOVA}F = 2.18$, $p < 0.01$) with the maximum concentration in Datmara series (214 mg/kg), followed by Beanibazar (122.70 mg/kg), Pahartoli (55.15 mg/kg), Khadimnagar (51.60 mg/kg), Pritimpasha (51.34 mg/kg), Panchagarh (47.40 mg/kg), Bijipur (44.23 mg/kg), Ranisonkail (43.63 mg/kg), Bhajonpur (42.36 mg/kg), Ruhia (41.80 mg/kg), Domar (41.16 mg/kg), Baliadangi (39.40 mg/kg), Ramghor (36.02 mg/kg), Baralekha (35.60 mg/kg), Shalban (34.72 mg/kg), Nolua (33.73 mg/kg), Mirsorai (28.89 mg/kg), Srimangal (28.12 mg/kg) and Rangamati (21.98 mg/kg) (Figure 4.3). The concentrations of zinc in the sub-surface soils were observed to be significantly higher in the soils of Pahartoli series (63.61 mg/kg) compared to other soil series (Figure A27).

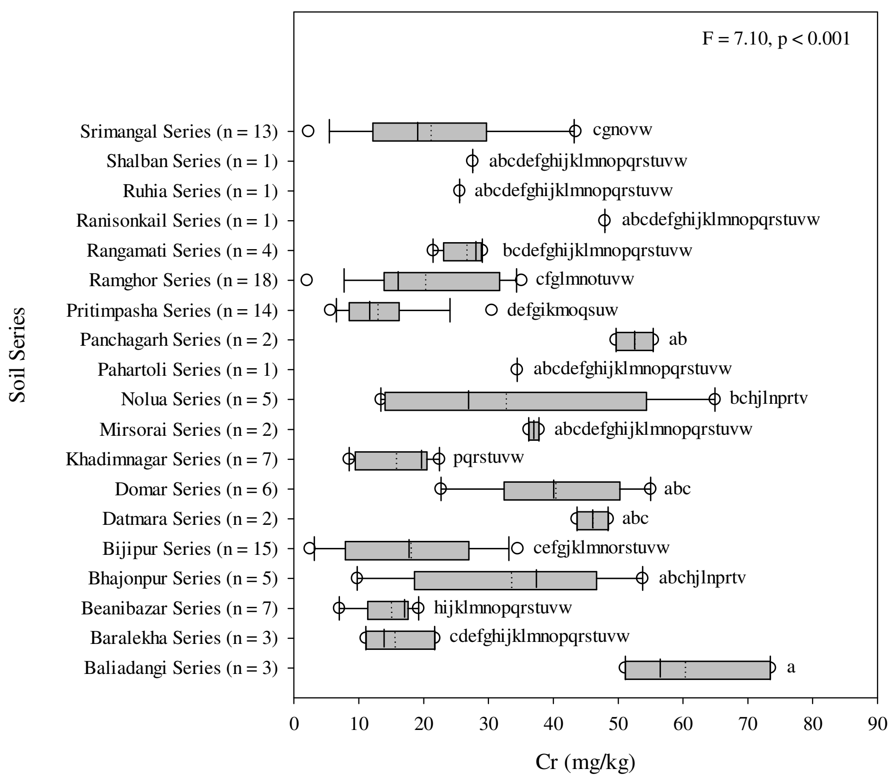



F = 3.40, p < 0.001
Srimangal Series (n = 13)
Shalban Series (n = 2)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 16)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
abc
abcd
abcd
abcd
abcd
abcd
abcd
abc
abcd
bcd
abcd
d
abcd
abcd
abcd
a
cd
abcd
abcd
Soil Series
0
1
2
3
4
5
OC (%)

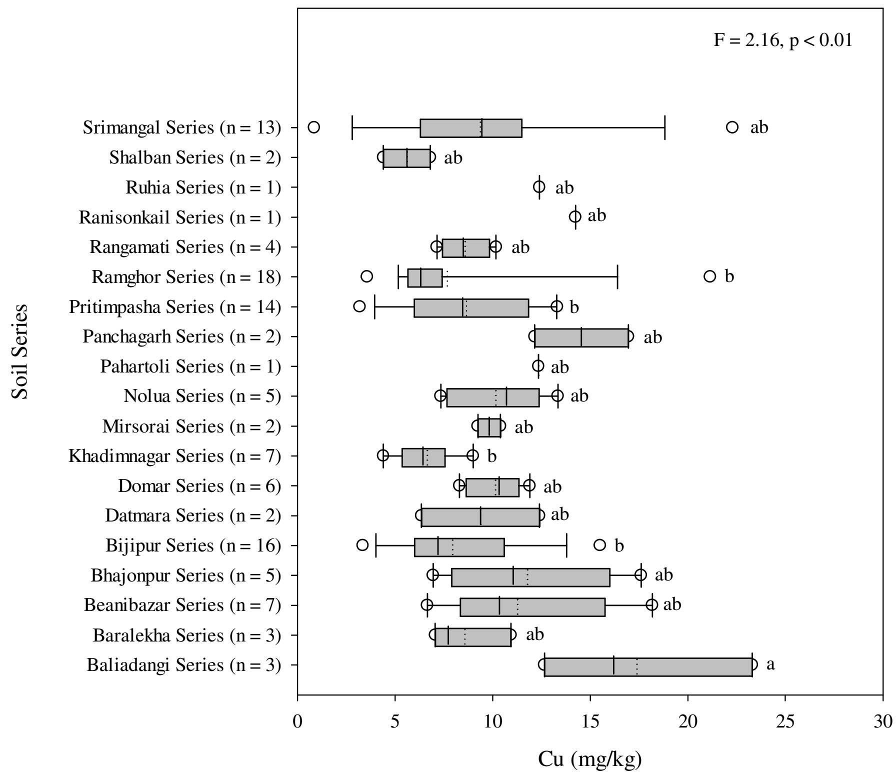

F = 3.44, p < 0.001
Srimangal Series (n = 13)
Shalban Series (n = 2)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 16)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
abcd
abcde
abcde
abcde
abcde
abcde
de
abcd
abcde
bcde
abcde
e
abcde
abcde
abcde
a
cde
abcde
abcde
Soil Series
0.0
0.1
0.2
0.3
0.4
Total N (%)

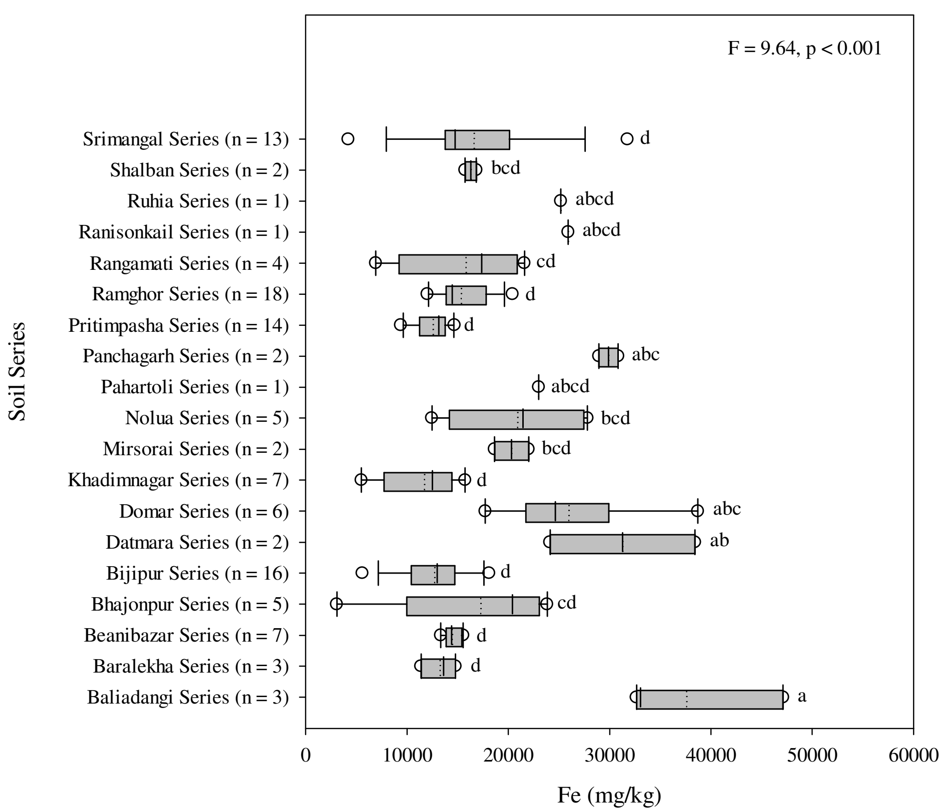


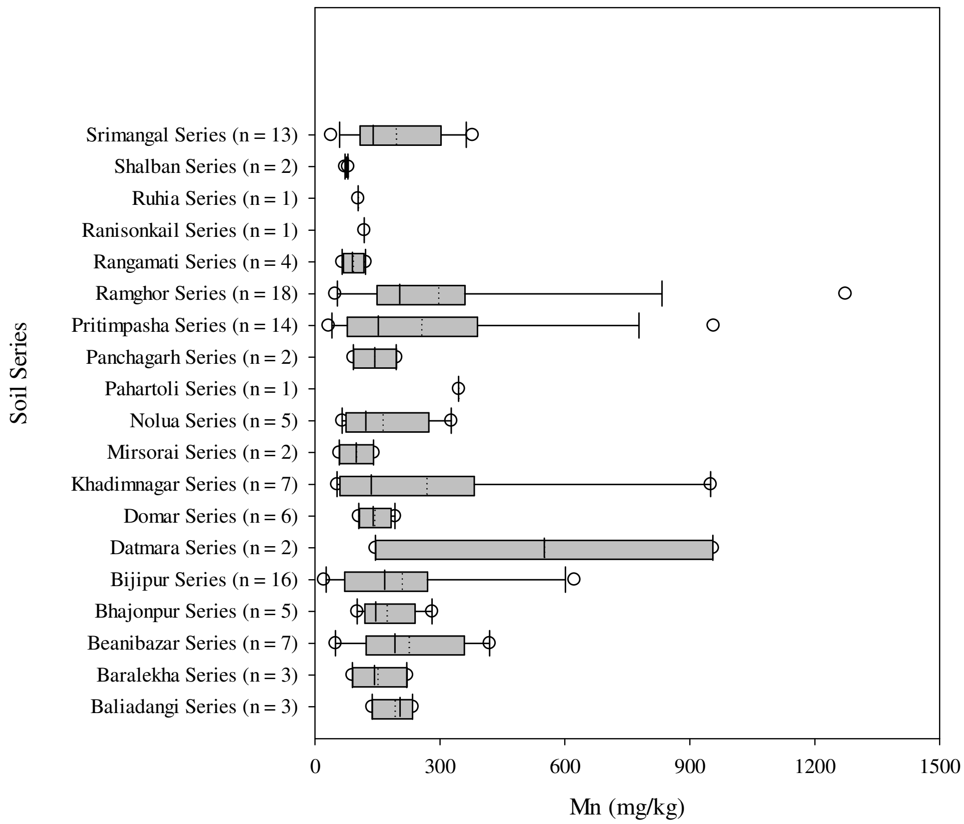
F = 2.92, p < 0.001
Soil Series
Srimangal Series (n = 13)
Shalban Series (n = 2)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 16)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
abcd
abcd
abcd
abcd
abcd
abcd
bcd
abcd
abcd
abcd
abcd
d
a
abcd
abcd
abcd
cd
abcd
abcd
0.0
0.5
1.0
1.5
2.0
2.5
3.0
B (mg/kg)



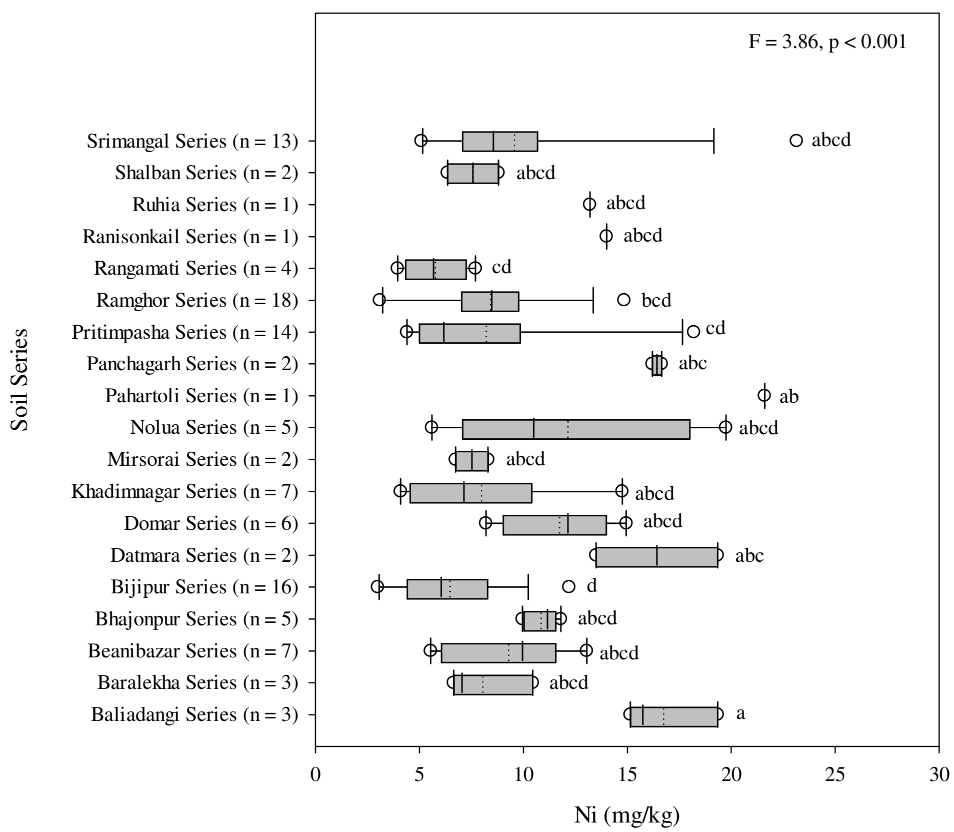
F = 7.10, p < 0.001
Soil Series
Srimangal Series (n = 13)
Shalban Series (n = 1)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 15)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
cgnovw
abcdefghijklmnopqrstuvw
abcdefghijklmnopqrstuvw
abcdefghijklmnopqrstuvw
bcdefghijklmnopqrstuvw
cfglmnotuvw
defgikmoqsuw
ab
abcdefghijklmnopqrstuvw
bchjlnprtv
abcdefghijklmnopqrstuvw
pqrstuvw
abc
abc
cefgjklmnorstuvw
abchjlnprtv
hijklmnopqrstuvw
cdefghijklmnopqrstuvw
a
0
10
20
30
40
50
60
70
80
90
Cr (mg/kg)


F = 2.16, p < 0.01
Soil Series
Srimangal Series (n = 13)
Shalban Series (n = 2)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 16)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
ab
ab
ab
ab
ab
b
b
ab
ab
ab
ab
b
ab
ab
b
ab
ab
ab
a
0
5
10
15
20
25
30
Cu (mg/kg)


F = 9.64, p < 0.001
Soil Series
Srimangal Series (n = 13)
Shalban Series (n = 2)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 16)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
d
bcd
abcd
abcd
cd
d
d
abc
abcd
bcd
bcd
d
abc
ab
d
cd
d
d
a
0
10000
20000
30000
40000
50000
60000
Fe (mg/kg)



Srimangal Series (n = 13)
Shalban Series (n = 2)
Ruhia Series (n = 1)
Ranisonkail Series (n = 1)
Rangamati Series (n = 4)
Ramghor Series (n = 18)
Pritimpasha Series (n = 14)
Panchagarh Series (n = 2)
Pahartoli Series (n = 1)
Nolua Series (n = 5)
Mirsorai Series (n = 2)
Khadimnagar Series (n = 7)
Domar Series (n = 6)
Datmara Series (n = 2)
Bijipur Series (n = 16)
Bhajonpur Series (n = 5)
Beanibazar Series (n = 7)
Baralekha Series (n = 3)
Baliadangi Series (n = 3)
Soil Series
0
300
600
900
1200
1500
Mn (mg/kg)



F = 3.86, p < 0.001
Srimangal Series (n = 13)
abcd
Shalban Series (n = 2)
abcd
Ruhia Series (n = 1)
abcd
Ranisonkail Series (n = 1)
abcd
Rangamati Series (n = 4)
cd
Ramghor Series (n = 18)
bcd
Pritimpasha Series (n = 14)
cd
Panchagarh Series (n = 2)
abc
Pahartoli Series (n = 1)
ab
Nolua Series (n = 5)
abcd
Mirsorai Series (n = 2)
abcd
Khadimnagar Series (n = 7)
abcd
Domar Series (n = 6)
abcd
Datmara Series (n = 2)
abc
Bijipur Series (n = 16)
d
Bhajonpur Series (n = 5)
abcd
Beanibazar Series (n = 7)
abcd
Baralekha Series (n = 3)
abcd
Baliadangi Series (n = 3)
a
Soil Series
0
5
10
15
20
25
30
Ni (mg/kg)

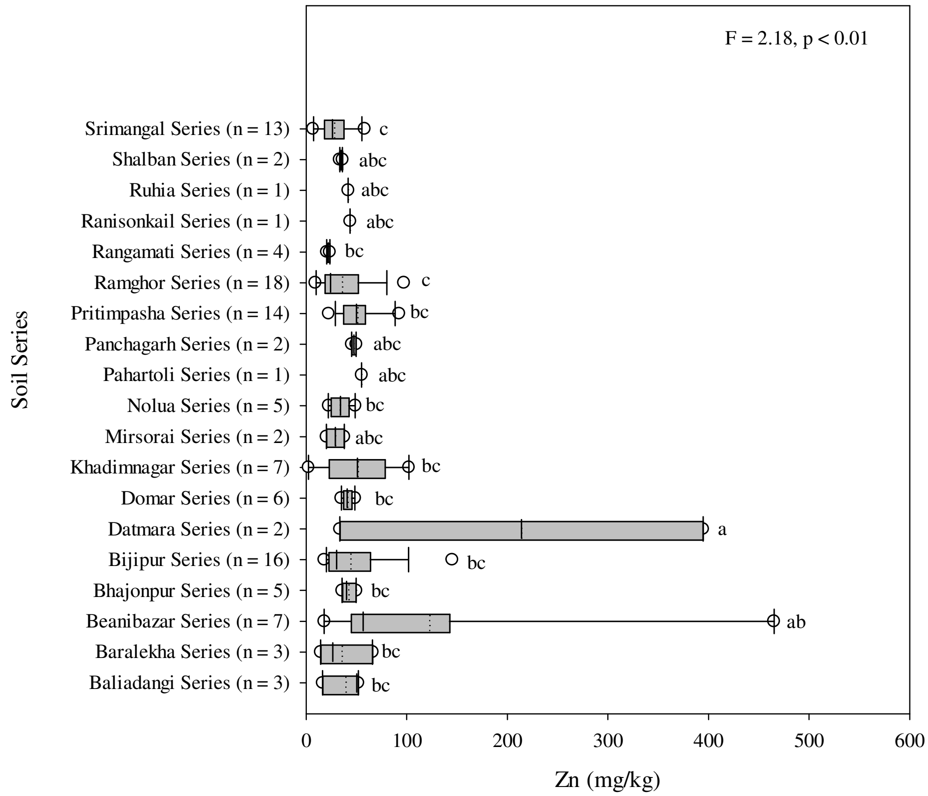


Figure 4.3: Box and whisker plots showing concentrations of different elements in various soil types (soil series) in tea garden soils. The boxplots indicate the lower and upper quartile (box), the median (solid line), the mean (dotted line), the 10th and 90th percentiles (whiskers) and the 5th and 95th percentiles (circles). The F-value and *p*-value represent the F-ratio that indicates the degree of variation among the soil types (soil series) and the level of significance of this variation, respectively, from one-way analysis of variance (ANOVA) test.

4.4.4 Variabilities in soil properties within different landforms

The pH in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 5.28$, $p < 0.001$), with the maximum mean pH in level ridge (5.36), followed by valley ridge (4.89), low hill (4.79), medium high hill (4.69) and terrace (4.60) landforms (Figure 4.4). The mean pH in the sub-surface soils of the tea gardens with different landforms also varied significantly with the maximum mean pH in level ridge (5.52) compared to other landforms (Figure A28). In the present study, surface and sub-surface soils of different landforms were acidic in nature which might be due to different pedogenic processes operating in different landforms (Reza *et al.*, 2022). The organic carbon contents in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 8.14$, $p < 0.001$) with the maximum mean organic carbon content in level ridge landform (2.00%), followed by low hill (1.27%), valley ridge (1.14%), terrace (1.05%) and medium high hill (0.92%) land forms (Figure 4.4). The organic carbon contents in the sub-surface soils were observed to be insignificantly higher in the soils of level ridge (1.69%) compared to other landforms.

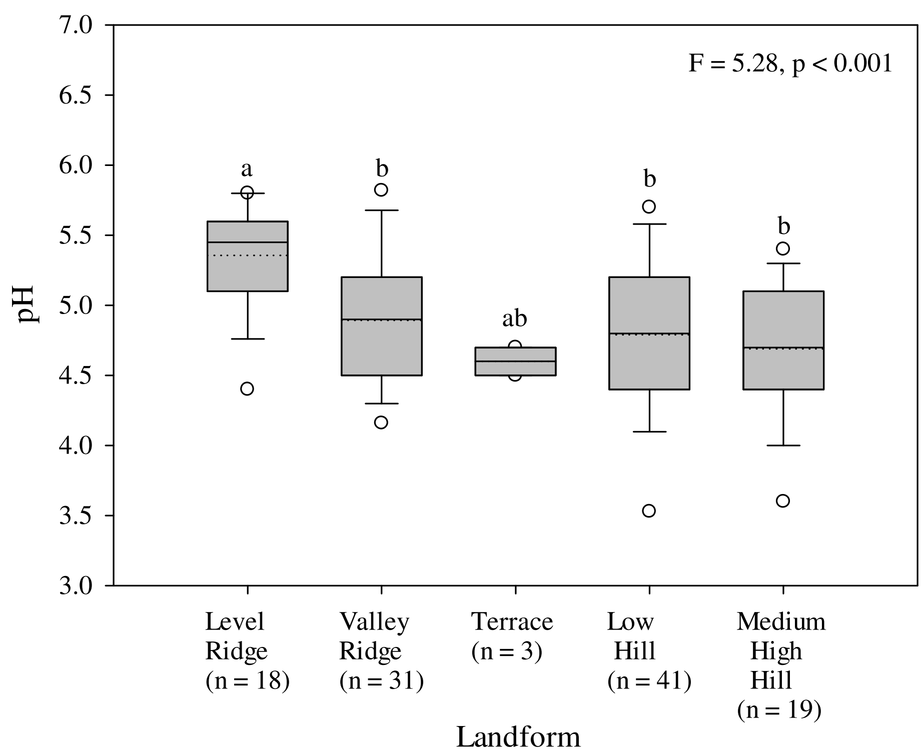
The total nitrogen contents in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 7.96$, $p < 0.001$) with the maximum mean total nitrogen content in level ridge landform (0.172%), followed by low hill (0.110%), valley ridge (0.099%), terrace (0.090%) and medium high hill (0.080%) landforms (Figure 4.4). The total nitrogen contents in the sub-surface soils were observed to be insignificantly higher in the soils of level ridge (0.146%) compared to other landforms. The concentrations of boron in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 3.55$, $p < 0.01$) with the maximum mean concentration of boron in level ridge landform (0.81 mg/kg), followed by low hill (0.53 mg/kg), valley ridge (0.39 mg/kg), medium high hill (0.27 mg/kg) and terrace (0.13 mg/kg) landforms (Figure 4.4). The concentrations of boron in the sub-surface soils were observed to be insignificantly higher in the soils of medium high hill (1.44 mg/kg) compared to other landforms.

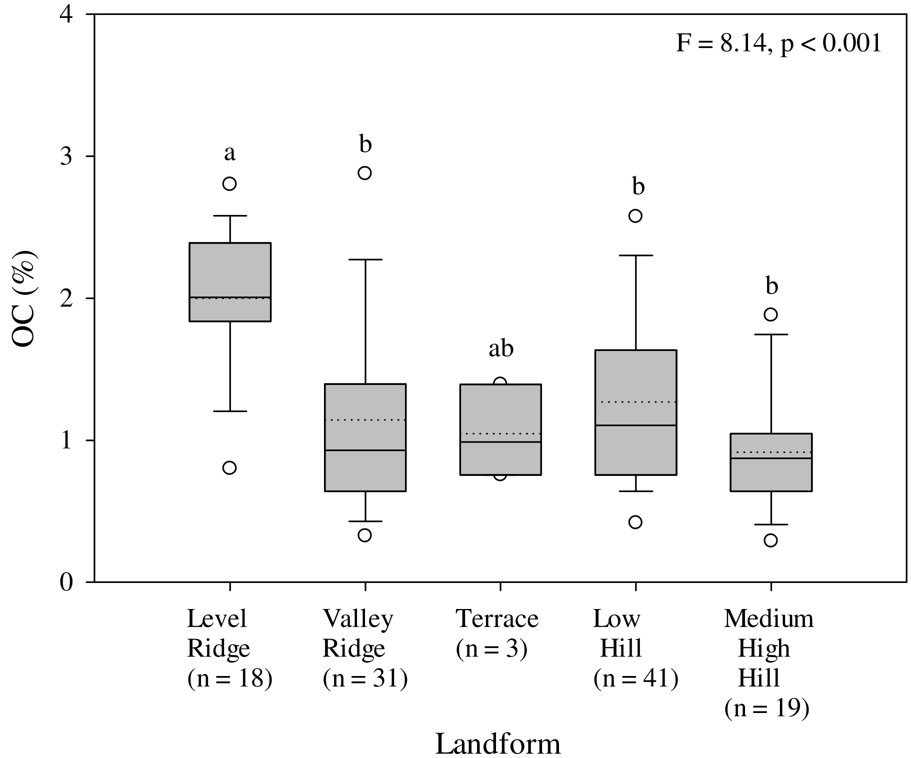
The concentrations of chromium in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 16.66$, $p < 0.001$) with the maximum mean concentration of chromium in level ridge landform (42.74 mg/kg), followed by low hill (24.28 mg/kg), valley ridge (18.05 mg/kg), medium high hill (16.79 mg/kg) and terrace (6.92 mg/kg) landforms (Figure 4.4). The concentrations of chromium in the sub-surface soils were also observed to be significantly higher in the soils of level ridge (49.86 mg/kg) compared to other landforms (Figure A29). The concentrations of copper in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 6.81$, $p < 0.001$) with the maximum mean concentration of copper in level ridge landform (12.64 mg/kg), followed by low hill (9.07 mg/kg), valley ridge (8.78 mg/kg), medium high hill (8.06 mg/kg) and terrace (2.97 mg/kg) landforms (Figure 4.4). The concentrations of copper in the sub-surface soils were observed to be significantly higher in the soils of level ridge (12.75 mg/kg) compared to other landforms (Figure A30).

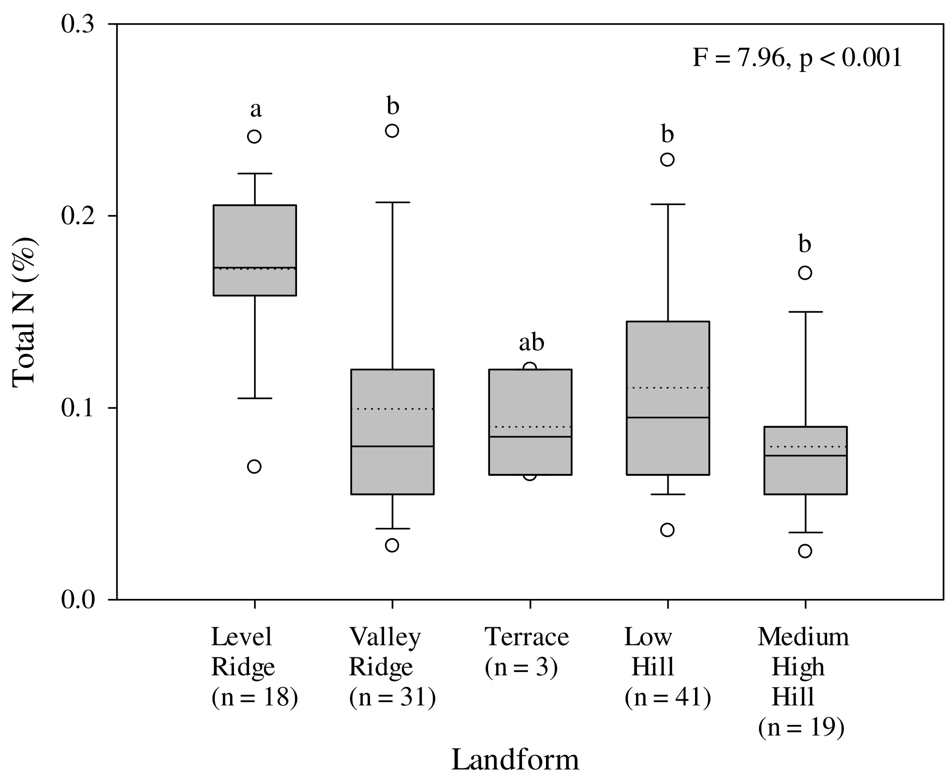
The concentrations of iron in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 15.99$, $p < 0.001$) with the maximum mean concentration of iron in level ridge landform (25882 mg/kg), followed by low hill (17065 mg/kg), medium high hill (14437 mg/kg), valley ridge (13594 mg/kg) and terrace (8531 mg/kg) landforms (Figure 4.4). The concentrations of iron in the sub-surface soils were observed to be significantly higher in the soils of level ridge (30353 mg/kg) compared to other landforms (Figure A31).

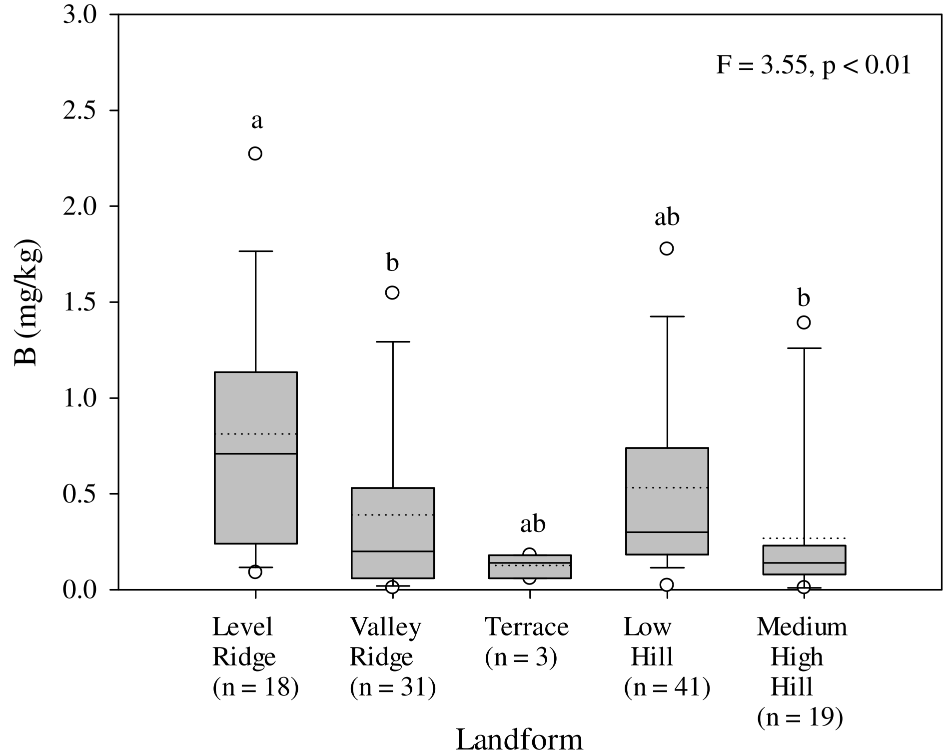
The concentrations of nickel in the surface soils of the tea gardens with different landforms varied significantly ($^{ANOVA}F = 5.38$, $p < 0.001$) with the maximum mean concentration of nickel in level ridge landform (13.06 mg/kg), followed by medium high hill (9.50 mg/kg), low hill (8.89 mg/kg), valley ridge (7.78 mg/kg) and terrace (7.13 mg/kg) landforms (Figure 4.4). The concentrations of nickel in the sub-surface soils were also observed to be significantly higher in the soils of level ridge (14.47 mg/kg) compared to other landforms (Figure A32).

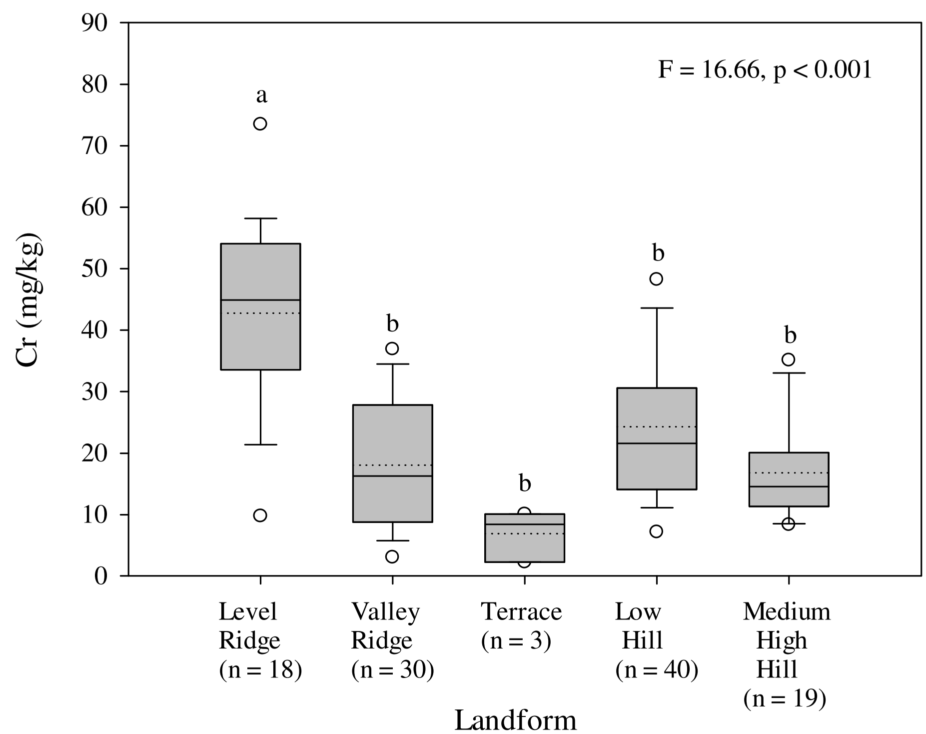
The occurrence and mobility of metals in soil is regulated by a complex interactions of different soil properties, particularly pH, organic matter and clay minerals which is further confounded by the topological characteristics of the landscape (Du Laing *et al*., 2009; Chowdhury *et al*., 2021; Sun *et al*., 2022).

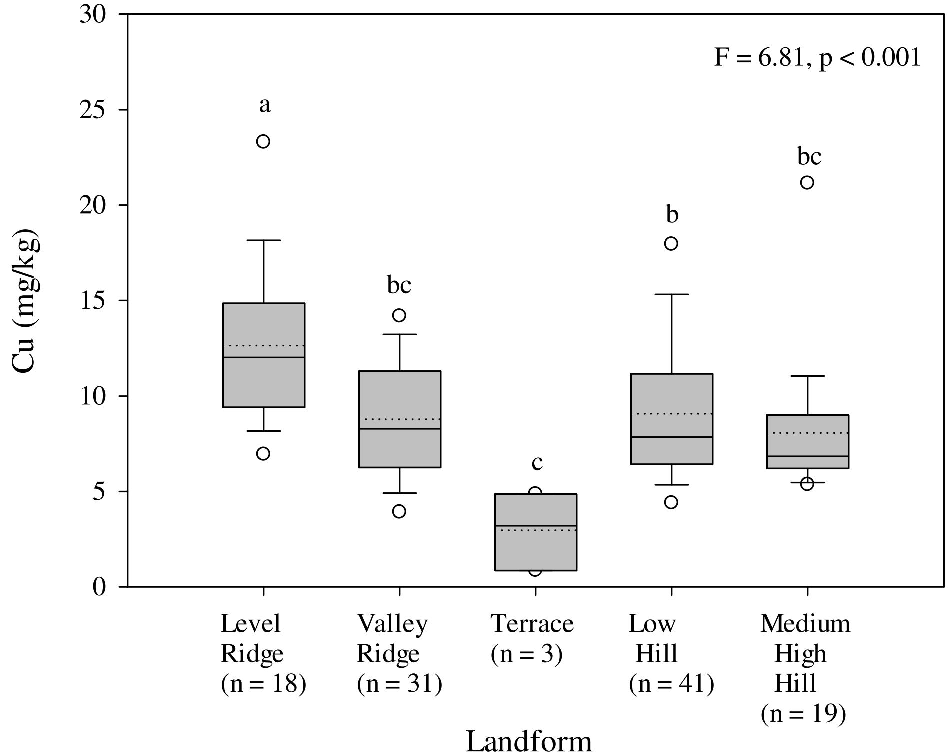



F = 8.14, p < 0.001
OC (%)
0
1
2
3
4
a
b
ab
b
b
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform

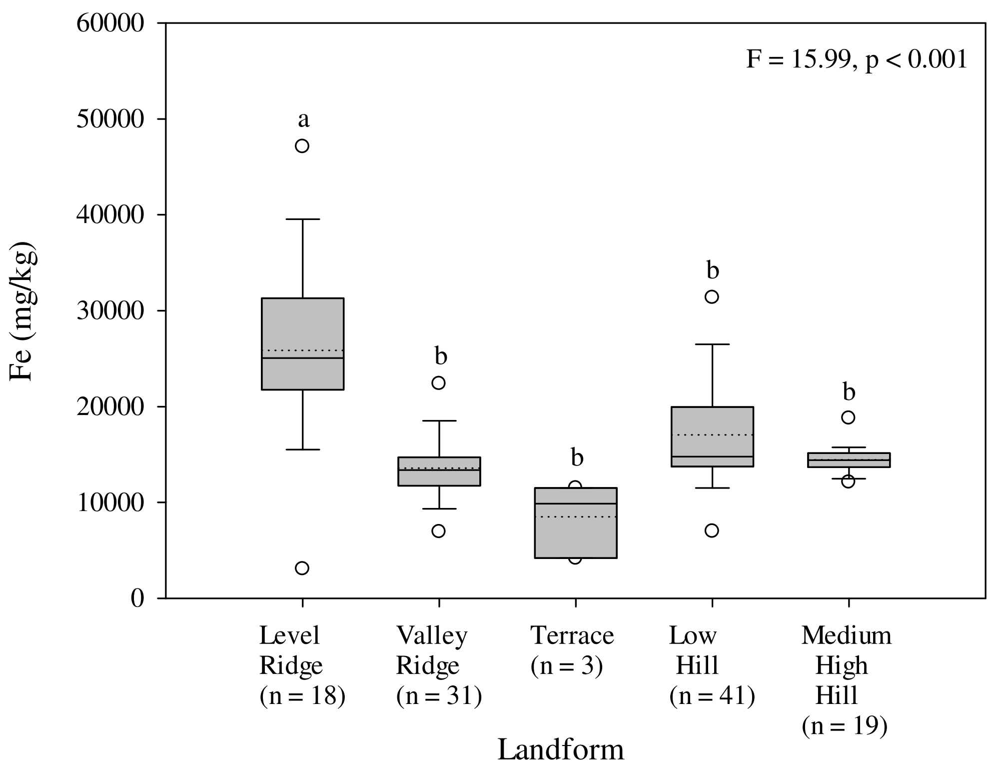

F = 7.96, p < 0.001
Total N (%)
0.0
0.1
0.2
0.3
a
b
ab
b
b
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform


3.0
2.5
2.0
1.5
1.0
0.5
0.0
B (mg/kg)
F = 3.55, p < 0.01
a
b
ab
ab
b
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform

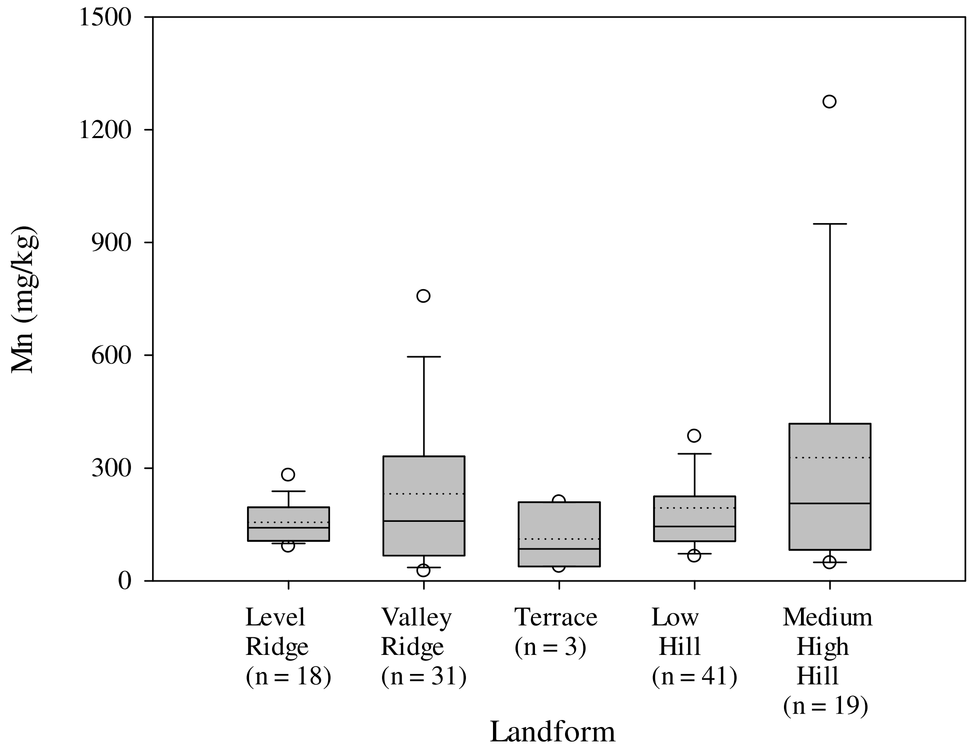

90
80
70
60
50
40
30
20
10
0
Cr (mg/kg)
F = 16.66, p < 0.001
a
b
b
b
b
Level Ridge (n = 18)
Valley Ridge (n = 30)
Terrace (n = 3)
Low Hill (n = 40)
Medium High Hill (n = 19)
Landform

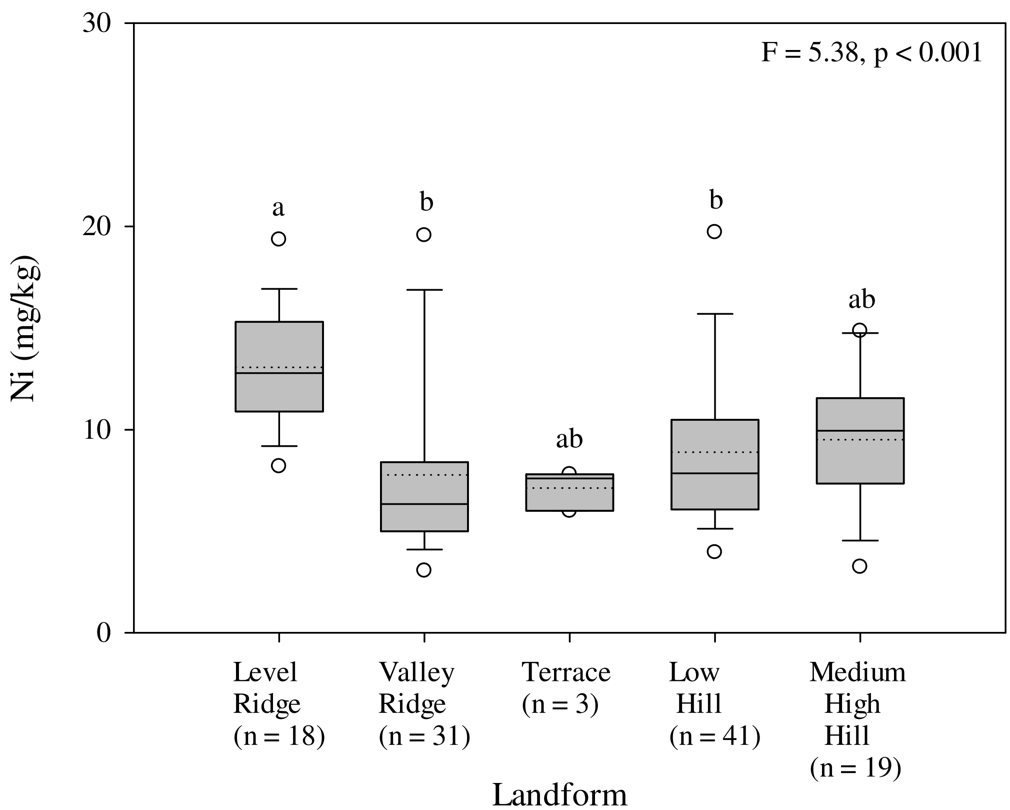


F = 6.81, p < 0.001
Cu (mg/kg)
30
25
20
15
10
5
0
a
bc
c
b
bc
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform



F = 15.99, p < 0.001
Fe (mg/kg)
60000
50000
40000
30000
20000
10000
0
a
b
b
b
b
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform


1500
1200
900
600
300
0
Mn (mg/kg)
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform


30
20
10
0
Ni (mg/kg)
F = 5.38, p < 0.001
a
b
ab
b
ab
Level Ridge (n = 18)
Valley Ridge (n = 31)
Terrace (n = 3)
Low Hill (n = 41)
Medium High Hill (n = 19)
Landform

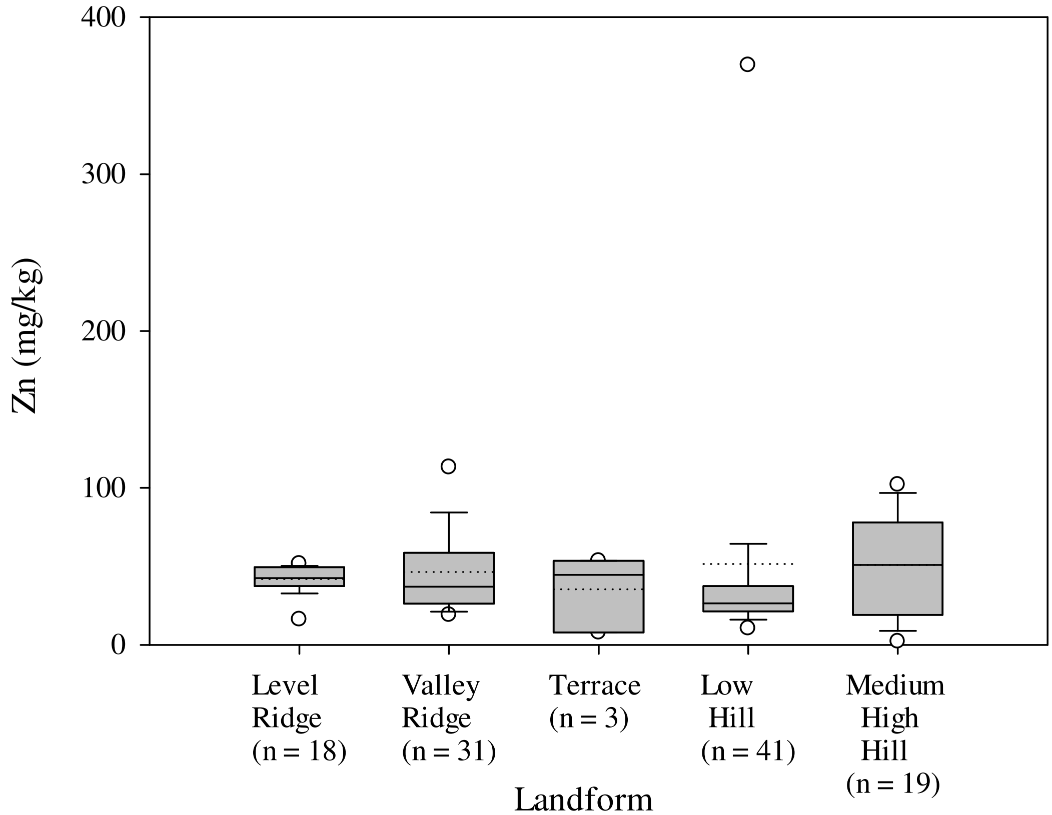


Figure 4.4: Box and whisker plots showing concentrations of different elements in tea garden soils of different land forms. The boxplots indicate the lower and upper quartile (box), the median (solid line), the mean (dotted line), the 10th and 90th percentiles (whiskers) and the 5th and 95th percentiles (circles). The F-value and *p*-value represent the F-ratio that indicates the degree of variation among the land forms and the level of significance of this variation, respectively, from one-way analysis of variance (ANOVA) test.

4.4.5 Variabilities in soil properties within different drainage class

The pH in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 2.75$, $p < 0.05$), with the maximum mean soil pH in moderately well drained condition (5.06), followed by imperfectly drained (4.89), well drained (4.79) and excessively drained (4.65) conditions (Figure 4.5). The mean pH in the sub-surface soils of the tea gardens with different drainage conditions also varied significantly with the maximum mean pH in moderately well drained (5.39) compared to other drainage conditions (Figure A33).

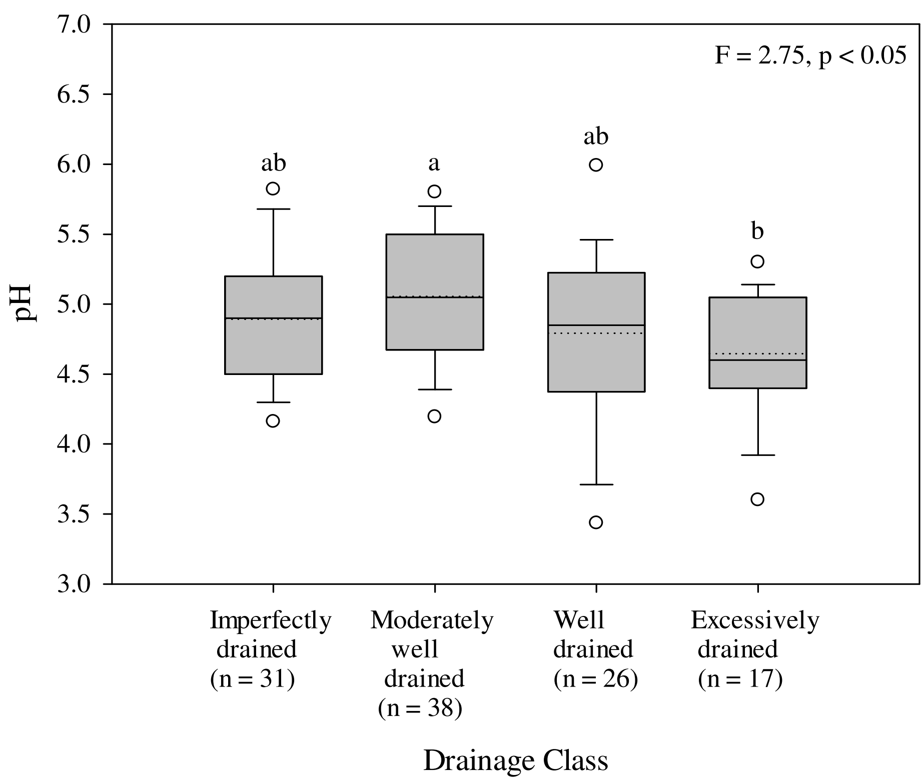
The organic carbon contents in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 4.52$, $p < 0.01$), with the maximum mean organic carbon content in moderately well drained condition (1.55%), followed by well drained (1.34%), imperfectly drained (1.14%) and excessively drained conditions (0.88%) (Figure 4.5). Raymond *et al*. (2012) reported that the largest soil organic carbon pools were found in soils with better drainage, not the poorly drained soils.

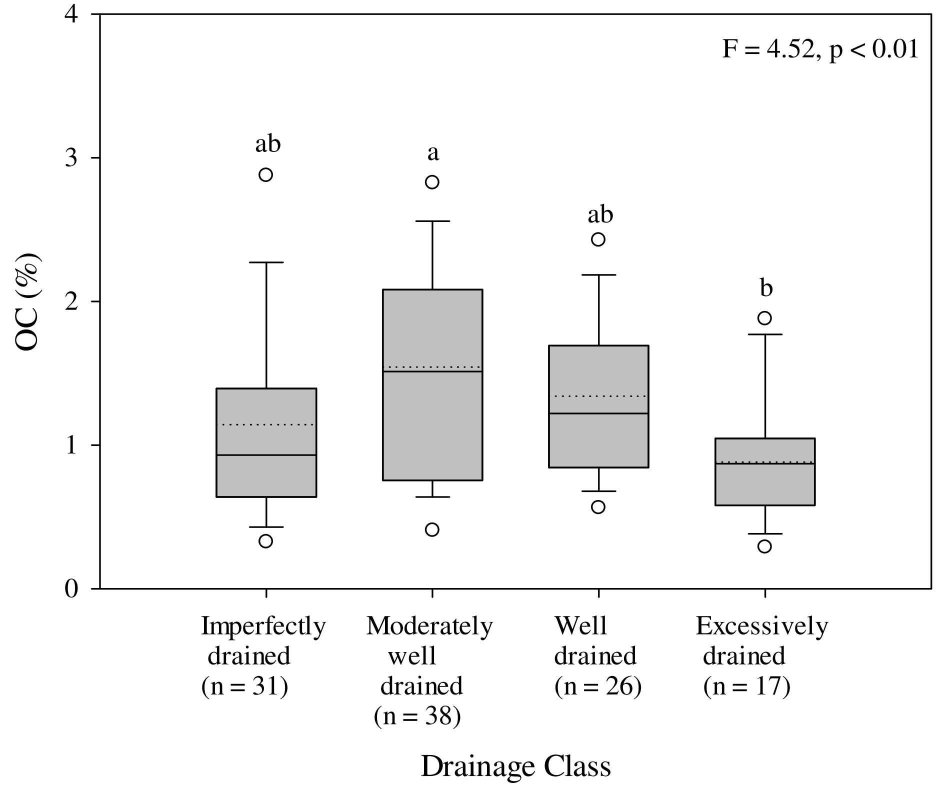
The total nitrogen contents in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 4.46$, $p < 0.01$), with the maximum mean total nitrogen content in moderately well drained condition (0.13%), followed by well drained (0.12%), imperfectly drained (0.10%) and excessively drained conditions (0.08%) (Figure 4.5). The concentrations of boron in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 3.25$, $p < 0.05$), with the maximum mean concentration of boron in well drained condition (0.65 mg/kg), followed by moderately well drained (0.57 mg/kg), imperfectly drained (0.39 mg/kg) and excessively drained conditions (0.21 mg/kg) (Figure 4.5).

The concentrations of chromium in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 6.66$, $p < 0.001$), with the maximum mean concentration of chromium in moderately well drained condition (30.66 mg/kg), followed by well drained (25.71 mg/kg), imperfectly drained (18.05 mg/kg) and excessively drained conditions (16.32 mg/kg) (Figure 4.5). The mean concentration of chromium in the sub-surface soils of the tea gardens with different drainage conditions also varied significantly with the maximum mean chromium in moderately well drained (45.75 mg/kg) compared to other drainage condition (Figure A34). In the present study the concentration of chromium in the sub-surface soil was observed more than surface soils in all types of drainage condition. It might be due to the leaching of chromium and variation of organic matter content in the surface and sub-surface soil as the addition of organic matter has the strongest influence on chromium mobility (Banks *et al*., 2006).

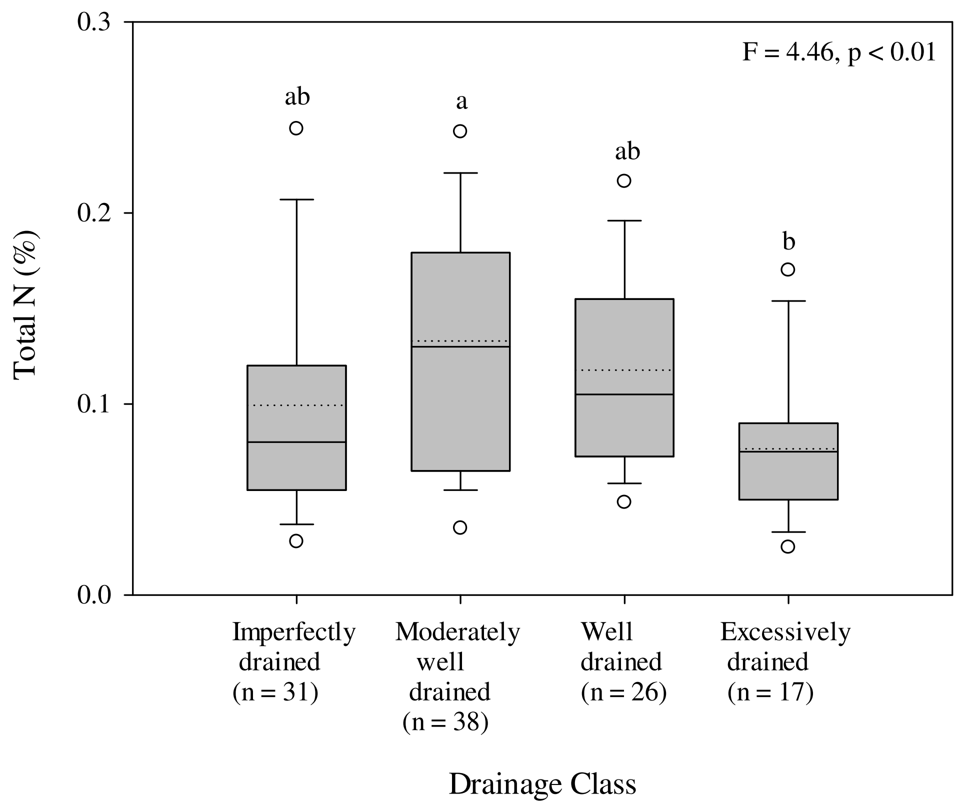
The concentrations of iron in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 7.10$, $p < 0.001$), with the maximum mean concentration of iron in moderately well drained condition (20442 mg/kg), followed by well drained (17139 mg/kg), excessively drained (14296 mg/kg) and imperfectly drained conditions (13594 mg/kg) (Figure 4.5). The mean concentration of iron in the sub-surface soils of the tea gardens with different drainage conditions also varied significantly with the maximum mean iron in moderately well drained (28251 mg/kg) compared to other drainage conditions (Figure A35). The concentrations of manganese in the surface soils of the tea gardens with different drainage conditions varied significantly ($^{ANOVA}F = 3.92$, $p < 0.05$), with the maximum mean concentration of manganese in excessively drained condition (348.60 mg/kg), followed by imperfectly drained (231.80 mg/kg), well drained (219.90 mg/kg) and moderately well drained conditions (149.30 mg/kg) (Figure 4.5).

The concentrations of chromium and iron in the moderately well drained soils were found to be significantly higher than that in the excessively drained and imperfectly drained soils (Figure 4.5). The comparatively higher pH and organic carbon content in the moderately well drained soils perhaps increased the accumulation and retention of heavy metals in the soils (Adriano, 2001; Bradl, 2004; Sherene, 2010; Rinklebe *et al*., 2016; Lasota *et al*., 2020).

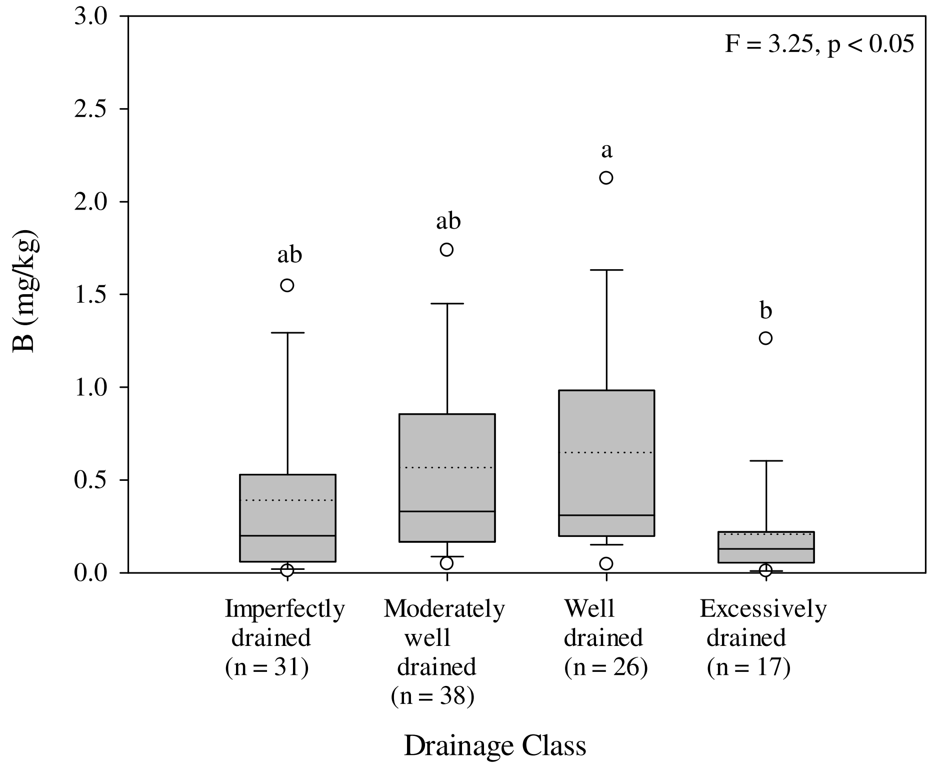


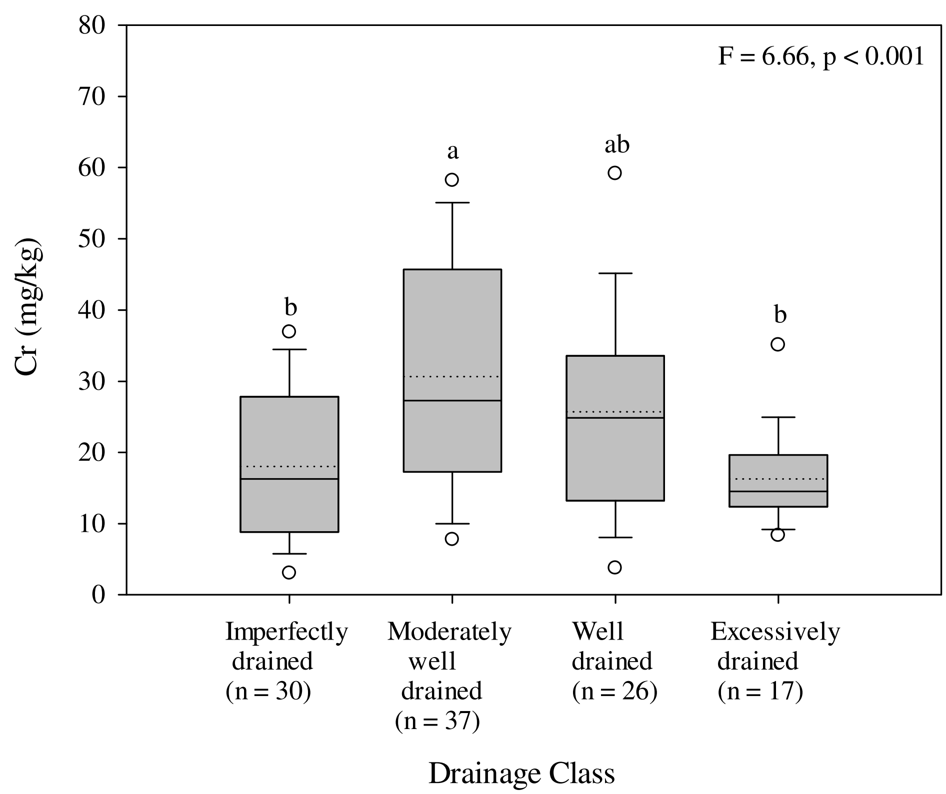


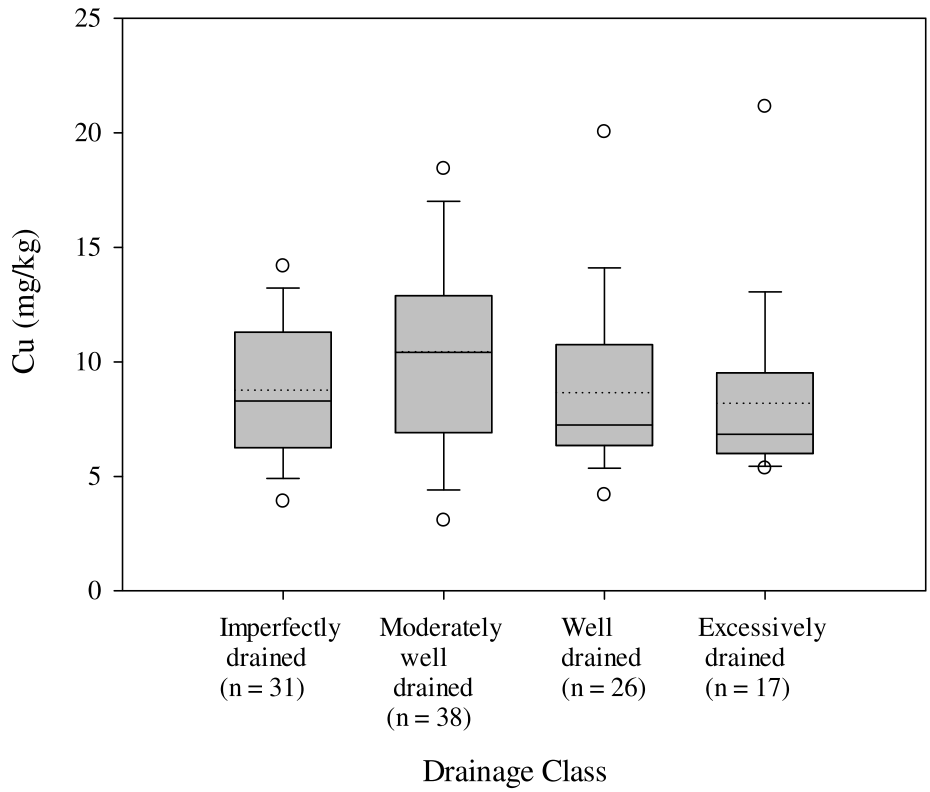
4
3
2
1
0
OC (%)
F = 4.52, p < 0.01
ab
a
ab
b
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class

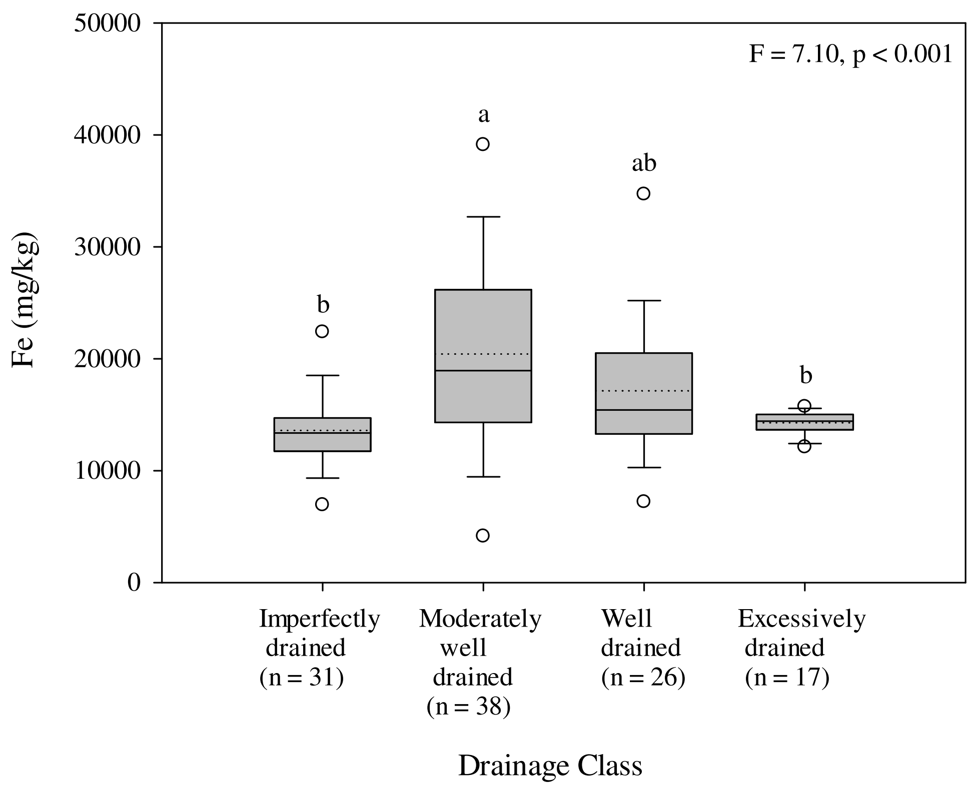


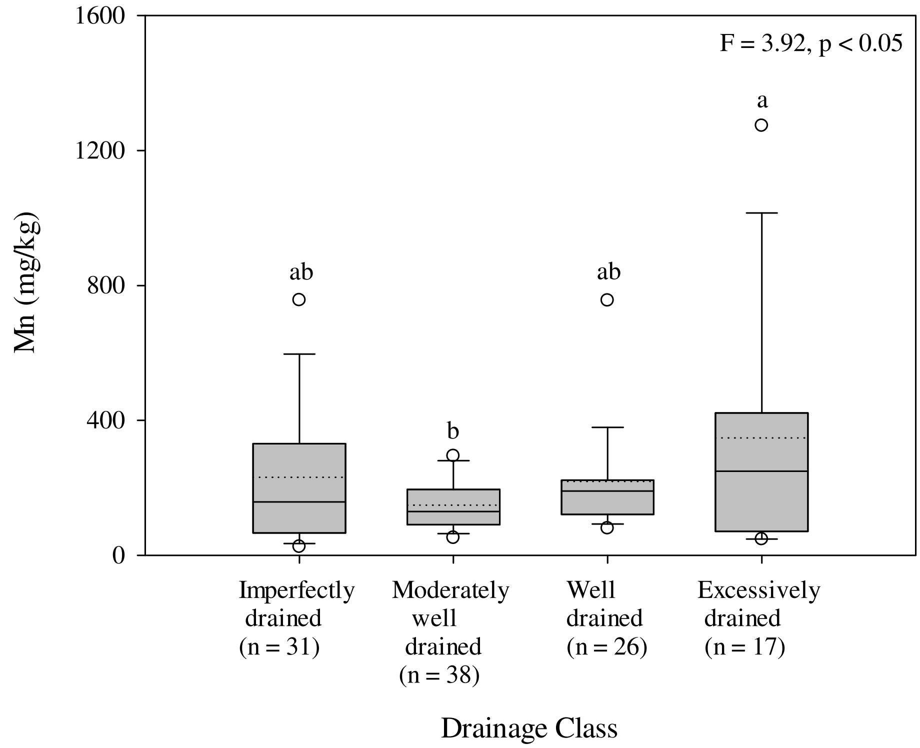
0.3
0.2
0.1
0.0
Total N (%)
F = 4.46, p < 0.01
ab
a
ab
b
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class

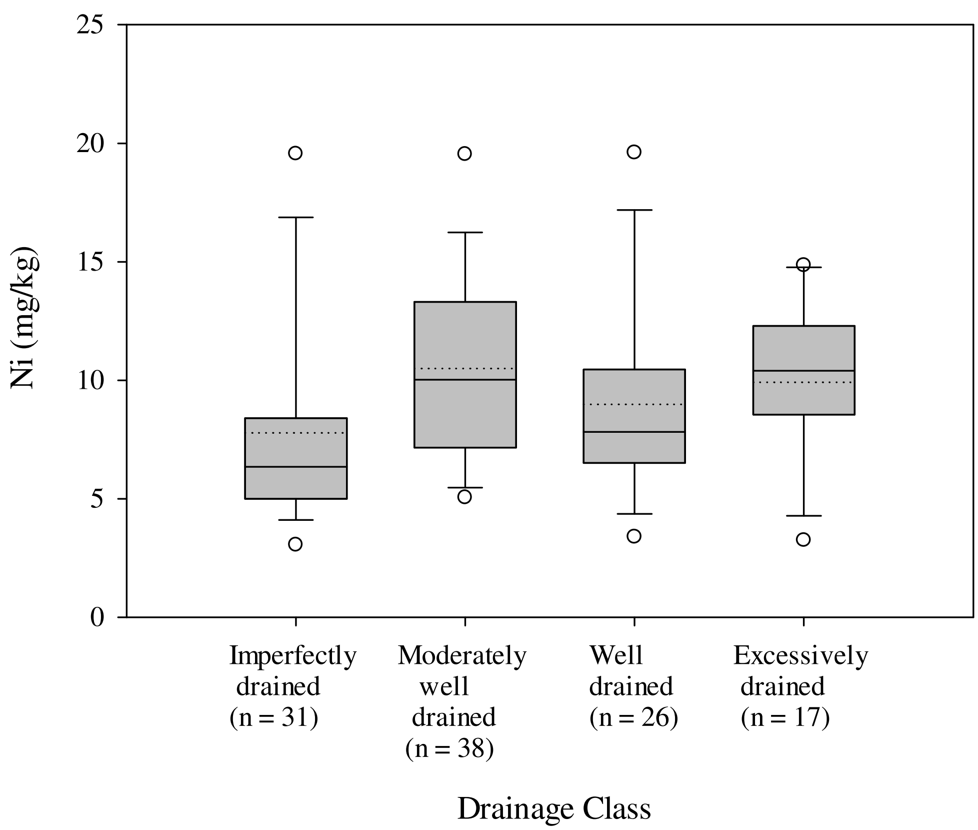

F = 3.25, p < 0.05
B (mg/kg)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
ab
ab
a
b
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class


F = 6.66, p < 0.001
Cr (mg/kg)
80
70
60
50
40
30
20
10
0
b
a
ab
b
Imperfectly drained (n = 30)
Moderately well drained (n = 37)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class


25
20
15
10
5
0
Cu (mg/kg)
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class


50000
40000
30000
20000
10000
0
Fe (mg/kg)
F = 7.10, p < 0.001
a
ab
b
b
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class


F = 3.92, p < 0.05
Mn (mg/kg)
1600
1200
800
400
0
ab
b
ab
a
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class


Ni (mg/kg)
25
20
15
10
5
0
Imperfectly drained (n = 31)
Moderately well drained (n = 38)
Well drained (n = 26)
Excessively drained (n = 17)
Drainage Class

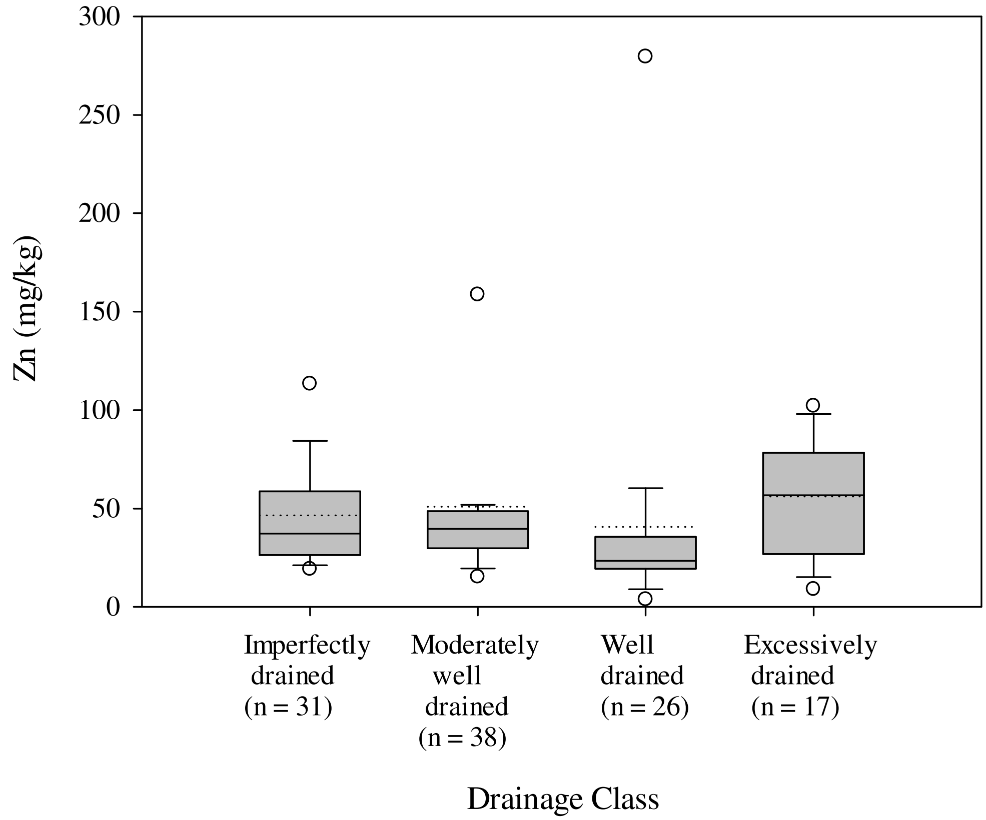


Figure 4.5: Box and whisker plots showing concentrations of different elements in tea garden soils of different drainage class. The boxplots indicate the lower and upper quartile (box), the median (solid line), the mean (dotted line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and *p*-value represent the F-ratio that indicates the degree of variation among the drainage class and the level of significance of this variation, respectively, from one-way analysis of variance (ANOVA) test.

4.4.6 Variabilities in soil properties within different soil textural classes

Table 4.2: Textural classes of the tea garden soils.

Name of tea garden	Soil textural class	%Sand	%Silt	%Clay
Allynugger	Sandy Clay Loam	58.07	16.53	25.40
	Clay Loam	33.08	34.81	32.11
Bilashcherra	Sandy Clay Loam	66.03	9.55	24.42
	Sandy Loam	74.35	9.29	16.36
BTRI	Sandy Clay Loam	54.15	21.84	24.01
	Sandy Loam	70.47	12.73	16.80
Jagcherra	Sandy Clay Loam	61.59	10.15	28.26
	Loam	54.66	17.41	27.93
Karnafuli	Sandy Clay Loam	64.22	11.75	24.03
	Sandy Loam	66.37	14.33	19.31
Karotoa	Sandy Clay Loam	58.04	18.42	23.55
	Sandy Loam	67.90	14.30	17.79
Kazi & Kazi	Sandy Clay Loam	59.86	17.90	22.24
	Sandy Loam	71.97	11.19	16.84
	Loamy Sand	82.77	8.08	9.15
Malnicherra	Sandy Clay Loam	65.42	14.09	20.49
	Sandy Loam	75.30	9.36	15.33
Neptune	Sandy Clay Loam	59.25	15.43	25.32
	Sandy Loam	67.4	17.02	15.58
	Clay Loam	44.62	19.84	35.55
	Loam	51.29	33.11	15.61
Oodaleah	Sandy Clay Loam	55.69	19.76	24.55
	Sandy Loam	68.03	18.68	13.28
	Clay Loam	44.67	19.82	35.51
	Clay	31.07	25.23	43.7
	Loam	39.78	42.79	17.43
Patrakhola	Sandy Clay Loam	53.42	22.53	24.05
	Clay Loam	37.94	28.33	33.73
	Loam	38.92	35.63	25.45
Rajghat	Sandy Clay Loam	55.42	17.64	26.94
	Sandy Loam	59.99	20.52	19.49
	Clay Loam	27.05	38.34	34.60
Sathgao	Sandy Clay Loam	62.36	13.19	24.45
	Sandy Loam	71.66	10.22	18.12
	Clay Loam	38.96	23.28	37.76

The pH in the surface soils of the tea gardens with different textural classes varied significantly ($^{ANOVA}F = 3.42$, $p < 0.01$), with the maximum mean pH in loamy sand texture (5.40), followed by sandy loam (5.11), sandy clay loam (4.82), loam (4.74), clay loam (4.56) and clay texture (3.80) soils (Figure 4.6). The pH in the sub-surface soils of the tea gardens with different textural classes also varied significantly with the maximum mean pH in loam (6.00) compared to other textural classes (Figure A36). In the present study lowest pH was found in the surface of the clay texture soil. It might be due to the high content of fine particles and high water absorption which reduced soil pH. Soil texture is significantly related to soil pH (Wang *et al*., 2021).

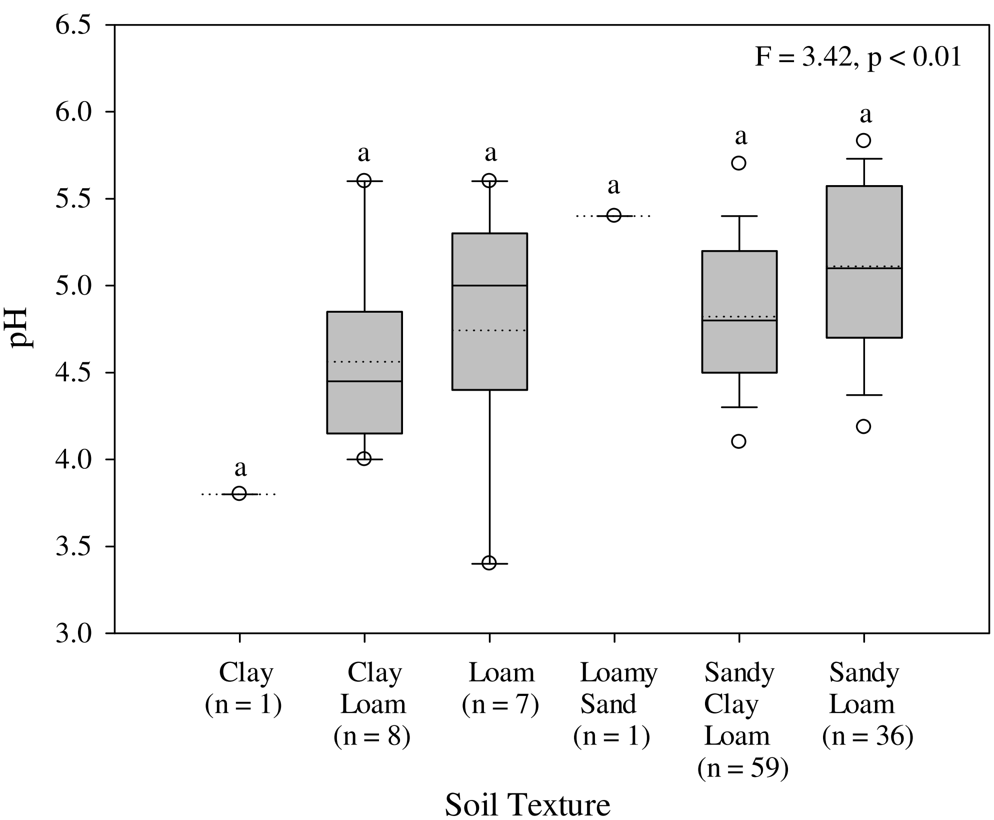
The concentrations of boron in the surface soils of the tea gardens with different textural classes varied significantly ($^{ANOVA}F = 2.42$, $p < 0.05$), with the maximum mean concentration of boron in loamy sand texture (1.12 mg/kg), followed by clay (0.70 mg/kg), sandy loam (0.67 mg/kg), sandy clay loam (0.42 mg/kg), clay loam (0.26 mg/kg) and loam texture (0.15mg/kg) soils (Figure 4.6). The concentrations of iron in the surface soils of the tea gardens with different textural classes ($^{ANOVA}F = 2.59$, $p < 0.05$), with the maximum mean concentration of iron in clay texture (38420 mg/kg), followed by loamy sand (22220 mg/kg), sandy clay loam (17258 mg/kg), clay loam (17242 mg/kg), sandy loam (16021 mg/kg) and loam texture (13333 mg/kg) soils (Figure 4.6). The concentrations of iron in the sub-surface soils of the tea gardens with different textural classes also varied significantly with the maximum mean iron in clay (52540 mg/kg) compared to other textural classes (Figure A37).

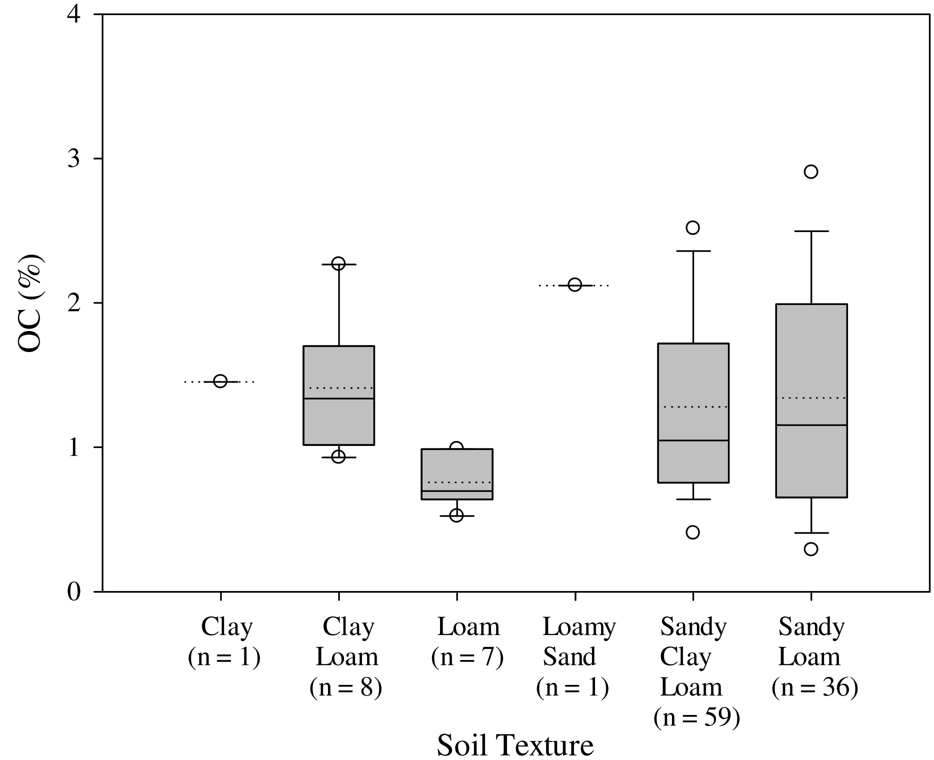
The concentrations of manganese in the surface soils of the tea gardens with different textural classes varied significantly ($^{ANOVA}F = 3.25$, $p < 0.01$), with the maximum mean concentration of manganese in clay (955 mg/kg), followed by clay loam (315 mg/kg), sandy clay loam (210.20 mg/kg), loamy sand (199 mg/kg), sandy loam (197.40 mg/kg) and loam texture (189.20 mg/kg) soils (Figure 4.6). The concentrations of manganese in the sub-surface soils of the tea gardens with different textural classes also varied significantly with the maximum mean manganese in clay (466 mg/kg) compared to other textural classes (Figure A38). In the present study the highest concentration of manganese was found in surface of the clay texture soil. It might be due to the strong correlation of manganese with the clay content of soil (Sharma *et al*., 2016).

The concentrations of nickel in the surface soils of the tea gardens with different textural classes varied significantly ($^{ANOVA}F = 3.00$, $p < 0.05$), with the maximum mean concentration of nickel in clay texture (19.35 mg/kg), followed by clay loam (12.87 mg/kg), loamy sand

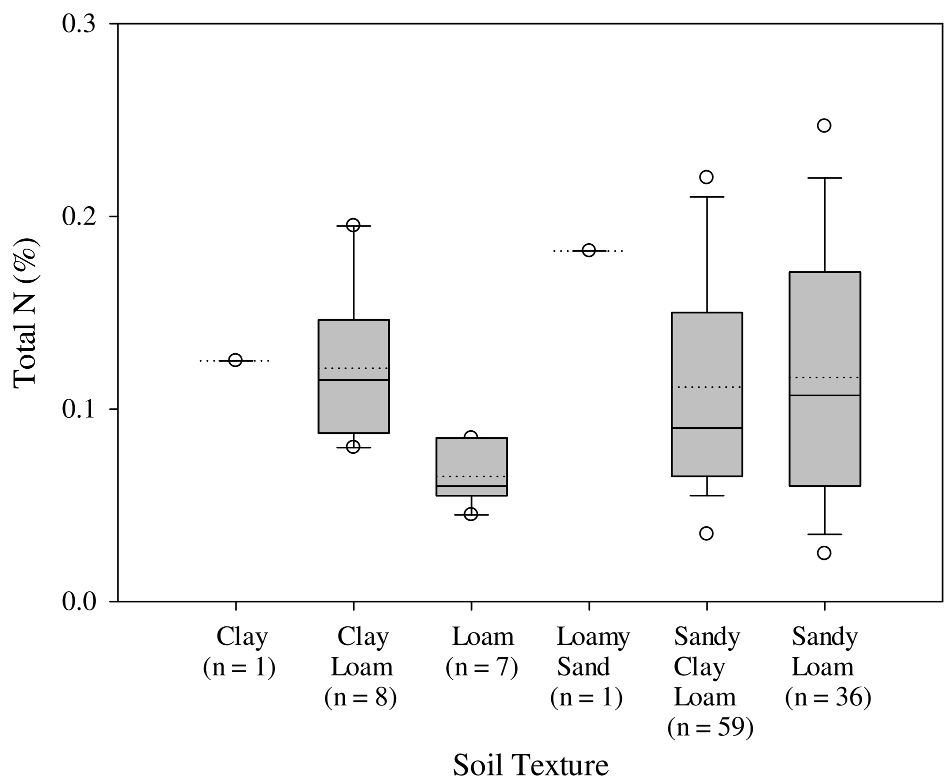
(11.15 mg/kg), sandy clay loam (9.31 mg/kg), sandy loam (8.61 mg/kg) and loam texture (7.13 mg/kg) soils (Figure 4.6). The concentrations of nickel in the sub-surface soils of the tea gardens with different textural classes also varied significantly with the maximum mean nickel in loam (23.70 mg/kg) compared to other textural classes (Figure A39). In the present study, the highest average concentration of nickel was found in the surface of clay texture soil. It might be due to the higher clay content, lower pH and higher organic matter content which are the major factor controlling solubility, mobility and sorption of nickel (Iyaka, 2021).

The concentrations of zinc in the surface soils of the tea gardens with different textural classes varied significantly ($^{ANOVA}F = 10.59$, $p < 0.001$), with the maximum mean concentration of zinc in clay texture (394.60 mg/kg), followed by loamy sand (49.59 mg/kg), clay loam (46.40 mg/kg), sandy clay loam (46.05 mg/kg), sandy loam (44.77 mg/kg) and loam texture (35.36 mg/kg) soils (Figure 4.6). The concentrations of zinc in the sub-surface soils of the tea gardens with different textural classes also varied significantly with the maximum mean zinc in loam (63.61 mg/kg) compared to other textural classes (Figure A40).

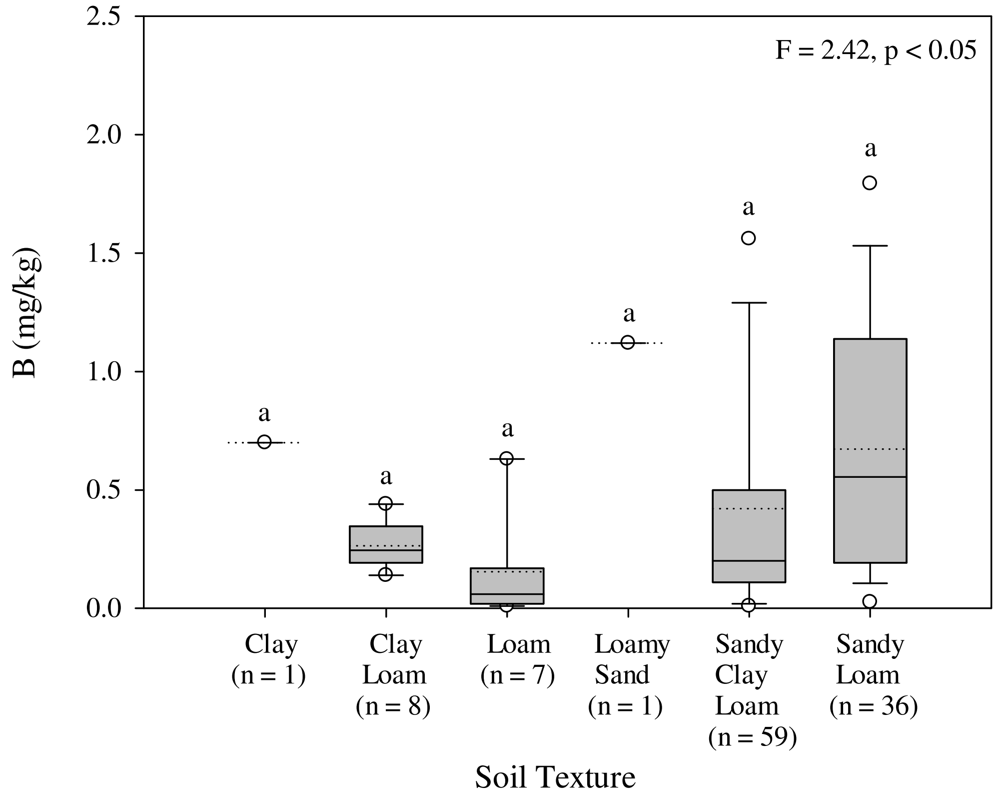


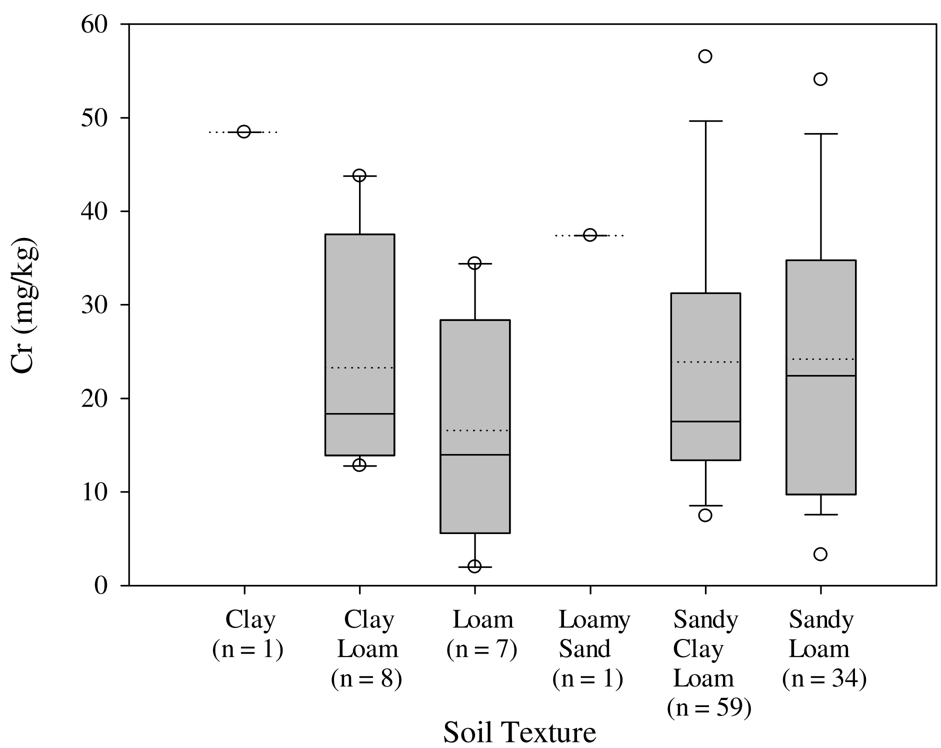


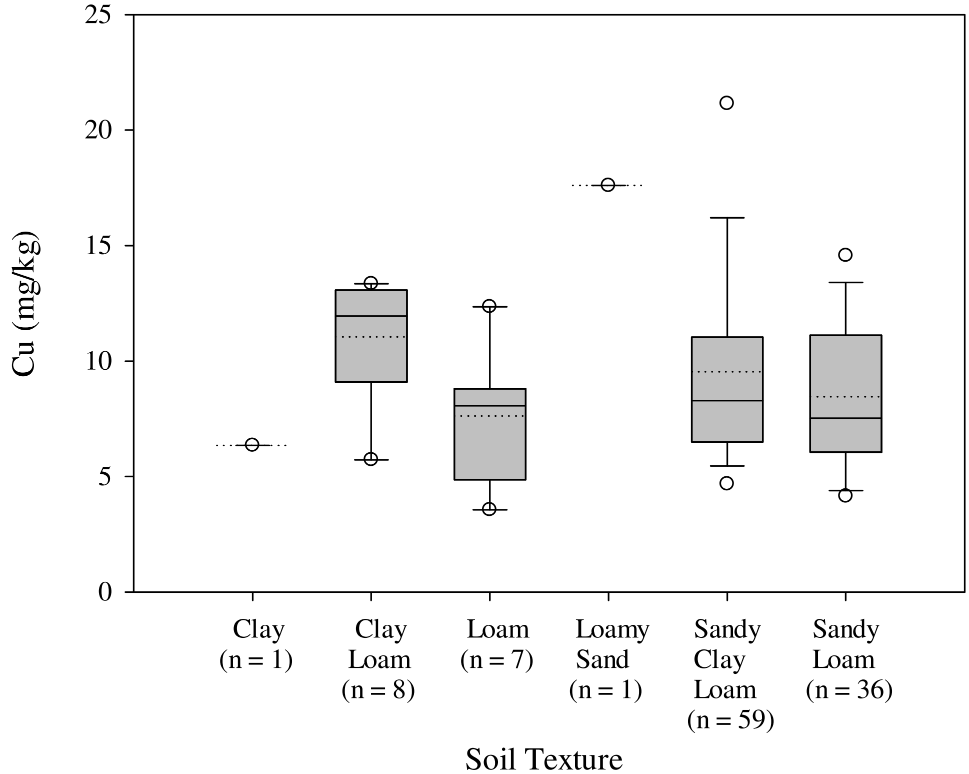
4
3
2
1
0
OC (%)
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture



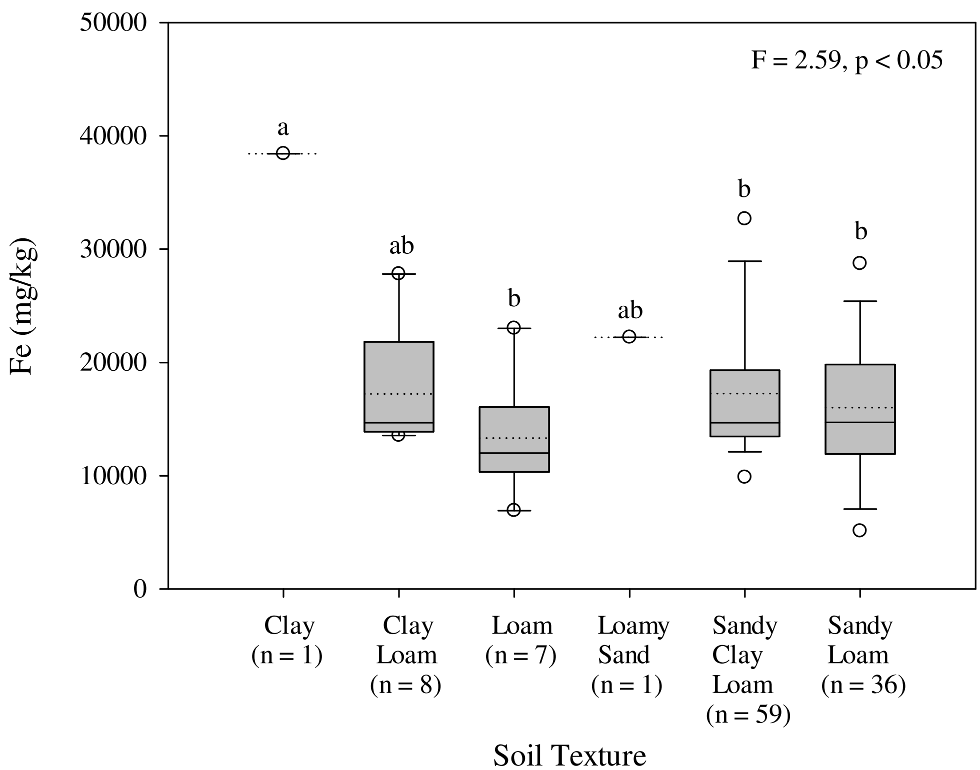
0.3
0.2
0.1
0.0
Total N (%)
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture


F = 2.42, p < 0.05
B (mg/kg)
2.5
2.0
1.5
1.0
0.5
0.0
a
a
a
a
a
a
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture

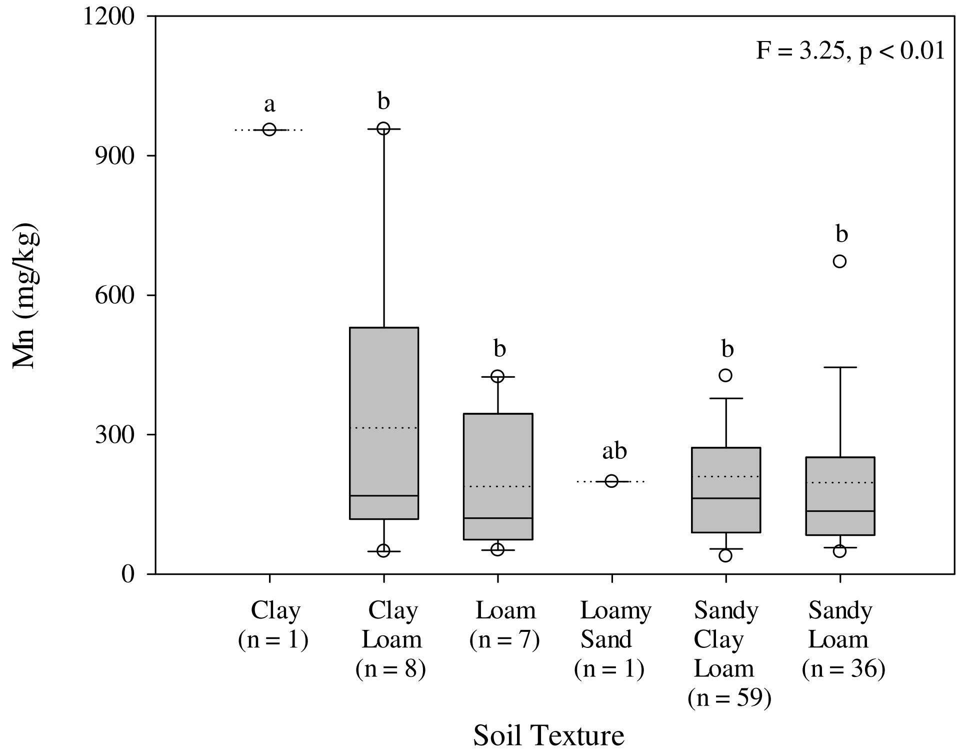

Cr (mg/kg)
60
50
40
30
20
10
0
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 34)
Soil Texture

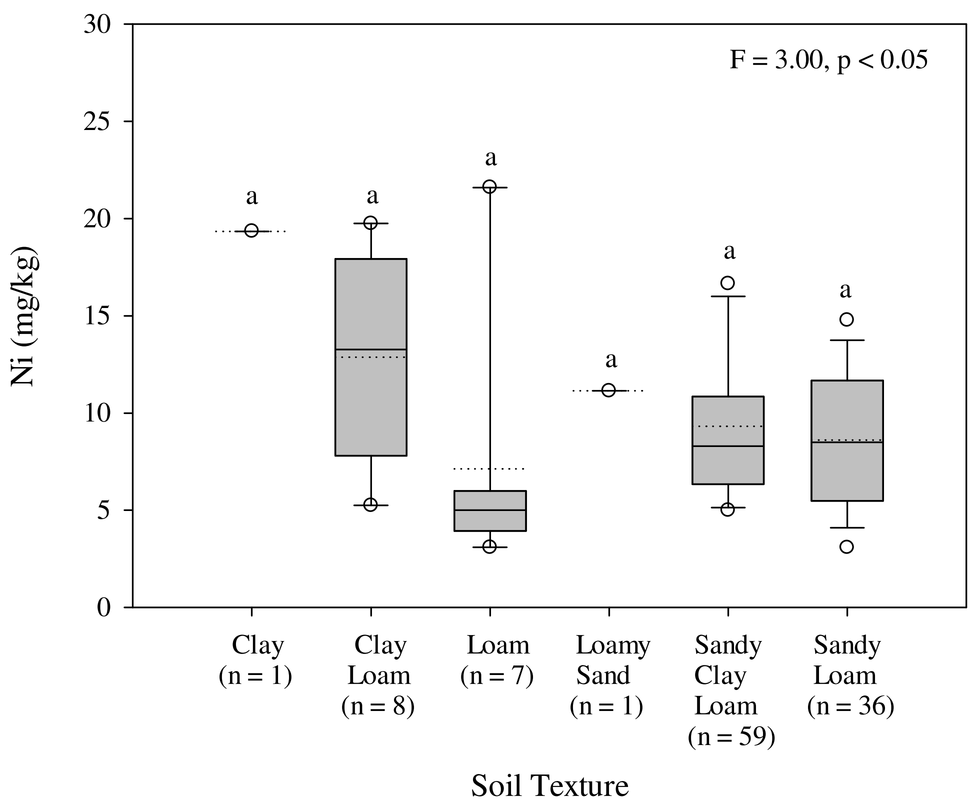


25
20
15
10
5
0
Cu (mg/kg)
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture



50000
40000
30000
20000
10000
0
Fe (mg/kg)
F = 2.59, p < 0.05
a
ab
b
ab
b
b
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture


F = 3.25, p < 0.01
Mn (mg/kg)
1200
900
600
300
0
a
b
b
ab
b
b
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture


F = 3.00, p < 0.05
Ni (mg/kg)
30
25
20
15
10
5
0
a
a
a
a
a
a
Clay (n = 1)
Clay Loam (n = 8)
Loam (n = 7)
Loamy Sand (n = 1)
Sandy Clay Loam (n = 59)
Sandy Loam (n = 36)
Soil Texture

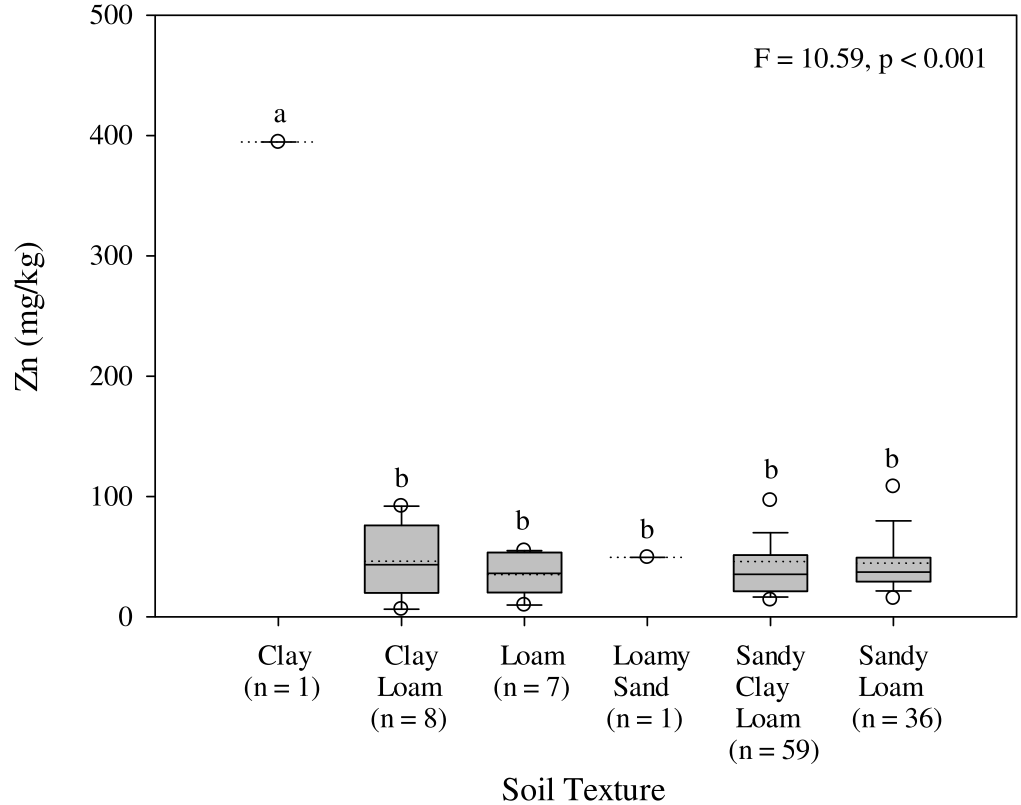


Figure 4.6: Box and whisker plots showing concentrations of different elements in tea garden soils of different textural class. The boxplots indicate the lower and upper quartile (box), the median (solid line), the mean (dotted line), the 10^{th} and 90^{th} percentiles (whiskers) and the 5^{th} and 95^{th} percentiles (circles). The F-value and *p*-value represent the F-ratio that indicates the degree of variation among the textural class and the level of significance of this variation, respectively, from one-way analysis of variance (ANOVA) test.

4.5 Conclusion

The concentrations of the heavy metals (chromium, copper, iron, manganese, nickel and zinc) varied in the tea garden soils of the three different physiographic regions. The concentrations of chromium, copper, iron and nickel are higher in Old Himalayan Piedmont Plain and varied significantly compared to other physiographic regions. The tea garden soils within different age of tea plants also have geochemical differences. The concentrations of heavy metals are higher in the soils of young tea compared to other age of tea plants. Among the heavy metals only chromium concentrations are varied significantly. In the soils of different soil series as well as within the soils with different landforms, drainage classes and textural classes also have geochemical differences. In the Baliadangi series the concentrations of chromium, copper and iron are higher compared to other soil series. The concentrations of manganese and zinc are higher in the Datmara series while nickel concentrations are higher in the Pahartoli series. In the level ridge landform of tea gardens the concentrations of chromium, copper, iron and nickel are higher compared to other landforms. The concentrations of manganese are higher in medium high hill while the concentrations of zinc are higher in low hill landforms of tea plantations. In the moderately well drained condition of the tea gardens, the concentrations of chromium, copper, iron and nickel are higher compared to other drainage classes. The concentrations of manganese and zinc are higher in excessively drained condition. In the clay texture soil of the tea gardens, the concentrations of chromium, iron, manganese, nickel and zinc are higher compared to other soil textural classes. The concentrations of copper are higher in the loamy sand texture of the tea garden soils. The present study revealed that the soils of the different tea gardens at the three different physiographic regions had variabilities in their elemental concentrations which were also related to other geomorphological differences. This study suggests further investigation to better appreciate whether the geomorphological variabilities have any effects on the accumulation of the heavy metals into tea plants.

Chapter Five

Assessing the effects of Heavy metals in tea garden soils on the Quality of tea in Bangladesh

Abstract

Tea leaves and made tea samples from the tea gardens of different tea growing areas of Bangladesh were analyzed to assess their qualities in terms of heavy metal concentrations. The effects of heavy metals on the concentrations of a range of biochemical parameters of made tea were also evaluated, and the relationships of heavy metals in the tea leaves and made tea were assessed. The mean concentrations of chromium, copper, iron, manganese, nickel and zinc in the tea leaves and made tea were 12.86, 21; 14.67, 17.51; 185, 373; 641, 613; 4, 6.29 and 43.79, 134 mg/kg, respectively. The concentrations of copper, manganese and zinc in the tea leaves were found to be significantly different ($p < 0.001$) among the tea gardens. The concentrations of iron, manganese, nickel, and zinc in the made tea were also found to be significantly different ($p < 0.001$) among the tea gardens. Significant and positive relationships of copper and zinc ($R^2 = 0.17$, $p < 0.001$), iron and nickel ($R^2 = 0.08$, $p < 0.05$), manganese and nickel ($R^2 = 0.04$, $p < 0.05$) were found in the tea leaves. Significant and positive relationships of iron and nickel ($R^2 = 0.22$, $p < 0.01$), manganese and nickel ($R^2 = 0.31$, $p < 0.001$), manganese and zinc ($R^2 = 0.22$, $p < 0.01$), zinc and caffeine ($R^2 = 0.27$, $p < 0.001$) were observed in the made tea. Significant and negative relationships of manganese and thearubigin ($R^2 = 0.41$, $p < 0.001$), nickel and total polyphenol ($R^2 = 0.201$, $p < 0.01$), zinc and theaflavin ($R^2 = 0.202$, $p < 0.01$), zinc and thearubigin ($R^2 = 0.28$, $p < 0.01$) were observed in the made tea. The concentrations of biochemical parameters in made tea such as caffeine ($p < 0.001$), protein ($p < 0.001$), total polyphenol ($p < 0.001$), theaflavin ($p < 0.05$) and thearubigin ($p < 0.001$) were found to be significantly different among the tea gardens of Bangladesh. Significant variations in the concentrations of copper ($p < 0.05$), iron ($p < 0.01$) and manganese ($p < 0.001$) in the tea leaves were observed among the physiographic regions of Bangladesh. The concentration of nickel was found to be significantly higher ($p < 0.05$) in the leaves of young tea plants. The soil to tea plant transfer factors for chromium, copper, iron, manganese, nickel and zinc were 0.72, 1.72, 0.009**,** 3.44, 0.44 and 1.15**,** respectively. The present study reveals that the concentrations of chromium and manganese in the tea leaves and made tea samples and zinc in the made tea samples were higher compare to the permissible limits in plant. The concentrations of copper, iron and nickel in the tea leaves and made tea samples and zinc in the tea leaves samples were within the permissible limits of heavy metals in plants. A larger scale investigation is also recommended to identify the origin and amount of different heavy metals in tea leaves across different tea plantations in Bangladesh and to relate this to the yield and quality of tea produced. This study also suggests further research to assess the release of toxic heavy metals from made tea into tea brew.

Key words: Heavy metals, tea leaf, made tea, tea quality, biochemical parameters.

5.1 Introduction

Tea (*Camellia sinensis L.*) is consumed by a large population in the world. Due to its medicinal properties and slight stimulating effects tea is one of the most admired drinks (Karak and Bhagat, 2010). Polyphenols in the tea leaves has numerous medicinal properties such as antioxidant (Li *et al.*, 2013), cholesterol-lowering (Srividhya *et al.*, 2011), hepatoprotective (Issabeagloo *et al.*, 2012) and anticancer activities (Dufresne and Farnworth, 2000). For the elimination of alcohol and toxins, its detoxifying properties are indispensable (Dufresne and Farnworth, 2000). The economic and social importance of tea is unparalleled considering its worldwide daily consumption of 18 billion cups of tea as drink (Achudume and Owoeye, 2010). There are different types of tea, such as black tea, green tea, oolong tea, white tea and yellow tea among which only black tea and green tea are produced in Bangladesh. Depending on the various technological processes, black tea recognized as fully fermented, while green and yellow tea recognized as non-fermented and oolong as well as white tea recognized as semi-fermented (de Oliveira *et al*., 2018). Theaflavins and thearubigins are the oxidation products produced by the fermentation of black tea during the oxidation process which represents the main difference between black and green tea. Approximately 20–30% polyphenols are contained in black tea (Mander and Lui, 2010). Phenolic compounds have antioxidant capacity and various health benefits which make it an important segment of human diet (Shahidi and Ambigaipalan, 2015).

Toxic levels of heavy metals may be increased through tea consumption (Bobková *et al.*, 2021). The non-decomposable character, extended biological half-lives, and obstinate gathering of heavy metals in various parts of human body create distress about the existence of heavy metals in tea (Sharma *et al.,* 2007). Tea from different countries, such as Sri Lanka (Pourramezani *et al*., 2019), Italy (Barone *et al*., 2016), Ghana (Nkansah *et al*., 2016), Brazil (Milani *et al*., 2016), India (Fung *et al*., 2009), and China (Wen *et al*., 2018; Peng *et al*., 2018) have been reported to contain higher concentrations of heavy metals. Tea plants have been found to be hyperaccumulator of manganese due to much higher contents of manganese in the tea leaves (Wen *et al*., 2018; Zhang *et al*., 2018). The concentrations and possible health risks of heavy metals such as chromium and nickel in tea leaves and made tea have been reported by several studies (Li *et al*., 2015; de Oliveira *et al*., 2018; Zhang *et al*., 2018; Sun *et al*., 2019). Contaminants such as heavy metals pose risks to human health and environment which leads to economic losses, and thus, have created a great interest of the safety of tea (Li *et al*., 2013). Cardiovascular, kidney and bone diseases may cause due to the deposition of heavy metals in the kidney and liver. Continued consumption of heavy metals increased risk of different cancers (Järup, 2003; Bower *et al*., 2005). While Bangladesh is one of the most important tea producing countries in the world and tea is consumed all over the country throughout the year (Nasir and Shamsuddoha, 2012), understanding the concentrations of heavy metals in tea produced in the country is highly necessary.

The sources of heavy metals in soil are from geological origin and anthropogenic activities. The deposition of toxic heavy metals by the use of chemical fertilizers, animal manures, sewage sludge, compost, pesticides; atmospheric deposition; and industrial processes may pollute soils. Heavy metals pollution may play an important role in influencing crop quality (Ngure and Kinuthia, 2020; Sungur *et al*., 2020). The age of the tea plants gradually increase the acidity of the tea garden soils. The application of nitrogen fertilizers, leaching of basic cations, root secretion of organic acids and decomposition of plant litters are perhaps responsible for the acidification of tea garden soils (Li *et al*., 2016; Yang *et al*., 2018; Yan *et al*., 2018; Li *et al*., 2019). In the soils of old tea plantations, lower pH was observed which may have potential to enhance the deposition of heavy metals in the tea plants (Zhang and Fang, 2007). The most important factors that can regulate the accumulations of heavy metals in tea plants are the physiological characteristics of the plant and process of metal absorption by tea plants, physicochemical properties of the soils and chemical diversification of metals in the soils (Li *et al*., 2017; Zhang *et al*., 2018). Besides geological origin and agricultural practices, heavy metals might also be introduced into the tea products during different technological processes such as CTC (cut, tear and curl), fermentation and drying processes of tea production (Seenivasan *et al*., 2008a; Brzezicha-Cirocka *et al*., 2016; Malik *et al*., 2013).

In the present study, we analysed 112 tea leaves and 39 made tea samples to assess the concentrations of heavy metals in tea leaves and made tea of different tea gardens of Bangladesh. The concentrations of heavy metals in leaves of different age of tea plants located at different sub-districts and physiographic regions of Bangladesh were also assessed. Made tea samples of different grades were analysed to determine the concentrations of heavy metals. The effects of heavy metals on the biochemical parameters of made tea as well as the relationships of the heavy metals in the tea leaves and made tea were also investigated. The transfer factors of chromium, copper, iron, manganese, nickel and zinc from soil to tea plants were also determined.

5.2. Objectives

The objectives of the research reported in this chapter are:

1. to determine the levels of heavy metals in tea leaves and made tea of different tea gardens of Bangladesh;
2. to assess the effects of heavy metals on the concentrations of biochemical parameters of made tea;
3. to determine the relationships of heavy metals in the tea leaves and made tea; and,
4. to estimate the transfer factors of heavy metals from soil to tea plants;

5.3 Materials and Methods

A detail of the *Materials and Methods* for this chapter was described in chapter two (General Methodology).

5.4 Results and Discussions

5.4.1 Heavy metals in the tea leaves and made tea of Bangladesh

The concentrations of heavy metals in the tea leaves and made tea of Bangladesh are presented in the Table 5.1. The highest concentration of chromium in the tea leaves was found to be 77.85 mg/kg while the mean concentration of chromium was found 12.86 mg/kg. The highest concentration of chromium in the made tea was found 174 mg/kg, while the mean concentration was found 20.99 mg/kg (Table 5.1). Rashid *et al.* (2016) reported that the levels of chromium in the fresh tea leaves were below the detection limit, while made tea contained 0.45 to 10.73 mg/kg of chromium in Bangladesh. Wolf (2021) observed the mean concentrations of chromium 8.8 and 9.6 mg/kg, respectively, in packet made tea and loose-leaf made tea in Bangladesh. Nargis and Chowdhury (2020) reported 30.50 mg/kg chromium in infused tea (made tea after use).

The concentration of chromium in Japanese, Chinese, Iranian and Thai green tea leaves were reported 0.024, 0.14, 0.05 and 0.06 mg/kg, respectively (Limmatvapirat *et al*., 2012; Cabrera *et al*., 2003). In black tea from India, China, Sri Lanka and Turkey, the concentrations of chromium were found to be 0.371, 0.155, 0.050 and 3.000 mg/kg, respectively, as reported by Cabrera *et al*. (2003) and Soylak and Aydin, (2011). Seenivasan *et al.* (2008a) reported that the CTC (cut, tear and curl) rollers used in the manufacturing of black tea are made of stainless-steel. The stainless-steel contain chromium (17% w/w) which is the source of chromium contamination in black tea. There is no specific critical limit for chromium in tea. Dabanović *et al.* (2016) reported that the permissible limit of chromium in plants is 1.3 mg/kg, whereas, in the present study, the mean concentrations of chromium in tea leaves and made tea were observed to be 12.86 and 20.99 mg/kg, respectively.

The concentrations of copper in the tea leaves were found to be ranged from 2.15 to 31.41 mg/kg (mean, 14.67 mg/kg). The concentrations of copper in the made tea were found ranged from 2.45 to 60.35 mg/kg (mean, 17.51 mg/kg) (Table 5.1).

Table 5.1: The concentration of heavy metals (mg/kg) in the tea leaves and made tea of Bangladesh.

Heavy metals	Sample	Sample No	Mean±*s.e.*	Min	Median	Maximum
Chromium (Cr)	Tea Leaf	112	12.86±1.04	BDL	11.40	77.85
	Made Tea	39	20.99±4.69	BDL	16.85	174
Copper (Cu)	Tea Leaf	112	14.67±0.44	2.15	14.48	31.41
	Made Tea	39	17.51±1.41	2.45	15.65	60.35
Iron (Fe)	Tea Leaf	112	185±68.3	15.00	84.10	7300
	Made Tea	39	373±84.5	3.00	205	2274
Manganese (Mn)	Tea Leaf	112	641±42.2	96	546	2006
	Made Tea	39	613±50.2	183	481	1240
Nickel (Ni)	Tea Leaf	112	3.99±0.21	0.50	3.55	11.50
	Made Tea	39	6.29±0.41	1.95	6.30	11.25
Zinc (Zn)	Tea Leaf	112	43.79±2.51	1.17	35.21	163
	Made Tea	39	134±22.0	14.40	40.40	541

BDL = Below detection limit.

Wolf (2021) reported the mean concentration of copper in the packet made tea and loose-leaf made tea was 23.00 mg/kg and 35.00 mg/kg, respectively in Bangladesh. Nargis and Chowdhury (2020) observed the concentration of copper in the infused tea was 17.40 mg/kg. The concentrations of copper in the tea leaves of Yuyao County, China were ranged from 8.05 to 33.50 mg/kg (Jin *et al*., 2008). Seenivasan *et al*. (2008b) observed the mean concentration of copper ranged from 15.9 to 32.2 mg/kg in South India. The permissible limit of copper in plants is 10 mg/kg reported by Dabanović *et al.* (2016). According to Food Safety & Standard Authority in India (FASSI), the standard limit of copper specific only for tea is 150 mg/kg (Jana *et al*., 2017), whereas, in the present study, the mean concentrations of copper in the tea leaves and made tea were observed to be 14.67 and 17.51 mg/kg, respectively. Use of copper containing fungicides in the tea gardens might be the reason for the presence of high levels of copper in the tea leaves and made tea (Seenivasan *et al*., 2008b).

The concentrations of iron in the tea leaves were found to be ranged from 15 to 7300 mg/kg (mean, 185 mg/kg). The concentrations of iron in the made tea were found ranged from 3 to 2274 mg/kg (mean, 373 mg/kg) (Table 5.1). Kotoky *et al*., (2013) reported that the iron concentration of tea leaves ranged from 69.20 to 85.60 mg/kg (mean, 80 mg/kg) in Assam, India. In Kenya, the concentrations of iron in tea leaves were observed to be ranged from 55 to 203 mg/kg (Moseti *et al*., 2013). Wolf (2021) reported that the mean iron concentrations in the packet made tea and loose-leaf made tea were 268 and 410 mg/kg, respectively in Bangladesh. Street *et al.* (2006) observed the concentrations of iron ranged from 103 to 523 mg/kg in made tea samples of different origins (black tea samples from India, Sri Lanka, China, Kenya; green tea samples from India, Japan, China, Vietnam, Indonesia; semi-fermented tea and white tea). In the current research, the higher concentration of iron in the tea leaves might be due to the higher concentration of iron in the tea garden soils and transfer of iron to the tea leaves. The permissible limit of iron in plants is 450 mg/kg reported by Dabanović *et al.* (2016), whereas, in the present study, the mean concentrations of iron in tea leaves and made tea were observed to be 185 and 373 mg/kg, respectively. Among the trace elements the major sources of iron is tea. Iron gets solubilized in the acid soil and due to the acidophilic nature of the tea plants iron potentially available to the tea plants (Karak *et al*., 2016).

The concentrations of manganese in the tea leaves were found to be ranged from 96 to 2006 mg/kg (mean, 641 mg/kg). The concentrations of manganese in the made tea were found to be ranged from 183 to 1240 mg/kg (mean, 613 mg/kg) (Table 5.1). AL-Oud (2003) reported that the concentrations of manganese in tea leaves were ranged from 390 to 900 mg/kg beacuse tea plant itself have the capacity to deposit higher concentrations of manganese. Chen *et al*. (2009) observed that the concentration of manganese varied within the range of 950 to 1224 mg/kg in eight different tea cultivars. Wolf (2021) reported that the mean concentrations of manganese in the packet made tea and loose-leaf made tea were 470 and 866 mg/kg, respectively in Bangladesh. The concentrations of manganese were observed to be varied from 371 to 758 mg/kg (mean, 575 mg/kg) in fifteen different brands of Indian black tea reported by Kumar *et al.* (2005). Fernandez-Caceres *et al*. (2001) observed that the concentrations of manganese ranged from 148 to 1595 mg/kg (mean, 825 mg/kg) in forty-six commercial made tea samples in Spain. The permissible limit of manganese in plants is 200 mg/kg reported by Dabanović *et al.* (2016), whereas, in the present study, the mean concentrations of manganese in tea leaves and made tea were observed to be 641 and 613 mg/kg, respectively.

The concentrations of nickel in the tea leaves were observed to be ranged from 0.50 to 11.50 mg/kg (mean, 3.99 mg/kg). The concentrations of nickel in the made tea were found to be ranged from 1.95 to 11.25 mg/kg (mean, 6.29 mg/kg) (Table 5.1). Zhang *et al.* (2018) reported that the concentrations of nickel in the tea leaves were varied from 3.43 to 14.90 mg/kg (mean, 9.44 mg/kg). In the tea leaves, the concentrations of nickel were varied from 4.26 to 24.08 mg/kg (mean, 14.03 mg/kg) reported by Wen *et al.* (2018). The concentration of nickel in the tea leaves of south India was found 2.59 mg/kg (Seenivasan *et al*., 2016). Wolf (2021) reported that the mean concentrations of nickel in the packet made tea and loose-leaf made tea were observed below detection limit in Bangladesh. Moreda-Piñeiro *et al.* (2003) reported that the concentrations of nickel in Chinese tea samples were 4.92 mg/kg, Indian tea and Sri Lankan tea samples were 4.08 mg/kg. In Asian and African tea, the mean concentration of nickel was found 4.24 and 4.76 mg/kg, respectively (Moreda-Piñeiro *et al*., 2003). The permissible limit of nickel in plants is 10 mg/kg reported by Dabanović *et al.* (2016), whereas, in the present study, the mean concentrations of nickel in tea leaves and made tea were observed to be 3.99 and 6.29 mg/kg, respectively. Since higher levels of nickel is toxic to plants and human health, the agrochemicals used in tea gardens should be analyzed for the element as well as any other toxic components prior to application.

The concentrations of zinc in tea leaves were found to be ranged from 1.17 to 163 mg/kg (mean, 43.79 mg/kg). The concentrations of zinc in the made tea were observed to be ranged from 14.40 to 541 mg/kg (mean, 134 mg/kg) (Table 5.1). Kotoky *et al.* (2013) reported that the concentrations of zinc in the tea leaves of Assam, India were ranged from 53.55 to 74.26 mg/kg (mean, 69.16 mg/kg). The concentrations of zinc in the tea leaves of Kenya were ranged from 15.4 to 32.6 mg/kg reported by Moseti *et al.* (2013). In the tea leaves of Puan tea area of southwestern China, the concentrations of zinc were ranged from 9.1 to 50.3 mg/kg (mean, 42.2 mg/kg) (Zhang *et al*., 2018). The concentrations of zinc in the made tea of Assam, India were ranged from 20.5 to 39.26 mg/kg (Tamuly *et al.,* 2016). Srividhya *et al.* (2011) reported that the mean concentration of zinc in the made tea of South India was 25.39 mg/kg. Street *et al*. (2006) observed the concentrations of zinc ranged from 21.5 to 82.2 mg/kg in made tea samples of different origins (black tea samples from India, Sri Lanka, China, Kenya; green tea samples from India, Japan, China, Vietnam, Indonesia; semi-fermented tea and white tea). Wolf (2021) reported that the mean zinc concentrations in the packet made tea and loose-leaf made tea were 34.00 and 45.00 mg/kg, respectively in Bangladesh. The permissible limit of zinc in plants is 50 mg/kg reported by Dabanović *et al.* (2016), whereas, in the present study, the mean concentrations of zinc in tea leaves and made tea were observed to be 43.79 and 134 mg/kg, respectively. The higher concentrations of zinc in the tea leaves and made tea might be due to the foliar application of zinc sulphate in the tea gardens of Bangladesh.

5.4.2 Heavy metals in the tea leaves and made tea of different tea gardens of Bangladesh

The concentration of heavy metals in the tea leaves and made tea of different tea gardens of Bangladesh are presented in table 5.2. The highest mean concentration of chromium was found in tea leaves of Rajghat tea garden (21.31 mg/kg) and the lowest mean chromium concentration was found in the tea leaves of Allynugger tea garden (4.18 mg/kg). The highest mean concentration of chromium was found in made tea of BTRI and Bilashcherra tea gardens (56.9 mg/kg) and the lowest mean chromium concentration was found in the made tea of Jagcherra tea garden (2.35 mg/kg) (Table 5.2). Significantly different ($p < 0.001$) concentrations of chromium were found in the soils of the tea gardens. No significant variations of the concentrations of chromium were observed in the tea leaves and made tea of the different tea gardens (Figure 5.1). Chromium was found to be significantly related with manganese ($R^2 = 0.09$, $p < 0.001$) in the tea leaves (Figure 5.7). No significant relationships of chromium with the biochemical parameters of made tea such as caffeine, protein, total polyphenol, theaflavin and thearubigin were observed (Figure 5.9 – 5.13).

The highest mean concentration of copper (26.68 mg/kg) was found in tea leaves of BTRI tea garden and the lowest mean copper concentration (10.56 mg/kg) was found in tea leaves of Oodaleah tea garden. The highest mean concentration of copper was found in made tea of Kazi & Kazi tea garden (25.80 mg/kg) and the lowest mean concentration of copper was found in the made tea of Neptune tea garden (10.47 mg/kg) (Table 5.2). Significantly different ($p < 0.001$) concentrations of copper were observed in the soils and leaves of the tea gardens. No significant variations of the concentrations of copper were observed in the made tea of the different tea gardens (Figure 5.2). Significant and positive relationships of copper with zinc ($R^2 = 0.17$, $p < 0.001$) were observed in the tea leaves (Figure 5.7). No significant relationships of copper with the biochemical parameters such as caffeine, protein, total polyphenol, theaflavin and thearubigin were observed in the made tea (Figure 5.9 – 5.13).

Table 5.2: Concentration of heavy metals in the tea leaves and made tea of different tea gardens of Bangladesh

Tea Estate	Sample		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Allynugger	Tea Leaf	*Min – Max*	2.10 – 6.95	12.42 – 17.99	65.12 – 99.75	231.8 – 738.2	2.25 – 11.50	28.30 – 121.60
	(n = 9)	*Mean±s.e.*	4.18±0.54	14.69±0.52	79±4.4	422.4±65.7	4.10±0.98	55.51±9.61
	Made Tea	*Min – Max*	6.70 –11.40	12.04 –13.57	188 – 1408	409.3 – 466	4.45 – 6.45	35 – 541
	(n = 3)	*Mean±s.e.*	8.97±1.36	12.69±0.46	610±399	434.9±16.7	5.67±0.62	267±148
Bilashcherra	Tea Leaf	*Min – Max*	1.15 – 19.70	2.15 –18.75	57.35 – 207.80	249.0 – 1144.0	2.45 – 10.30	1.17 – 43.24
	(n = 18)	*Mean±s.e.*	9.24±1.36	12.29±1.16	113.22±7.50	732.6±53.6	5.55±0.56	27.31±2.42
	Made Tea	*Min – Max*	10.0 – 173.5	2.45 – 16.15	36.0 – 87.0	183.0 – 313.0	2.55 – 4.10	21.97 – 27.18
	(n = 4)	*Mean±s.e.*	56.9±39.0	12.24±3.28	68.3±11.8	228.0±29.0	3.26±0.32	25.34±1.19
BTRI Farm	Tea Leaf	*Min – Max*	6.65 – 25.20	20.77 – 31.41	27.3 – 130.3	150.3 – 698.0	1.75 – 8.55	53.85 – 99.85
	(n=5)	*Mean±s.e.*	14.30±4.36	26.68±2.43	92.7±17.3	358.8±99.8	3.84±1.23	78.67±8.69
	Made Tea	*Min – Max*	10.0 – 173.5	2.45 – 16.15	36.0 – 87.0	183.0 – 313.0	2.55 – 4.10	21.97 – 27.18
	(n = 4)	*Mean±s.e.*	56.9±39.0	12.24±3.28	68.3±11.8	228.0±29.0	3.26±0.32	25.34±1.19
Jagcherra	Tea Leaf	*Min – Max*	9.45 –15.15	13.53 –19.99	46.77 – 90.25	347 – 1880	2.30 – 10.80	24.75 – 86.95
	(n = 9)	*Mean±s.e.*	12.26±0.63	16.62±0.72	72.28±4.67	1287±172	4.66±0.87	55.13±8.10
	Made Tea	*Min – Max*	2.05 – 2.90	19.06 – 23.71	106.1 – 281.4	479.4 – 761.5	2.45 – 4.40	294.8 – 395.5
	(n = 3)	*Mean±s.e.*	2.35±0.28	20.89±1.43	176.6±53.4	666±93	3.60±0.59	331±32.5
Karnafuli	Tea Leaf	*Min – Max*	BDL – 14.80	8.05 – 12.80	69.60 –138.25	233 – 1047	1.30 – 4.30	23.82 – 48.38
	(n=7)	*Mean±s.e.*	4.59±2.37	10.61±0.62	95.40±8.60	528±105	2.11±0.39	34.79±3.65
	Made Tea	*Min – Max*	17.75 –26.80	13.15 – 29.45	16.0 – 214.0	737 – 806.0	5.50 – 7.45	20.19 – 33.91
	(n = 3)	*Mean±s.e.*	22.20±2.61	18.93±5.27	97.7±59.7	778±21	6.62±0.58	25.50±4.25
Karotoa	Tea Leaf	*Min – Max*	6.50 –11.35	8.45 – 19.30	18 –2363	143 – 266	2.25 –7.35	22.41 – 35.30
	(n=6)	*Mean±s.e.*	8.48±0.68	13.02±1.63	611±389	195±19.3	3.86±0.78	28.89±1.85
	Made Tea	*Min – Max*	BDL – 45.3	12.10 –28.70	926 – 2274	442.0 – 477.0	8.75 – 9.75	36.87 – 44.64
	(n = 3)	*Mean±s.e.*	30.0±15.0	18.23±5.26	1791±434	461.3±10.3	9.13±0.31	40.63±2.24
Kazi & Kazi	Tea Leaf	*Min – Max*	4.65 – 77.85	5.95 – 17.35	15 – 7300	143.0 – 715.0	2.00 –9.45	28.95–37.03
	(n=12)	*Mean±s.e.*	16.32±6.24	14.21±0.91	748±600	305.8±48.5	3.71±0.60	32.43±0.87
	Made Tea	*Min – Max*	0.55 –31.30	15.15 – 60.35	102.4 –394.0	207 – 312	1.95 –7.75	28.38 – 40.78
	(n = 5)	*Mean±s.e.*	15.47±5.43	25.80±8.72	201.7±51.7	254.8±19.5	4.66±1.03	35.45±2.04

Table 5.2: Continued.

Tea Estate	Sample		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Malnicherra	Tea Leaf	*Min – Max*	14.60 –18.75	12.75 – 23.27	39.56 – 133.44	468 – 1849	1.75 –7.45	11.25 – 89.20
	(n = 9)	*Mean±s.e.*	17.04±0.46	15.78±1.10	73.14±9.28	997±166	4.16±0.57	45.42±8.83
	Made Tea	*Min – Max*	10.90 –12.50	14.72 – 20.07	326 – 653	792.75–814.45	9.95–11.25	165.8 –272.7
	(n = 3)	*Mean±s.e.*	11.43±0.53	18.10±1.70	450±102	802.17±6.43	10.58±0.38	221.1±30.9
Neptune	Tea Leaf	*Min – Max*	1.40 – 54.15	7.90 –15.90	31.00 –102.30	229.0 –872.0	2.05 –5.85	19.57 – 33.01
	(n = 8)	*Mean±s.e.*	13.92±5.89	12.34±0.86	75.10±7.39	424.8±74.5	3.58±0.57	25.95±1.49
	Made Tea	*Min – Max*	23.40 –26.80	7.95 – 13.00	35.0 – 92.0	221.0 – 481.0	3.90 –7.05	14.40 –24.23
	(n = 3)	*Mean±s.e.*	25.10±1.70	10.47±1.46	65.7±16.6	366.0±76.5	5.82±0.97	20.02±2.92
Oodaleah	Tea Leaf	*Min – Max*	1.25 – 51.85	6.50 – 14.50	64.20 –113.50	96 –1387	0.50 – 9.35	19.66 – 42.75
	(n= 8)	*Mean±s.e.*	15.81±9.29	10.56±0.78	85.99±5.46	765±158	4.14±0.97	29.07±3.67
	Made Tea	*Min – Max*	16.15 –18.60	9.44 –14.75	3.00 –28.00	466.0 –711.0	3.05 – 6.30	20.75 – 25.17
	(n = 3)	*Mean±s.e.*	17.38±1.23	12.13±1.53	12.00±8.02	620.3±77.6	4.62±0.94	23.42±1.36
Patrakhola	Tea Leaf	*Min – Max*	7.45 –11.15	17.88 – 19.81	56.28 – 105.63	404.9 – 884.5	1.10 – 4.150	35.05 – 83.35
	(n = 5)	*Mean±s.e.*	9.31±0.74	18.62±0.32	82.15±8.22	664.6±76.4	3.01±0.52	56.57±9.23
	Made Tea	*Min – Max*	17.55 –25.95	15.65–16.06	315.4 –555.1	1176.6 –1240	8.60 – 9.90	128.0 –241.0
	(n = 3)	*Mean±s.e.*	22.40±2.51	15.86±0.12	410.7±73.4	1217±20.1	9.18±0.38	171.8±35.0
Rajghat	Tea Leaf	*Min – Max*	18.25 –25.05	10.26 –20.33	47.10 –85.14	206 –2006	0.75 – 6.60	34.8 –163.4
	(n= 8)	*Mean±s.e.*	21.31±0.90	16.05±1.17	65.49±5.88	987±242	3.79±0.74	62.6±15.6
	Made Tea	*Min – Max*	7.80 –18.95	15.57–21.70	202 –1051	932.7 – 998.8	5.30 – 9.00	153.9 –344.8
	(n = 3)	*Mean±s.e.*	11.68±3.64	19.50±1.97	505±273	958.6±20.4	6.57±1.22	246.0±55.2
Sathgao	Tea Leaf	*Min – Max*	12.20 –17.25	13.68 –20.55	52.41 –106.34	145.6 –749.8	1.15 – 5.10	28.0 –133.9
	(n= 8)	*Mean±s.e.*	14.63±0.62	17.16±0.96	76.17±6.22	397.6±71.4	2.88±0.50	73.1±12.8
	Made Tea	*Min – Max*	10.60 –15.00	19.63 –22.53	233.7 – 400.8	838.4 –1098.1	7.15 –8.55	275.6 –324.7
		Mean±s.e.	12.83±1.27	21.48±0.93	299.3±51.5	938.8±80.6	7.80±0.41	298.6±14.3
	Tea Leaf	F value	1.66	10.09	0.93	7.37	1.61	8.68
		p value	0.09	< 0.001	0.53	<0.001	0.10	< 0.001
	Made Tea	F value	0.81	1.11	7.45	51.05	10.65	7.91
		p value	0.64	0.39	< 0.001	< 0.001	< 0.001	<0.001

BDL = Below detection limit.

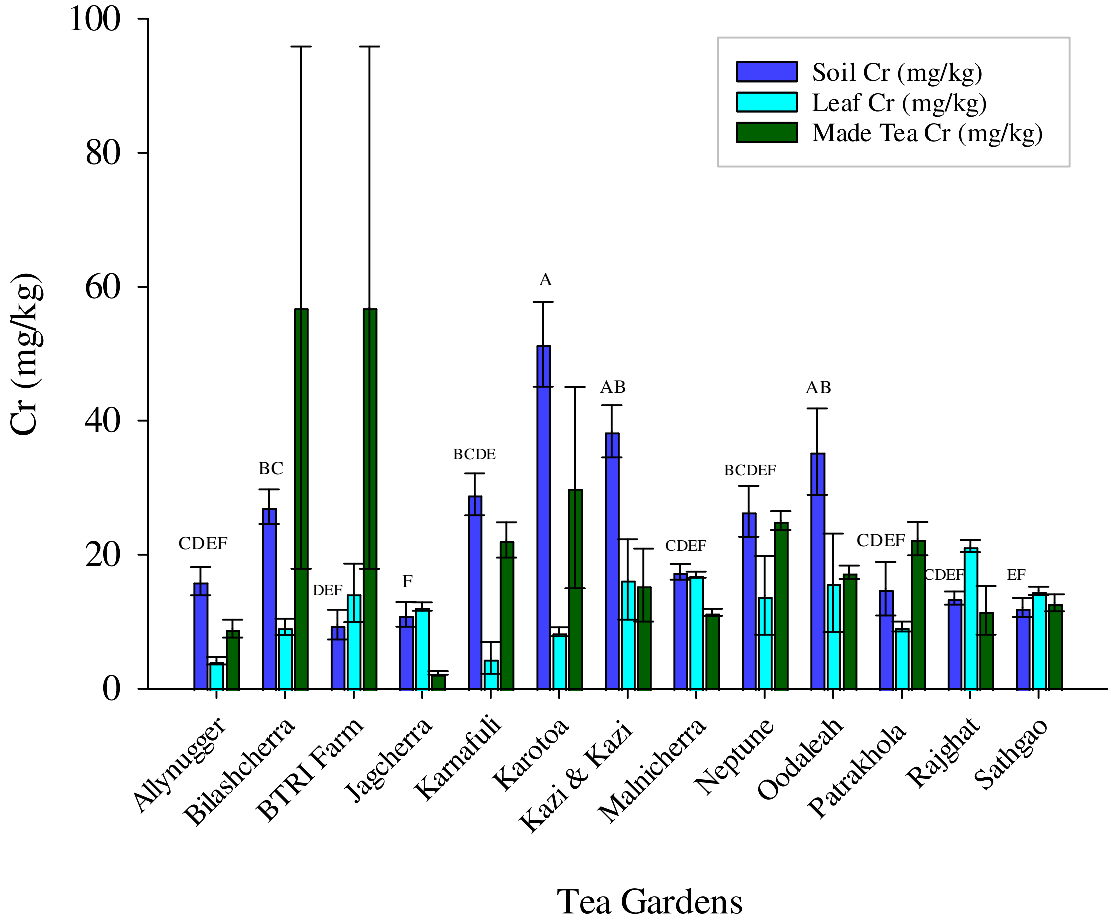


Figure 5.1: The concentrations of chromium in the soils, tea leaves and made tea of different tea gardens of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of chromium concentration in the tea garden soils, tea leaves and made tea. Tea gardens that share the same letter (A – F, a – b) are not significantly different. The letters indicate Tukey groupings for the tea gardens with respect to their mean concentrations of chromium in the soils, tea leaves and made tea. Bars with different capital letters (A-F) indicate that concentrations of chromium in the soils are significantly ($p < 0.001$) different among the tea gardens. The bars are mean±standard error of the mean.

The highest mean concentration of iron (748 mg/kg) was found in tea leaves of Kazi & Kazi tea garden and the lowest mean concentration of iron (65.49 mg/kg) was found in the tea leaves of Rajghat tea garden. The highest mean concentration of iron (1791 mg/kg) was found in made tea of Karotoa tea garden and the lowest mean concentration of iron (12.00 mg/kg) was found in made tea of Oodaleah tea garden (Table 5.2).

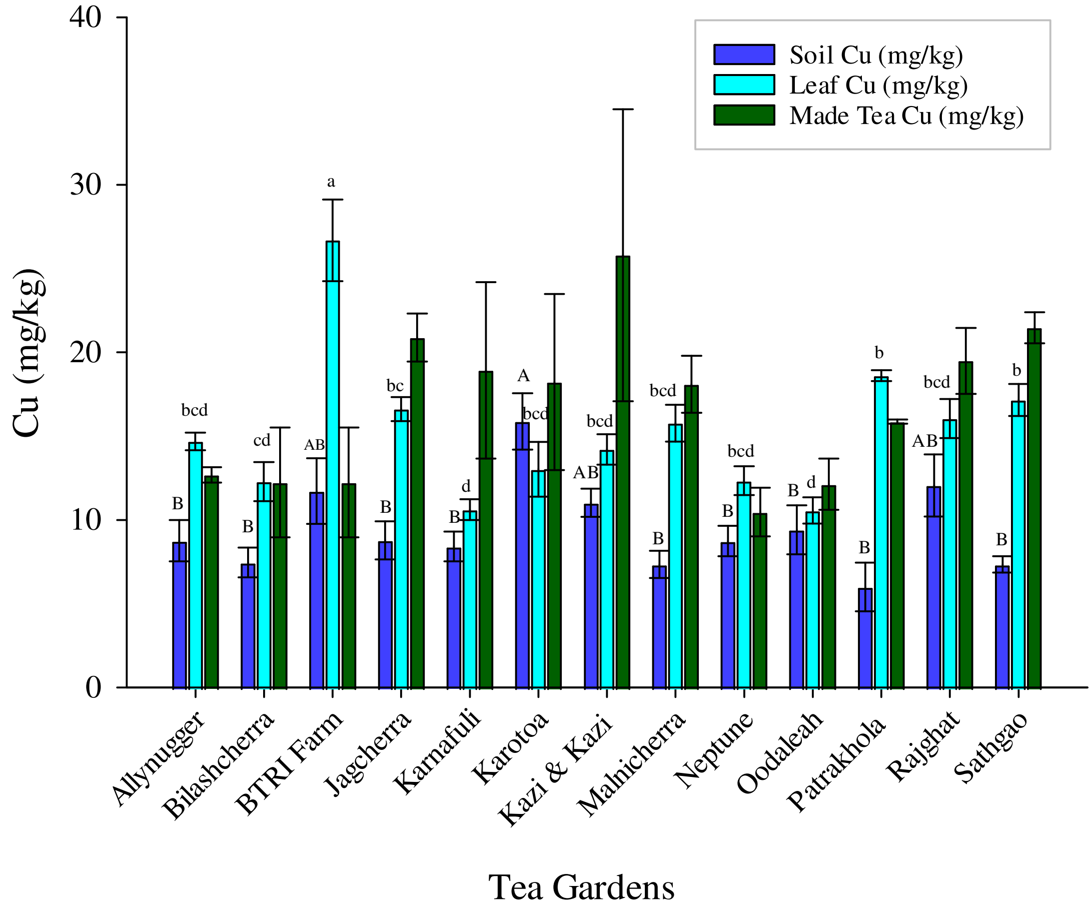


Figure 5.2: The concentrations of copper in the soils, tea leaves and made tea of different tea gardens of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of copper concentration in the tea garden soils, tea leaves and made tea. Tea gardens that share the same letter (A – B, a – d) are not significantly different. The letters indicate Tukey groupings for the tea gardens with respect to their mean concentrations of copper in the soils, tea leaves and made tea. Bars with different capital letters (A-B) indicate that concentrations of copper in the soils are significantly different among the tea gardens ($p < 0.001$), while bars followed by different lower-case letters (a-d) indicate that concentrations of copper in the tea leaves are significantly different among the tea gardens ($p < 0.001$). The bars are mean±standard error of the mean.

Significantly different ($p < 0.001$) concentrations of iron were observed in the soils of the tea gardens. The variations of the concentrations of iron in the tea leaves were found insignificant while concentrations of iron in the made tea were observed to be significantly different ($p < 0.001$) among the tea gardens (Figure 5.3).

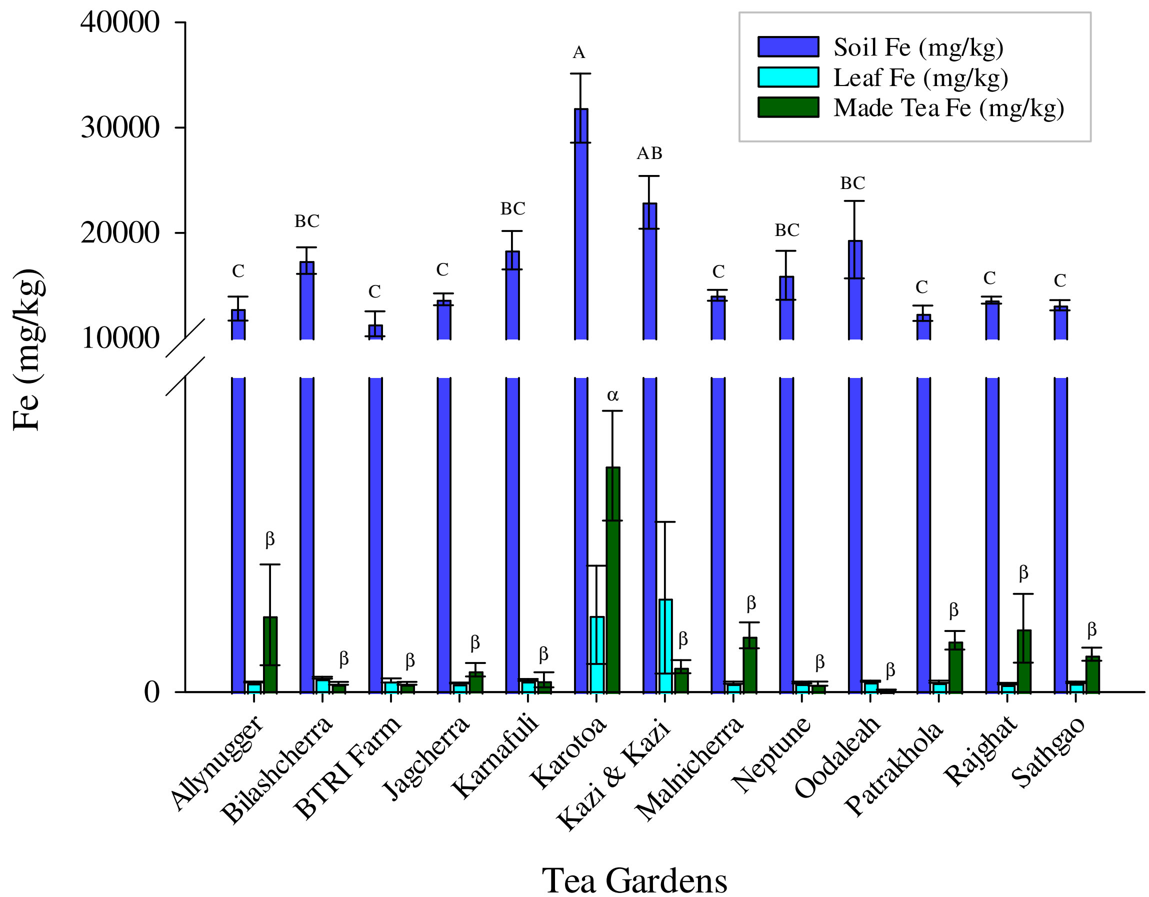


Figure 5.3: The concentrations of iron in the soils, tea leaves and made tea of different tea gardens of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of iron concentration in the tea garden soils, tea leaves and made tea. Tea gardens that share the same letter (A – C, α - β) are not significantly different. The letters indicate Tukey groupings for the tea gardens with respect to their mean concentrations of iron in the soils, tea leaves and made tea. Bars with different capital letters (A-C) indicate that concentrations of iron in the soils are significantly different among the tea gardens ($p < 0.001$), while Bars with Greek letters (α – β) indicate that concentrations of iron in the made tea are significantly different among the tea gardens ($p < 0.001$). The bars are mean±standard error of the mean.

Significant and positive relationships between iron and nickel (R^2 = 0.08, p < 0.05) were observed in the tea leaves (Figure 5.7). In the made tea, significant and positive relationships were observed between iron and nickel (R^2 = 0.22, p < 0.01) (Figure 5.8). No significant relationships of iron with the biochemical parameters such as caffeine, protein, total polyphenol, theaflavin and thearubigin were observed in the made tea (Figure 5.9 – 5.13).

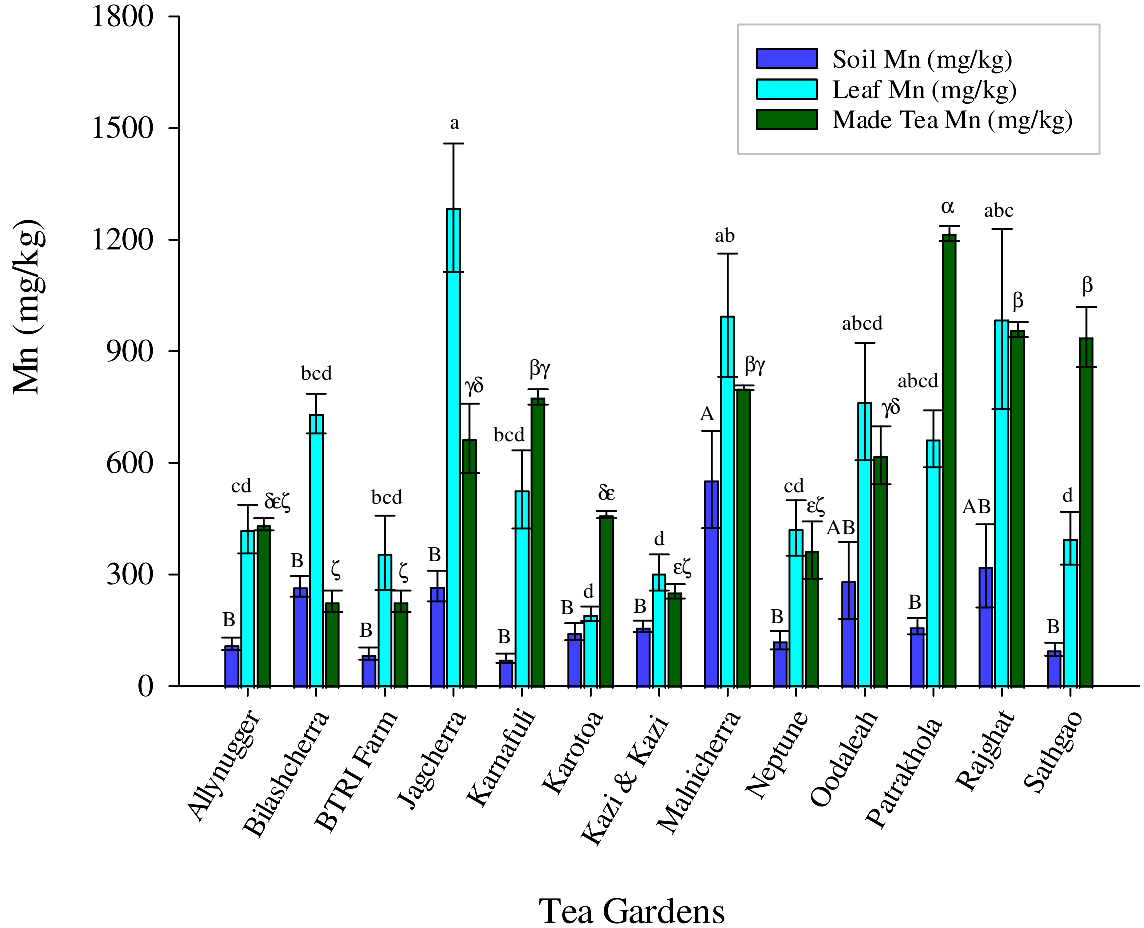


Figure 5.4: The concentrations of manganese in the soils, tea leaves and made tea of different tea gardens of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of manganese concentration in the tea garden soils, tea leaves and made tea. Tea gardens that share the same letter (A – B, a – d, α - ζ) are not significantly different. The letters indicate Tukey groupings for the tea gardens with respect to their mean concentrations of manganese in the soils, tea leaves and made tea. Bars with different capital letters (A-B) indicate that concentrations of manganese in the soils are significantly different among the tea gardens ($p < 0.001$), while bars followed by different lower-case letters (a-d) indicate that concentrations of manganese in the tea leaves are significantly different among the tea gardens ($p < 0.001$). Bars with Greek letters (α-ζ) indicate that concentrations of manganese in the made tea are significantly different ($p < 0.001$) among the tea gardens. The bars are mean±standard error of the mean.

The highest mean concentration of manganese was found in the tea leaves of Jagcherra tea garden (1287 mg/kg) and the lowest mean concentration of manganese was found in the tea leaves of Karotoa tea garden (195 mg/kg). The highest mean concentration of manganese

(1217 mg/kg) was found in made tea of Patrakhola tea garden and the lowest mean concentration of manganese (228 mg/kg) was found in the made of Bilashcherra and BTRI tea garden (Table 5.2). Significantly different (p < 0.001) concentrations of manganese were found in the soils of the tea gardens. The concentrations of manganese in the tea leaves and made tea were also found to be significantly different (p < 0.001) among the tea gardens (Figure 5.4).

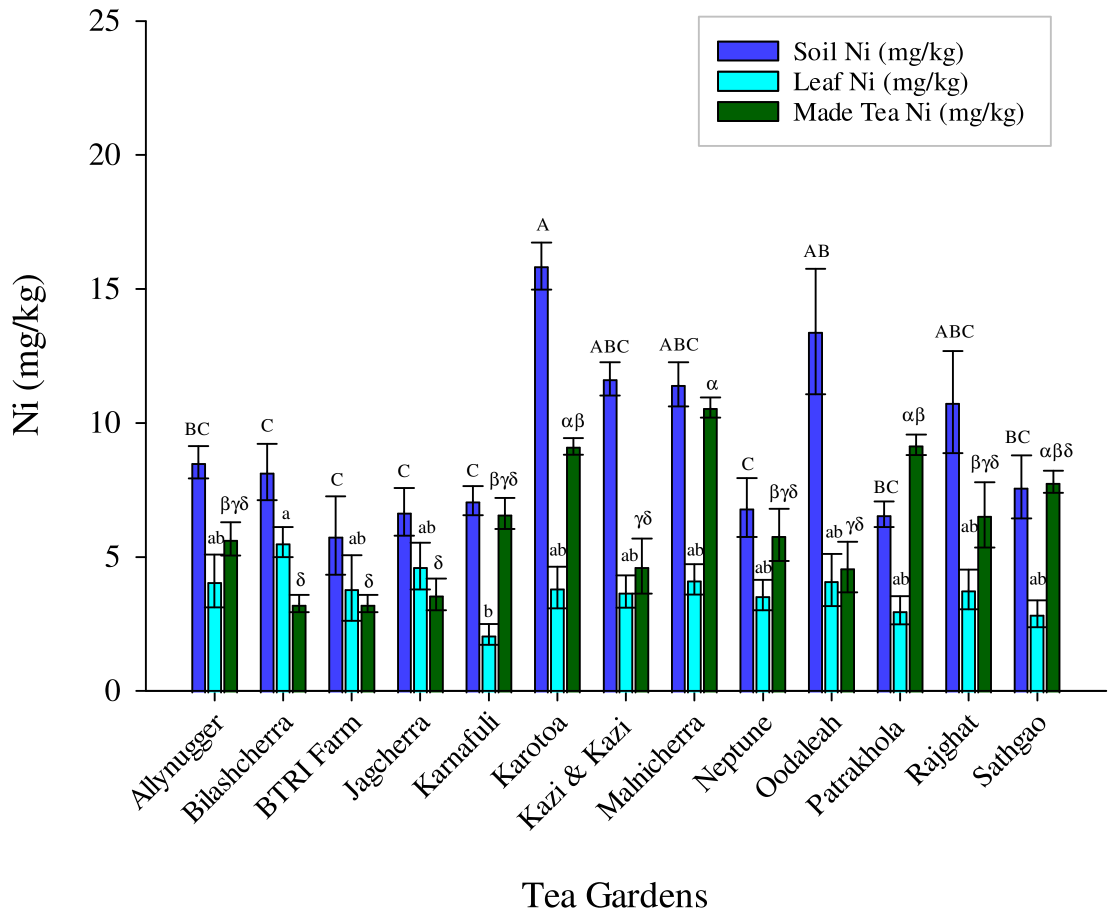


Figure 5.5: The concentrations of nickel in the soils, tea leaves and made tea of different tea gardens of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of nickel concentration in the tea garden soils, tea leaves and made tea. Tea gardens that share the same letter (A – C, α - δ) are not significantly different. The letters indicate Tukey groupings for the tea gardens with respect to their mean concentrations of nickel in the soils, tea leaves and made tea. Bars with different capital letters (A-C) indicate that concentrations of nickel in the soils are significantly different among the tea gardens ($p < 0.001$), while bars with Greek letters (α-δ) indicate that concentrations of nickel in the made tea are significantly different ($p < 0.001$) among the tea gardens. The bars are mean±standard error of the mean.

Significant and positive relationships were observed between manganese and chromium (R^2 = 0.09, $p < 0.001$) in the tea leaves (Figure 5.7). A positive and significant relationship was observed between manganese and nickel ($R^2 = 0.31$, $p < 0.001$) in the made tea (Figure 5.8). A negative and significant relationships was observed between manganese and thearubigin (R^2 = 0.41, $p < 0.001$) in the made tea (Figure 5.13). A sharp decline in polyphenol content in tea due to excess manganese concentration was also observed by Venkatesan *et al.* (2007). No significant relationships of manganese with the other biochemical parameters such as caffeine, protein, total polyphenol and theaflavin were observed in the made tea (Figure 5.9 – 5.12).

The highest mean concentration of nickel was found in tea leaves of Bilashcherra tea garden (5.55 mg/kg) and the lowest mean concentration of nickel was found in tea leaves of Karnafuli tea garden (2.11 mg/kg). The highest mean concentration of nickel was found in made tea of Malnicherra tea garden (10.58 mg/kg) and the lowest mean concentration of nickel was found in the made tea of Bilashcherra and BTRI tea garden (3.26 mg/kg) (Table 5.2). Significantly different ($p < 0.001$) concentrations of nickel were observed in the soils of the tea gardens. The concentrations of nickel in the made tea were found to be significantly different ($p < 0.001$) among the different tea gardens (Figure 5.5). Significant and negative relationships were observed between nickel and total polyphenol ($R^2 = 0.201$, $p < 0.01$) in the made tea. (Figure 5.11). No significant relationships of nickel with the biochemical parameters such as caffeine, protein, theaflavin and thearubigin were observed in the made tea (Figure 5.9 – 5.13). In the present study, significant and negative relationships between total polyphenol and nickel indicated that with increase nickel concentration total polyphenol content was decreased in the made tea. Bobková *et al.* (2021) also reported negative correlation between nickel and total polyphenol content in tea. Total polyphenol or phenolic compounds which are present in plants have antioxidant capacity and various health benefits. These are an indispensible segment of human nutrition (Shahidi and Ambigaipalan, 2015). Therefore, excess nickel might decrease the quality of tea through decreasing the total polyphenol content.

The highest mean concentration of zinc (78.67 mg/kg) was found in the tea leaves of BTRI tea garden and the lowest mean concentration of zinc (25.95 mg/kg) was found in the tea leaves of

Neptune tea garden. The highest mean concentration of zinc (331 mg/kg) was found in made tea of Jagcherra tea garden and the lowest mean concentration of zinc (20.02 mg/kg) was found in made tea of Neptune tea garden (Table 5.2). Significantly different ($p < 0.05$) concentrations of zinc were observed in the soils of the tea gardens. The concentrations of zinc in the tea leaves and made tea were also found to be significantly different ($p < 0.001$) among the tea gardens (Figure 5.6).

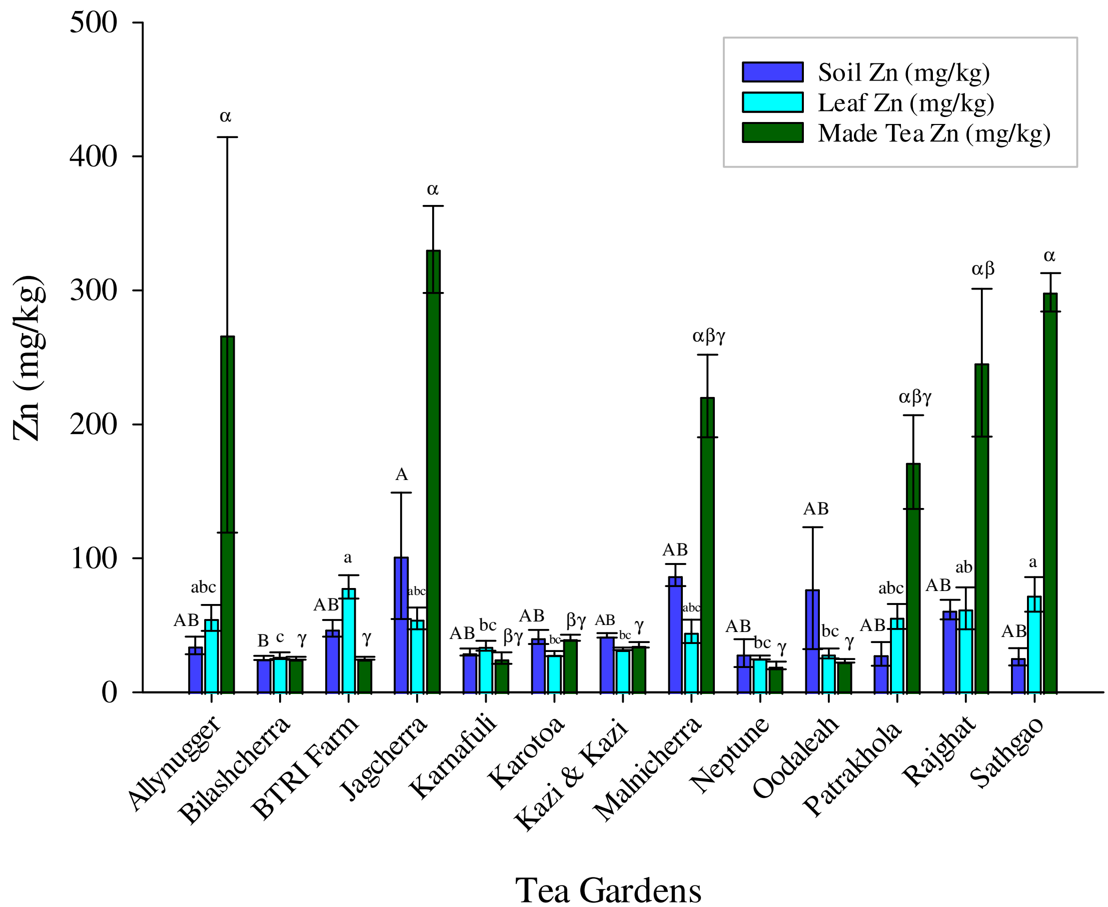


Figure 5.6: The concentrations of zinc in the soils, tea leaves and made tea of different tea gardens of Bangladesh. One-way analysis of variance was used in order to compare pair-wise the means of zinc concentration in the tea garden soils, tea leaves and made tea. Tea gardens that share the same letter (A – B, a – c, α - γ) are not significantly different. The letters indicate Tukey groupings for the tea gardens with respect to their mean concentrations of zinc in the soils, tea leaves and made tea. Bars with different capital letters (A-B) indicate that concentrations of zinc in the soils are significantly different among the tea gardens ($p < 0.05$), while bars followed by different lower-case letters (a-c) indicate that concentrations of zinc in the tea leaves are significantly different among the tea gardens ($p < 0.001$). Bars with Greek letters (α-γ) indicate that concentrations of zinc in the made tea are significantly different among the tea gardens ($p < 0.001$). The bars are mean±standard error of the mean.

In the made tea significant and positive relationships were observed between caffeine and zinc (R^2 = 0.27, p < 0.001) (Figure 5.9). Significant and negative relationships were observed between theaflavin and zinc (R^2 = 0.202, p < 0.01), thearubigin and zinc (R^2 = 0.28, p < 0.01) in the made tea (Figure 5.12 – 5.13). Significant and positive relationships between caffeine and zinc indicated that with increase zinc concentration caffeine content also increased in the made tea. Caffeine is a naturally occurring chemical stimulant which can negatively affect human health if it is consumed excessively.

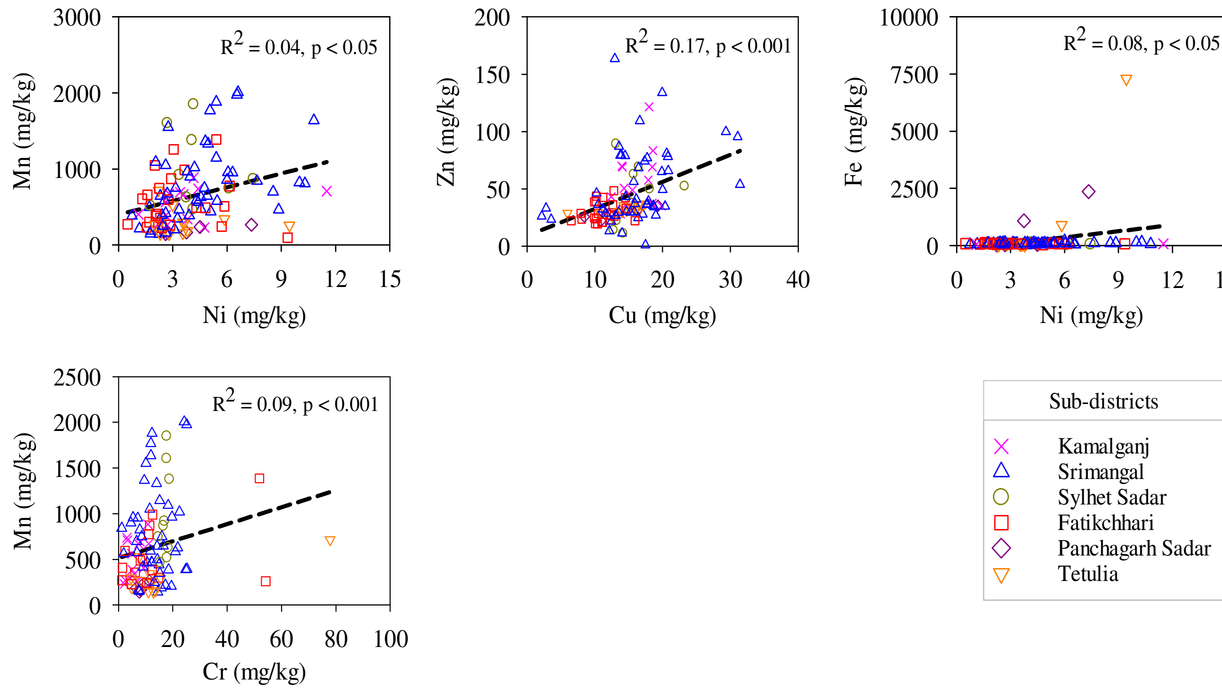


Figure 5.7: Relationships of heavy metals in the tea leaves.

Lacerda *et al.* (2018) reported that increased doses of zinc increased caffeine levels in coffee. Due to the mild diuretic effect of caffeine leads increase in urination which is responsible for the depletion of water-soluble vitamin B6 and vitamin C through fluid loss. Absorption of essential nutrients such as calcium, iron, magnesium and B vitamins can be affected by caffeine (Wolde, 2014).

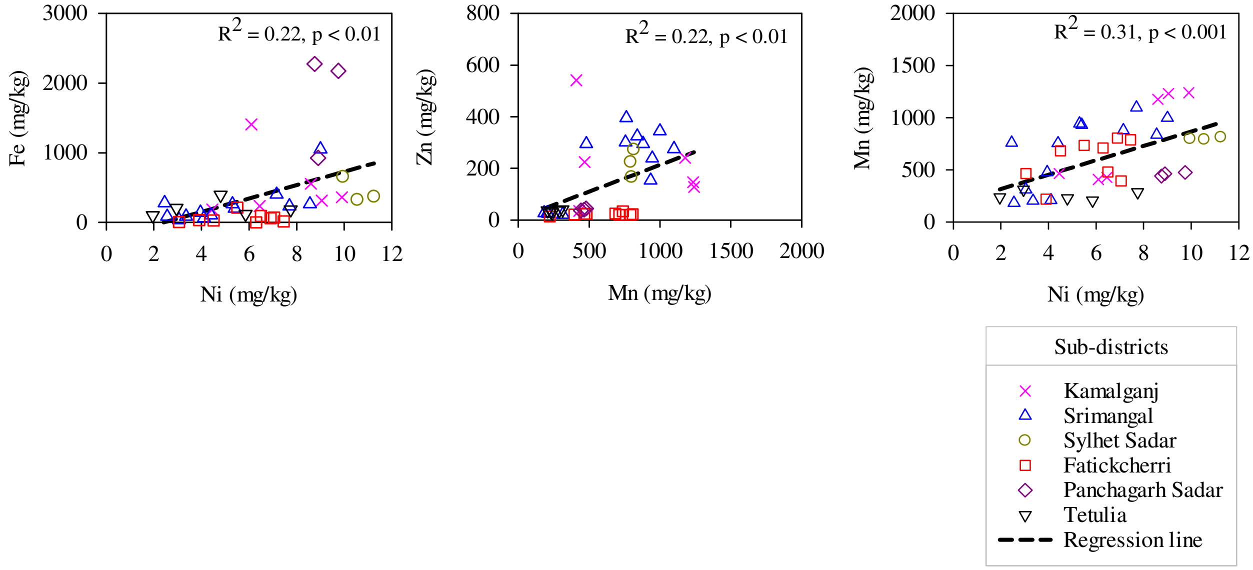


Figure 5.8: Relationships of heavy metals in the made tea.

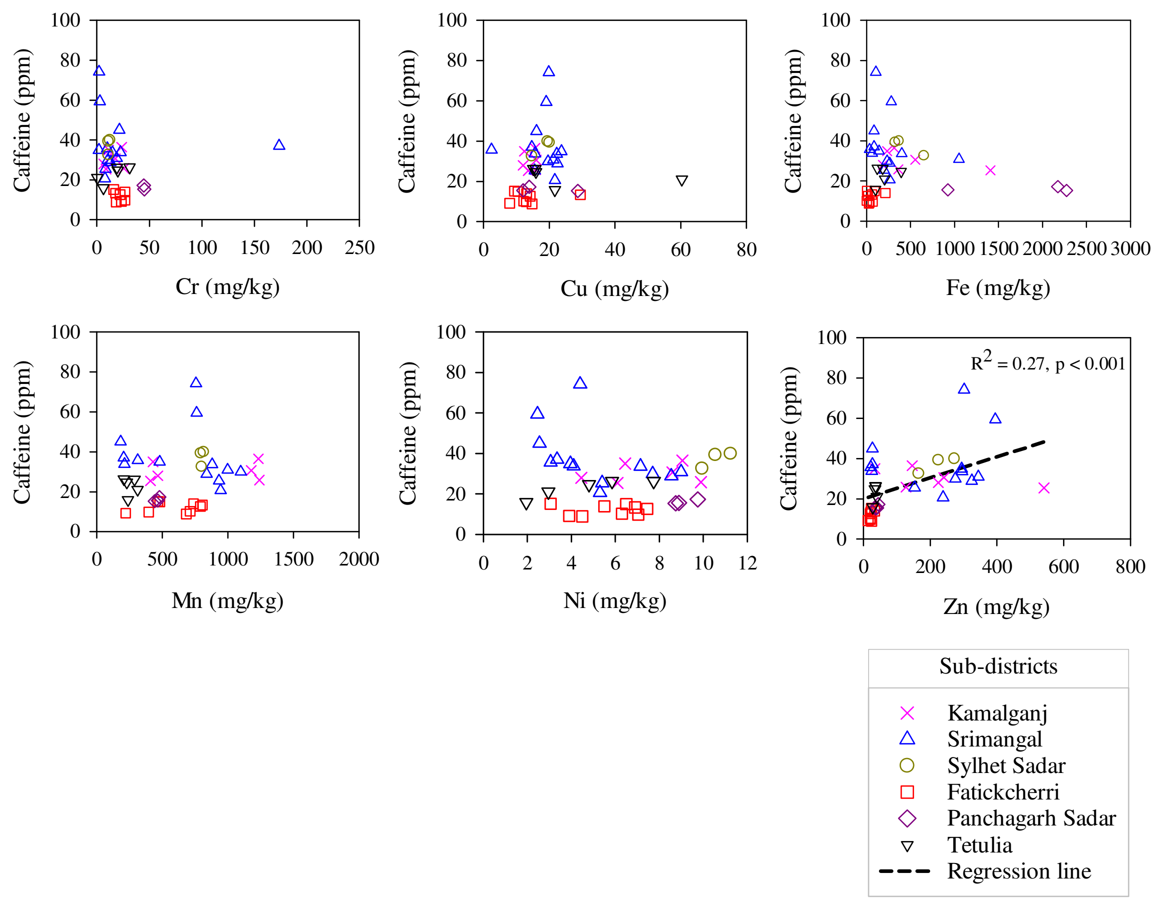


Figure 5.9: Relationships of caffeine with heavy metals in the made tea.

Significant and negative relationships between theaflavin and zinc as well as thearubigin and zinc indicated reductions in the contents of theaflavin and thearubigin in tea due to increased zinc concentrations.

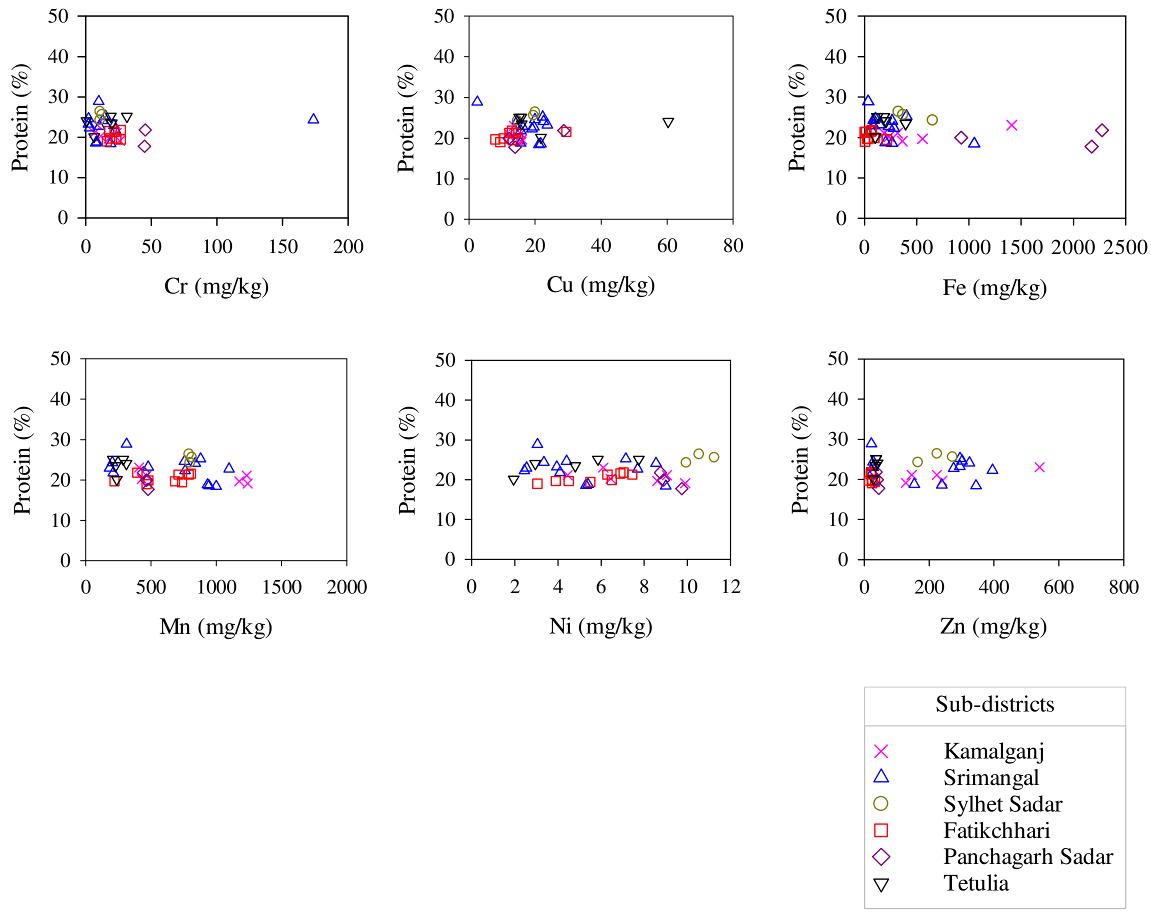


Figure 5.10: Relationships of protein with heavy metals in the made tea.

Zhang *et al*. (2016) also reported reductions in the levels of flavonoids with increasing zinc content in tea. Theaflavin and thearubigin are the biological compounds that belong to flavonoid family (Bond and Derbyshire, 2020). The definite color and taste of black tea brew depends on the orange or orange-red pigment which is called theaflavin. The taste and aroma of black tea depends on prime polyphenolic compound named thearubigin (Koch, 2020). Theaflavin and thearubigin also have strong antioxidant capacity (Koch, 2020). Therefore, excess zinc might decrease the quality of tea through decreasing theaflavin and thearubigin contents.

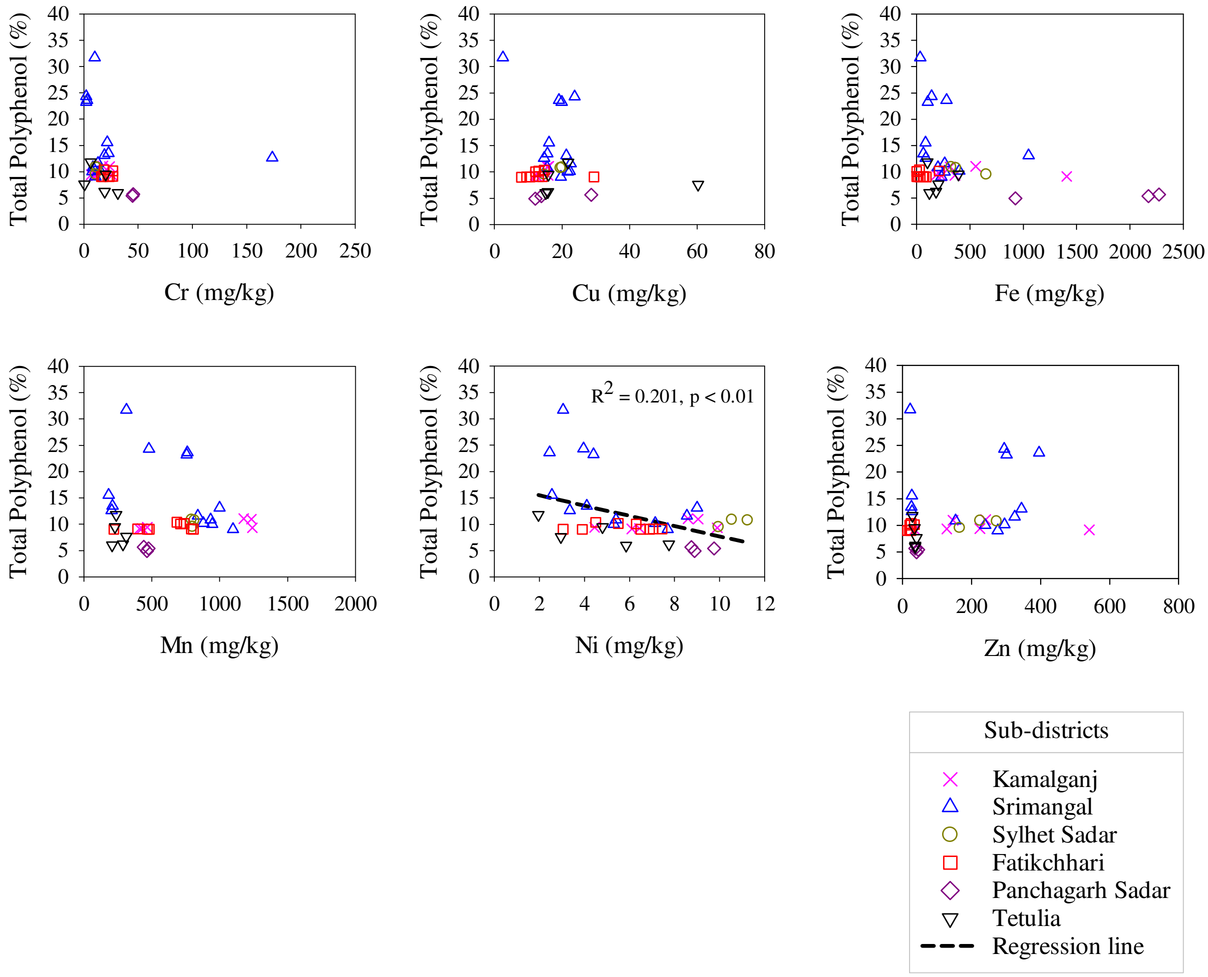


Figure 5.11: Relationships of total polyphenol with heavy metals in the made tea.

In the made tea significant and positive relationships were observed between protein and caffeine ($R^2 = 0.23$, $p < 0.01$), total polyphenol and caffeine ($R^2 = 0.39$, $p < 0.001$), thearubigin and protein ($R^2 = 0.22$, $p < 0.01$), total polyphenol and protein ($R^2 = 0.16$, $p < 0.05$), thearubigin and theaflavin ($R^2 = 0.99$, $p < 0.001$) (Figure 5.14). Significant and negative relationships were observed between theaflavin and caffeine ($R^2 = 0.19$, $p < 0.01$), theaflavin and total polyphenol ($R^2 = 0.51$, $p < 0.001$) in the made tea (Figure 5.14).

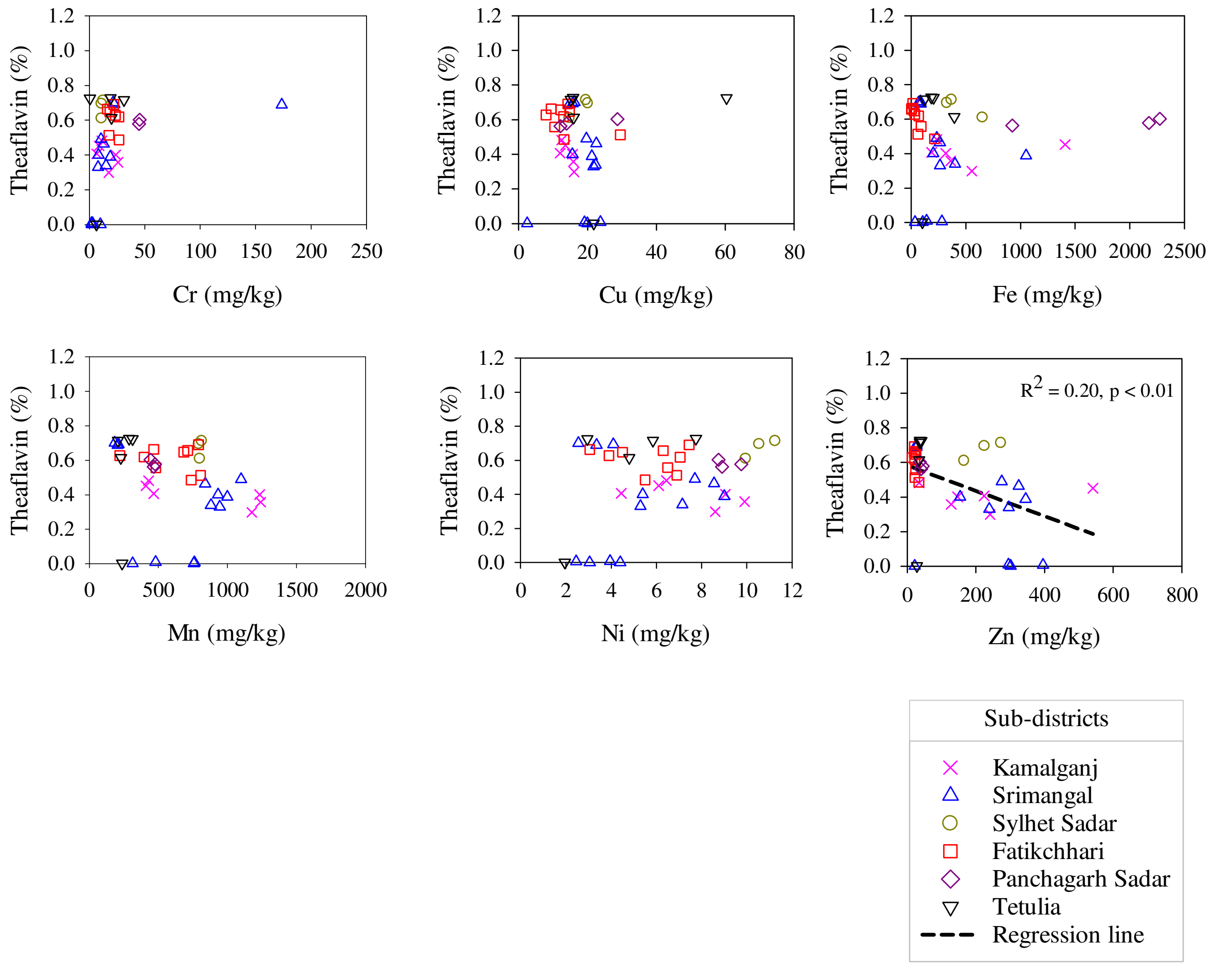


Figure 5.12: Relationships of theaflavin with heavy metals in the made tea.

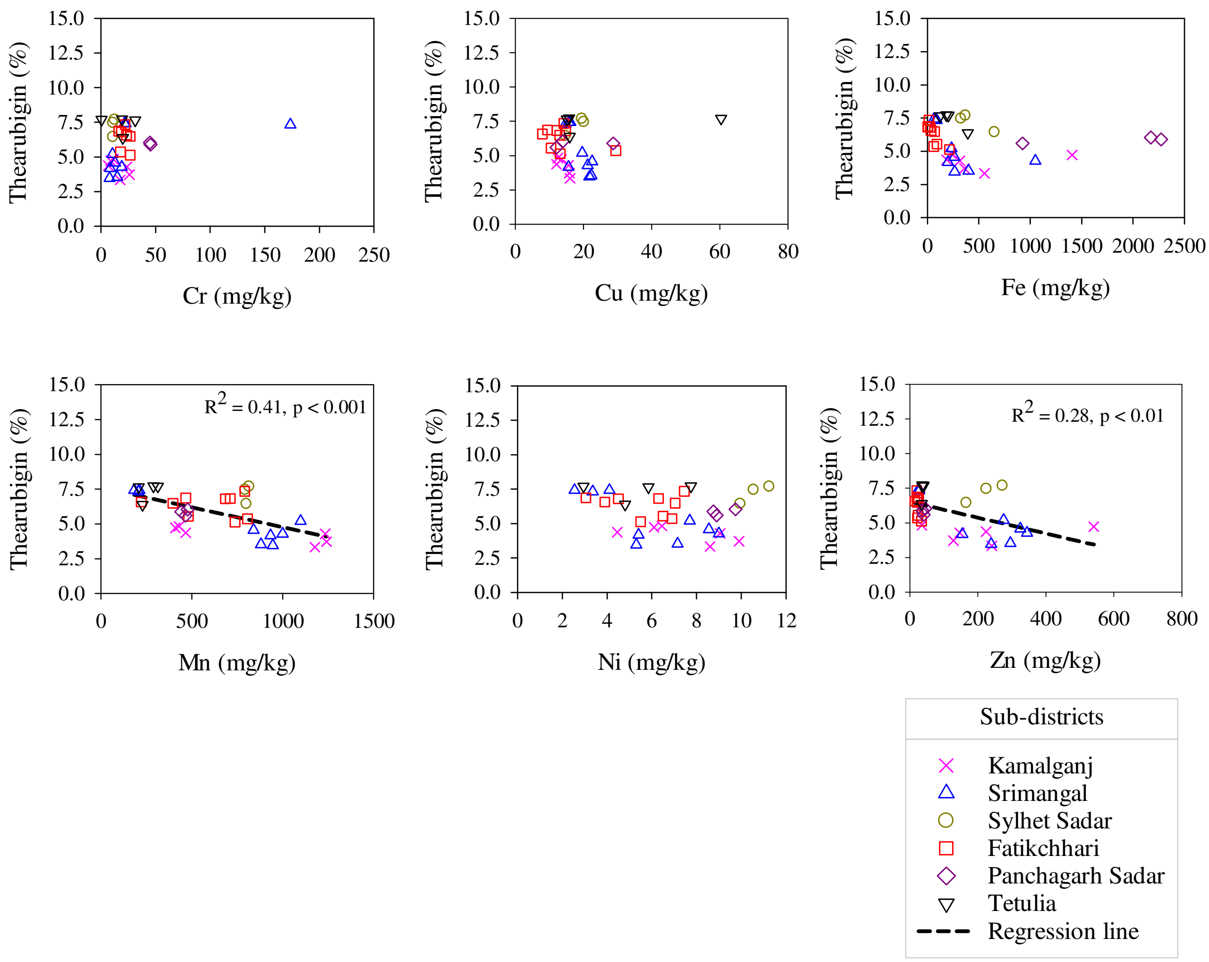


Figure 5.13: Relationships of thearubigin with heavy metals in the made tea.

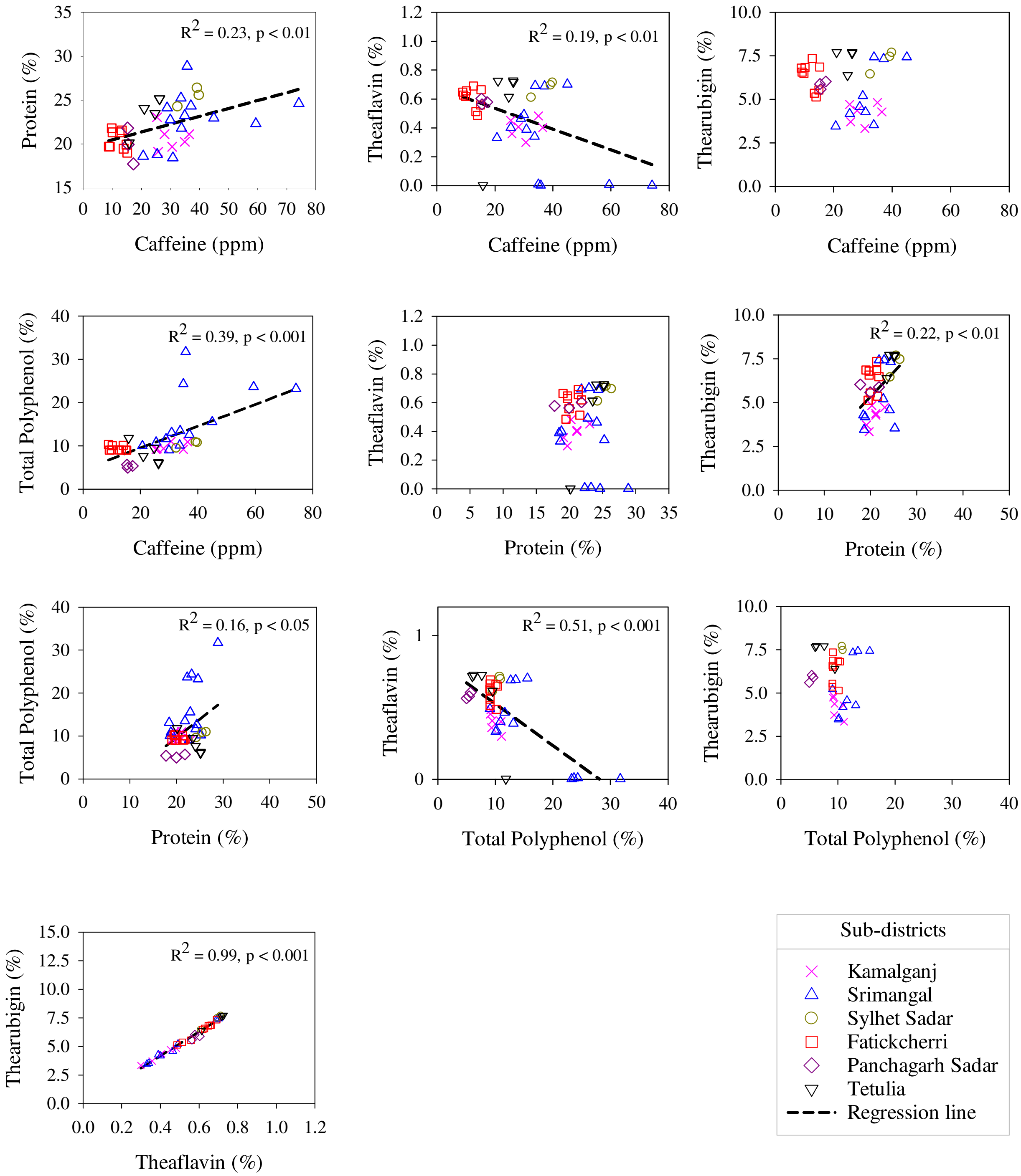


Figure 5.14: Relationships of biochemical parameters in the made tea.

5.4.3 Heavy metals in the tea leaves and made tea of tea gardens located at different sub-districts of Bangladesh

The concentrations of heavy metals in the tea leaves and made tea of tea gardens located at different sub-districts of Bangladesh are presented in table 5.3. The highest mean concentration of chromium (17.04 mg/kg) in the tea leaves was found in the Sylhet Sadar sub-district and the lowest mean concentration of chromium (6.01 mg/kg) in the tea leaves was found in the Kamalganj sub-district. Significantly different ($^{ANOVA}F = 2.34$, $p < 0.05$) concentrations of chromium were observed in the tea leaves (Table 5.3). The highest mean concentration of chromium (30.00 mg/kg) in the made tea sample was found in the Panchagarh Sadar sub-district and the lowest mean concentration of chromium (11.43 mg/kg) in the made tea sample was found in the Sylhet Sadar sub-district. No significant differences of the concentrations of chromium were found in the made tea samples (Table 5.3). The highest mean concentration of copper (16.09 mg/kg) in the tea leaves was found in the Kamalganj sub-district and the lowest mean concentration of copper (11.19 mg/kg) in the tea leaves was found in the Fatikchhari sub-district. Significantly different ($^{ANOVA}F = 4.57$, $p < 0.001$) concentrations of copper were observed in the tea leaves (Table 5.3).

The highest mean concentration of copper (25.80 mg/kg) in the made tea samples was found in the Tetulia sub-district and the lowest mean concentration of copper concentration (13.84 mg/kg) was found in the Fatikchhari sub-district. No significant differences of the concentrations of copper were found in the made tea samples (Table 5.3). The highest mean concentration of iron (748 mg/kg) in the tea leaves was found in the Tetulia sub-district and the lowest mean concentration of iron (73.14 mg/kg) was found in the Sylhet Sadar sub-district. Significantly different ($^{ANOVA}F = 2.37$, $p < 0.05$) concentrations of iron were observed in the tea leaves (Table 5.3). The highest mean concentrations of iron (1791 mg/kg) in the made tea were found in the Panchagarh Sadar sub-district and the lowest mean concentration of iron (58.4 mg/kg) in the made tea was found in the Fatikchhari sub-district. Significantly different ($^{ANOVA}F = 15.46$, $p < 0.001$) concentrations of iron were observed in the made tea samples (Table 5.3).

Table 5.3: Concentration of heavy metals in the tea leaves and made tea in different sub-districts of Bangladesh

Sub-districts	Sample		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Fatikchhari	Tea Leaf (n = 23)	*Min – Max*	BDL – 54.15	6.50 –15.90	31.00 –138.25	96.0 –1387.0	0.50 – 9.35	19.57–48.38
		Mean±s.e.	9.68±3.05	11.19±0.46	85.07±4.31	574.7±72.5	3.33±0.43	29.73±1.85
	Made Tea (n = 9)	*Min – Max*	16.15 –26.80	7.95 –29.45	3.0 –214.0	221.0 –806.0	3.05 –7.45	14.40 –33.91
		Mean±s.e.	21.65±1.63	13.84±2.09	58.4±21.9	588.0±68.0	5.68±0.52	22.98±1.74
Kamalganj	Tea Leaf (n = 14)	*Min – Max*	2.10 –11.15	12.42 –19.81	56.28 –105.63	231.8 –884.5	1.10 –11.50	28.30 –121.60
		Mean±s.e.	6.01±0.80	16.09±0.63	80.19±3.92	508.9±58.2	3.71±0.65	55.89±6.78
	Made Tea (n = 6)	*Min – Max*	6.70 – 25.95	12.04 –16.06	188 –1408	409 –1240	4.45–9.90	34.9 – 541.0
		Mean±s.e.	15.68±3.26	14.27±0.74	510±187	826±175	7.43±0.85	219.3±71.1
Panchagarh Sadar	Tea Leaf (n= 6)	*Min – Max*	6.50–11.350	8.45 – 19.30	18 – 2363	143.0 – 266.0	2.25–7.35	22.41 – 35.30
		Mean±s.e.	8.48±0.68	13.02±1.63	611±389	195±19.3	3.86±0.78	28.89±1.85
	Made Tea (n = 3)	*Min – Max*	BDL –45.3	12.10 –28.70	926 –2274	442.0 –477.0	8.75–9.75	36.87 – 44.64
		Mean±s.e.	30.0±15.0	18.23±5.26	1791±434	461.3±10.3	9.13±0.31	40.63±2.24
Srimangal	Tea Leaf (n = 48)	*Min – Max*	1.15–25.20	2.15 –31.41	27.28 –207.80	145.6 –2006	0.75 –10.80	1.17 –163.40
		Mean±s.e.	13.51±0.91	16.04±0.82	89.27±4.60	784.1±71.5	4.47±0.35	51.39±4.70
	Made Tea (n = 13)	*Min – Max*	2.00 –173.5	2.45–23.71	36.0 –1050.5	183.0 –1098.1	2.45 – 9.00	22.0 –395.5
		Mean±s.e.	23.7±12.6	18.04±1.54	247.5±73.1	661.7±93.1	5.15±0.63	209.8±38.7
Sylhet Sadar	Tea Leaf (n= 9)	*Min – Max*	14.60 –18.75	12.75 –23.27	39.56 –133.44	468 – 1849	1.75 –7.45	11.25 –89.20
		Mean±s.e.	17.04±0.46	15.78±1.10	73.14±9.28	997±166	4.16±0.57	45.42±8.83
	Made Tea (n = 3)	*Min – Max*	10.90 –12.50	14.72 –20.07	326 – 653	792.75 –814.45	9.95–11.25	165.8 –272.7
		Mean±s.e.	11.43±0.53	18.10±1.70	450±102	802.17±6.43	10.58±0.38	221±30.9
Tetulia	Tea Leaf (n= 12)	*Min – Max*	4.65 –77.85	5.95–17.35	15 –7300	143.0 –715.0	2.00 –9.45	28.95–37.03
		Mean±s.e.	16.32±6.24	14.21±0.91	748±600	305.8±48.5	3.71±0.60	32.43±0.87
	Made Tea (n = 5)	*Min – Max*	0.55–31.30	15.15 –60.35	102.4 – 394.0	207.0 –312.0	1.95 –7.75	28.38 – 40.78
		Mean±s.e.	15.47±5.43	25.80±8.72	201.7±51.7	255±19.5	4.66±1.03	35.45±2.04
	Tea Leaf	*F value*	2.34	4.57	2.37	6.22	0.93	3.95
		p value	< 0.05	< 0.001	< 0.05	< 0.001	0.462	< 0.01
	Made Tea	*F value*	0.21	1.47	15.46	2.84	6.37	5.51
		p value	0.955	0.228	< 0.001	< 0.05	< 0.001	< 0.001

The highest mean concentration of manganese (997 mg/kg) in the tea leaves was found in the Sylhet Sadar sub-district and the lowest mean concentration of manganese (195 mg/kg) in the tea leaves was found in the Panchagarh Sadar sub-district. Significantly different ($^{ANOVA}F$ = 6.22, $p < 0.001$) concentrations of manganese were observed in the tea leaves (Table 5.3). The highest mean concentration of manganese (826 mg/kg) in the made tea was found in the Kamalganj sub-district and the lowest mean concentration of manganese (255 mg/kg) in the made tea was found in the Tetulia sub-district. Significantly different ($^{ANOVA}F$ = 2.84, $p <$ 0.05) concentrations of manganese were observed in the made tea samples (Table 5.3). The highest mean concentration of nickel (4.47 mg/kg) in the tea leaves was found in the Srimangal sub-district and the lowest mean concentration of nickel (3.33 mg/kg) in the tea leaves was found in the Fatikchhari sub-district. No significant differences in the concentrations of nickel were observed in the tea leaves (Table 5.3).

The highest mean concentration of nickel (10.58 mg/kg) in the made tea was found in the Sylhet Sadar sub-district and the lowest mean concentration of nickel (4.66 mg/kg) was found in the Tetulia sub-district. Significantly different ($^{ANOVA}F$ = 6.37, $p < 0.001$) concentrations of nickel were observed in the made tea samples (Table 5.3). The highest mean concentration of zinc (55.89 mg/kg) in the tea leaves was found in the Kamalganj sub-district and the lowest mean concentration of zinc (28.89 mg/kg) was found in the Panchagarh Sadar sub-district. Significantly different ($^{ANOVA}F$ = 3.95, $p < 0.01$) concentrations of zinc were observed in the tea leaves (Table 5.3). The highest mean concentration of zinc (221 mg/kg) in the made tea was found in the Sylhet Sadar sub-district and the lowest mean concentration of zinc (22.98 mg/kg) in the made tea was found in the Fatikchhari sub-district. Significantly different ($^{ANOVA}F$ = 5.51, $p < 0.001$) concentrations of zinc were observed in the made tea samples (Table 5.3).

5.4.4 Estimation of translocation factor/transfer factor (TF) of heavy metals in tea plants

Total concentration of heavy metal in tea leaves was divided by the total concentration of heavy metal in soil to measure the translocation factor or transfer factor of heavy metals (Kachenko and Singh, 2006). The food chain is the main process to bring the human beings to the exposure of heavy metals by the transfer of heavy metals from soil to plant. The translocation of heavy metals from soils to the plant body is described by the transfer factor (TF). Higher transfer factor (TF) values (≥1) indicated that higher absorption of metal from soil by the tea plant which means accumulation of heavy metals in the plants. On the contrary, TF values around 1 indicated that the plants are not influenced by the metals. The lower TF values (<1) indicated poor response of plants towards metal absorption which means plants abstained to uptake and accumulation of heavy metals (Olowoyo *et al*., 2010; Rangnekar *et al*., 2013). The permissible limit of chromium in plants is 1.3 mg/kg (Dabanović *et al*., 2016). In the present study, the chromium content in the tea leaves collected from different tea gardens were above the permissible limit. The transfer factor for chromium was 0.72 (Table 5.4). Dabanović *et al*. (2016) reported that, the permissible limit of copper in plants is 10 mg/kg. The copper content in the tea leaves collected from different tea gardens were above the permissible limit. The transfer factor for copper was 1.72 (Table 5.4). At low pH, the transfer process of copper from soil to tea leaves could be related to the redox reaction which is commonly caused by Fe/Mn hydroxides and organic matter (Grybos *et al*., 2007). The permissible limit of iron in plants is 450 mg/kg (Dabanović *et al*., 2016). Except Karotoa tea garden and Kazi and Kazi tea garden, the iron content in the tea leaves collected from different tea gardens were below the permissible limit. The transfer factor for iron was 0.009 (Table 5.4). The permissible limit of manganese in plants is 200 mg/kg (Dabanović *et al*., 2016). Except Karotoa tea garden, the manganese content in the tea leaves collected from different tea gardens were above the permissible limit. The transfer factor for manganese was 3.44 (Table 5.4). Geochemistry of iron and manganese hydroxides in soil could be related to enrichment of Mn in tea (Karak and Bhagat, 2010; Xu *et al*., 2015; Karak *et al*., 2017). The permissible limit of nickel in plants is 10 mg/kg (Dabanović *et al*., 2016). The nickel content in the tea leaves collected from different tea gardens were below the permissible limit. The transfer factor for nickel was 0.44 (Table 5.4). The permissible limit of zinc in plants is 50 mg/kg (Dabanović *et al*., 2016). The zinc content in the tea leaves collected from Allynugger, BTRI farm, Jagcherra, Patrakhola, Rajghat and Sathgao tea gardens were above the permissible limit. The transfer factor for zinc was 1.15 (Table 5.4). In the studied tea leaf samples, the translocation factors of heavy metals were reduced as follows: Mn > Cu > Zn > Cr > Ni > Fe. Generally, with decreasing the concentrations of metal in soils the transfer factors increased. Thus, uptake saturation can explain the lower transfer factors in tea plants (Karak *et al*., 2014). In another research, the transfer factors of manganese, zinc and copper in lettuce, spinach, radish and carrot were found to be reduced in the order: Mn > Zn > Cu (Intawongse and Dean, 2006).

Table 5.4: Transfer factors of heavy metals from tea garden soils to tea leaves

Tea Estate	Chromium	Copper	Iron	Manganese	Nickel	Zinc
Allynugger	0.260	1.677	0.0062	3.705	0.481	1.585
Bilashcherra	0.340	1.647	0.0065	2.727	0.679	1.065
BTRI Farm	1.496	2.276	0.0082	4.091	0.662	1.647
Jagcherra	1.105	1.893	0.0053	4.784	0.698	0.540
Karnafuli	0.158	1.260	0.0052	7.031	0.297	1.159
Karotoa	0.165	0.820	0.019	1.329	0.243	0.698
Kazi & Kazi	0.425	1.289	0.033	1.902	0.318	0.764
Malnicherra	0.975	2.150	0.0052	1.796	0.364	0.519
Neptune	0.526	1.412	0.0047	3.431	0.523	0.889
Oodaleah	0.446	1.122	0.0044	2.684	0.308	0.374
Patrakhola	0.624	3.098	0.0066	4.120	0.457	1.977
Rajghat	1.573	1.331	0.0048	3.056	0.352	1.013
Sathgao	1.207	2.338	0.0058	4.000	0.378	2.758
Mean	**0.72**	**1.72**	**0.009**	**3.44**	**0.44**	**1.15**

Geochemical behaviours of chromium, copper, nickel and zinc are mainly related to soil pH and iron oxides in tea gardens. Soil texture, soil organic matter and fertilizer application are the main factors influencing transfer factors of heavy metals from soil to tea. So geological background and natural factors are the main sources of heavy metals in tea. Beside minerals properties, soil pH is thought as the main factor that influence the transfer and accumulation of heavy metals from soil to crop plant (Huang *et al*., 2019; Zhang *et al*., 2020). Tea plants have higher potential risk of heavy metal accumulation from soil under low soil pH (Fung and Wong, 2002). Tea plants can accumulate heavy metals such as zinc, copper and nickel easily (de Oliveira *et al*., 2018; Malik *et al*., 2008) and concentrations of these heavy metals in tea could reach 2 to dozens of times higher (Karak and Bhagat, 2010). Concentrations of various heavy metals in the tea leaves indicated that the accumulation process of metals from soil could be influenced by both tea plant metabolism and heavy metals concentration in soil directly under acid soil condition (Chen *et al*., 2009; Yemane *et al*., 2008).

5.4.5 Heavy metals in the tea leaves of different tea gardens located at different physiographic regions

The concentration of heavy metals in the tea leaves of tea gardens located at different physiographic regions of Bangladesh are presented in table 5.5. The highest mean concentration of chromium was found in the tea leaves of Old Himalayan Piedmont Plain (13.55 mg/kg). No significant variations of the concentrations of chromium were observed in the tea leaves of the different physiographic regions (Table 5.5). The highest mean concentration of copper was found in the tea leaves of Northern and eastern piedmont plains (16.34 mg/kg). Significant variations ($^{ANOVA}F = 3.15$, $p < 0.05$) in the concentrations of copper were observed in the tea leaves of the different physiographic regions (Table 5.5). The highest mean concentration of iron was found in the tea leaves of Old Himalayan Piedmont Plain (702 mg/kg). Significant variations ($^{ANOVA}F = 6.00$, $p < 0.01$) in the concentrations of iron were observed in the tea leaves of the different physiographic regions (Table 5.5). The highest mean concentration of manganese was found in the tea leaves of Northern and eastern hills (732 mg/kg). Significant variations ($^{ANOVA}F = 8.68$, $p < 0.001$) in the concentrations of manganese were observed in the tea leaves of the different physiographic regions (Table 5.5).

The highest mean concentration of nickel was found in the tea leaves of Northern and eastern hills (4.11 mg/kg). No significant variations in the concentrations of nickel were observed in the tea leaves of the different physiographic regions (Table 5.5). The highest mean concentration of zinc was found in the tea leaves of Northern and eastern piedmont plains (47.02 mg/kg). No significant variations in the concentrations of zinc were observed in the tea leaves of the different physiographic regions (Table 5.5).

Table 5.5: Concentrations of heavy metals in the tea leaves of different physiographic regions

Physiography	Sample		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Northern and Eastern Hills	Tea Leaf (n = 61)	*Min – Max*	BDL – 54.15	2.15 – 20.82	39.56 – 208	145.6 – 1880	0.75 – 11.50	1.17 –163
		Mean±s.e.	11.59±1.31	14.02±0.51	86.69±3.67	732±54.0	4.11±0.29	45.74±3.95
Northern and Eastern Piedmont plains	Tea Leaf (n = 33)	*Min – Max*	BDL – 25.05	7.90 – 31.41	27.28 – 136	96.0 – 2006	0.50 – 10.80	11.25 – 99.85
		Mean±s.e.	12.00±1.25	16.34±1.03	82.86±4.23	675.8±87.1	3.92±0.42	47.02±4.05
Old Himalayan Piedmont Plain	Tea Leaf (n= 18)	*Min – Max*	4.65 –77.85	5.95 –19.30	15 –7300	143 – 715	2.00 – 9.45	22.41–37.03
		Mean±s.e.	13.55±4.09	13.81±0.80	702±413	268.7±34.8	3.76±0.47	31.25±0.91
	Tea Leaf	*F value*	0.22	3.15	6.00	8.68	0.20	2.49
		p value	0.801	< 0.05	< 0.05	< 0.001	0.817	0.088

5.4.6 Heavy metals concentration in the tea leaves of different age of tea plants

The concentration of heavy metals in the tea leaves of different aged tea plants of the tea gardens are presented in table 5.6. Among the heavy metals, only nickel was observed to be significantly varied (${}^{ANOVA}F = 3.65$, $p < 0.05$) in the leaves of different age of tea plants. The highest mean concentration of nickel was found in the tea leaves of the young tea (4.73 mg/kg) followed by mature tea (3.87 mg/kg) and old tea (3.34 mg/kg). The concentrations of chromium, copper, iron, manganese and zinc were observed to be insignificantly varied in the leaves of different age of tea plants (Table 5.6).

5.4.7 Heavy metals concentration in the made tea of different grades

The concentrations of heavy metals in the made tea of different grades are presented in table 5.7. Among the heavy metals, copper and nickel were observed to be significantly varied (${}^{ANOVA}F = 17.36$, $p < 0.001$ for copper and ${}^{ANOVA}F = 3.44$, $p < 0.05$ for nickel, respectively) in the made tea of different grades. Considering the mean concentrations of copper in the made tea, the grades were found to be in the following order: Orthodox black tea, 60.35 mg/kg > PF, 18.39 mg/kg > Green Tea, 17.37 mg/kg > CD, 15.68 mg/kg > GBOP, 14.62 mg/kg. The mean concentrations of nickel in the made tea of different grades were observed to be in the following order: CD, 6.89 mg/kg > GBOP, 6.85 mg/kg > PF, 6.84 mg/kg > Green Tea, 3.16 mg/kg > Orthodox, 2.95 mg/kg. The concentrations of chromium, iron, manganese and zinc were observed to be insignificantly varied in the made tea of different grades (Table 5.7).

Table 5.6: Concentrations of heavy metals in the tea leaves of different age of tea plants

Plant age	Sample		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Mature Tea (11-40yrs)	Tea Leaf (n = 42)	*Min – Max*	BDL – 77.85	5.95 – 31.41	31.0 – 2363.0	162.8 – 1880.1	1.15 – 10.80	11.25 – 133.90
		Mean±s.e.	12.75±2.17	15.24±0.75	185.5±61.3	682.7±68.2	3.87±0.30	37.83±3.48
Old Tea (41-60yrs)	Tea Leaf (n = 33)	*Min – Max*	BDL –54.15	2.15 –20.82	47.10–133.44	145.6–1849.1	0.50–8.85	11.41–163.40
		Mean±s.e.	12.82±1.73	13.86±0.79	87.61±3.11	677.4±78.1	3.34±0.31	46.56±5.22
Young Tea (6-10 yrs)	Tea Leaf (n= 37)	*Min – Max*	BDL – 25.05	3.55 –31.07	15–7300	96.0–2006.4	1.10–11.50	1.17–121.60
		Mean±s.e.	11.46±1.06	14.75±0.75	270±195	561.3±74.1	4.73±0.45	48.08±4.42
	Tea Leaf	*F value*	0.18	0.82	0.55	0.88	3.65	1.75
		p value	0.839	0.444	0.579	0.418	< 0.05	0.179

Table 5.7: Concentration of heavy metals in the made tea of different grades

Made Tea Grade	Sample		Cr (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
CD	Black Tea (n = 11)	*Min – Max*	8.80 – 44.75	10.45 –22.53	28 –2174	183.0 – 1176.6	2.55 – 9.95	24.2 –541.0
		Mean±s.e.	20.07±3.21	15.68±1.05	629±202	637.6±94.2	6.89±0.73	164.2±53.3
GBOP	Black Tea (n = 11)	*Min – Max*	BDL – 25.95	7.95 – 22.30	5.00 – 926.0	211–1240	3.05 –11.25	14.4 –295.5
		Mean±s.e.	16.05±2.32	14.62±1.24	254.6±80.2	612±102	6.85±0.78	95.2±31.4
Green Tea	Green Tea (n = 5)	*Min – Max*	2.05 – 9.95	2.45 – 23.71	36.0 – 281.4	238 –762	1.95 – 4.40	22.0 –395.5
		Mean±s.e.	4.67±1.54	17.37±3.82	133.7±40.7	510±109	3.16±0.46	208.4±76.9
Orthodox	Black Tea (n =1)	*Min – Max*	0.55	60.35	205.00	312.00	2.95	40.78
		Mean±s.e.	0.55	60.35	205.00	312.00	2.95	40.78
PF	Black Tea (n = 11)	*Min – Max*	6.70 – 173.5	12.04 – 29.45	3.00 – 2274	205 – 1233	3.35 – 10.55	20.7 –275.6
		Mean±s.e.	35.4±15.8	18.39±1.87	359±194	664±105	6.84±0.64	116.0±32.0
	Made Tea	*F value*	1.36	17.36	1.08	0.43	3.44	0.87
		p value	0.269	< 0.001	0.382	0.788	< 0.05	0.492

5.4.8 Biochemical parameters in the made tea of different tea gardens of Bangladesh

The concentration of biochemical parameters in the made tea of different tea gardens of Bangladesh are presented in table 5.8. The mean concentrations of moisture in the made tea samples of different tea gardens were observed to be varied from 2.22 to 5.87%. Vankatesan *et al*. (2006) reported that for better quality of tea concentration of moisture should be controlled at 2.5 to 6.5%. The highest mean concentration of protein (25.39%) in the made tea was found in the Malnicherra tea garden and the lowest mean concentration of protein (18.62%) in the made tea was found in the Rajghat tea garden. Significant variations ($^{ANOVA}F = 4.68$, $p < 0.001$) in protein contents were observed in the made tea of different tea gardens (Table 5.7). Zhang *et al*. (2015) reported that the concentration of protein in tea was around 21–28%, whereas, in the present study, the mean protein contents in the made tea were observed to be varied from 18.62 to 25.39%.

The highest mean concentration of caffeine (56.20 ppm) in the made tea was found in Jagcherra tea garden and the lowest mean concentration of caffeine (11.32 ppm) in the made tea was found in the Neptune tea garden. Significant variations ($^{ANOVA}F = 12.64$, $p < 0.001$) in the concentrations of caffeine were observed in the made tea of different tea gardens (Table 5.8). Bdullahi *et al*. (2019) reported that the caffeine concentrations ranged from 22.29 to 49.91 ppm in tea of different brands available in the local market in Nigeria. Ali *et al*. (2012) reported that the concentration of caffeine in tea samples ranged from 440 to 473 ppm (mean, 458.6 ppm), whereas, in the present study, the mean concentrations of caffeine in the made tea were observed to be varied from 11.32 to 56.20 ppm. The concentration of caffeine might be affected due to the growing conditions and processing of tea as well as certain types of tea contains somewhat more caffeine than other teas (Wanyika *et al*., 2010).

The highest mean concentration of total polyphenol (23.75%) in the made tea was found in the Jagcherra tea garden and the lowest mean concentration of total polyphenol (5.35%) in the made tea was found in the Karotoa tea garden. Significant variations ($^{ANOVA}F = 5.00$, $p < 0.001$) in the concentrations of total polyphenol were observed in the made tea of different tea gardens (Table 5.8).

Table 5.8: Concentration of biochemical components in the made tea of different tea gardens of Bangladesh.

Tea Estate	Sample	Moisture (%)	Protein (%)	Caffeine (ppm)	Total Polyphenol (%)	Theaflavin (%)	Thearubigin (%)	TF/TR Ratio
Allynugger	Made Tea (n = 3)	2.52±0.13	21.48±0.83	29.47±2.85	9.25±0.08	0.45±0.02	4.64±0.14	1:10.31
Bilashcherra	Made Tea (n = 4)	4.64±0.04	24.50±1.55	37.91±2.45	18.35±4.50	0.52±0.17	7.39±0.03	1:14.21
BTRI Farm	Made Tea (n = 4)	4.64±0.04	24.50±1.55	37.91±2.45	18.35±4.50	0.52±0.17	7.39±0.03	1:14.21
Jagcherra	Made Tea (n = 3)	5.87±0.84	23.39±0.67	56.20±11.4	23.75±0.32	0.006±0.002	BDL	-
Karnafuli	Made Tea (n = 3)	4.26±1.60	20.79±0.66	13.31±0.39	9.43±0.36	0.56±0.06	5.95±0.70	1:10.63
Karotoa	Made Tea (n = 3)	4.32±0.46	19.84±1.18	16.04±0.62	5.35±0.21	0.58±0.01	5.84±0.13	1:10.07
Kazi & Kazi	Made Tea (n = 5)	3.57±0.83	23.61±0.92	22.84±2.00	8.22±1.09	0.56±0.14	7.37±0.33	1:13.16
Malnicherra	Made Tea (n = 3)	4.68±0.29	25.39±0.62	37.22±2.36	10.37±0.44	0.67±0.03	7.20±0.38	1:10.75
Neptune	Made Tea (n = 3)	2.22±0.06	20.49±0.67	11.32±1.82	9.06±0.03	0.60±0.02	6.20±0.33	1:10.33
Oodaleah	Made Tea (n = 3)	2.79±0.25	20.02±0.69	11.43±1.92	9.83±0.40	0.66±0.005	6.84±0.02	1:10.36
Patrakhola	Made Tea (n =3)	3.93±2.06	19.98±0.60	30.96±3.08	10.45±0.57	0.35±0.03	3.78±0.28	1:10.80
Rajghat	Made Tea (n =3)	3.74±0.83	18.62±0.11	25.66±2.95	11.33±0.92	0.37±0.02	3.98±0.26	1:10.76
Sathgao	Made Tea (n =3)	2.40±0.36	24.04±0.72	30.84±1.44	10.27±0.74	0.43±0.046	4.44±0.49	1:10.32
	F value	1.66	4.68	12.64	5.00	2.37	18.00	
	p value	0.126	< 0.001	< 0.001	< 0.001	< 0.05	< 0.001	

The data are mean ± standard error

TF/TR ratio = Ratio of Theaflavin and Thearubigin

Anesini *et al*. (2008) reported that the concentrations of total polyphenol in green tea and black tea were ranged from 14.32 to 21.02% and 8.42 to 17.62%, respectively, whereas, in the present study, the mean concentrations of total polyphenol were observed to be varied from 5.35 to 23.75%. The highest mean concentration of total polyphenol was found in the green tea of Jagcherra tea garden. It might be due to that being non-fermented tea, higher concentration of total polyphenols was observed in green tea (Nibir *et al.*, 2017).

The highest mean concentration of theaflavin (0.67%) in the made tea was found in the Malnicherra tea garden and the lowest mean concentration of theaflavin (0.006%) in the made tea was found in the Jagcherra tea garden. Significant variations ($^{ANOVA}F = 2.37$, $p < 0.05$) in the concentrations of theaflavin were observed in the made tea of different tea gardens (Table 5.8).

The highest mean concentration of thearubigin (7.39%) in the made tea was found in the BTRI and Bilashcherra tea garden while the lowest mean concentration of thearubigin (3.78%) in the made tea was found in the Patrakhola tea garden. Significant variations ($^{ANOVA}F = 18.00$, $p < 0.001$) in the concentration of thearubigin were observed in the made tea of the different tea gardens (Table 5.8). Vankatesan *et al*. (2006) reported that the optimum ratio of theaflavin/thearubigin (TF/TR) to be 1:10, whereas, in the present study, in general, the ratio of the mean concentrations of theaflavin and thearubigin were observed around 1:10 in all the made tea samples.

The moisture content of tea not only determines the shelf life of tea, but also affects the physical and chemical reactions in tea processing, so measurement of moisture content is an important task for producing high-quality tea (Okamura, 2000). Caffeine, which is found in tea imparts bitterness and acts as a flavor constituent (Leo, 1992). Earlier studies revealed that caffeine content is associated to origin, genetic and environmental variability, harvest time and processing manner of plant material (Athayde *et al*., 2000). Polyphenols are the most significant group of tea components and have a wide range of pharmaceutical properties including antioxidative, anticarcinogenic and antiarteriosclerotic (Atoui *et al.*, 2005). Depending on how the tea is harvested, handled, processed, and brewed, the polyphenol level in the tea can vary. During black tea processing, catechins (flavan-3-ol) in tea shoots undergo oxidative changes to form theaflavins (TFs) and thearubigins (TRs), which are largely responsible for brightness, strength and colour of black tea (Kottawa-Arachchi *et al*., 2014). Thearubigins are red in colour and are responsible for much of the staining effect of tea. The taste of tea liquor depends on the ratio of TF, TR concentrations. This ratio dominates the two important taste attributes like briskness and strength (Yasin *et al*., 2020). However, these chemicals also determine the brightness and colour of tea liquor (Roberts, 1962). It is said that the ratio should be 10 for good quality tea. Less or more than 10 is not good for the quality of the tea (Yasin *et al*., 2020). In the present study, except Bilashcherra, BTRI Farm and Kazi & Kazi tea garden in all the made tea samples the ratio of theaflavin and thearubigin is around 1:10 which is good for the quality of tea. In the black tea samples of Karotoa tea garden, the ratio of theaflavin and thearubigin was very closest to 1:10. The made tea samples of Jagcherra tea garden was green tea which contained higher amount of total polyphenol than the made tea of other tea gardens. During green tea processing, oxidation step is absent which could not convert total polyphenol to theaflavin and thearubigin.

5.4.9 Biochemical parameters in the made tea of different grades

The concentrations of different biochemical parameters in the made tea of different grades are presented in table 5.9. The highest mean concentration of moisture (5.78%) was found in green tea and the lowest mean concentration of moisture (2.43%) was found in orthodox black tea. Significant variations ($^{ANOVA}F = 4.03$, $p < 0.01$) in the moisture contents were observed in the made tea of different grades (Table 5.9). The highest mean concentration of caffeine (44 ppm) was found in green tea and the lowest mean concentration of caffeine (21 ppm) was observed in orthodox black tea. Significant variations ($^{ANOVA}F = 2.62$, $p < 0.05$) in the concentrations of caffeine were observed in the made tea of the different grades (Table 5.9). The highest mean concentration of total polyphenol (22.96%) was found in the green Tea and the lowest mean concentration of total polyphenol (7.62%) was found in the orthodox black tea. Significant variations ($^{ANOVA}F = 18.48$, $p < 0.001$) in the concentrations of total polyphenol were observed in the made tea of different grades (Table 5.9). The highest mean concentration of theaflavin (0.725%) was found in the orthodox black tea and the lowest mean concentration of theaflavin (0.0045%) was found in the green Tea. Significant variations ($^{ANOVA}F = 21.13$, $p < 0.001$) in the concentrations of theaflavin were observed in the made tea of different grades (Table 5.9). No significant variations were observed in the concentrations of protein and thearubigin in the made tea of different grades.

Table 5.9: Concentration of different biochemical parameters in the made tea of different grades.

Made Tea Grade	Sample	% Moisture	Protein (%)	Caffeine (ppm)	Total Polyphenol (%)	Theaflavin (%)	Thearubigin (%)
CD	(n =11)	4.27±0.64	21.17±0.73	24.83±3.13	10.40±0.77	0.53±0.04	5.52±0.37
GBOP	(n = 11)	3.15±0.32	21.45±0.80	24.76±3.11	9.29±0.68	0.57±0.04	5.95±0.49
Green Tea	(n = 5)	5.78±0.58	23.84±1.45	44.0±10.2	22.96±3.19	0.0045±0.001	BDL
Orthodox	(n = 1)	2.43	24.06	20.99	7.62	0.72	7.71
PF	(n = 11)	3.07±0.30	22.37±0.66	24.22±3.31	9.35±0.62	0.56±0.041	5.85±0.43
	F value	4.03	1.31	2.62	18.48	21.13	0.76
	p value	< 0.01	0.29	< 0.05	< 0.001	< 0.001	0.52

5.5 Conclusion

The highest mean concentration of chromium, iron, manganese and nickel in the tea leaves were found in the Rajghat, Kazi and Kazi, Jagcherra and Bilashcherra tea gardens, respectively. Copper and zinc concentrations were observed to higher in the tea leaves of BTRI tea garden. The highest mean concentrations of chromium, copper, iron, manganese, nickel and zinc in the made tea were observed in the Bilashcherra and BTRI, Kazi and Kazi, Karotoa, Patrakhola, Malnicherra and Jagcherra tea gardens, respectively. In general, there are no critical limits of heavy metals specifically for tea. Since tea is a widely used popular beverage in Bangladesh, in the present study we compare the concentration of heavy metals in the tea leaves and made tea with the permissible limits of heavy metals for plants. The concentrations of chromium and manganese in the tea leaves and made tea samples and zinc in the made tea samples were higher compared to the permissible limits in plant. The concentrations of copper, iron and nickel in the tea leaves and made tea samples and zinc in the tea leaves samples were within the permissible limits of heavy metals in plants. The concentrations of chromium, copper, iron, manganese and zinc in the tea leaves were observed to be significantly different among the sub-districts. The concentrations of iron, manganese, nickel and zinc in the made tea samples were also observed to be significantly different among the sub-districts. In the tea leaves the concentrations of copper, iron and manganese were observed to be significantly varied among the different physiographic regions of Bangladesh. Among the heavy metals, only nickel was observed to be significantly varied in the leaves of different age of tea plants. In the different grades of made tea, the concentrations of copper and nickel were found to be significantly different. The concentration of biochemical parameters of made tea such as caffeine, protein, total polyphenol, theaflavin and thearubigin was found to be significantly different among the tea gardens of Bangladesh. In the different grades of made tea, the concentration of moisture, total polyphenol and theaflavin was found to be significantly different. Significant and positive relationships between manganese and nickel, manganese and chromium, zinc and copper, iron and nickel were observed in the tea leaves. Significant and positive relationships between manganese and nickel, zinc and manganese were observed in the made tea. There was found a significant and positive relationship between zinc and caffeine in made tea. Significant and negative relationships of zinc were observed with theaflavin and thearubigin. Significant and positive relationships between caffeine and zinc indicated that with increase zinc concentration caffeine content also increased in the made tea. Excessive consumption of caffeine can negatively affect human health in various ways such as increased urination, depleted water-soluble vitamin B6, vitamin C through fluid loss and inhibited the absorption of essential minerals, including calcium, iron, magnesium and B vitamins. Theaflavin and thearubigin are biological compounds belong to flavonoid family. Theaflavin and thearubigin are responsible for the specific color, taste, aroma of black tea and have strong antioxidant capacity. Excess zinc might decrease the quality of tea through decreasing theaflavin and thearubigin contents. There were also found significant and negative relationships between nickel and total polyphenol as well as manganese and thearubigin in the made tea. Total polyphenol or phenolic compounds present in tea have antioxidant properties and potential health benefits. Excess nickel might decrease the quality of tea through decreasing the total polyphenol content. The translocation factors of heavy metals in the studied tea leaves samples were reduced as follows: Manganese > Copper > Zinc > Chromium > Nickel > Iron. The chemical fertilizers, pesticides and other agrochemicals used in the tea gardens might increase the concentrations of heavy metals in the tea gardens. Therefore, it is necessary to analyze the contents of heavy metals in the fertilizers and other agrochemicals used in the tea gardens. It is also important to periodic monitoring to assess the degree of the concentration of heavy metals in the tea garden soils, tea leaves and made tea of Bangladesh. This study also suggests further investigation to assess the release of toxic heavy metals from made tea into tea brew. However, to assess the quality of made tea and tea brew, it is also important to develop a separate standard limits of toxic metal elements in tea.

Chapter Six

General Discussion and Suggestions for Future Research

The present study has been conducted with a large-scale tea garden soil geochemical survey comprising 160 tea garden soils (surface soil = 112 and sub-surface soil = 48) and 32 non-tea garden soils (non-tea garden soils were adjacent to the corresponding tea garden soils) from the geographically different tea gardens of Bangladesh were collected and analysed for a range of geochemical elements such as nitrogen, boron, chromium, copper, iron, manganese, nickel and zinc to assess their variabilities with respect to physiographic regions; land topographic features such as landforms; soil texture; drainage class; soil series as well as the age of tea plants. In the present study, 112 tea leaves and 39 made tea samples were also analysed to assess the concentrations of heavy metals. The made tea samples were also analysed to assess the biochemical parameters. The concentrations of heavy metals in leaves of different age of tea plants located at different sub-districts and physiographic regions of Bangladesh were also assessed. Made tea samples of different grades were analysed to determine the concentrations of heavy metals. The effects of heavy metals on the biochemical parameters of made tea as well as the relationships of the heavy metals in the tea leaves and made tea were also investigated. The transfer factors of chromium, copper, iron, manganese, nickel and zinc from soil to tea plants were also determined.

In Chapter 3 it was shown that tea garden soils in general, had higher mean concentrations of heavy metals (chromium, copper, iron, manganese, nickel and zinc) than the adjacent non-tea garden soils. The highest mean concentrations of chromium, copper, iron and nickel were found in the surface and sub-surface soils of the Karotoa tea garden. Chromium, copper, iron and nickel concentrations were found to be highest in the surface and sub-surface soils of the tea garden located at Panchagarh Sadar sub-district. The mean concentrations of the heavy metals were found within the world-soil average concentrations and the typical concentrations of the heavy metals in soil.

In Chapter 4 it was shown that the concentrations of the heavy metals varied in the tea garden soils of the three different physiographic regions. The concentrations of chromium, copper, iron and nickel were found higher in Old Himalayan Piedmont Plain and varied significantly compared to other physiographic regions. The tea garden soils within different age of tea plants also had geochemical differences. The concentrations of heavy metals were found higher in the soils of young tea compared to other age of tea plants. Among the heavy metals only chromium concentrations were varied significantly. In the soils of different soil series as well as within the soils with different landforms, drainage classes and textural classes also had geochemical differences. The concentrations of heavy metals in the tea garden soils of three physiographic regions were varied with their geochemical differences.

In Chapter 5, it was shown that the highest mean concentration of chromium, iron, manganese and nickel in the tea leaves were found in the Rajghat, Kazi and Kazi, Jagcherra, Bilashcherra and Malnicherra tea gardens, respectively. Copper and zinc concentrations were observed to higher in the tea leaves of BTRI tea garden. The highest mean concentrations of chromium, copper, iron, manganese, nickel and zinc in the made tea were observed in the Bilashcherra and BTRI, Kazi and Kazi, Karotoa, Patrakhola, Malnicherra, Allynugger and Jagcherra tea gardens, respectively. In the tea leaves the concentrations of copper, iron and manganese were observed to be significantly varied among the different physiographic regions of Bangladesh. Among the heavy metals, only nickel was observed to be significantly varied in the leaves of different age of tea plants. The concentration of biochemical parameters of made tea such as caffeine, protein, total polyphenol, theaflavin and thearubigin was found to be significantly different among the tea gardens of Bangladesh. There was found a significant and positive relationship between zinc and caffeine in made tea. Significant and negative relationships of zinc were observed with theaflavin and thearubigin. There were also found significant and negative relationships between nickel and total polyphenol as well as manganese and thearubigin in the made tea. The transfer factors for heavy metals in the investigated tea leaves samples were decreased as follows: Manganese > Copper > Zinc > Chromium > Nickel > Iron.

From the understanding of the objectives and findings of the present study, some suggestions can be proposed for future research on the tea garden soils of Bangladesh:

1. Further large-scale investigation is highly recommended to assess the origin and bioavailability of different heavy metals in the geomorphologically different tea garden soils across Bangladesh as well as to relate this to the yield and nutritional quality of the produced tea.
2. Further investigation is recommended to study other heavy metal elements such as cadmium, lead, mercury and others, which 'were not tested in the present study, in the tea garden soils, tea leaves and made tea.

3. This study also suggests further investigation to assess the release of toxic heavy metals from made tea into tea brew.
4. The present study also suggests research to assess the effects of heavy metals on individual biochemical parameters of tea.
5. As tea is a popular drink, considering the health security issues it is also important to develope separate permissible limits of toxic metal elements in tea to assess the quality of made tea and tea brew.
6. The present study also suggests further research to assess the uptake and transfer factors of heavy metals of the 23 tea clones of BTRI.
7. This study highly recommends further investigation on the toxicity symptoms and effects of higher concentrations of heavy metals on the tea plants as well as the quality of tea.

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Appendix

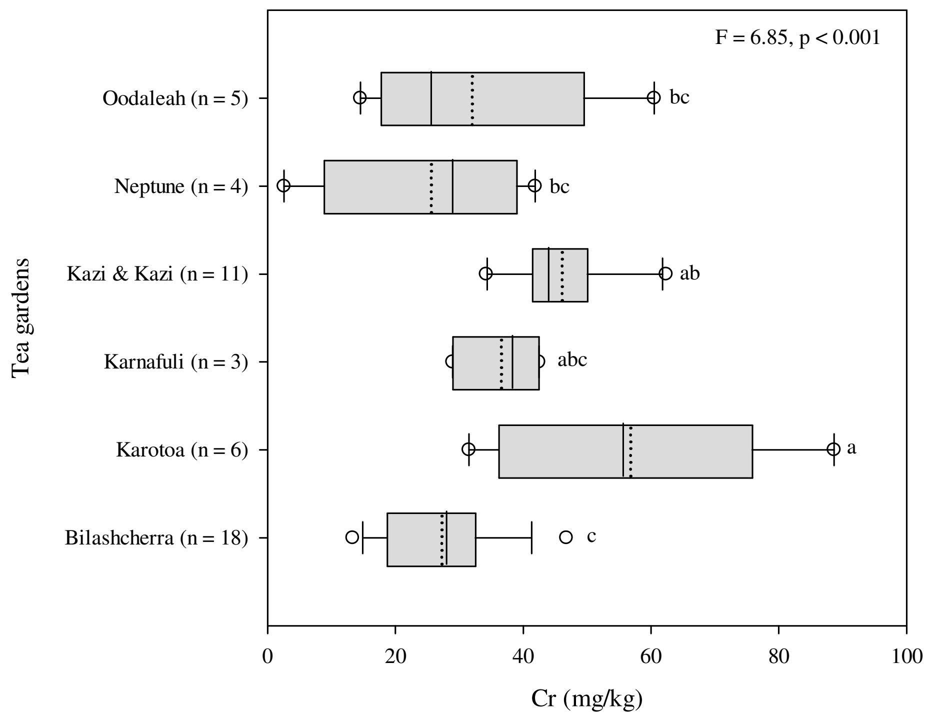

Figure A1: Variability of chromium (mg/kg) concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

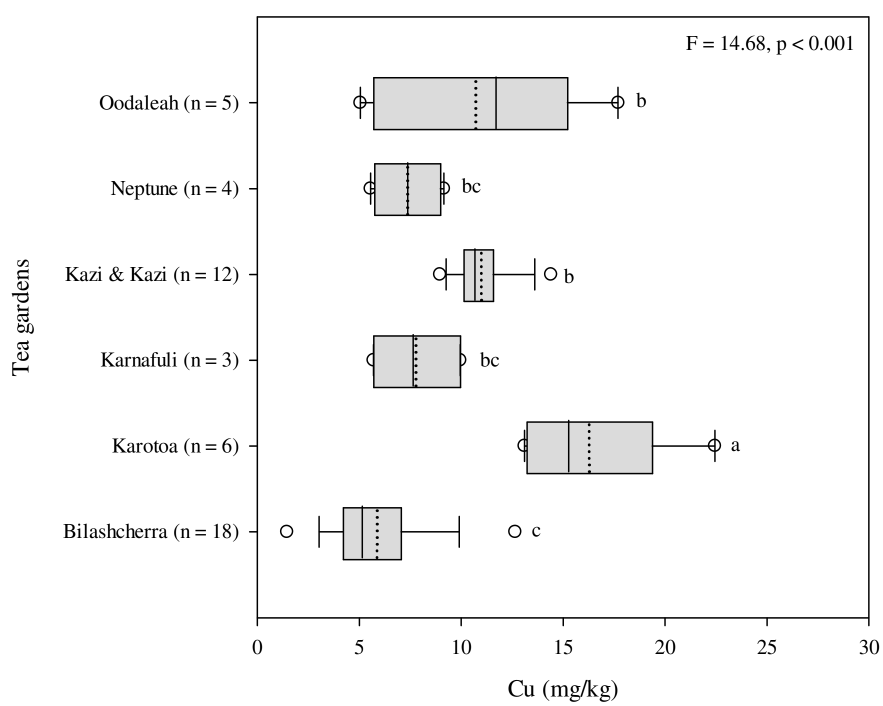

Figure A2: Variability of copper (mg/kg) concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

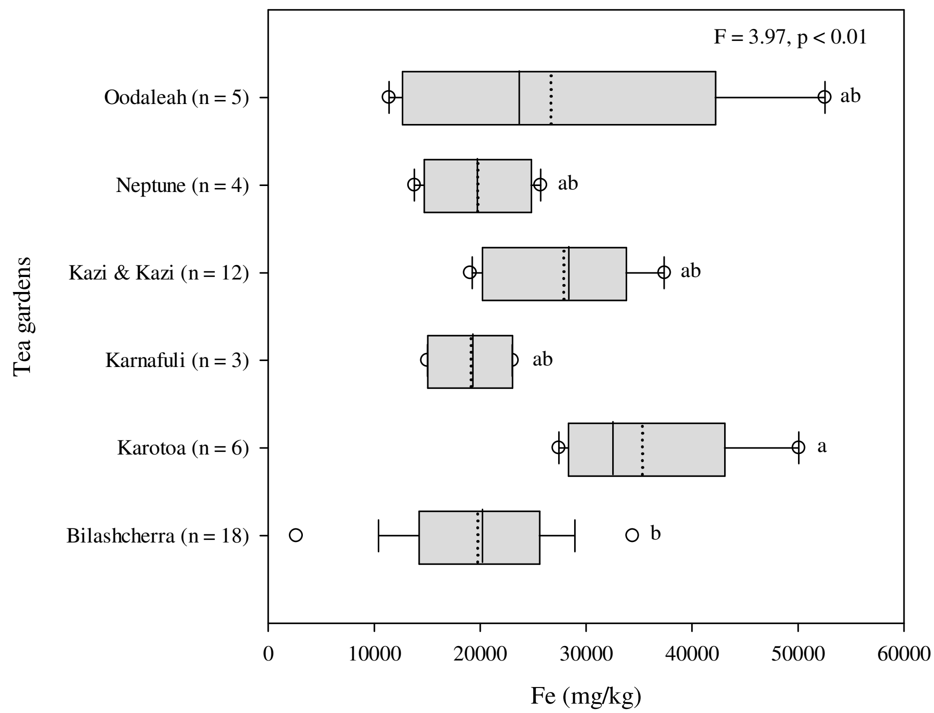

Figure A3: Variability of iron (mg/kg) concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

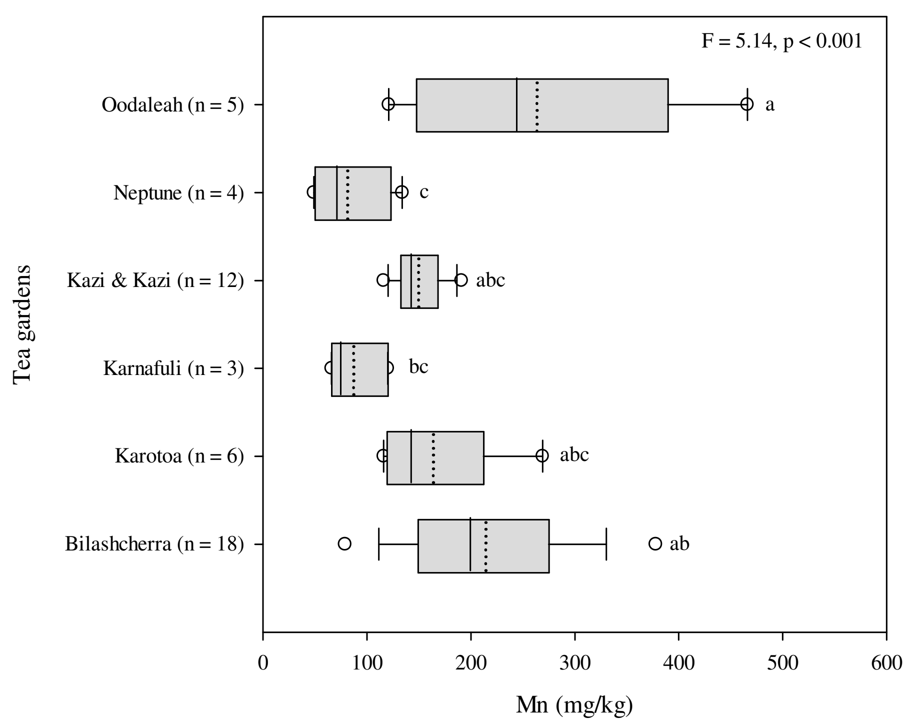

Figure A4: Variability of manganese (mg/kg) concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

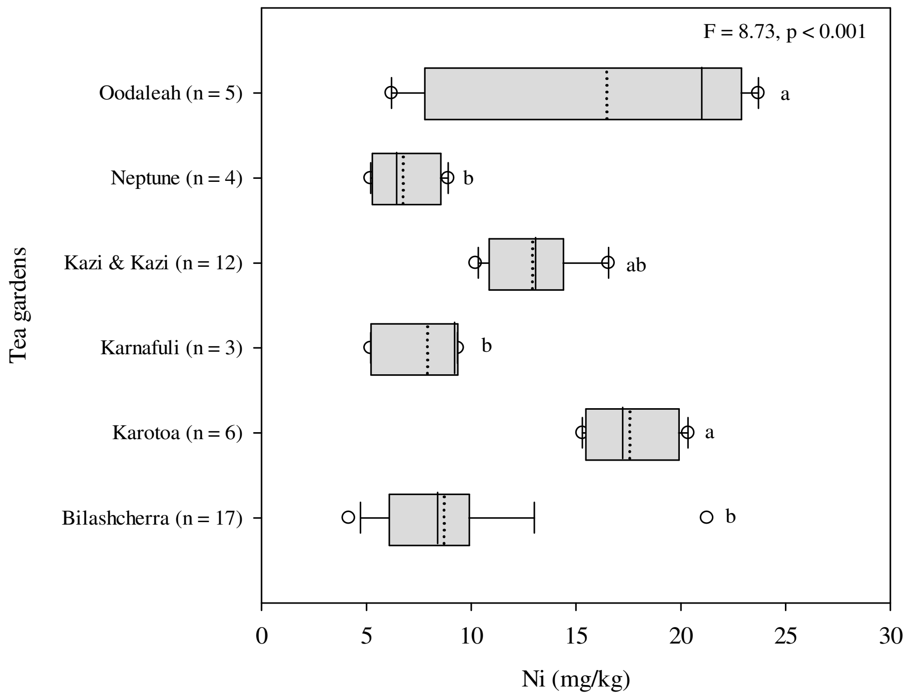

Figure A5: Variability of nickel (mg/kg) concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

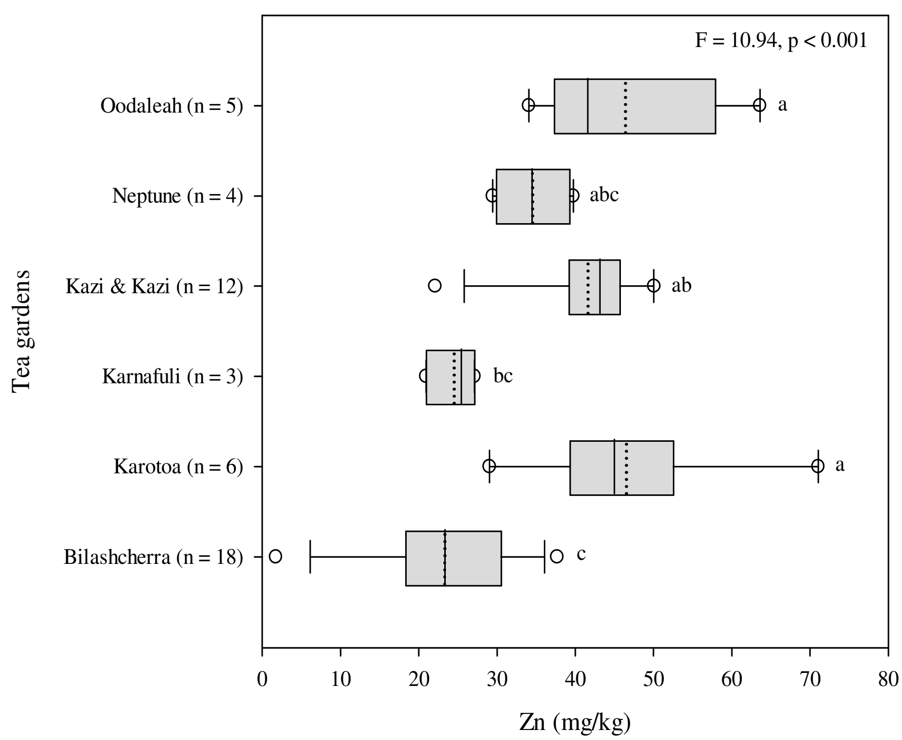

Figure A6: Variability of zinc (mg/kg) concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

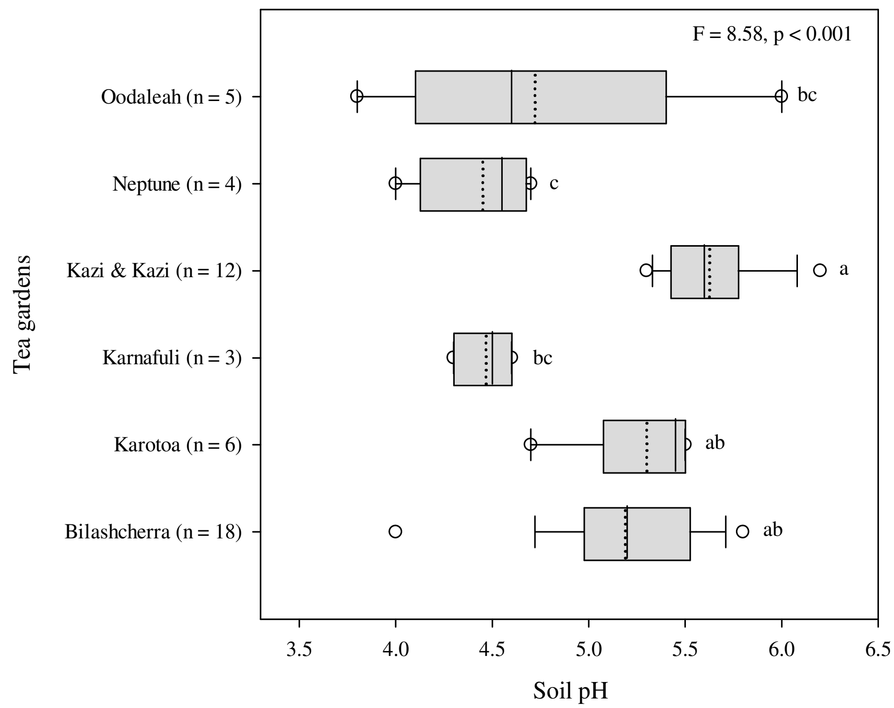

Figure A7: Variability of pH in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

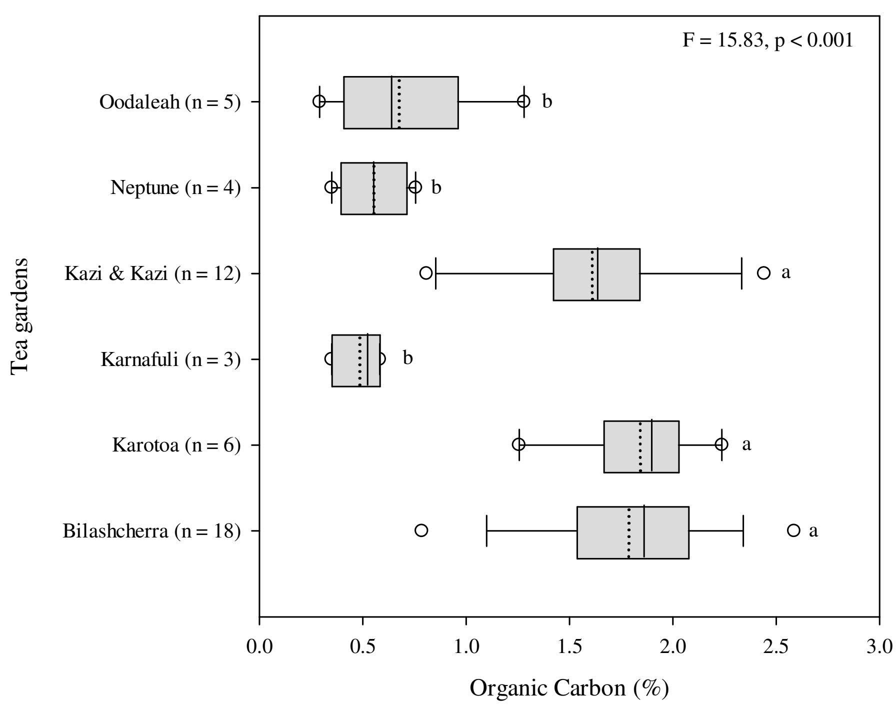

Figure A8: Variability of organic carbon content in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

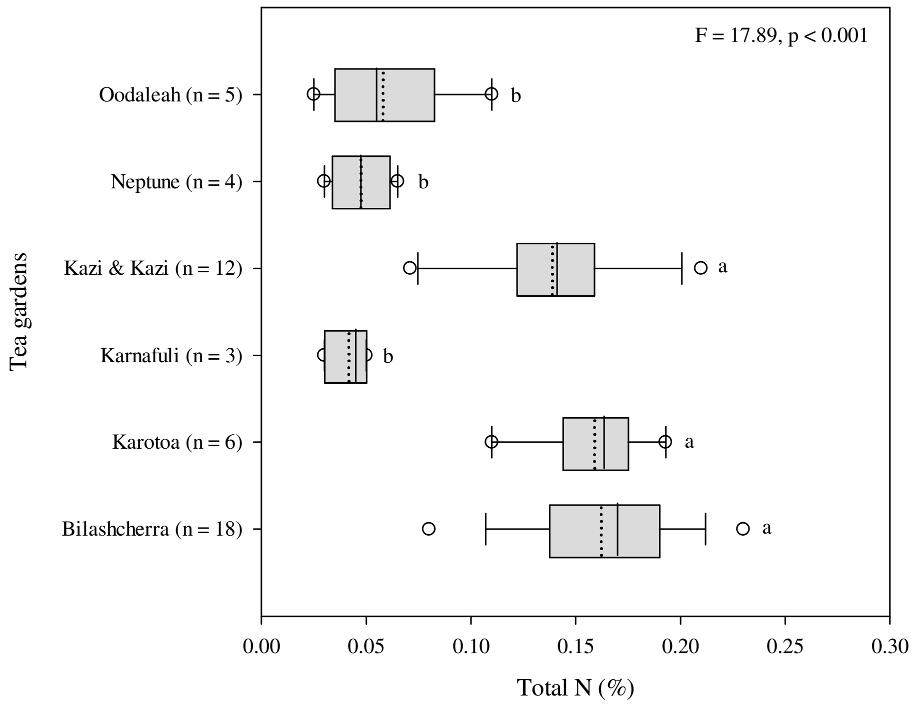

Figure A9: Variability of total nitrogen content in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

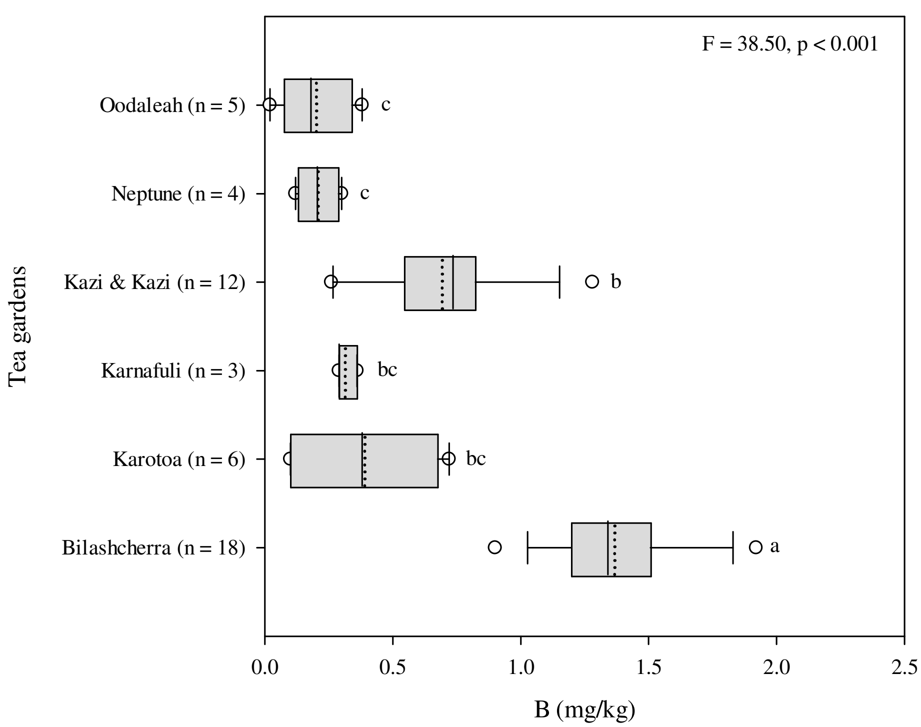

Figure A10: Variability of boron concentrations in the sub-surface soils (23-46 cm depth) of different tea gardens of Bangladesh.

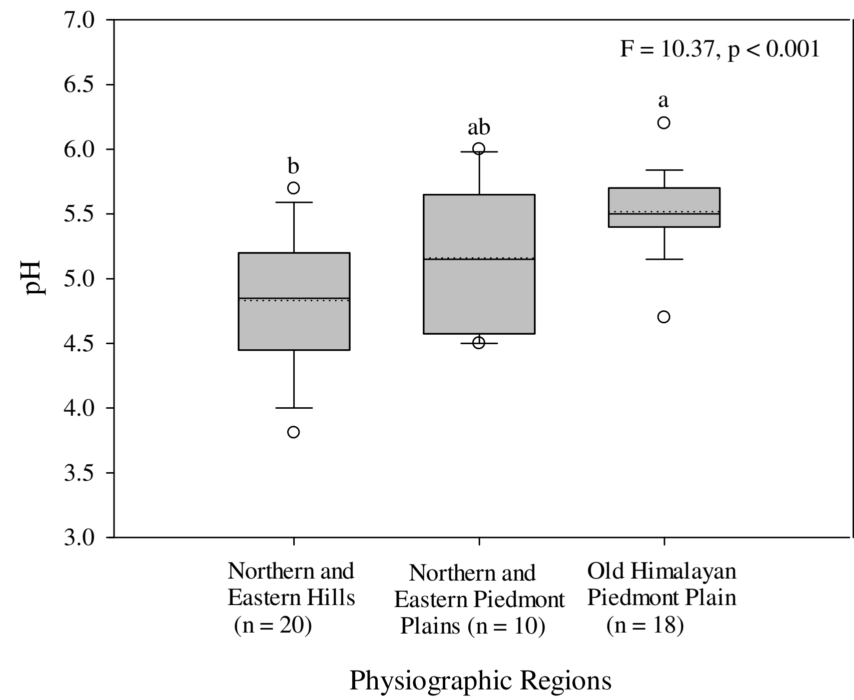

Figure A11: pH in sub-surface soils of the tea gardens located at different physiographic regions.

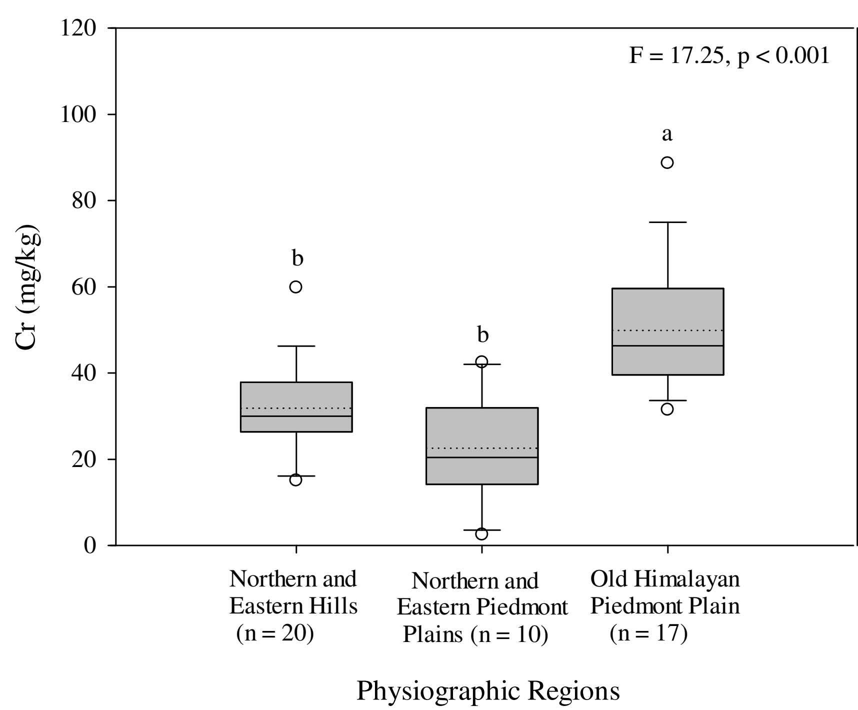

Figure A12: Concentrations of chromium in sub-surface soils of the tea gardens located at different physiographic regions.

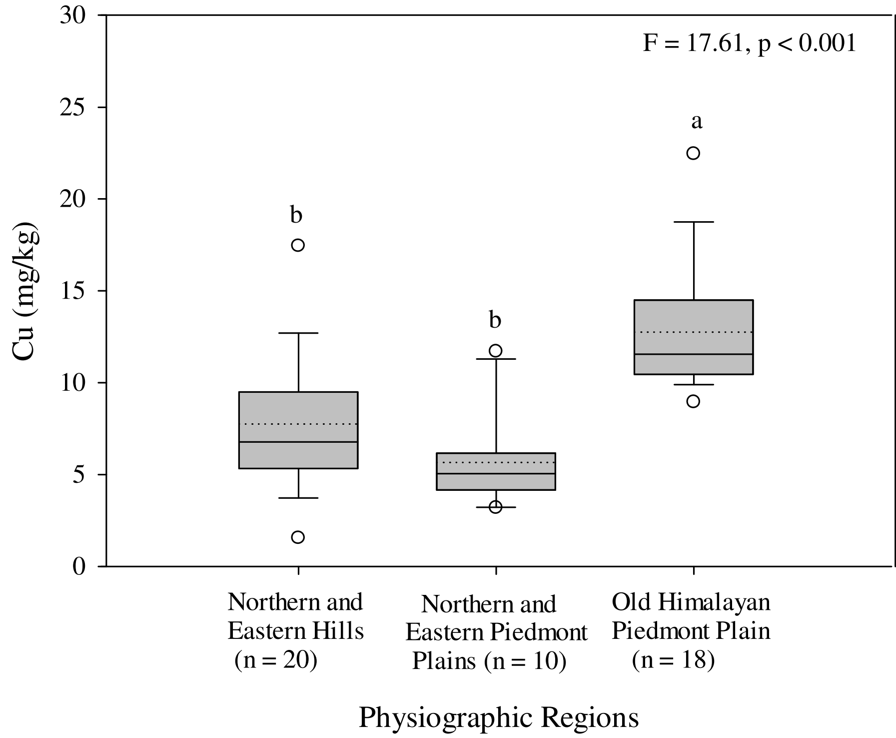

Figure A13: Concentrations of copper in sub-surface soils of the tea gardens located at different physiographic regions.

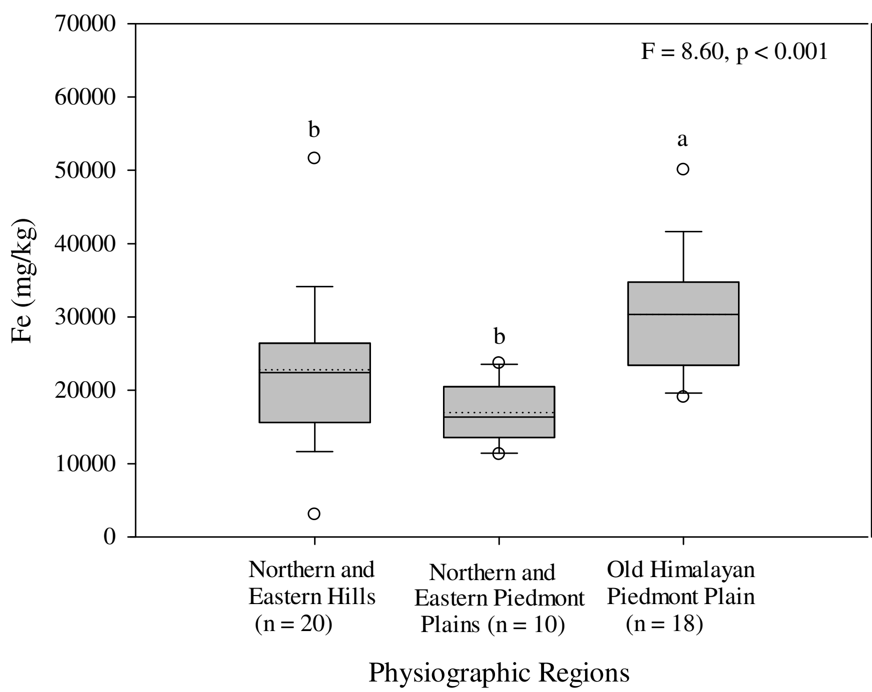

Figure A14: Concentrations of iron in sub-surface soils of the tea gardens located at different physiographic regions.

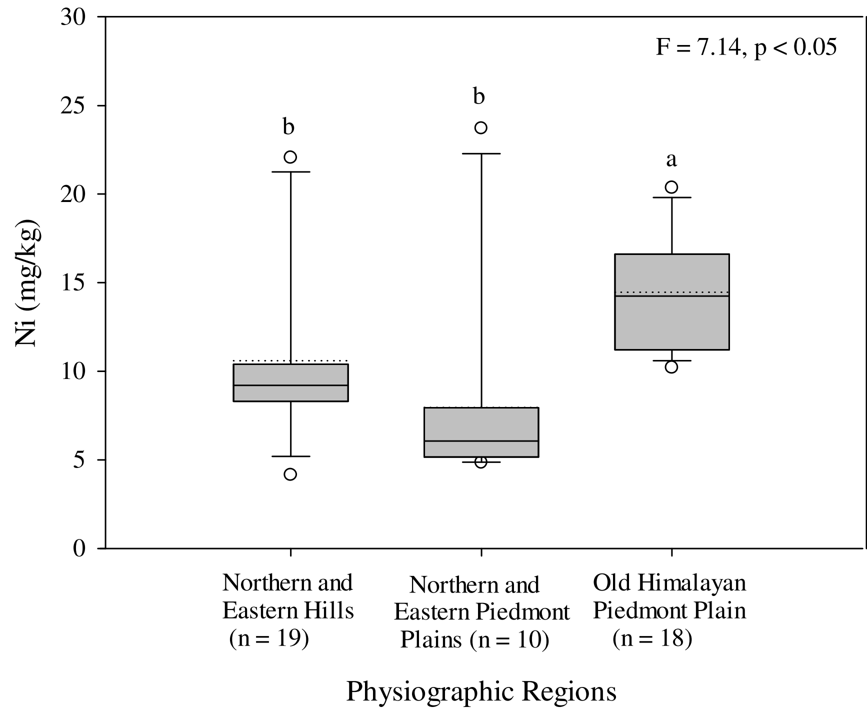

Figure A15: Concentrations of nickel in sub-surface soils of the tea gardens located at different physiographic regions.

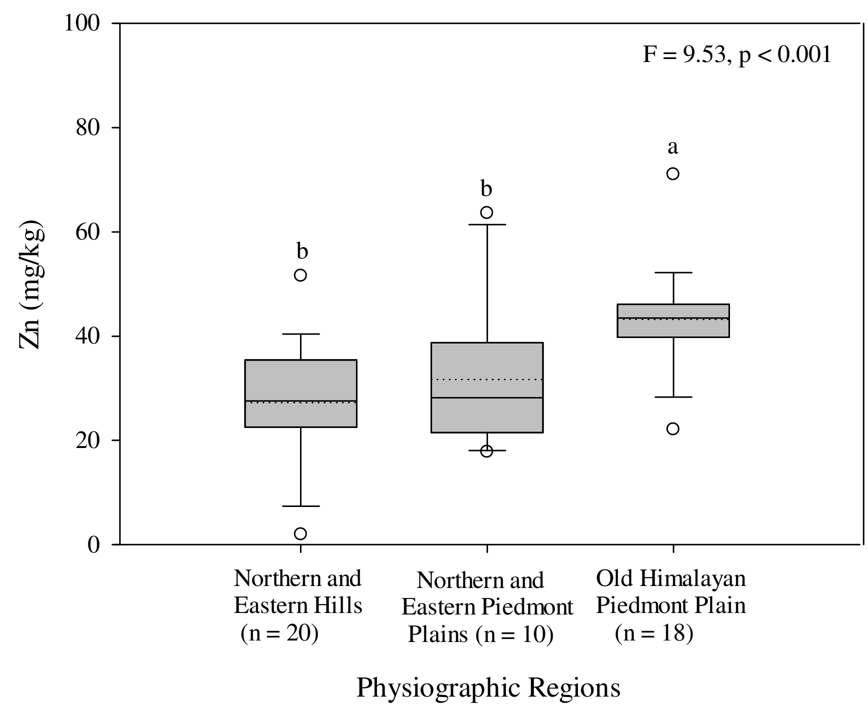

Figure A16: Concentrations of zinc in sub-surface soils of the tea gardens located at different physiographic regions.

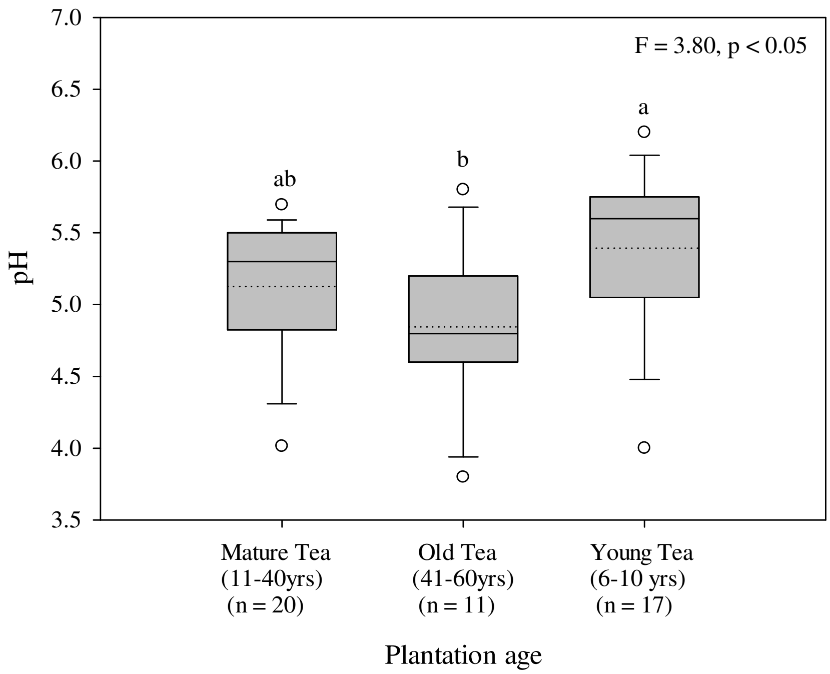

Figure A17: pH in sub-surface soils under the different age of tea plantations.

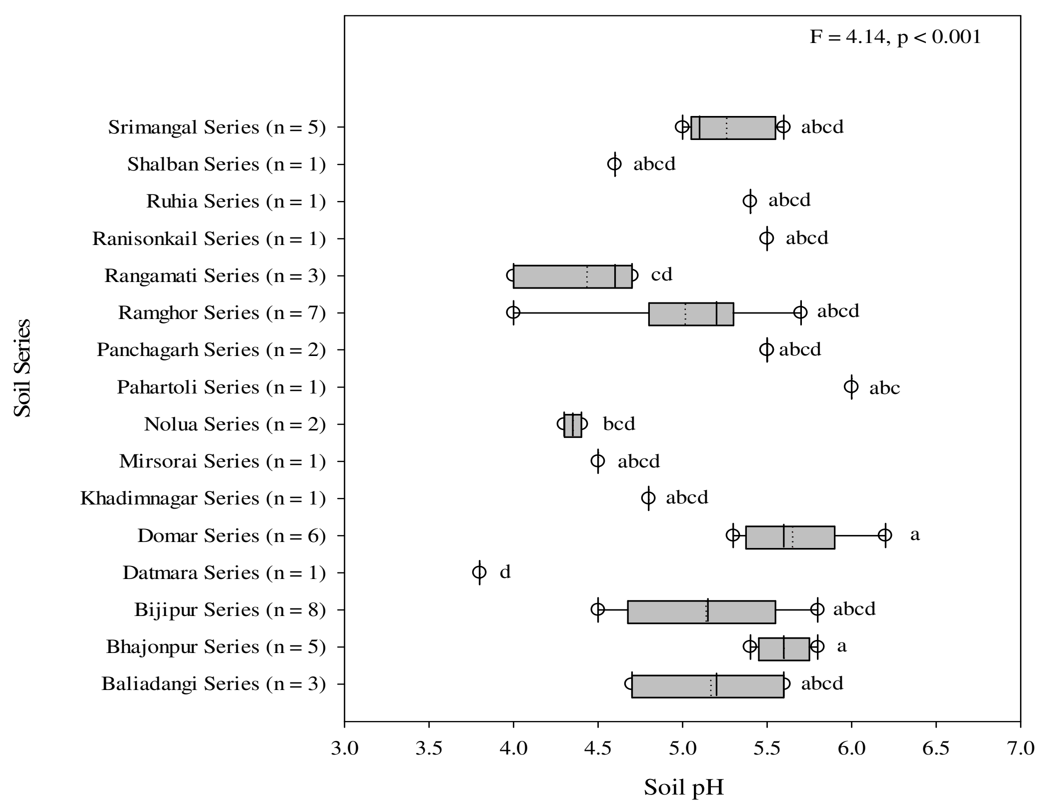

Figure A18: pH in sub-surface soils of different soil series in the tea gardens.

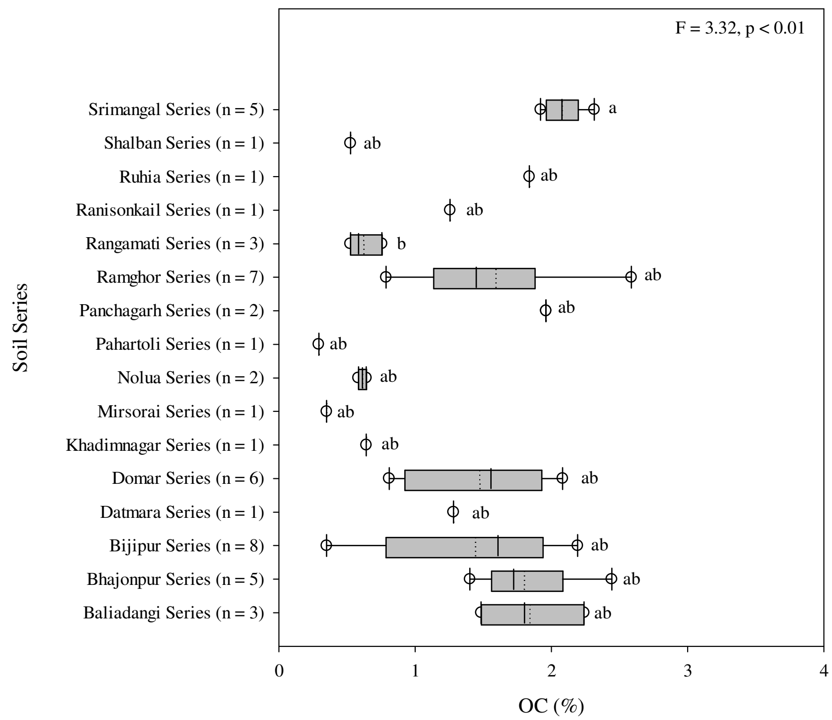

Figure A19: Organic carbon contents in sub-surface soils of different soil series in the tea garden

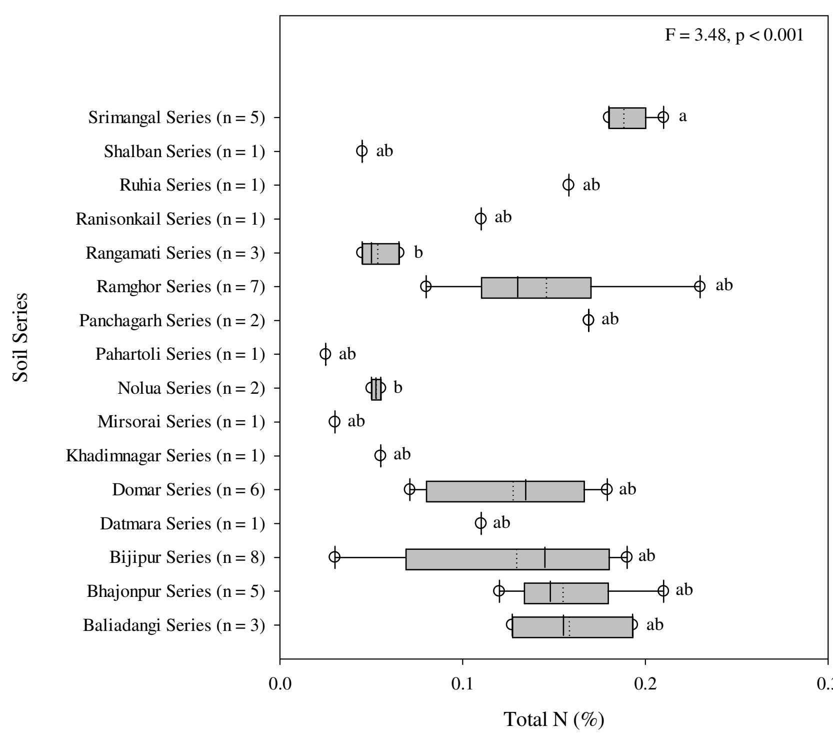

Figure A20: Total nitrogen contents in sub-surface soils of different soil series in the tea gardens.

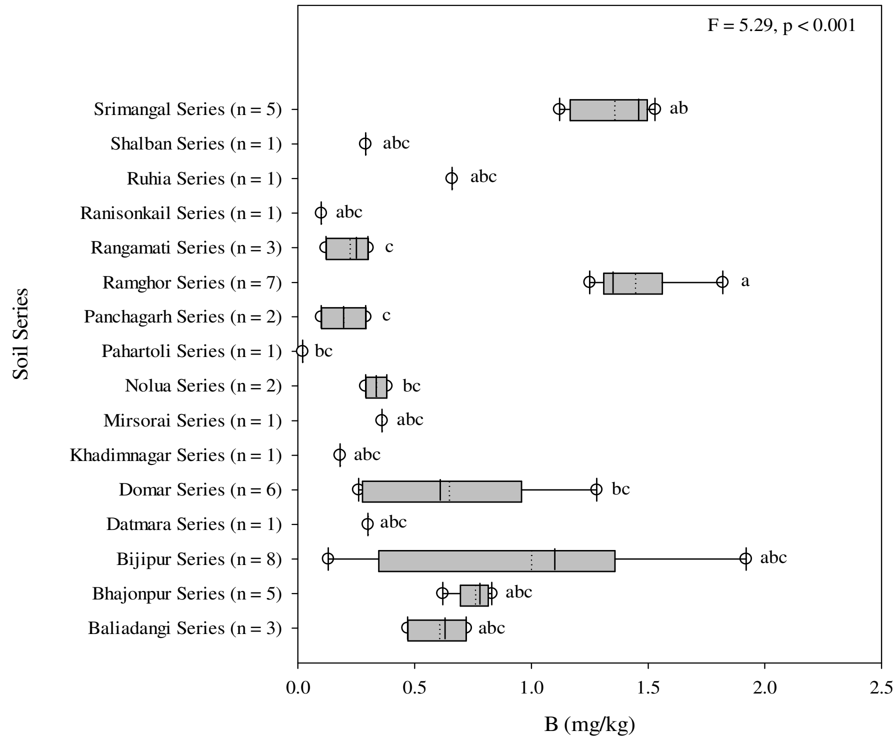

Figure A21: Concentrations of boron in sub-surface soils of different soil series in the tea gardens.

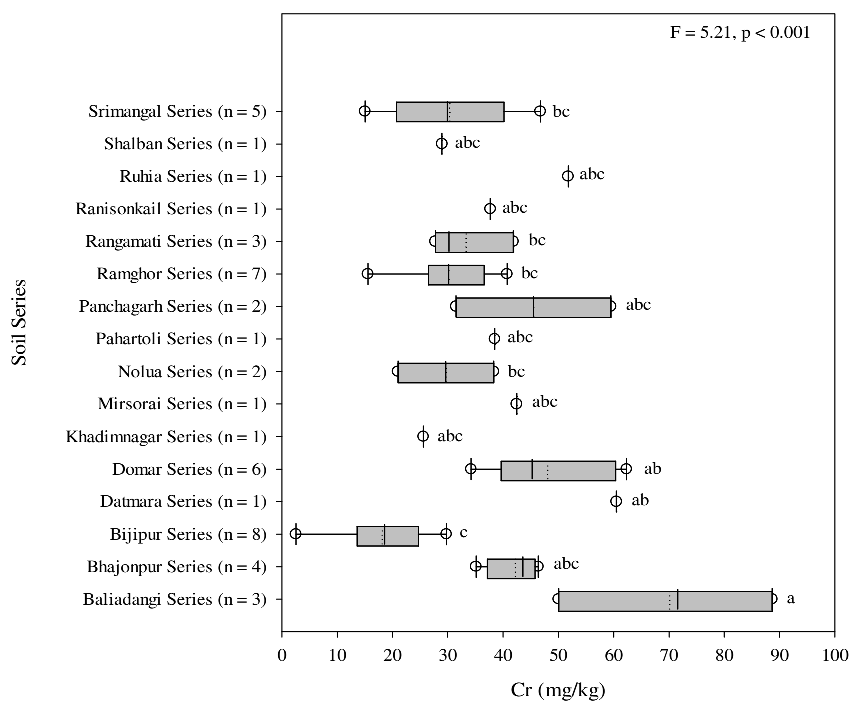

Figure A22: Concentrations of chromium in sub-surface soils of different soil series in the tea gardens.

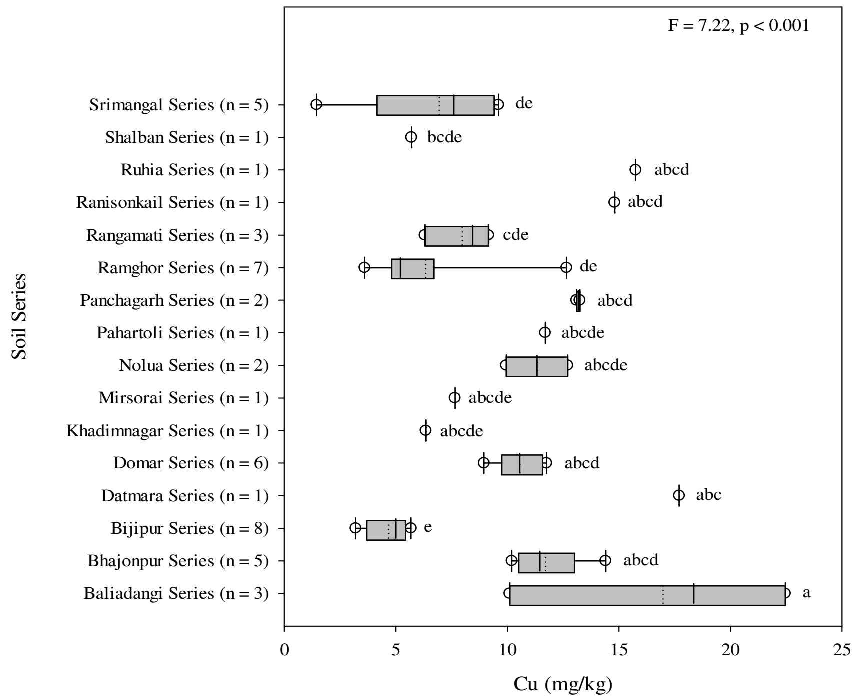

Figure A23: Concentrations of copper in sub-surface soils of different soil series in the tea gardens.

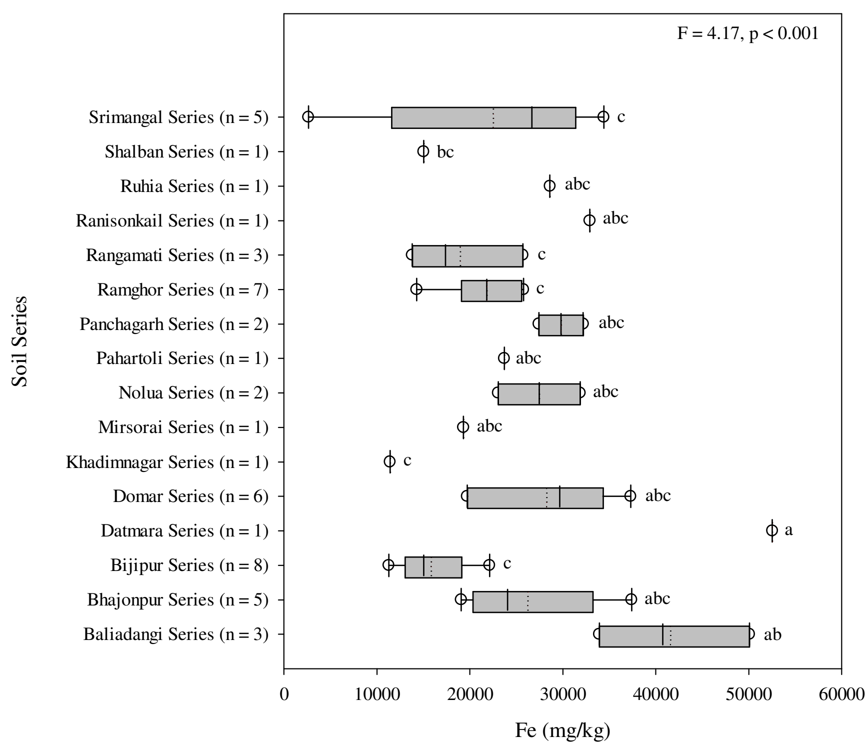

Figure A24: Concentrations of iron in sub-surface soils of different soil series in the tea gardens.

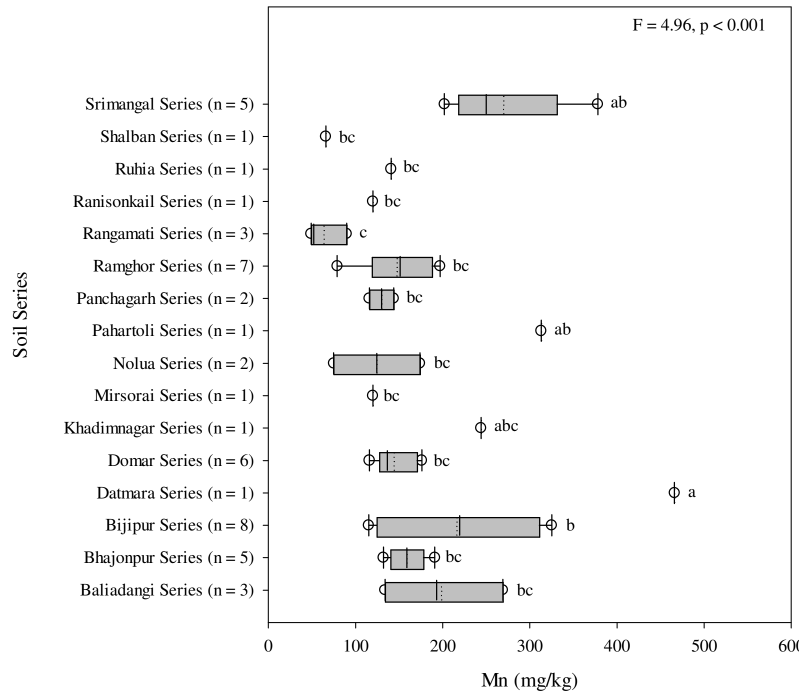

Figure A25: Concentrations of manganese in sub-surface soils of different soil series in the tea gardens.

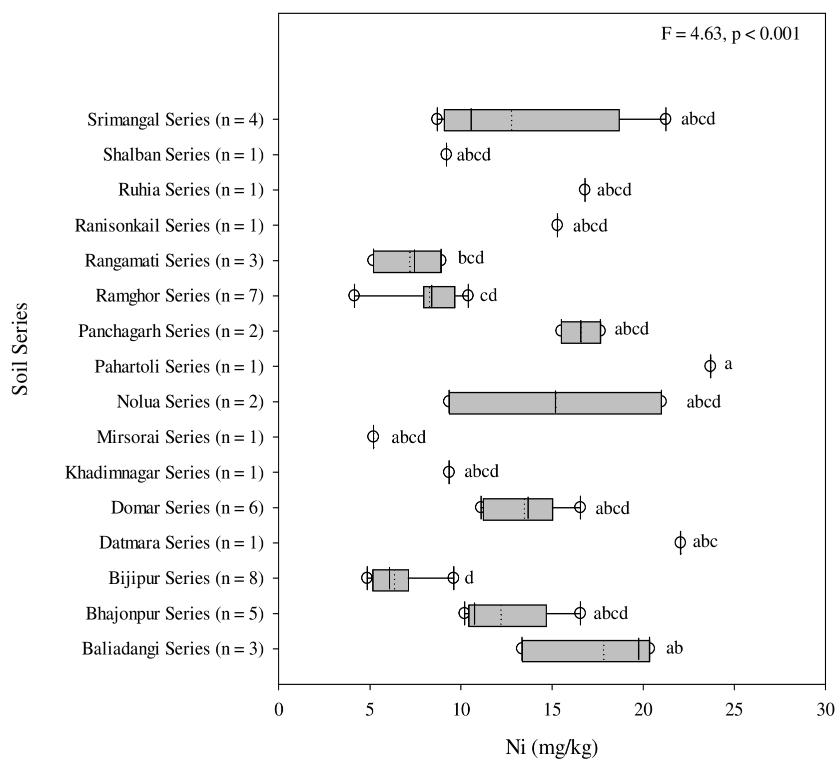

Figure A26: Concentrations of nickel in sub-surface soils of different soil series in the tea gardens.

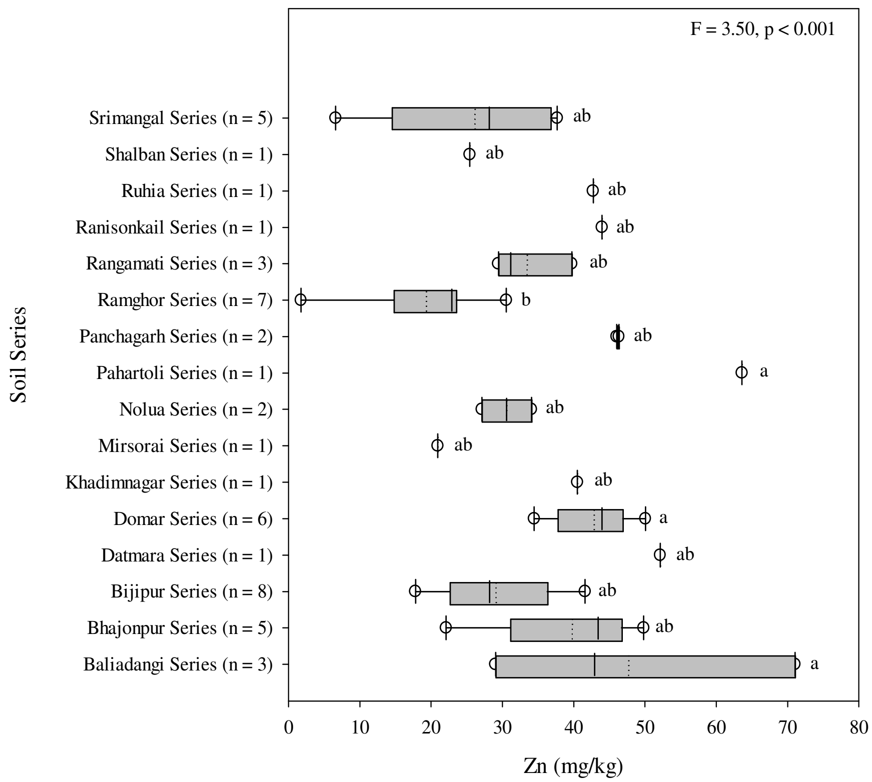

Figure A27: Concentrations of zinc in sub-surface soils of different soil series in the tea gardens.

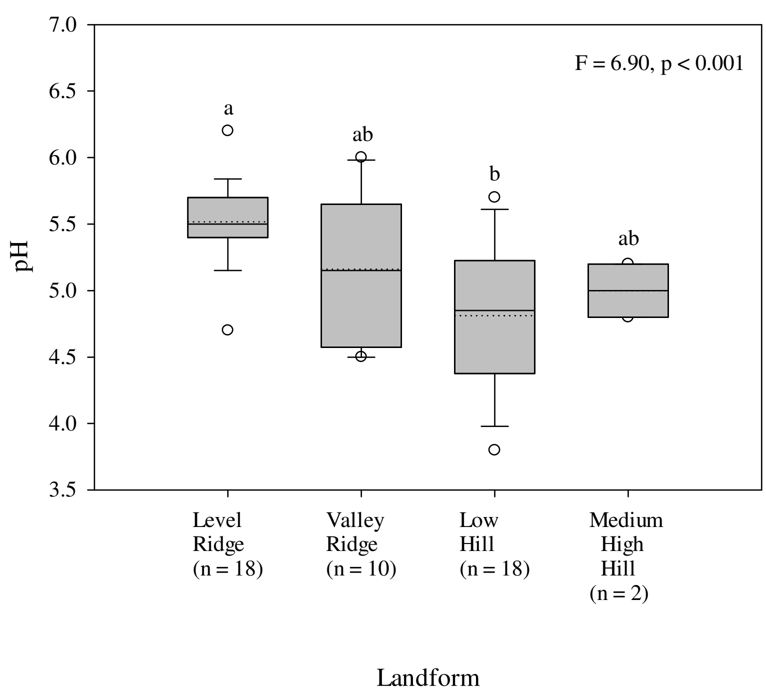

Figure A28: pH in sub-surface soils of different landforms in the tea gardens.

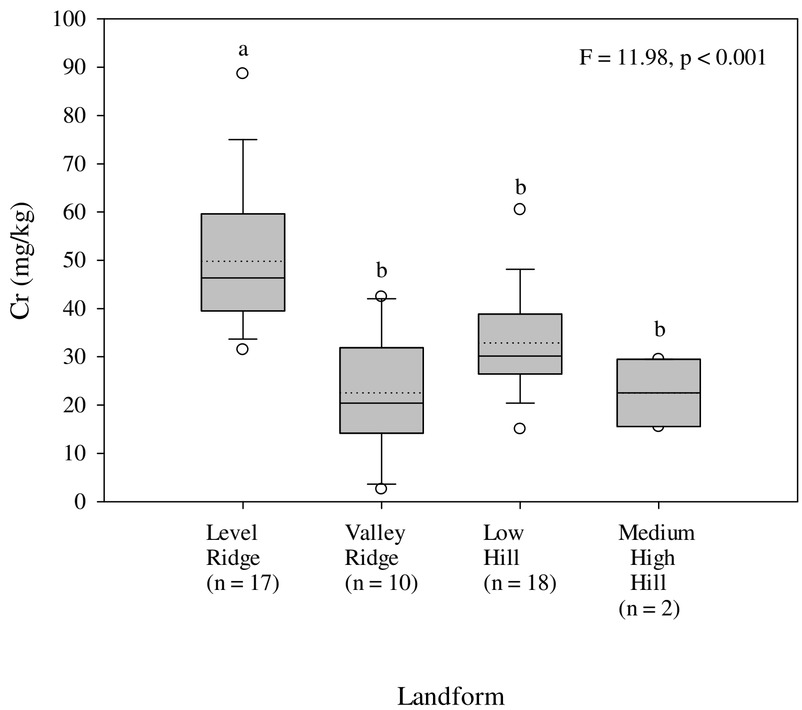

Figure A29: Concentrations of chromium in sub-surface soils of different landforms in the tea gardens.

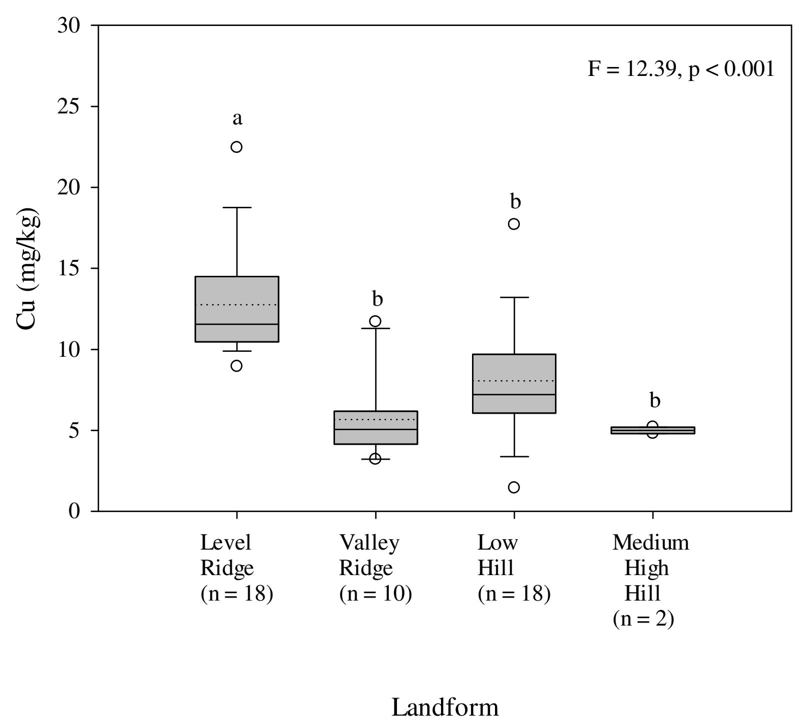

Figure A30: Concentrations of copper in sub-surface soils of different landforms in the tea gardens.

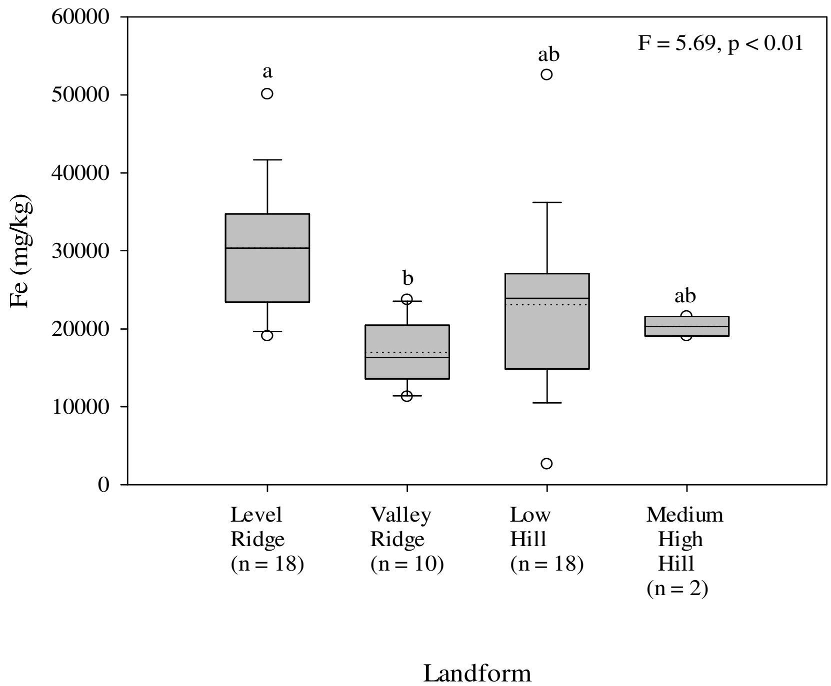

Figure A31: Concentrations of iron in sub-surface soils of different landforms in the tea gardens.

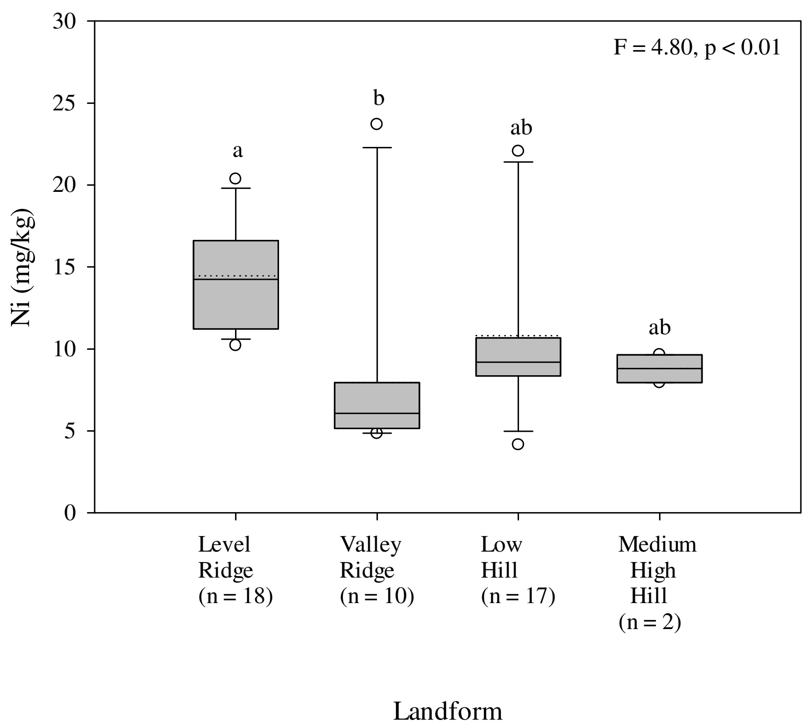

Figure A32: Concentrations of nickel in sub-surface soils of different landforms in the tea gardens.

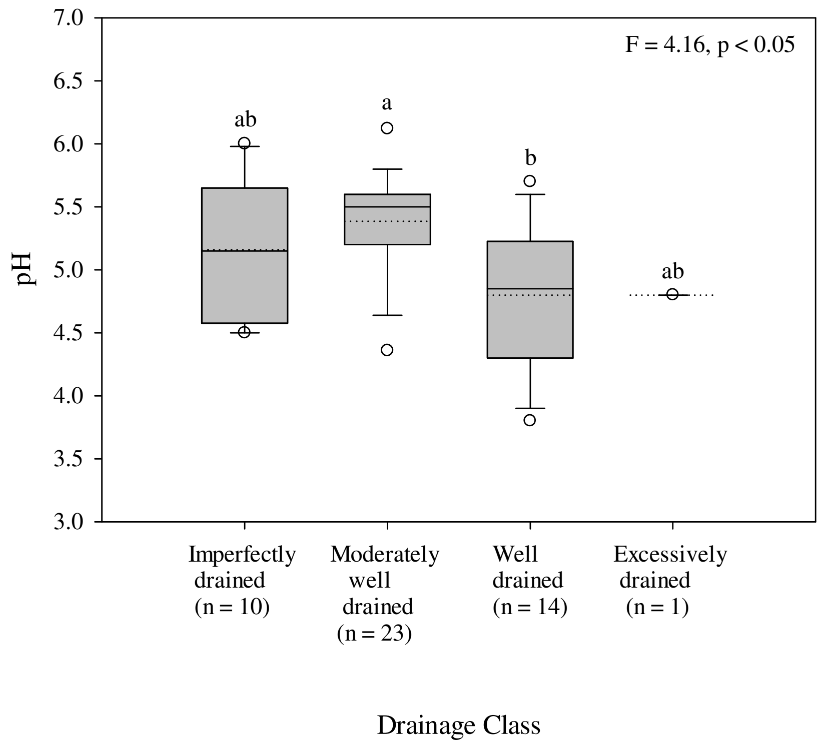

Figure A33: pH in sub-surface soils of different drainage conditions in the tea gardens.

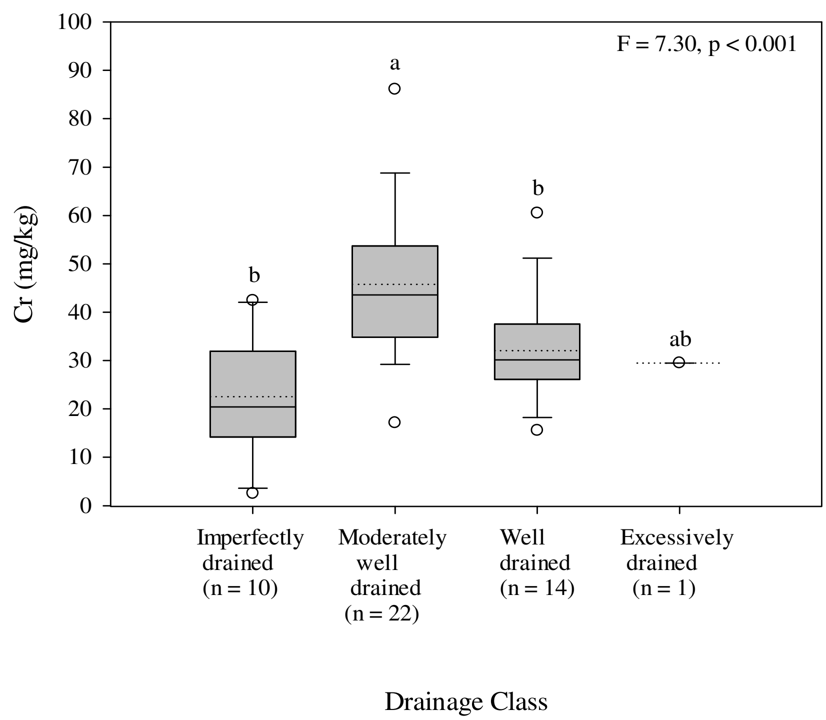

Figure A34: Concentration of chromium in sub-surface soils of different drainage conditions in the tea gardens.

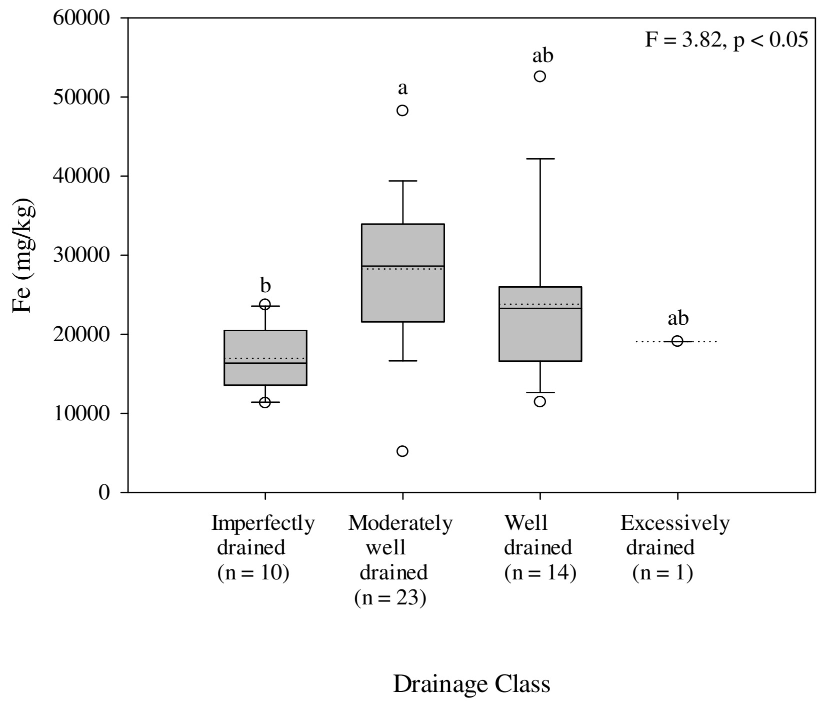

Figure A35: Concentration of iron in sub-surface soils of different drainage conditions in the tea gardens.

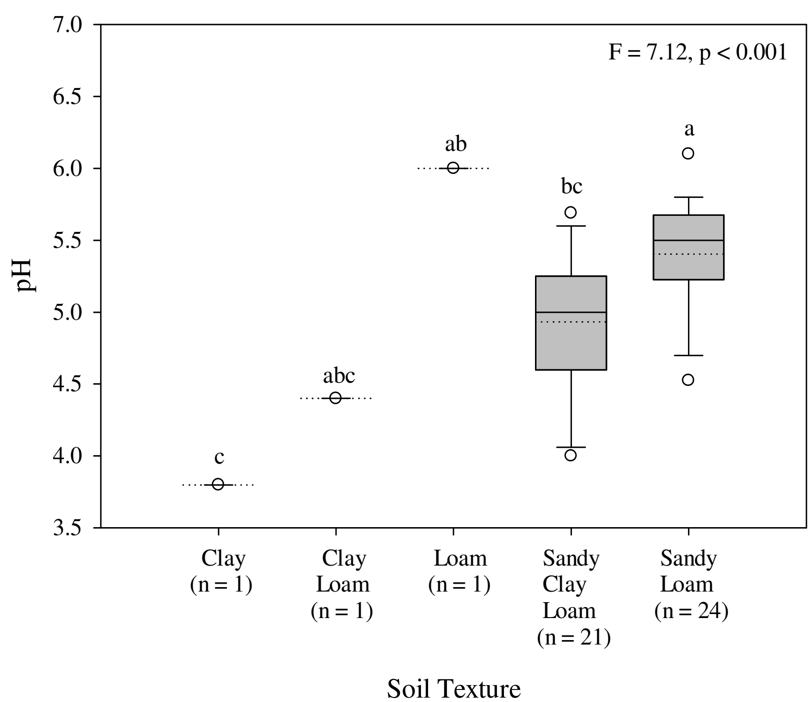

Figure A36: pH in sub-surface soils of different textural classes in the tea gardens.

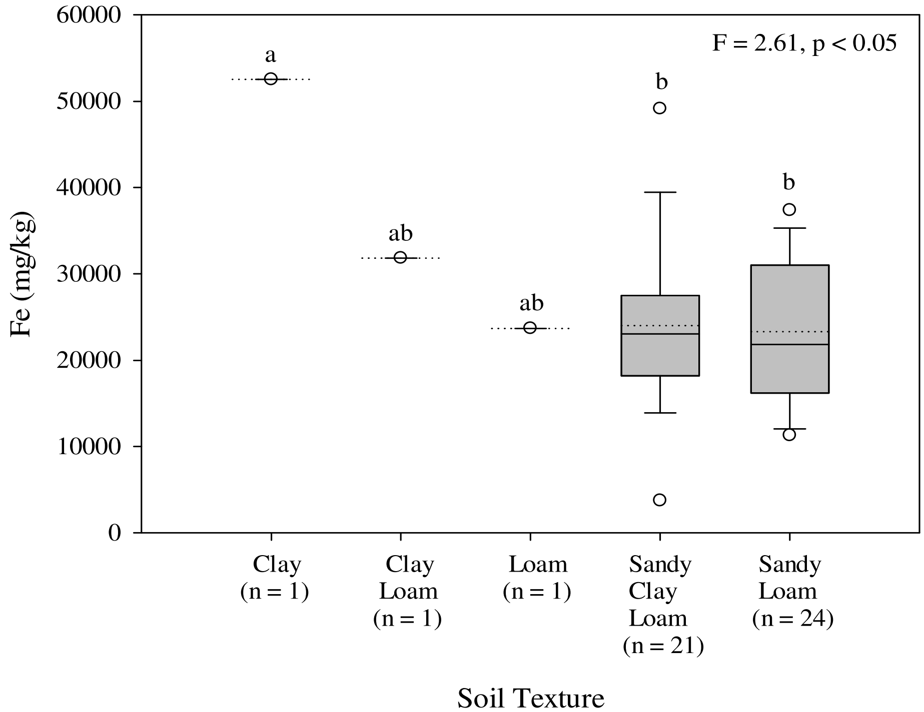

Figure A37: Concentrations of iron in sub-surface soils of different textural classes in the tea gardens.

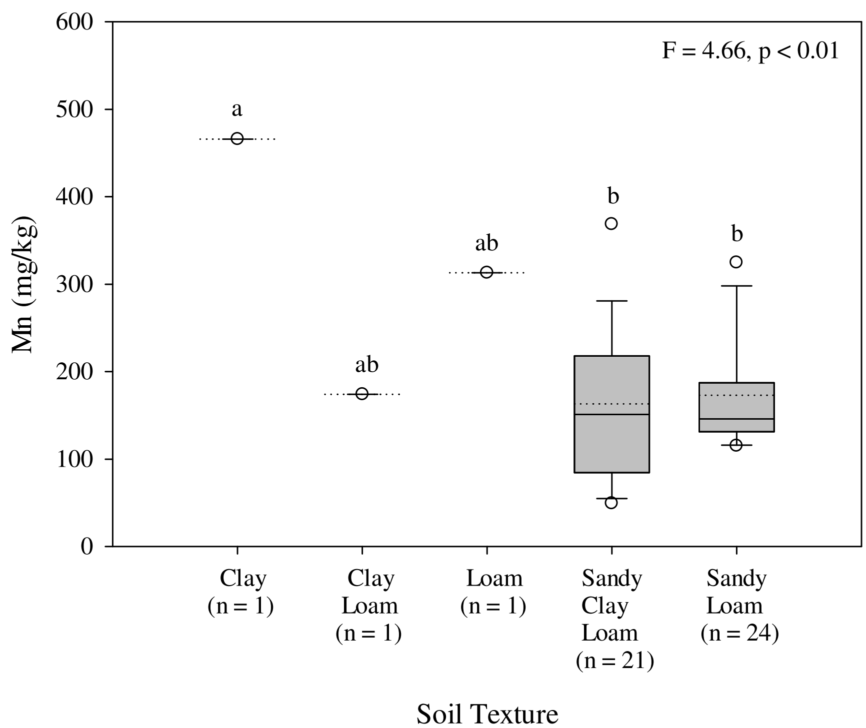

Figure A38: Concentrations of manganese in sub-surface soils of different textural classes in the tea gardens.

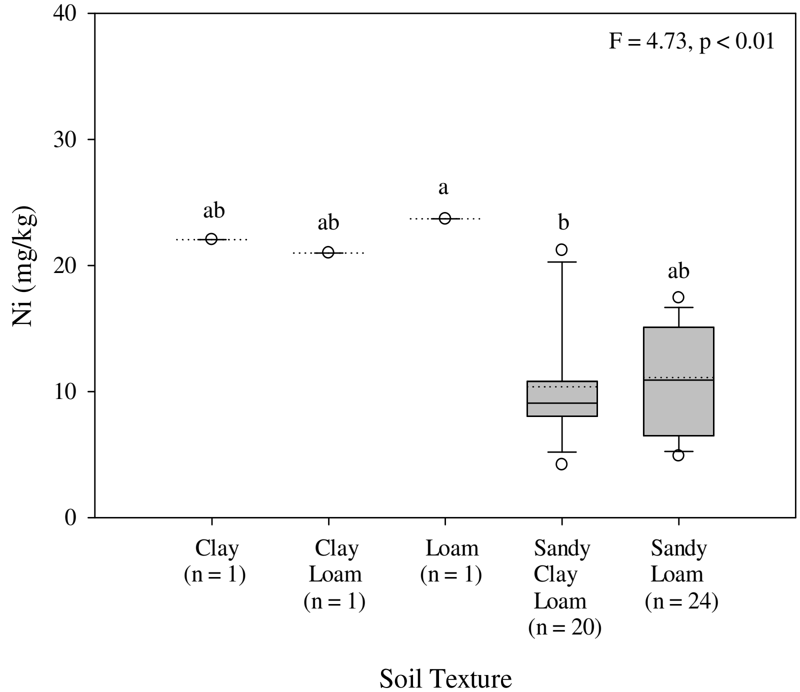

Figure A39: Concentrations of nickel in sub-surface soils of different textural classes in the tea gardens.

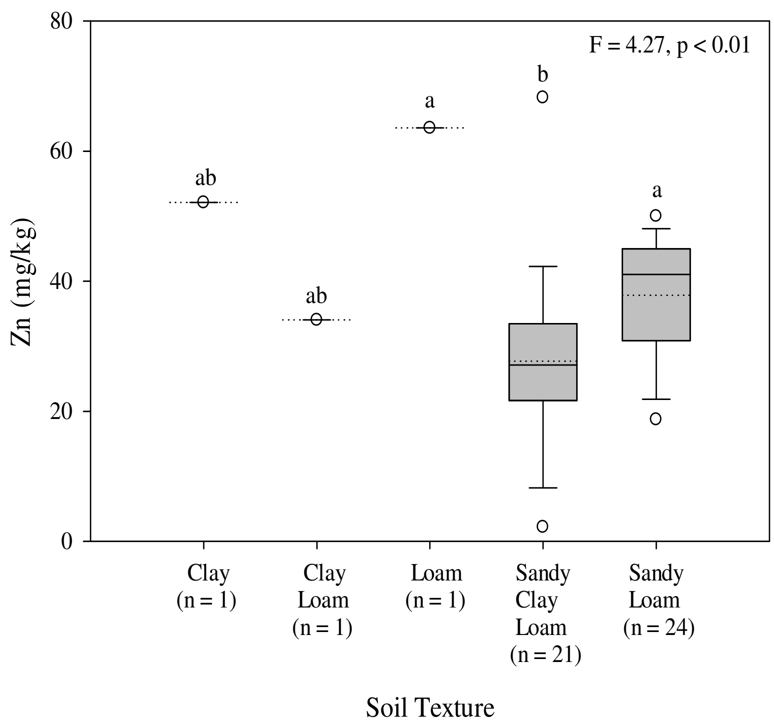

Figure A40: Concentrations of zinc in sub-surface soils of different textural classes in the tea gardens.

Table A1: Geographic locations of the collected soil samples from the tea gardens located at different sub-districts of Bangladesh.

Samples No.	Name gardens	Upazila	District	Latitude	Longitude
1	Karnafuli	Fatickcherri	Chattogram	22.707905	91.848095
2	Karnafuli	Fatickcherri	Chattogram	22.704522	91.84805
3	Karnafuli	Fatickcherri	Chattogram	22.697867	91.844082
4	Karnafuli	Fatickcherri	Chattogram	22.699557	91.846701
5	Karnafuli	Fatickcherri	Chattogram	22.6927778	91.848888
6	Karnafuli	Fatickcherri	Chattogram	22.6916667	91.843611
7	Karnafuli	Fatickcherri	Chattogram	22.6738889	91.837222
8	Karnafuli	Fatickcherri	Chattogram	22.6727778	91.856944
9	Oodaleah	Fatickcherri	Chattogram	22.630907	91.761461
10	Oodaleah	Fatickcherri	Chattogram	22.613611	91.783055
11	Oodaleah	Fatickcherri	Chattogram	22.6405556	91.785277
12	Oodaleah	Fatickcherri	Chattogram	22.594722	91.777777
13	Oodaleah	Fatickcherri	Chattogram	22.6411111	91.749166
14	Oodaleah	Fatickcherri	Chattogram	22.632778	91.788611
15	Oodaleah	Fatickcherri	Chattogram	22.6172222	91.762777
16	Oodaleah	Fatickcherri	Chattogram	22.6425	91.750277
17	Oodaleah	Fatickcherri	Chattogram	22.6008333	91.746944
18	Neptune	Fatickcherri	Chattogram	22.8319444	91.732777
19	Neptune	Fatickcherri	Chattogram	22.8422222	91.7325
20	Neptune	Fatickcherri	Chattogram	22.8588889	91.7
21	Neptune	Fatickcherri	Chattogram	22.8530556	91.703611
22	Neptune	Fatickcherri	Chattogram	22.841667	91.800277
23	Neptune	Fatickcherri	Chattogram	22.815	91.760555
24	Neptune	Fatickcherri	Chattogram	22.835	91.748611
25	Neptune	Fatickcherri	Chattogram	22.842778	91.800833
26	Neptune	Fatickcherri	Chattogram	22.8158333	91.761666
27	Neptune	Fatickcherri	Chattogram	22.844167	91.801666
1	Allynugger	Kamalganj	Moulvibazar	24.3598611	91.90725
2	Allynugger	Kamalganj	Moulvibazar	24.3598611	91.90786
3	Allynugger	Kamalganj	Moulvibazar	24.3597778	91.90877
4	Allynugger	Kamalganj	Moulvibazar	24.3415833	91.9015
5	Allynugger	Kamalganj	Moulvibazar	24.3415556	91.90083
6	Allynugger	Kamalganj	Moulvibazar	24.3544722	91.896388
7	Allynugger	Kamalganj	Moulvibazar	24.3540833	91.896222
8	Allynugger	Kamalganj	Moulvibazar	24.3544444	91.89575
9	Allynugger	Kamalganj	Moulvibazar	24.3530556	91.897611
10	Allynugger	Kamalganj	Moulvibazar	24.3527222	91.897972
11	Allynugger	Kamalganj	Moulvibazar	24.3525833	91.898305
12	Patrakhola	Kamalganj	Moulvibazar	24.2380278	91.82575
13	Patrakhola	Kamalganj	Moulvibazar	24.2376389	91.826055
14	Patrakhola	Kamalganj	Moulvibazar	24.2389444	91.823055
15	Patrakhola	Kamalganj	Moulvibazar	24.2399722	91.813
16	Patrakhola	Kamalganj	Moulvibazar	24.2396389	91.813194
1	Karotoa	Panchagarh Sadar	Panchagarh	26.3958333	88.546666

Table A1: Continued.

Samples No.	Name gardens	Upazila	District	Latitude	Longitude
2	Karotoa	Panchagarh Sadar	Panchagarh	26.4097222	88.5375
3	Karotoa	Panchagarh Sadar	Panchagarh	26.4094444	88.537222
4	Karotoa	Panchagarh Sadar	Panchagarh	26.3975	88.543611
5	Karotoa	Panchagarh Sadar	Panchagarh	26.4091667	88.533888
6	Karotoa	Panchagarh Sadar	Panchagarh	26.3763889	88.520833
7	Karotoa	Panchagarh Sadar	Panchagarh	26.3819444	88.522222
8	Karotoa	Panchagarh Sadar	Panchagarh	26.374722	88.524722
9	Karotoa	Panchagarh Sadar	Panchagarh	26.3594444	88.561388
1	Sathgao	Srimangal	Moulvibazar	24.2867696	91.632354
2	Sathgao	Srimangal	Moulvibazar	24.284065	91.62652
3	Sathgao	Srimangal	Moulvibazar	24.284579	91.62829
4	Sathgao	Srimangal	Moulvibazar	24.297028	91.643252
5	Sathgao	Srimangal	Moulvibazar	24.290918	91.636
6	Sathgao	Srimangal	Moulvibazar	24.292574	91.639826
7	Sathgao	Srimangal	Moulvibazar	24.319266	91.649134
8	Sathgao	Srimangal	Moulvibazar	24.3100278	91.634027
9	Sathgao	Srimangal	Moulvibazar	24.3103056	91.633027
10	Sathgao	Srimangal	Moulvibazar	24.3103333	91.632861
11	Rajghat	Srimangal	Moulvibazar	24.1703056	91.699916
12	Rajghat	Srimangal	Moulvibazar	24.1710278	91.699833
13	Rajghat	Srimangal	Moulvibazar	24.171	91.699833
14	Rajghat	Srimangal	Moulvibazar	24.1649722	91.696750
15	Rajghat	Srimangal	Moulvibazar	24.1634722	91.695777
16	Rajghat	Srimangal	Moulvibazar	24.1632222	91.694500
17	Rajghat	Srimangal	Moulvibazar	24.1631111	91.693361
18	Rajghat	Srimangal	Moulvibazar	24.1665	91.68925
19	Rajghat	Srimangal	Moulvibazar	24.1675556	91.688444
20	BTRI	Srimangal	Moulvibazar	24.2916667	91.752694
21	BTRI	Srimangal	Moulvibazar	24.2920278	91.75375
22	BTRI	Srimangal	Moulvibazar	24.2925278	91.752111
23	BTRI	Srimangal	Moulvibazar	24.2920278	91.750694
24	BTRI	Srimangal	Moulvibazar	24.2946667	91.746805
25	Jagcherra	Srimangal	Moulvibazar	24.3432778	91.773611
26	Jagcherra	Srimangal	Moulvibazar	24.343	91.773805
27	Jagcherra	Srimangal	Moulvibazar	24.3444722	91.7745
28	Jagcherra	Srimangal	Moulvibazar	24.3448889	91.77825
29	Jagcherra	Srimangal	Moulvibazar	24.3448889	91.778583
30	Jagcherra	Srimangal	Moulvibazar	24.3445833	91.777916
31	Jagcherra	Srimangal	Moulvibazar	24.3451944	91.772638
32	Jagcherra	Srimangal	Moulvibazar	24.3451667	91.772555
33	Jagcherra	Srimangal	Moulvibazar	24.3394167	91.758805
34	Jagcherra	Srimangal	Moulvibazar	24.3394444	91.759027
35	Jagcherra	Srimangal	Moulvibazar	24.3404722	91.758611
36	Bilashcherra	Srimangal	Moulvibazar	24.244988	91.771633

Table A1: Continued.

Samples No.	Name gardens	Upazila	District	Latitude	Longitude
37	Bilashcherra	Srimangal	Moulvibazar	24.240077	91.776545
38	Bilashcherra	Srimangal	Moulvibazar	24.238021	91.780599
39	Bilashcherra	Srimangal	Moulvibazar	24.237564	91.78631
40	Bilashcherra	Srimangal	Moulvibazar	24.243731	91.783397
41	Bilashcherra	Srimangal	Moulvibazar	24.249556	91.775574
42	Bilashcherra	Srimangal	Moulvibazar	24.237507	91.790022
43	Bilashcherra	Srimangal	Moulvibazar	24.247386	91.77683
44	Bilashcherra	Srimangal	Moulvibazar	24.2487	91.77072
45	Bilashcherra	Srimangal	Moulvibazar	24.245273	91.779114
46	Bilashcherra	Srimangal	Moulvibazar	24.240476	91.789108
47	Bilashcherra	Srimangal	Moulvibazar	24.248357	91.766551
48	Bilashcherra	Srimangal	Moulvibazar	24.2475	91.784539
49	Bilashcherra	Srimangal	Moulvibazar	24.242018	91.787852
50	Bilashcherra	Srimangal	Moulvibazar	24.247672	91.763353
51	Bilashcherra	Srimangal	Moulvibazar	24.25264	91.760555
52	Bilashcherra	Srimangal	Moulvibazar	24.239277	91.794533
53	Bilashcherra	Srimangal	Moulvibazar	24.247386	91.787509
54	Bilashcherra	Srimangal	Moulvibazar	24.233909	91.796875
55	Bilashcherra	Srimangal	Moulvibazar	24.231967	91.791564
56	Bilashcherra	Srimangal	Moulvibazar	24.249785	91.756843
1	Malnicherra	Sylhet Sadar	Sylhet	24.9255	91.858611
2	Malnicherra	Sylhet Sadar	Sylhet	24.9272778	91.858666
3	Malnicherra	Sylhet Sadar	Sylhet	24.9283333	91.857638
4	Malnicherra	Sylhet Sadar	Sylhet	24.9484167	91.908555
5	Malnicherra	Sylhet Sadar	Sylhet	24.9483333	91.909
6	Malnicherra	Sylhet Sadar	Sylhet	24.9469167	91.908055
7	Malnicherra	Sylhet Sadar	Sylhet	24.9451111	91.890194
8	Malnicherra	Sylhet Sadar	Sylhet	24.9452778	91.890861
9	Malnicherra	Sylhet Sadar	Sylhet	24.9448333	91.889277
10	Malnicherra	Sylhet Sadar	Sylhet	24.9408056	91.885333
1	Kazi & Kazi	Tetulia	Panchagarh	26.5316667	88.449166
2	Kazi & Kazi	Tetulia	Panchagarh	26.5319444	88.449722
3	Kazi & Kazi	Tetulia	Panchagarh	26.5322222	88.45
4	Kazi & Kazi	Tetulia	Panchagarh	26.5425	88.436388
5	Kazi & Kazi	Tetulia	Panchagarh	26.4808333	88.434444
6	Kazi & Kazi	Tetulia	Panchagarh	26.4811111	88.435277
7	Kazi & Kazi	Tetulia	Panchagarh	26.5363889	88.435555
8	Kazi & Kazi	Tetulia	Panchagarh	26.5480556	88.405
9	Kazi & Kazi	Tetulia	Panchagarh	26.5516667	88.419166
10	Kazi & Kazi	Tetulia	Panchagarh	26.5361111	88.431111
11	Kazi & Kazi	Tetulia	Panchagarh	26.5302778	88.428333
12	Kazi & Kazi	Tetulia	Panchagarh	26.5330556	88.428888
13	Kazi & Kazi	Tetulia	Panchagarh	26.5338889	88.429166
14	Kazi & Kazi	Tetulia	Panchagarh	26.5302778	88.428611

Table A2: Summary of PCA analysis of the tea garden soils from different sub-district of Bangladesh.

	PC1	PC2	PC3	PC4	PC5
Eigenvalue	3.01	1.71	1.12	0.66	0.60
Proportion (%)	37.6	21.4	14.0	8.20	7.40
Cumulative (%)	37.6	59.0	73.0	81.2	88.7
N	0.35	-0.39	0.27	0.26	0.41
B	0.25	-0.45	0.47	0.23	-0.19
Ni	0.45	0.28	-0.05	-0.38	0.13
Cr	0.49	-0.11	-0.12	-0.09	-0.33
Fe	0.49	0.04	-0.12	-0.24	-0.33
Mn	0.15	0.46	0.58	-0.21	0.42
Cu	0.33	0.12	-0.52	0.49	0.46
Zn	0.09	0.56	0.23	0.62	-0.41