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Application of Natural Materials to Alternative Pond Water Treatment in a Rural Area of Bangladesh

(バングラデシュ農村における自然素材による溜池浄化への検討)

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Abstract

This thesis was designed to investigate pond water quality and categorization in a rural area of Bangladesh, to developed modified natural material (rice husk) by immobilization (Bangladesh clay soil (80%) and rice husk charcoal (RHC, 20%)) and to apply alternative pond water treatment in a rural area of Bangladesh using simple ceramic filter (SCF) made of natural material. Ponds in the rural area around Khulna and Bagerhat Districts in Bangladesh were surveyed and categorized into domestic use (denoted as D), wastewater storage use (W), agriculture use (A) and fisheries use (F) for the rural ponds and urban domestic use (U: divided into large (Ul) and small groups (Us)). The main survey was carried out during the wet season and a part of the surveyed ponds was also investigated during the dry season. The area and volume of the ponds were large in Ul and F. The transparency was low in the category F, W and U ponds and the SS, TOC were higher in the category F and U ponds than the others. On the other hand the T-N and T-P were higher in category W and U ponds than the others. The size of all the ponds was decreased and the pond water changed more turbid during the dry season caused by increase of particulate matter in the water. The principal component analysis showed that the first component indicating the progress of eutrophication was largely contributed. The water quality of in rural domestic ponds especially during the dry season should be improved. Immobilization of rice husk charcoal (IRHC) was successfully developed both in muffle furnace and laboratory design furnace. By immobilization RHC, release of dissolved solid was decreased. Moreover using IRHC was easy to handle and 100% involvement in treatment was possible, although their active surface area was decreased. Application of IRHC on Cr (VI) reduction as an example of the usage of the IRHC was investigated and found that IRHC could be used as reductant. The effectiveness of suspended solid (SS), total coliforms (TC), and Escherichia coli (E. coli) removal from pond waters in a rural area of Bangladesh using alternative treatment by a filtration unit made of simple ceramic filter (SCF) was investigated. The SCF was manufactured using Bangladesh local clay soil and rice bran, and the filter unit was made by attaching the SFC to a 15 L clay flower pot. Laboratory testing with artificial pond water was performed in Japan followed by a field test with actual pond water in Bangladesh. The log removal of TC and E. coli by the filtration unit in the laboratory test was 2 to 3 log or more, and in the field test, it was 1 to 2 log or more during the wet season and approximately 1.5 log during the dry season. The filter unit also had good removal efficiency for SS (primarily algae) from the raw pond water. The effect of additional devices (iron net, rice husk charcoal and immobilized rice husk charcoal) was not apparent, but the addition of an iron net decreased dissolved phosphorus (D-P). The filter unit can be applied to actual pond water and is an alternative treatment system for pond water.

Table of contents

List of Tables

List of Figures

List of Abbreviations

%	Percentage
μm	Micrometer
A	Agriculture
APSU	Arsenic Policy Support Unit
B-filter	Basic type unit
C:N:P	Carbon : Nitrogen : Phosphorous
C_e	Final concentration
C-filter	Combination type unit
CFU	Colony-forming unit
C_i	Initial concentration
cm	Centimeter
D	Domestic
D-N	Dissolved nitrogen
DO	Dissolved oxygen
DOC	Dissolved organic carbon
D-P	Dissolved phosphorous
DPHE	Department of Public Health Engineering
DS	Dissolved solids
E.coli	*Escherichia coli*
EC	Electrical conductivity
ECR	Environment Conservation Rules
F	Fisheries
g	Gram
g/cm^3	Gram per centimeter cubed
H-filter	Hard-charcoal-added type unit
I-filter	Iron-covered type unit
IRHC	Immobilized rice husk charcoal
JIS	Japanese Industrial Standard
KUET	Khulna University of Engineering & Technology
L	Liter
M	Mass of adsorbent

m	Meter
m/day	Meter per day
m^2	Square meter
m^2/gm	Square meter per gram
m^3	Meter cubed
MDG	Millennium Development Goal
mg/100gm	Milligram per 100 gram
mg/L	Milligram per liter
mgN/L	Milligram nitrogen per liter
mgP/L	Milligram phosphorous per liter
min	minute
mL	Milliliter
mm	Millimeter
MS	Mild steel
mS/cm	Millisiemens per meter
mV	Millivolt
NTU	Nephelometric turbidity units
^oC	Degree celsius
ORP	Oxidation reduction potential
PO_4-P	Phosphate phosphorous
POU	Point of use
PSF	Pond sand filter
R-filter	Charcoal-added type unit
RHC	Rice husk charcoal
SCF	Simple ceramic filter
SS	Suspended solids
TC	Total coliforms
T-N	Total nitrogen
TOC	Total organic carbon
T-P	Total phosphorous
U	Urban domestic
Ul	Urban domestic large
UN	United Nation

UNICEF	United Nations Children's Fund
Us	Urban domestic small
W	Wastewater storage
WHO	World Health Organization
WSSCC	Water Supply and Sanitation Collaborative Council
WSSD	World Summit on Sustainable Development
Z	Principal component

Chapter 1

Introduction

1.1 Background

Safe water and proper sanitation is the essential part of human life. But access to the safe water and using proper sanitation are not easy for all the people around the world. Although our planet has sufficient fresh water, but millions of people die from diseases associated with inadequate water supply, sanitation and hygiene due to bad economics or poor infrastructure. In 2000, the World Health Organization (WHO) reported that 1.1 billion people and 2.4 billion people around the world had no access to “improved water supply” and “improved sanitation”, respectively. In Asia the number of people that had no access to “improved water supply” and “improved sanitation” were 0.7 billion people (19%) and 1.9 billion people (52%), respectively (WHO and UNICEF, 2000). To improve these condition in the same year, United Nation (UN) declare “United Nations Millennium Declaration” after three days summit in New York where the goal was set as the proportion of people who are unable to reach or to afford safe drinking water will be reduced to be halve by the year 2015 (United Nations, 2000). Also “provide water, sanitation and hygiene for all by 2025” goal was set in 2000 in the Second World Water Forum held in Hague (WSSCC, 2000). In 2002, millennium development goal for sanitation was set at the World Summit on Sustainable Development (WSSD) held in Johannesburg same as safe drinking water goal (United Nations, 2002). It was reported that global population using an improved drinking water source achieved 91% by 2015 which met Millennium Development Goal (MDG) five years ahead of the schedule (United Nations, 2015). Although 0.7 billion people still not gained access to improved drinking water source and at least 1.8 billion people around the world use fecal contaminated water as a source of drinking water. And 2.4 billion people are far away from the access to basic sanitation services, such as toilets or latrines, which caused nearly 1000 children; die each day due to due to preventable water and sanitation-related diarrhoeal diseases.

Water scarcity, poor water quality and inadequate sanitation affect the human life especially poor people. Due to the climate change, drought is increasing all over the world which leads worsening hunger and malnutrition to some of the world's poorest countries people [http://www.un.org]. According to the WSSD, the main reason for lack of safe water is either scarcity of water or contamination of water sources, which is due to both lack of investment in water systems and inadequate maintenance of existing systems (United Nations, 2002). It was reported that, more than 40% of global population were affected by water scarcity, which is projected to rise (United Nations, 2015). Besides discharging wastewater resulting from human activities directly into the water bodies (more than 80%, http://www.un.org), continue population growth and urbanization, rapid industrialization, agricultural application and contamination by human activities leads decreeing the amount of 1% of easily accessible fresh water (Hasan, 2014 and Hotlos, 2008). It was assumed that at least one in four people would be affected by shortage of fresh water at year 2050 (http://www.un.org).

In 2015 UN adopt a new sustainable development agenda and a new global agreement on climate change in UN climate change conference held in Paris. According to new sustainable development agenda it was set that by 2030, universal and equitable access to safe and affordable drinking water for all should be achieved and it means global population using an improved drinking water source should be raised from 91% to 100% (http://www.un.org).

Bangladesh is low-lying riverine country which formed as the largest deltaic plain located in southern Asia (Frenken K., 2012). A huge area of water resources of 4.575 million ha complies with many rivers, canals, depression and oxbows, ponds, lakes and floodplains are the part of Bangladesh. Among this, 45.75 lakh ha used for inland fisheries, which includes inland water bodies, ponds, ox-bow lakes and coastal shrimp farms (Department of Fisheries, 2011). Although Bangladesh had huge water resource, but scarcity of the safe water is present due to arsenic contamination, groundwater depletion, flooding, drought, salinity etc. (Abdin *et al*., 2014; Shafiquzzaman *et al.*, 2009; Khan *et al.*, 2011 and http://siteresources.worldbank.org). Northern part of Bangladesh was specially affected by drought during dry season (Islam *et al*., 2014). According to Costal Zone Policy 2005, 76 sub-districts of 19 coastal area of Bangladesh are affected by salinity due to rise in sea level (Ministry of Water Resources, Bangladesh, 2005). Generally people use both surface water and ground water but ground water is the most reliable source of safe water. However safe ground water amount was decreasing due to decreasing water table, arsenic contamination and salinity (DPHE and UNICEF, 1994 and http://siteresources.worldbank.org). These enforce people to be more dependent on traditional unprotected surface water sources such as ponds or ditches or walking to distant wells, which lead to increase the risk of acute bacteriological contamination, leading to greater outbreaks of water-related diseases. To reduce the health risk treatment of this unprotected surface water is necessary.

There are many treatment options used for water treatment such as pre-treatment, chemical coagulation, and filtration to remove different impurities (particulate matter, bacteria, chemical constituent etc.). Many of these treatment options are expensive to install and maintenance due to chemical cost, advanced technology that cannot be affordable to the rural people most of the time. However some treatment options used natural material for the treatment, which reduces the cost. Another major problem of these treatments is unsustainability. Presently researchers are working on low-cost treatment option using different natural materials as they are easily available and could be maintained easily resulting long-term sustainability. Among all the treatment options, filtration is widely used. In modern water filtration systems, several natural materials such as stone, gravel, sand, soil, carbon, diatomaceous earth, agricultural by-products etc. are incorporated (Nkwonta and Ochieng, 2009; Asano *et al.*, 2007 and Huisman and Wood, 1974).

In Bangladesh, pond is the most important water resources used for productive purposes, such as irrigation and fish cultivation and non-productive purposes, such as cooking, bathing, ablution, washing cloths and kitchens and sometimes for drinking purpose (Abedin *et al.*, 2014; Karäzlin, 2000; Milton *et al.*, 2006 and http://unpan1.un.org). Moreover, most of the pond water is contaminated with various impurities, such as algae and pathogenic bacteria, which cause serious health effects (Knappett *et al.* 2011). However ponds could be reconsidered as alternative water sources after applying easy treatment as they are traditionally and culturally accepted by the Bangladesh people (Karäzlin, 2000). However there is no certain data that can explain proper categorization of the ponds in Bangladesh and difference in their water quality, which could be important data to understand the water quality of different categories. Filtration technology using locally available material and skills could be one of the easy treatment options for pond water treatment. In Bangladesh, community-based pond water treatment system named pond sand filter (PSF) and point of use (POU) system for households using filters (Safi filter, Bishuddhaya filter) and three pitcher filter were used for pond water treatment. But there are many disadvantages such as for PSF system has high installation and maintenance cost (Jakariya, 2005). Most of the users are not satisfied with its performance (Harun and Kabir, 2013) and also most of POU systems are too expensive for much rural population and questionable performance. A low-cost simple ceramic filter (SCF) was developed in our laboratory using natural material of rice bran and clay (Shafiquzzam *et al.,* 2011). This SCF was successfully applied for arsenic removal from ground water for one year (Hasan *et al.* 2012). This SCF could be applicable for pond water treatment to obtain drinkable water quality as porous ceramic filter was reported to be reduced turbidity and bacteria (Sobsey, 2002).

1.2 Purposes of the study

1.2.1 Broad objective

The principle objective of this study is to introduce a low cost water purification unit made of natural materials for pond water treatment focusing on developing countries.

1.2.2 Specific objectives

Targeting the main objective, this study was carried out with the following objectives in mind:

1. To study of pond water categorization and its characteristics in a rural area of Bangladesh.
2. To manufacture immobilize rice husk charcoal (IRHC) and it's possible application.
3. To development of pond water purification unit using simple ceramic filter (SCF) and additional device.
4. To clarify the practical application of the pond water purification unit in water sectors.

1.3 Content of this thesis

This thesis combined and showed the results obtained from different studies, which were conducted in different stages during doctoral degree tenure at Ritsumeikan University, Japan. A brief outline of each study is shown in Figure 1.1.

Chapter 1 describes an introduction of this research including research background, objectives and framework of the study.

Chapter 2 reviews the natural materials used in water treatment sector, their modification and applications.

Chapter 3 investigates pond water condition in rural area of Bangladesh. Therefore, this chapter describes the categorization of rural area ponds according to their usages and pond water quality during wet season and dry season. The factors affecting the water quality in the ponds and any issues that need improvement also described in this chapter.

Chapter 4 describes the development and manufacturing of IRHC with its possible application in water sector.

Chapter 5 investigates applicability of pond water purification unit for pond water treatment. Therefore, this chapter describes pond water purification unit assembly and studied applicability of pond water purification unit to evaluate the effectiveness of suspended solid (SS) and bacteria (total coliforms (TC) and *Escherichia coli* (*E. coli*)) removal.

Chapter 6 draws the conclusions regarding the potential application of pond water purification unit to treat surface water (pond water) for ensuring safe water supply in rural area of developing countries.

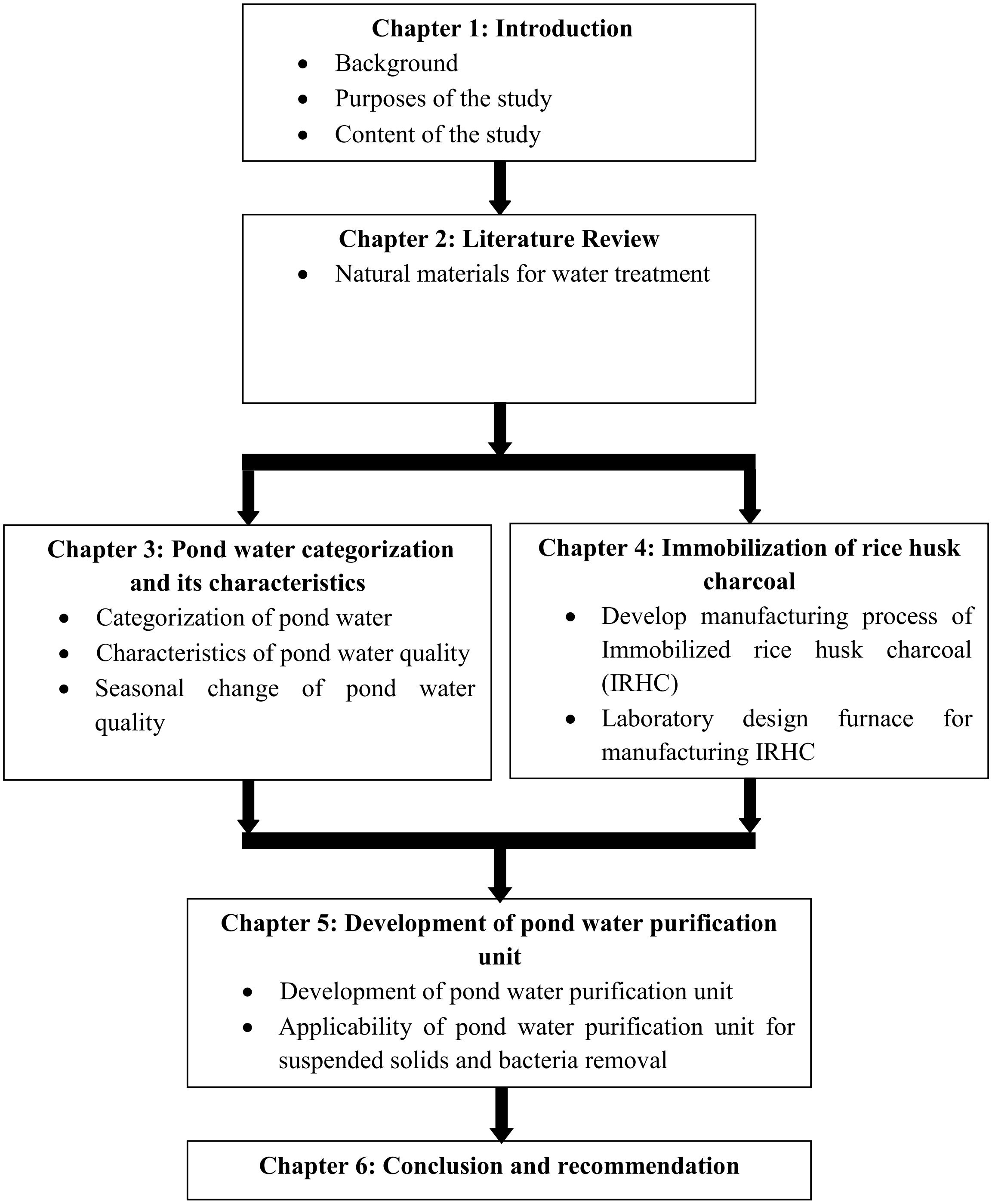

Figure 1.1 – Framework of the research and this thesis

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Chapter 2

Literature review on natural material uses in water treatment

2.1 Introduction

Any product or physical matter that comes from plants, animals or the ground is called natural materials. Also minerals and the metals that can be extracted from them without further modification called natural material. Contaminants such as disease-causing microorganisms or toxin-dissolved metals present in drinking water, can pose a serious health risk. Also non-hazardous contaminants can make bad taste of water. Several natural materials such as stone, gravel, sand, soil, carbon, diatomaceous earth, agricultural by-products etc. are incorporated into modern water filtration systems to help in removing potential contaminants to get safer and tastier water. Stone, gravel and sand widely used as a filter media in different treatment system such as roughing filter (Nkwonta and Ochieng, 2009), depth filtration (Asano *et al.*, 2007), slow sand filtration (Huisman and Wood, 1974), multi-layer filtration (http://www.gewater.com) etc. to remove suspended particulate matters as well as pathogenic bacteria. Activated carbon or charcoal is a very porous type of carbon, which can be made from a variety of natural substances and helps to purify water in a water filter by trapping contaminants as well as carbon's positive charge attract contaminants to the carbon. Carbon generally found in nature or prepared by simple modification of natural material widely used to remove heavy metals like chromium, lead, zinc, copper etc. as well as dissolved chemicals and some types of parasites (Monser and Adhoum, 2002; Narayanan and Gansen, 2009; Mohan *et al.*, 2006; Mohanty *et al.*, 2005 and Fahim *et al.*, 2006).

2.2 Different natural material used for water treatment

2.2.1 Granular natural materials

Stone, gravel and sand are the most useful granular natural material used for water and wastewater treatment that found abundance in the world. They are widely used for filtration in different conventional filters such as slow sand filtration, rapid sand filtration, multilayer filtration, moving-bed filtration, direct filtration etc. (Lorch, 1987; APSU, 2006 and Asano, 2007). Traditionally stone or gravel was used for roughing filters, but recently many alternative filter media such as broken burnt clay bricks, plastic material, burnt charcoal, coconut fiber (Nkwonta and Ochieng, 2009). Gravel also used in trickling filter for biological treatment (Dermuo and Vayenas, 2007).

2.2.2 Zeolites

Zeolites are naturally occurring silicate minerals that also could be produced synthetically known for more than 200 years. They generally used as adsorbent catalyst in water and wastewater treatment for their ion exchange capability and sorption properties (Barkat, 2011 and Bailey *et al.*, 1999). Many natural zeolites identified in the world of which clinoptilolite, chabazite, phillipsite, stilbite, analcime, mordenite and laumontite are very common forms whereas offretite, barrerite, paulingite and mazzite are much rarer (Wang and Peng, 2010). The most abundant natural zeolites species is clinoptilite that widely used in the world. The chemical composition of clinoptilite zeolite mainly composed of SiO_2 (60 – 70%), Al_2O_3 (9 – 12%), K_2O (<1 – 8%), CaO (<1 – 6%), Fe_2O_3 (<1 – 4%), MgO (<1 – 2%), Na_2O (<1 – 4%), MnO (<0.05%), TiO_2 (<0.01 – 0.26%) and P_2O_5 (<0.05%) (Wang and Peng, 2010 and Barlokova, 2008). Zeolites are widely used in drinking water, wastewater, grey water, surface water, underground water, ground water treatment (http://www.intechopen.com). Natural zeolites were explored as an effective adsorbent for ammonium adsorption (Cooney *et al*., 1999; Wang *et al*., 2006 and Wen *et al*., 2006), heavy metal ion adsorption (Panayotoya, 2001), inorganic anions sorption (Wang and Peng, 2010), dye adsorption (Wang *et al.*, 2005), humic substances removal (Wang *et al.*, 2006) with modification or without modification.

2.2.3 Clay

Clay is another natural material that widely used in water and wastewater treatment. Clays are composed of extremely fine crystals or particles of clay minerals that had good chemical properties of colloids. Generally they are classified by the number of tetrahedral and octahedral layers that made sandwiches shaped sheet structure (Rozïicâ *et al.,* 2000). There are basic classes of clays named kaolinite, micas (such as illite) and semecities (such as monotmorillonite). They also have high surface area (up to 800 m^2/g) and high cation exchange capacity (Bailey *et al.*, 1999).

Clay is the main raw materials for manufacturing ceramics. Clay mixed with various types of combustible/burn-out material, sewage sludge, red mud was used for manufacturing ceramic filters (He *et al.*, 2012; Bielefeldt *et al.*, 2010 and Han *et al.*, 2009). However ceramic filters sometimes treated with silver to kill virus in the water (Simonis *et al.*, 2014 and Dies, 2003). Clay also had adsorption capabilities due to net negative charge on the structure of fine-grain silicate minerals that neutralized positively charged species (Bailey *et al.*, 1999). For these properties clay was used as adsorbent for removal of heavy metal (Potgieter *et al.*, 2006 and Vhahangwele and Mugera, 2015). They also used for gas separation (Zulhairun *et al.*, 2014), fluoride removal (Vinati *et al.*, 2015), dye removal (Zhao *et al*, 2013), heavy metal removal by electrocoagulation (Mountassir *et al.*, 2015). Clay was also used for Fe(III) reduction where bacteria plays important roles (Mueller, 2015).

2.2.4 Agricultural by-products

Agriculture by-products that contain cellulose showed potential sorption capacity. Their basic components were hemicellulose, lignin, lipids, proteins, simple sugar, water, hydrocarbons and starch. They are economic and eco-friendly due to their unique chemical composition, availability in abundance, renewable nature and low cost (Bhatnagar and Sillanpää, 2010). There were many agricultural by-products such as rice husk, rice bran, saw dust, coconut shell, sugarcane bagasse, soybean hulls, corn cob etc. that used for water and wastewater treatment (Deepika *et al.*, 2013 and Sharma *et al.*, 2013).

2.2.4.1 Rice husk and rice bran

Rice husk and rice bran are the agricultural by-products come from rice production. Annually about 571 million tonnes rice produced around the world which results approximately 140 million tonnes of rice husk and about 50-60 million tonnes rice bran (Kalderis *et al.*, 2008 and Devi and Arumughan, 2007). The physical properties and chemical composition of rice husk were shown in Table 2.1 and Table 2.2, respectively. It was seen from Table 2.1 and 2.2, that rice husk had high porosity, surface area, fixed carbon amount and high silica content. The content of hemicellulose, cellulose and lignin are 24.3, 34.4 and 19.2 %, respectively. The monoromeric components of hemicellulose present in rice husk are responsible for producing large-scale commodities like activated carbon, silicon oxide (Soltani *et al.*, 2015).

Table 2.1 Physical properties of rice husk (Ye *et al.*, 2012)

Parameter	Unit	Content
Surface area	m^2/g	438.05
Bulk density	g/cm^3	0.3086
Porosity	fraction	0.38
Moisture content	%	8.25
Volatile content	%	42.8
Fixed carbon	%	30.56
Ash	%	18.39

Table 2.2 – Typical chemical composition of rice husk (Ghosh and Bhattacherjee, 2013)

Constituents	Weight (%)
Silica (SiO_2)	94.50
Calciaum oxide (CaO)	0.48
Manganese oxide (MnO)	1.09
Magnesium oxide (MgO)	0.23
Iron oxide (Fe_2O_3)	0.54
Aluminum oxide (Al_2O_3)	0.21
S, P_2O_5, K_2O, Na_2O	Traces

The proximate composition and mineral composition of rice bran were shown in Table 2.3 and 2.4, respectively. Rice bran contains high amount of carbohydrate, fat and around 1% of potassium and phosphorus. The proximate and mineral composition of rice bran had potential for food use.

2.2.4.1.1 Application of rice husk

Rice husk was not used effectively in the last decade due to lack of awareness of its potentiality by farmers and industry persons, socio-economic problems, penetration of technology, lack of environmental concerns etc. (Soltani *et al.*, 2015). Unmodified and modified rice husk was widely used as adsorbent in water and wastewater treatment. They were used to remove heavy metals such as copper, zinc, mercury, chromium etc. (Bansal *et al.*, 2000; El-Shafey E.I., 2010; Ye *et al.*, 2012; Khan *et al.*, 2014 and Xu *et al.*, 2013), cationic or organic dyes (Leng *et al.*, 2015 and Tolba *et al.*, 2015), for phenol adsorption (Daffalla *et al.*, 2010) and for remediation of COD (Mohan *et al.*, 2008).

Table 2.3 – Proximate composition of rice bran* (Rosniyana *et al*. 2007)

Compositon	(% wet weight)
Moisture	9.99
Protein	12.81
Fat	24.14
Carbohydrate	30.06
Crude fiber	12.49
Ash	10.76

* Calculated by taking average of different millig degree value

Table 2.4 – Mineral composition of rice bran* (Rosniyana *et al*. 2007)

Compositon	(mg/100gm)
Calcium	49.9
Iron	5.5
Magnesium	743.5
Sodium	25.8
Potassium	1075.0
Phosphorous	1022.5

* Calculated by taking average of different millig degree value

However, rice husk was also used for silica extraction (Zulfiqar *et al.*, 2015 and Kumar *et al.*, 2015), production of bio-oil (Qiang *et al.*, 2008) etc.

2.2.4.1.2 Application of rice bran

Rice bran currently used as animal food and extracted oil for cooking or industrial applications (Devi and Arumughan, 2007 and Rosniyana *et al.*, 2007). In Japan, rice bran found to be used for many purposes other than oil such as mushroom production, pickle preservation, fertilization and waste. Rice bran also used as fuel, stuck to commercial ice blocks to hinder them from melting etc (Pourali *et al.*, 2009). It also used as adsorbent to immobilization of microbes (Chitra *et al.*, 1996), to remove dye (Ogata *et al.*, 2015) and also to remove Cr (VI) (Singh *et al.*, 2005) Recently rice bran was used for making simple ceramic filter which was used for different sector of water and wastewater (Shafiquzzam *et al.,* 2011 and Hasan *et al.* 2012).

2.2.4.2 Other agricultural by-products

Wheat husk and wheat bran were also agricultural products, which used for removal of cadmium (Singh *et al.*, 2006), lead (Bulut and Baysal, 2006) and dye (Gupta *et al.*, 2007 and Fatma *et al.*, 2007). Coconut coir also used for removal of chromium (Chaudhuri and Azizan, 2012), cobalt, nickel (Parab *et al.*, 2006), dye (Namasivayam *et al.,* 2001) etc. Agricultural by-products from tea, coffee, peanuts, peels of different agricultural product was used for removal of different dye and heavy metal (Ahuwalia and Goyal, 2005; Hameed, 2009; Franca *et al*., 2009; Namasivayam and Perismy, 1993; Ajmal *et al.*, 2000; Memon *et al.*, 2009 and Memon *et al.*, 2008)

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Chapter 3

Pond water categorization and its characteristics

3.1 Introduction

In developing Asian countries, a large number of artificial water storage ponds are used for different purpose. These artificial water storage ponds where water is stored are usually used directly on-site (Meester *et al*., 2005). To ensure safe use of these ponds water for its intended purpose, maintaining certain pond water quantity and quality is important issue. Normally pond water is not used for drinking, but it is used for other domestic purposes including as an agriculture water resource in aquaculture areas (Kazi N.M., 2003).

Bangladesh possesses enormous wetland areas equal to roughly 11 percent of the total area of the country (Rasheed, 2008). The land is sometimes flooded during the monsoon season. People construct artificial storage ponds by simply building embankments to enclose water during the dry season so that they can use the pond water for various purposes, especially in rural areas. Pond water is considered as one of the most important water resources in these areas.

The Khulna and Bagerhat Districts are located on a delta in the southwestern part of Bangladesh near a mangrove forest and the Bay of Bengal. The districts are supposed to be very vulnerable to climate change (Asian Development Bank, 2011). The water resource system in the area must be improved to be more able to adapt to the water crisis caused by climate change. Rural people use pond water as well as groundwater in these districts. They also possess wide area ponds that are used for aquaculture (Dey *et al*., 2008; Department of Fisheries, Bangladesh, 2011). Although the pond water in the district plays an important role as a water resource in the area, little research has been conducted to determine its water quality, characteristics and the seasonal changes caused by the differences in precipitation during the wet and dry seasons (Bangladesh Bureau of Statistics, 2013).

The purpose of this chapter is to understand the water qualities and problems with water usage of the ponds in the rural areas around the Khulna and Bagerhat Districts in Bangladesh. The water storage ponds categorization according to their water usages, the size and water quality of these ponds as well as their seasonal changes was described in this chapter. Also the factors affecting the water quality in the ponds and any issues that need improvement were identified. The main part of this chapter has been published in Journal of Water and Environment Technology (Rahman et al., 2015).

3.2 Materials and methods

3.2.1 Surveyed area

Pond water characteristics were surveyed in the urban and rural areas near Khulna City, Bangladesh (Khulna City and Rupsa, Fakirhat and Mollahat villages (Figure 3.1)). A significant number of natural wetlands and artificial ponds cover the area (Chowdhury and Mamun, 2006). The ponds in the villages are used for many purposes: rural domestic uses including dish washing, laundry, bathing and cooking (Sultana and Crow, 2000), wastewater storage use, agriculture use (paddy fields) especially in Rupsa and Mollahat and fishery use (prawns and fish) in Fakirhat. The village people use well water (which is partially contaminated with arsenic) for drinking and cooking (Shafiquzzaman *et al*., 2009). Tap and bottled water are also used in urban areas (Khulna City) where people use both pond and well water for their domestic use. The ponds in the urban areas are also used for vehicle washing, recreation and wastewater discharge. Monthly precipitation is almost 100-400 mm from the period from May to October (during the wet season) and is less than 100 mm from the period from November to April (during the dry season, Bangladesh Bureau of Statistics, 2013). The daily maximum temperature is less than 30^{o}C in only December, January and February (Shahid, 2011) and is higher than 30^{o}C during all the other months (Bangladesh Bureau of Statistics, 2013).

3.2.2 Pond categorization

The surveyed rural ponds were clearly divided into four kinds of usage: domestic water use, wastewater storage use, seasonal agriculture use and fishery use. So we defined 4 pond categories in the surveyed area by their usages as follows and denoted them as D, W, A and F. The ponds in the urban areas were divided into domestic use (denoted as U) and other various usages (wastewater storage, vehicle washing, and recreational use). Because the number of ponds in urban area which are not for domestic use was small, we defined only 1 pond category denoted as U by adding to the rural pond categories as follows.

The definitions of 5 pond categories of the surveyed area in this paper:

(1) D: A pond that is used for bathing, laundry, dish-washing or other domestic use (Figure 3.2 (a)).

(2) W: A pond that is used for storage of wastewater (Figure 3.2 (b)).

(3) A: A pond that is used for paddy cultivation during dry season (Figure 3.2 (c)). (Most of them are used for fish cultivation during wet season in the area.)

(4) F: A pond that is used for fish and prawn cultivation during all seasons (Figure 3.2 (d)).

(5) U: A pond that is situated in urban area and used for the same purpose as D (Figure 3.2 (e)).

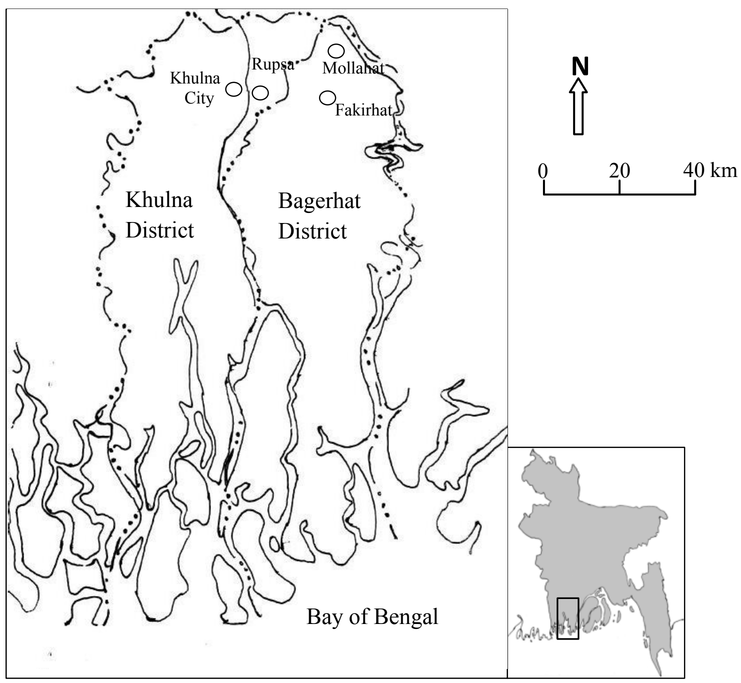


Figure 3.1 - Study area location (Rahman *et al.*, 2015)

(a) Domestic pond (b) Wastewater storage pond

(c) Agriculture pond (d) Fisheries pond

(e) Urban domestic pond

Figure 3.2 – Different categories ponds

3.2.3 Wet and dry season survey

3.2.3.1 Number of different categories surveyed ponds

The numbers of surveyed ponds in the five categories corresponding to D, W, A, F and U were 35, 12, 19, 20 and 20. The survey was carried out during the wet season during September 2012, as shown in Table 3.1. The percentage of the surveyed ponds was higher for D category (31%) and lower in W category (11%). The percentages of other surveyed ponds were almost same (around 18%). To take into account the differences between the wet and dry seasons, a portion of the surveyed ponds (49 ponds) were also investigated during February and March 2013 (dry season, see Table 3.1). The number of ponds used during the dry season was almost half that as during the wet season with the exception of a single W pond because the rest of these ponds lost water and dried out.

3.2.3.1 Number of surveyed ponds in different area

The number of surveyed ponds and their percentages during both wet and dry season in Khulna City and in the three villages are shown in Figure 3.3. The percentages of the surveyed ponds were 31, 18, 21 and 29% during wet season and 27, 12, 20 and 41% during dry season in Khulna, Rupsha, Fakirhat and Mollahat, respectively. The percentage of surveyed ponds in the four surveyed area were almost the same during both season except Mollahat during dry season.

Table 3.1 – Numbers of the different categories of surveyed ponds (Rahman *et al.*, 2015)

	Rural area				Urban area	
	Domestic (D)	Wastewater Storage (W)	Agriculture (A)	Fisheries (F)	Domestic (U)	Others*
Wet season	35	12	19	20	20	6
Dry season	16	1	14	10	8	-

* Wastewater, vehicle wash, recreational use

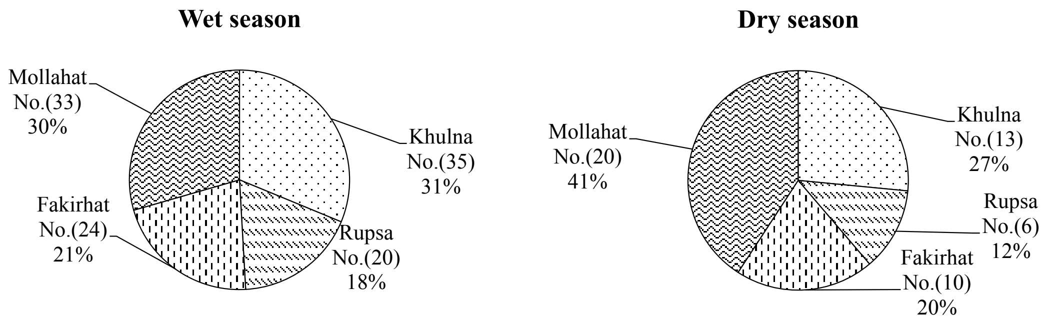


Figure 3.3 – Numbers and percentages of the surveyed ponds in different areas

3.2.4 Pond size and water quality measurement

The surveyed ponds were all artificial ponds which normally made in rectangular form. The length and width of the surveyed ponds were measured using a laser distance meter (Leica DISTO X310). Their water depth was directly measured or estimated by interviewing the owners or local people. The areas and volumes were estimated from their lengths, widths and depths. Because the area and the depth of the pond largely decreased during the dry season, the remaining surface area and decreased depth were measured.

Because it was difficult to take sample water inside of the ponds, surface water that seemed uninfluenced by bank and bottom mud, water weed and floating matters was carefully selected and taken from the pond side. Temperature, pH, DO, ORP, EC and transparency of the sample water were measured by portable meters (HORIBA D-54SE and HACH HQ30D) on-site. The collected sample in a 1L plastic bottle was brought to the laboratory in Khulna University of Engineering and Technology where SS was measured immediately. Two hundred and fifty mL samples were transferred in other plastic bottles and they were stored in a refrigerator until the transportation to Japan. No acid was added to the sample during the one day transportation. In Japan rest of all laboratory analysis (including examinations of TOC, T-N, T-P, PO_4-P and alkalinity using the JIS K0102 (Japanese standard association, 2013), Shimadzu TOC-V WP and Hitachi U-2900 was carried out. Statistical analysis (normality, F-test, t-test etc.) also carried out to compare different categories of pond groups.

3.2.5 Principal component analysis

The correlation matrix of the 12 water quality items (on-site tests and laboratory analysis) derived from the samples was calculated. Eigenvalues and eigenvectors of the correlation matrix were estimated using the power method (Hoffman, 1992). Factor loadings for the principal components and their contribution ratios were then obtained from the eigenvalues and eigenvectors. Finally, the principal component scores of the samples were calculated.

3.3 Results and discussion

3.3.1 Pond sizes during the wet season

The areas and volumes of the ponds varied according to the different categories. The average and standard deviation of volume, area and depth of the category D, W, A, F and U ponds are shown in Figure 3.4. Although the standard deviation of each pond category was large, the differences between the five categories were statistically clear. Because U ponds were divided into two (large and small) groups, they were statically analyzed by dividing into the large group (Ul) and the small group (Us). The area was largest in Ul ponds and secondarily large in F and A ponds. The area of Ul of which average was greater than 10,000 m^2 showed significant difference from the area of category F and A ponds ($p < 0.001$). Ul ponds were used for not only domestic water usage but also recreational and landscape use in the urban area. That the reason why Ul ponds possessed such large area. No difference between category F and A ponds ($p > 0.05$) was found and their averages were greater than 2,000 m^2. Because A ponds were used as paddy field mainly in dry season, they had suitable size for paddy cultivation. The average area of F ponds was slightly smaller than the reported area in the same district (Chowdhury and Ahmed, 2012) and within the reported area in eastern Bangladesh that ranged between 2,000 to 5,000 m^2 (Siddique *et al.*, 2012). Rice was already harvested in the paddy fields in category A ponds at the time of the survey and these ponds were already covered with water. Moreover, fish were cultivated in some of the category A ponds as reported by Islam *et al*. (2004) and Roy *et al*. (2013). They reported that category A ponds cultivate fish during wet season and cultivate paddy during dry season in the same area. The area of A ponds was larger than category D ponds ($p < 0.05$) and the area of D ponds (an average of < 1,000m^2) was larger than W ponds ($p < 0.001$). The Us ponds and D ponds showed similar distribution and the averages area were not significantly different ($p > 0.05$). The average depth of the five categories did not differ significantly ($p > 0.05$) and was approximately 1 m as shown in Figure 3.4.

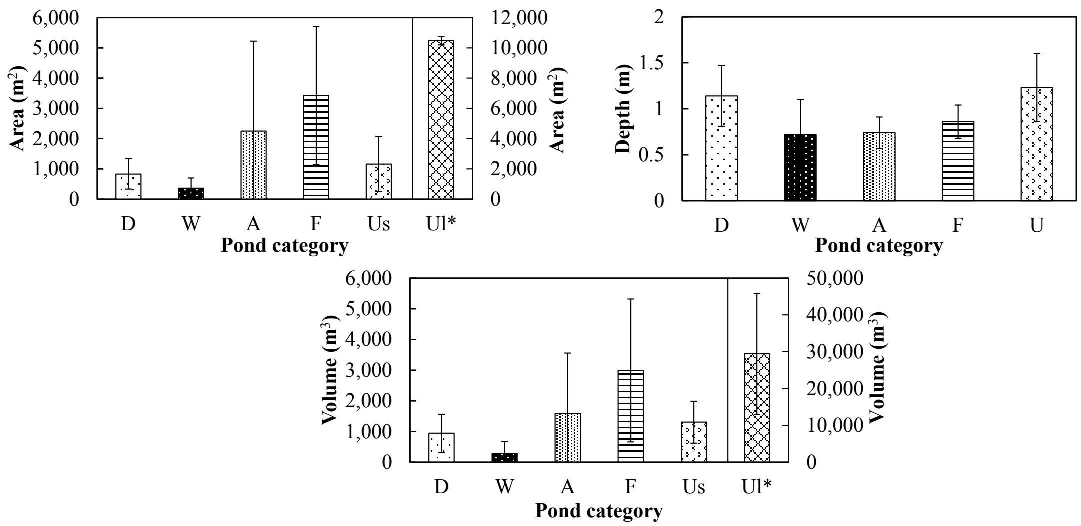


Figure 3.4 - Pond size during the wet season (*Ul uses the secondary axis (right side); error bar shows standard deviation) (Rahman *et al.*, 2015)

The volumes of the pond were large for the category F and Ul ponds (average > 2000 m^3) and have no significant difference between them ($p > 0.05$). The volume of F category pond was significantly larger than category D ($p < 0.001$), A ($p < 0.05$), W ($p < 0.001$) and Us ($p < 0.005$) ponds. Fishery ponds required enough volume for the cultivation of prawns and fish. Additionally, their depth needed to be shallow enough for the fishermen to walk in the ponds to maintain and catch fish. Large domestic ponds in urban areas (Ul) were large compared to the rural domestic ponds (D). Most of the Ul ponds were used by many people and families at the same time, whereas most of the D ponds were used by only one family. The pond volumes for category A, Us and D ponds had no significant difference between them ($p > 0.05$) and their averages being 1,600, 1,300 and 940 m^3, respectively. The D pond water amount decreases during dry season (November to April). So the D pond volume during the wet season must be larger than the volume that can keep water at the end of the dry season after the water decrease during dry season. The pond volume for category W ponds was significantly small (average < 300 m^3, $p < 0.005$) compared to the other categories.

3.3.2 Water quality during the wet season

The average, standard deviation, maximum and minimum values for the temperature, pH, DO, EC, transparency, SS, TOC, T-N and T-P are shown in Table 3.3. The average temperatures in the five pond categories were all approximately 30^{o}C. The average pH values ranged from 7.4 – 7.7. Although the differences by category were small, the pH values were rather highest ($p < 0.01$) in category A ponds than others. The DO of W category ponds was lower than the other categories, although DO results showed no significant difference ($p > 0.05$) between different categories. Also ORP results had no significant difference ($p > 0.05$) between different categories. These DO and ORP results showed aerobic conditions in all the ponds. However, the aerobic conditions were less in category W ponds than the others.

The averages of the EC for category D, W, A, F and U ponds were 1.3, 12, 0.54, 2.1 and 0.68 mS/cm, respectively. The EC in category W ponds was high because these were contaminated with toilet waste containing high salinity. Category F ponds were divided into two groups that exhibited an EC of 0.30 - 1.4 mS/cm (0.84±0.33 mS/cm; average±standard deviation) and 2.0 - 5.7 mS/cm (3.1±1.0 mS/cm) (Figure 3.5). The average EC of the 15 shallow tube wells water in the area (Rupsa, Fakirhat and Mollahat) ranged from 0.7 to 1.2 mS/cm except one well (Hasan *et al*., 2012). Therefore EC of the former group was close to the well water. So we considered the former group as fresh water ponds in this paper, while the latter pond group seemed to contain partly saline water were called as blackish water. Culture fishery includes a variety of fish, shrimp and prawn harvests from ponds in the greater Khulna district using both fresh and saline water (Rasheed, 2008). The EC exhibited a positive correlation (r = 0.63) to the pond sizes except one large freshwater pond (Figure 3.5). The average volume of blackish water ponds was 3,318±1,189 m^3 which was significantly larger ($p < 0.001$) than the average volume of fresh water ponds (1,364±735 m^3). This corresponded to the fact that the average farm size for bagda (Tiger Shrimp) grown in saline conditions was larger than that for golda (Giant Prawns) grown in freshwater (Rasheed, 2008).

The average and minimum transparencies were small ($p < 0.001$) in category F, W and U ponds. The average SS was significantly large ($p < 0.01$) in category F and U ponds where the water was greenish colored and the low transparency appeared to be the result of significant algal growth. The low transparency of the category W ponds may be caused by the polluted wastewater containing human excreta. The transparency was significantly larger ($p < 0.05$) and the SS was low in category A and D ponds, suggesting a cleaner water quality compared to the other categories. The TOC was also significantly high ($p < 0.05$) in category F and U ponds, whereas it was low ($p < 0.05$) in category A and D ponds.

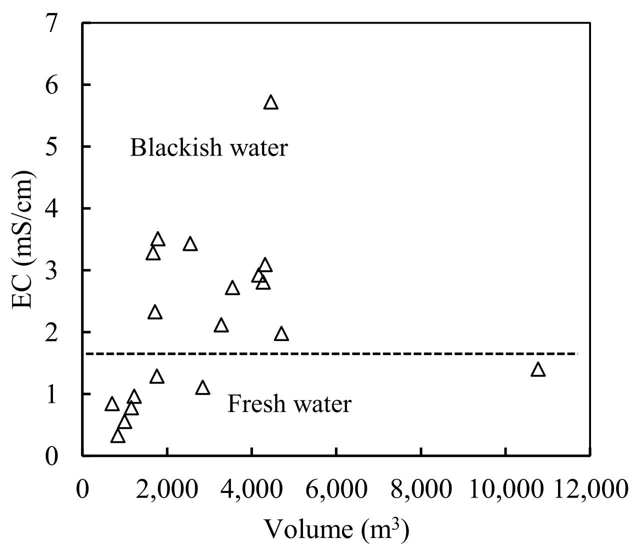


Figure 3.5 – Relation between pond volume and EC for F category ponds

The T-N and T-P were significantly high in category W and U ponds than other categories ponds ($p < 0.05$). Whereas T-N and T-P were significantly lower in category A ponds ($p < 0.05$) compared to the others. Wastewater, especially human excreta containing high amounts of nitrogen and phosphorus would explain the increase in the T-N and T-P in category W ponds. Only small amounts of wastewater were found to be discharged directly into the domestic ponds in the urban areas (U). U ponds were situated in urban area where presence of livestock was little. However, some ponds were very near to the toilet as well as drainage. These ponds have high risk of contamination by toilet or drainage channel wastewaters which increase T-P and T-N of pond water. Moreover, wastewater discharging into drainage channels in the city often flowed out or infiltrated into the groundwater. Polluted water containing nitrogen and phosphorus would increase the T-N and T-P concentrations in category U ponds. Such pollution of the pond by wastewater suggested the pathogenic contamination, although it was not measured in this survey. People directly use U ponds water (as same as D pond) for hand washing, washing of cloths and dishes, bathing etc. These water uses make direct contact with their skin and affect their health by waterborne disease like skin disease, diarrhea, dysentery, cholera, fever etc. (Haque *et al.*, 2010). They reported that skin disease, diarrhea, dysentery were the first rank disease in Khulna district where 77.14% people use pond water as cooking water, washing and bathing purpose.

The T-N and T-P values in category F and D ponds were also greater than the eutrophication level. The nitrogen and phosphorus seemed to be added into category F ponds as fish feed or chemicals used during cultivation. Actually it is reported that people input fertilizer into F ponds in the area (Islam *et al*., 2004 and Roy *et al*., 2013). The T-N and T-P concentrations in category A ponds were very low compared to the others.

3.3.3 Changes in pond size and water quality during the dry season

The rainfall amount entering the ponds during the dry season was very small compared to the wet season. The decrease in precipitation changed the pond size (depth, area and volume) significantly. The volumes of all the ponds surveyed during both seasons exhibiting a large decrease ($p < 0.05$) were compared, as shown in Figure 3.6. The decrease of volume for category A and W ponds was especially large. For category W, only one pond was surveyed and it retained no water. Water was removed in the central parts of category A ponds where rice was planted. Pond water in category A remained only in the surroundings of the rice fields. The ratio of the volume of category A ponds during the dry season to that during the wet season was 0.304±0.288 (average ± standard deviation). The volume of category D, F and U ponds during the dry season decreased to less than half of the volume during the wet season (0.421±0.175, 0.477±0.277 and 0.495±0.197, respectively).

Table 3.3 - Pond water quality during the wet season

		Domestic (D)	Wastewater Storage (W)	Agriculture (A)	Fisharies (F)	Urban Domestic (U)
Temperature (^oC)	Avg±SD	30.8±0.9	30.3±1.7	32.0±1.4	30.3±0.6	30.7±1.3
	Max	33.7	33.6	34.9	31.5	33.2
	Min	29.3	27.8	29.9	29.3	28.8
pH	Avg±SD	7.5±0.2	7.4±0.3	7.7±0.2	7.5±0.3	7.6±0.2
	Max	8.3	8.0	8.1	8.4	8.0
	Min	7.2	7.2	7.5	7.0	7.2
DO (mg/L)	Avg±SD	6.3±2.2	4.5±3.1	6.3±1.7	5.6±1.7	6.3±3.3
	Max	10.5	9.1	9.6	8.1	16.3
	Min	2.3	0.1	3.5	1.9	1.3
ORP (mV)	Avg±SD	169±70	150±72	189±68	171±89	145±92
	Max	270	272	289	290	298
	Min	44	66	60	33	-58
EC (mS/cm)	Avg±SD	1.30±1.47	0.50±0.15	0.54±0.21	2.09±1.35	0.68±0.25
	Max	5.79	0.79	1.11	5.72	1.36
	Min	0.25	0.31	0.24	0.33	0.34
Transperancy (cm)	Avg±SD	47±19	27±15	51±21	26±8	40±29
	Max	80	60	110	45	110
	Min	20	10	25	15	16
SS (mg/l)	Avg±SD	12±10	14±16	7.8±6.5	28.7±18.6	22±22
	Max	53	55	24	75	100
	Min	0	0	0	6	0
TOC (mg/l)	Avg±SD	8±3	10±10	7.3±3.3	12±4	10±3
	Max	16	41	16.5	21	17
	Min	3	4	3.7	5	4
TN (mg/l)	Avg±SD	1.03±0.56	2.16±3.43	0.59±0.28	1.61±1.01	2.11±1.16
	Max	2.83	13.18	1.37	4.68	4.28
	Min	0.31	0.42	0.34	1.12	1.53
TP (mg/l)	Avg±SD	0.17±0.25	0.88±0.74	0.03±0.03	0.13±0.25	0.64±0.45
	Max	1.1	2.73	0.09	1.12	1.53
	Min	0.01	0.01	0	0.01	0

The temperature is low from November to February in the area surveyed. Because the survey during the dry season was carried out in February and March, the pond water temperature was a slightly lower (approximately 4^oC lower) than during the wet season. A comparison of the transparency, SS, TOC, T-N and T-P during the wet and dry seasons is shown in Figure 3.7. The transparency was significantly decreased ($p < 0.001$) and the concentration of SS, TOC, T-N and T-P were significantly increased ($p < 0.001$, $p < 0.005$, $p < 0.001$ and $p < 0.01$, respectively) during the dry season. The transparency during the dry season drastically decreased to almost half of the transparency during the wet season with the exception of a few ponds in categories D and U. The SS increased by more than 10, 20 and 30 mg/L in 75%, 63% and 25% of the ponds, respectively. Such an increase in SS and a decrease in the transparency meant that the pond water became more turbid during the dry season, which seemed to be caused by the increase of algae in the water.

The comparison of SS, TOC, T-N and T-P during the two seasons shown in Figure 3.7 exhibited the regression lines having the slopes of 2.37, 2.23, 1.60 and 2.17, respectively. The result that the slopes were almost same and around 2 suggested that the increase of TOC, T-N and T-P was related to the increase of SS. The average weight ratio of C:N:P was 100:13:2 during the wet season and 100:16:3 during the dry season. Those ratios were close to the weight ratio (100:17.6:2.4) of nominal composition of plankton such as $C_{106}N_{16}P$ (Redfield ratio; Redfield, 1934). Although chlorophyll pigment was not measured, the greenish color of the pond water suggested that the particulate matter mainly consisted of phytoplankton community. Chowdhury and Mamum (2006) reported high abundance of Cyanophyceae in two fishponds in Khulna during dry season. Affan *et. al.* (2005) reported that chlorophyll a concentration in a pond of central Bangladesh became higher in March (dry season) than the concentration in September (wet season). Hossain *et al.* (2008) also reported that total phytoplankton abundance was higher in dry season (January-March) in northern part of Bangladesh.

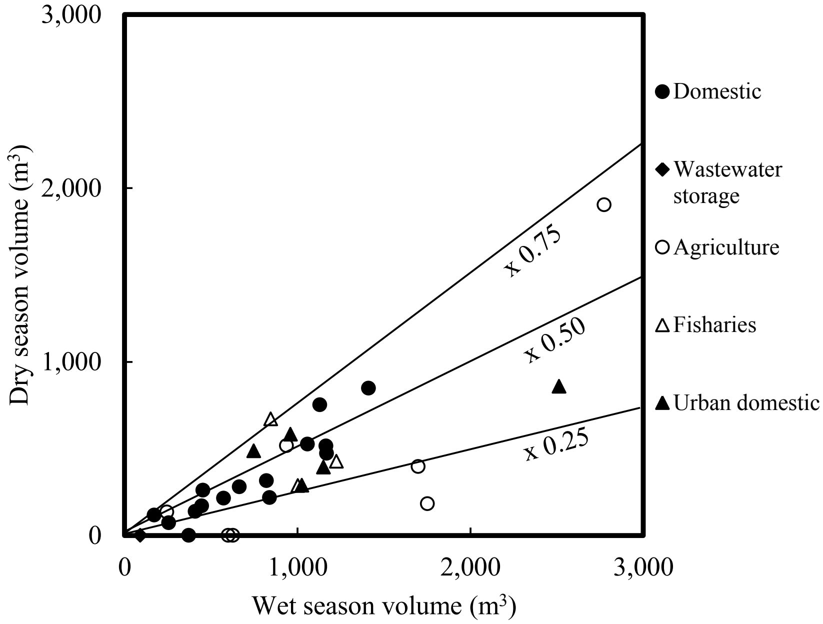


Figure 3.6 – Pond volume comparison during the dry and wet seasons (Rahman *et al.*, 2015)

The increase in the TOC during the dry season was not as drastic as for the SS. The difference between the two seasons was less than 10 mg/L with the exception of one category D and category U pond each. The average ratio of the TOC during the dry season to that in the wet season was 1.4. The increase in T-N was also small. The average ratio of T-N during the dry season to of that during the wet season was 1.8. The TOC and T-N were almost completely in a dissolved form (the average DOC/TOC and D-N/T-N were 0.86 and 0.94 in both seasons, respectively, as shown in Table 3.4) and therefore did not correspond to the large increase in the SS experienced during the dry season. Contrary to that, the T-P values largely increased and 79%, 55% and 39% of ponds experienced a T-P increase greater than 0.05, 0.1 and 0.2 mg/L, respectively. The average D-P/T-P ratio was significantly different during the wet (0.78) and dry seasons (0.41), as shown in 3.4. This suggested that the large amount of phosphorus accumulated in particulate form was likely from the phytoplankton abundance during the dry season. The fact that the D-N/T-N ratio remained high and the D-P/T-P ratio decreased during the dry season shows that phosphorus was a limiting factor controlling the phytoplankton growth in the pond water.

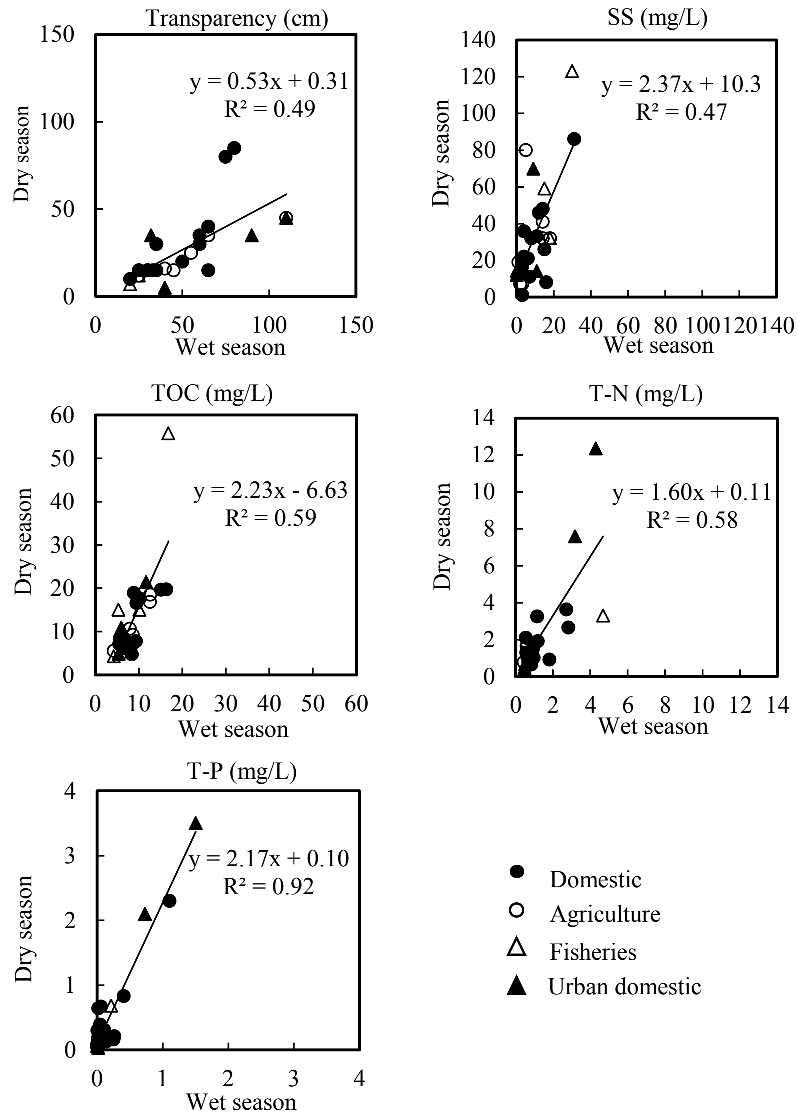


Figure 3.7 – Comparison of transparency, SS, TOC, T-N and T-P during the wet and dry seasons (Rahman *et al.*, 2015)

The increase of SS and phytoplankton in the surveyed ponds during the dry season seemed to be related to the change of their sizes. It was suggested that the decrease of volume of the ponds caused the increase of nutrients concentration and productivity resulting in the high algal growth in the ponds. Moreover bottom sediment would flow up because of the water turbulence in some shallow ponds when their depth decreased during the dry season.

Table 3.4 - DOC/TOC, D-N/T-N and D-P/T-P ratio in the surveyed ponds

		DOC/TOC	D-N/T-N	D-P/T-P
Total	Avg	0.86	0.94	0.58
	SD	0.19	0.08	0.30
Wet season	Avg	0.80	0.95	0.78
	SD	0.16	0.05	0.23
Dry season	Avg	0.92	0.92	0.41
	SD	0.20	0.10	0.24

3.3.4 Principal component analysis of the water quality

The contribution ratios from the first, the second, the third and the forth principal components (Z1, Z2, Z3 and Z4) were 0.38, 0.13, 0.11 and 0.10, respectively, as shown in Fig. 3.8. The contribution ratio of Z1 was large compared to the others and Z1 was responsible for almost 40% of the variances in the water qualities. The cumulative contribution ratio of Z1, Z2, Z3 and Z4 was 0.72, showing that two thirds of the variances in the 12 items examined were described by the four principal components.

The factor loadings for Z1, Z2, Z3 and Z4 are shown in Table 3.5. The Z1 factor loadings were large for the pH, DO, transparency (negative), SS, TOC, T-N, T-P, and PO_4-P. This suggested that Z1 indicated the trophic states of the ponds showing the eutrophication progress including the nutrient increase and algal growth (SS increase and transparency decrease) caused by photosynthesis.

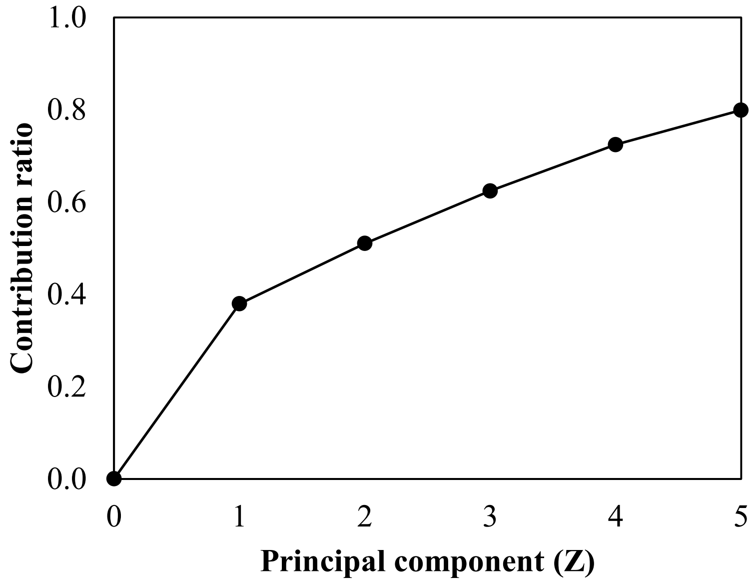


Figure 3.8 - Cumulative contribution ratio (Rahman *et al.*, 2015)

Table 3.5 - Factor loadings

Factor loading	Z1	Z2	Z3	Z4
pH	0.543	-0.403	0.343	0.284
Temp	-0.306	-0.029	0.747	-0.341
Alkalinity	0.379	0.112	-0.127	0.405
DO	0.561	-0.422	0.466	-0.249
ORP	-0.167	-0.066	0.397	0.706
EC	0.309	-0.456	-0.352	-0.434
Transp	-0.501	0.367	0.384	-0.183
SS	0.745	-0.274	-0.081	0.184
TOC	0.892	-0.132	0.089	-0.107
T-N	0.924	0.139	0.093	-0.055
T-P	0.808	0.557	0.075	-0.062
PO_4-P	0.689	0.669	0.046	-0.077

The Z2 factor loading was large for the EC (negative) and phosphorus. Figure 3.9 showed the scores for each sample on the Z1-Z2 plane. It was clear that the Z2 scores were positive for category W and U ponds and was negative for category F ponds during the dry season. As mentioned, category W and U ponds were influenced by wastewater contamination, resulting in high T-N and T-P values. Therefore, Z2 appeared to represent the influence of wastewater contamination.

The Z3 factor loading was large for the temperature. Because the temperatures were higher during the wet season and lower during the dry season, the sample scores for the two seasons were clearly separated in the Z1-Z3 plane, as shown in Figure 3.10. Therefore, Z3 seemed to respond to the seasonal influences.

The Z4 factor loading was large for the alkalinity. Figure 3.10 showed the scores for each sample on the Z1-Z4 plane. Because the alkalinity was lower during the wet season and higher during the dry season, the sample scores for the two seasons were clearly separated in the Z1-Z4 plane. Therefore, Z4 appeared to represent the influence of alkalinity.

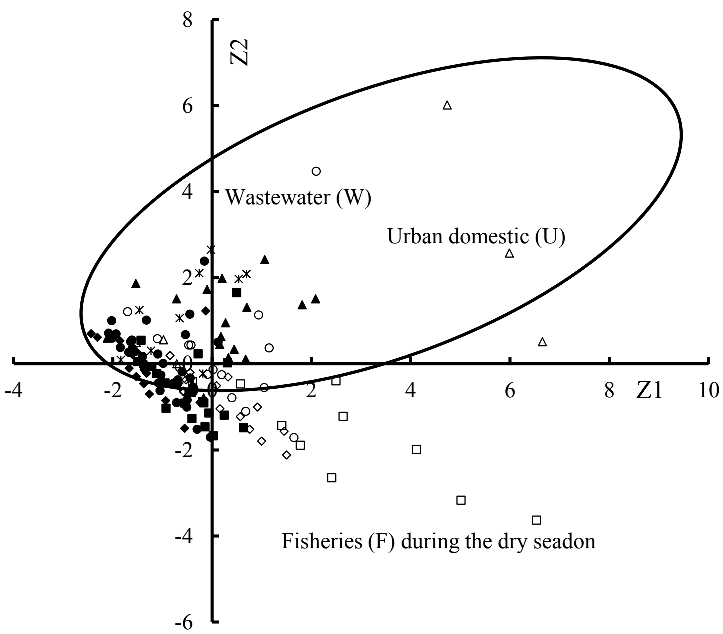


Figure 3.9 - Scores for the pond water on the Z1-Z2 plane (Circle: D, Rhombus: A, Square: F, Asterisk: W, Triangle: U, Black: wet season and White: dry season) (Rahman *et al.*, 2015)

Figure 3.12 shows the average and standard deviations for the Z1 scores in the five pond categories during the wet and dry seasons. The increase in Z1 was clear during the dry season and suggested an increase in the trophic state in ponds D, A, F and U. Nutrients are generally necessary for paddy and fish production. Nutrients such as cow dung and urea are also added in

F ponds to increase fish production in the same area (Chowdhury and Mamun, 2006). Therefore progress of the eutrophication during dry season seems to matter little to A and F ponds. For these reasons the increase of Z1 score didn't have serious problems on A and F ponds.

According to the increase of Z1 during dry season indicating the increase of SS and turbidity in the D and U ponds, the risk of contamination of pathogenic microorganisms would increase during the dry season and cause human health risk because some of them grow on particulate matter (Tortora *et al.*, 1992) and get longer survival (Maier *et al.*, 2009). Bacterial abundance has positive correlation with primary production in water bodies (Cole, 1982) and microbial attachment with SS is common in aquatic ecosystems (Pearl, 1975). For these reasons, the water quality in D and U ponds should therefore be improved especially during the dry season.

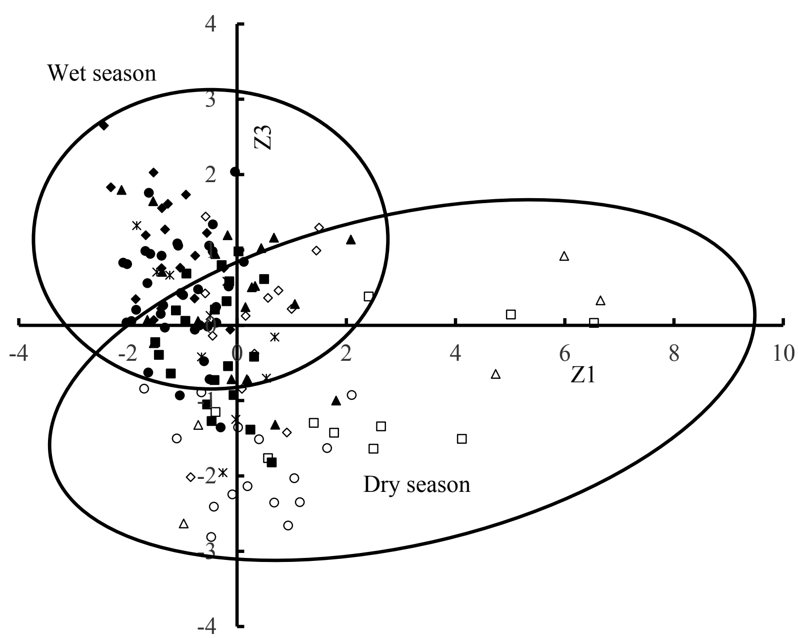


Figure 3.10 - Scores for the pond water on the Z1-Z3 plane (Circle: D, Rhombus: A, Square: F, Asterisk: W, Triangle: U, Black: wet season and White: dry season) (Rahman *et al.*, 2015)

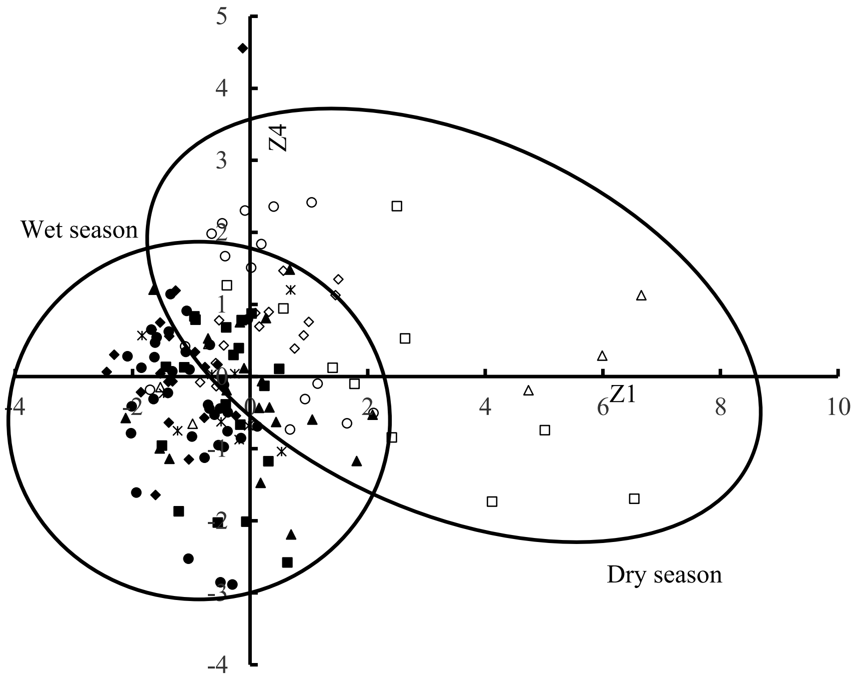
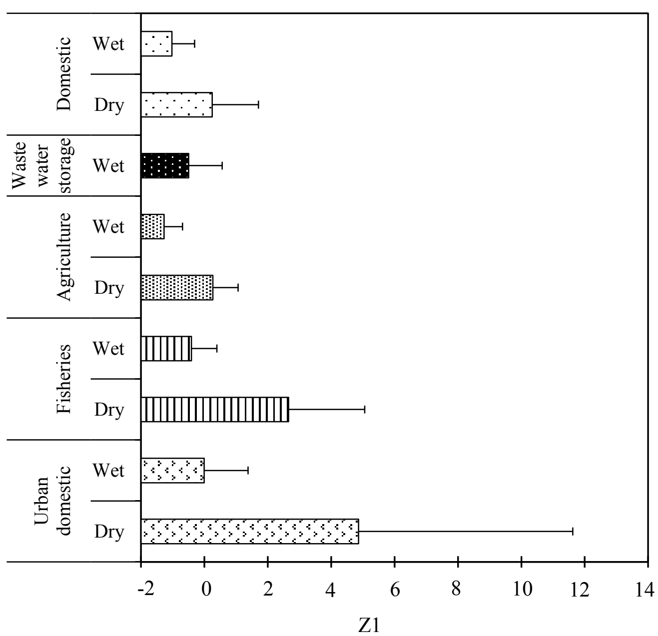


Figure 3.11 - Scores for the pond water on the Z1-Z4 plane (Circle: D, Rhombus: A, Square: F, Asterisk: W, Triangle: U, Black: wet season and White: dry season) (Rahman *et al.*, 2015)



.Figure 3.12 - Average and SD of the Z1 scores (Rahman *et al.*, 2015)

However, it has been difficult to introduce any technologies to improve the water quality of the ponds. According to our survey results the high turbulence or SS is the most serious problem of the pond water quality. Therefore filtration process seemed to be effective to separate them. Instead of improvement of the pond water quality, the government and NGOs have installed pond sand filter (PSF) which is used for purification of D ponds water (Harun and Kabir, 2013). But this system also has high installation and maintenance cost (UNEP, 1998) and most of the present users of PSF are not satisfied (50% partially dissatisfied and 30% dissatisfied) with this system due to poor PSF water quality for lack of maintenance, long waiting time, long distance from home etc. (Shafiquzzaman *et al.,*2009; Harun and Kabir, 2013). Low cost treatment system therefore need to explore which can treat the pond water with low maintenance and affordable to the local people.

3.4 Summary

This chapter successfully categorized the rural area ponds of Bangladesh. The following conclusions were found out during field survey.

(1) The surveyed ponds were categorized into domestic use (D), wastewater storage (W), agricultural use (A), and fisheries (F) in rural area and domestic use (U) wastewater storage, vehicle washing and recreational use in urban areas. U ponds was divided into two group as urban domestic large (Ul) and urban domestic small (Us)

(2) The area of the ponds was Ul > F > A > (D = Us) > W and volume of the ponds (Ul = F) > (A = D = Us) > W during the wet season. The average depth of the five categories did not change significantly and was approximately 1 m.

(3) The transparency was low in the category F, W and U ponds and the SS, TOC were high in the category F and U ponds than the others. On the other hand the T-N and T-P were higher in category W and U ponds than the others. The low transparency in the F and U category ponds seemed to be caused by high rates of algal growth.

(4) The size of all the ponds decreased during the dry season. Transparency during the dry season drastically decreased to almost half that during the wet season. The pond water became more turbid during the dry season resulting from an increase of primary production.

(5) The principle component analysis showed that the contribution ratio of the first component (Z1) was large compared to the others. The Z1 factor loadings suggested that it described the eutrophication progress (trophic states).

(6) The increase of Z1 during the dry season in D and U ponds suggested that water quality improvement is necessary.

Based on the above summaries, it was confirmed that pond water quality of D and U ponds need to be improve especially during dry season.

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Chapter 4

Immobilization of rice husk charcoal for water treatment

4.1 Introduction

Rice husk charcoal (RHC) generally prepared from rice husk, which a natural material, is found from agricultural waste by-product and generally used as adsorbent. Rice husks are accounting for approximately one-fifth of the annual gross of rice in the world (545 million metric ton) (Mansaray, and Ghaly, 1998). According to Bailey *et al*. (1990), an adsorbent can be considered as cheap or low-cost if it is abundant in nature, requires little processing and is a by-product of waste material from waste industry. Plant wastes are inexpensive as they have no or very low economic value. Some of the advantages of using plant wastes for wastewater treatment include simple technique, requires little processing, good adsorption capacity, selective adsorption of heavy metal ions, low cost, free availability and easy regeneration (Ahmaruzzaman and Gupta, 2011). Natural materials that are available in large quantities, or certain waste products from industrial or agricultural operations, may have potentiality as inexpensive adsorbents. The abundance, availability and low cost of agricultural byproducts make them good adsorbents for the removal of various pollutants from wastewater. Agricultural waste biomass currently is gaining importance. In this perspective rice husk, has emerged as an invaluable source for the utilization in the wastewater treatment (Ahmaruzzaman and Gupta, 2011). It consists of cellulose (32.24%), hemicelluloses (21.34%), lignin (21.44%) and mineral ash (15.05%) (Rahman *et al.*, 1997), with high percentage of silica (96.34%) in the mineral ash (Rahman and Ismail, 1993). However, the rice husk needs to be modified or treated before being applied for treatment (Chakraborty *et al.*, 2011). Some researchers reported that the pretreatments of rice husk increased the cellulose of the solid fraction by virtue of lignin removal and hemicelluloses solubilization. In addition pretreatment also caused the cellulose crystallinity to decrease and the porosity of the adsorbent to increase (Ngah, and Hanafiah , 2008). Many researchers found that the modified rice husk exhibited higher adsorption capacities than unmodified rice husk (El-Shafey, 2007; Sahu *et al.*, 2009 and Daffala *et al.*, 2010). Chemical or thermal treatment reduces cellulose, hemicelluloses and lignin crystalinity, leading to an increase of specific surface area for adsorption (Daffala *et al.*, 2010).

It was well known that RHC used as an adsorbent prepared by acid treatment, alkali treatment, carbonization (El-Shafey, 2004; Bishnoi *et al.*, 2004 and Shindhu *et al.*, 2012) for the removal of heavy metal like cadmium, copper, zinc, lead, nickel, chromium (Kumar and Bandhopadhay, 2006; Daifullah *et al.*, 2003 and Guo *et al.,* 2002) and many other impurities from wastewater. RHC also used as a reductant to reduce Cr(VI) to Cr(III) by ion exchange (Iwanaga *et al.,* 2013). RHC normally used either as granular type or as powder type. Using RHC in water and wastewater treatment facilities faced some difficulties. The main problem is floating RHC (especially powder type) on the water or wastewater, which could not participate fully in the treatment process that leads decrease in treatment efficiency. Another problem is to handling RHC in the maintenance process like removing used RHC for filter unit and installs new RHC. A certain amount of RHC was lost in the handling process. Moreover granular type RHC is become partially powdered in the treatment process. It would be more effective if RHC have suitable handling shape without changing their properties. By immobilizing RHC mixing with clay could be a good option to overcome the floating, broken and handling problem.

This chapter aims to develop immobilization rice husk charcoal (IRHC) using Bangladesh clay soil and rice husk charcoal (RHC) prepared in laboratory design furnace in Bangladesh. The details manufacturing procedure of IRHC and comparison of RHC and IRHC was described in this chapter. Moreover application of IRHC on Cr (VI) reduction as an example of the usage of the IRHC was investigated and described in Appendix.

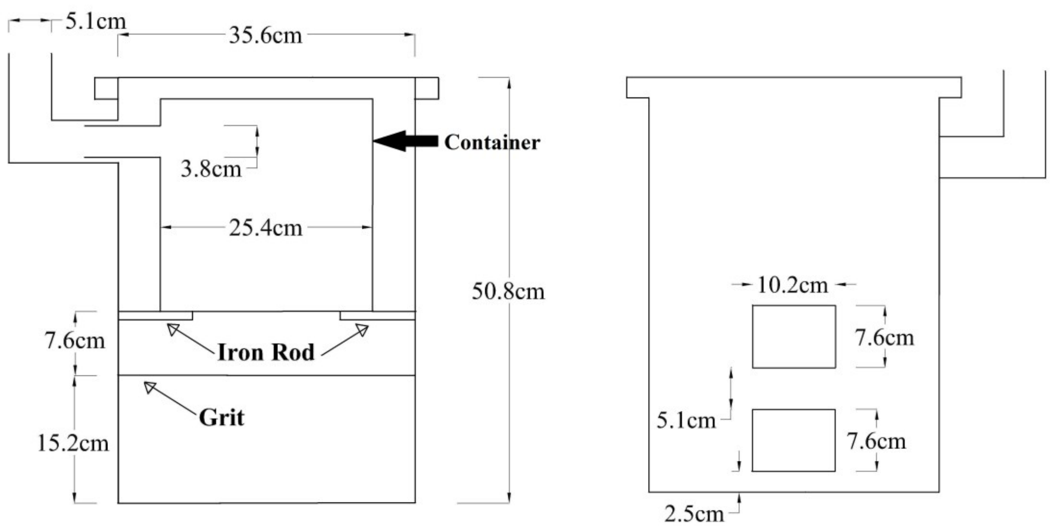
4.2 Materials and methods

4.2.1 Ingredients

Locally available materials – rice husk and clay soil were used as primary ingredients for manufacturing of IRHC. Rice husk was brought from local market and clay was collected from the paddy field of Khulna city in Bangladesh. Laboratory designed furnace was used for manufacturing RHC in Bangladesh. Muffle furnace and laboratory designed furnace was used for manufacturing IRHC.

4.2.2 Furnace

Schematic diagram of laboratory designed furnace (designed in environmental laboratory, department of civil engineering, Khulna University of Engineering & Technology, Bangladesh) was shown in Figure 4.1 designed in previous study (Rahman and Bari, 2010). The furnace was made by 1.25 mm MS steel sheet with filling of clay and chalk powder mixture that worked as an insulator in the furnace. The outer side of the furnace was coiled with asbestos rope. The furnace had a diameter of 35.56 cm and height of 50.8 cm with two 7.6 cm X 10.2 cm openings at the bottom side. Between these two openings grit was placed. The fuel was placed on the top of the grit. The top opening used to put fuel at the time of operation and bottom opening used to remove ash from the furnace. There was another small hole of 5.1 cm diameter at the topside of the furnace used as smoke outlet. The top of the furnace was open. This opening was closed in the time of operation with a separate cover made with 1.25 mm MS steel sheet.



(a) Cross-section view (b) Front view

Figure 4.1 – Schematic diagram of locally made furnace

A container made of 1.00 mm MS steel sheet with 25.4 cm in diameter and 25.4 cm in height was placed in the furnace. This container had two openings, one at the bottom and other at the top. The bottom-opening dimension was 5.08 cm X 7.62 cm. This opening was closed to a iron wire net. The top opening was joined with the furnace topside opening. Inside the container two trays of iron wire net were placed on which rice husk was placed.

4.2.3 RHC manufacturing

Figure 4.2 showed the flow chart of RHC manufacturing procedure using laboratory design furnace. RHC was prepared by carbonizing rice husk in locally made furnace in Bangladesh. Around 4 kg of rice husk was laid in three layers in a 25.4 cm diameter container. There was 2.54 cm gap between consequent two layers. Rice husk was put in loose condition in every layer. Then the container was closed, sealed and put in the furnace. Then the cover of the furnace was closed and also sealed with clay. Then the fuel was placed on the grit and burning started. When the process was going on, the smoke produced in the furnace entered the container through the bottom opening and passed out through the top opening which was connected with furnace smoke outlet. This heated air and the produced heat were used for producing activated carbon. The furnace was fueled around 4.5 hours until the smoke color turned into white from black that came out from the furnace outlet. When the smoke color turned into white all the openings of the furnace were sealed with clay. Then the sealed furnace was kept for 24 hours and opened to bring out the produced RHC. Continuous monitoring of sealing was provided because clay layer was cracked after some hours by propagation of heat. Figure 4.3 describes the different stages of RHC preparation.

4.2.4 Manufacturing of the IRHC

To select the ingredient ratio and manufacturing condition, IRHC cube (10x10 mm) were made manually with different ratios of clay soil and rice bran (90:10, 80:20 and 60:40) on dry weight basis and burned at predetermined time and temperature.

Figure 4.4 showed the flow chart of IRHC manufacturing procedure using muffle furnace. The clay soil was dried and grind by hammer and sieved by 0.5 mm mesh. The RHC was also grind and sieved by 0.2 mm mesh. The dried and sieved clay soil and rice husk were mixed with homogeneously with a ratio of 80:20 (weight basis). Sufficient amount of water was added to make dough with the homogeneous mixer. Square shaped IRHC of approximate size 10x10 mm was manually prepared by the dough using flat board, scale and knife. Then the prepared IRHC cube was put in the oven for 12 hours at 105^{o}C for drying. Figure 4.5 showed different steps of manufacturing oven dry IRHC cube. After drying the IRHC cube was transferred to the porcelain dish, covered with aluminum foil and then burned in at 500^{o}C for 90 min. Figure 4.6 showed the resulting IRHC cube manufactured in muffle furnace.

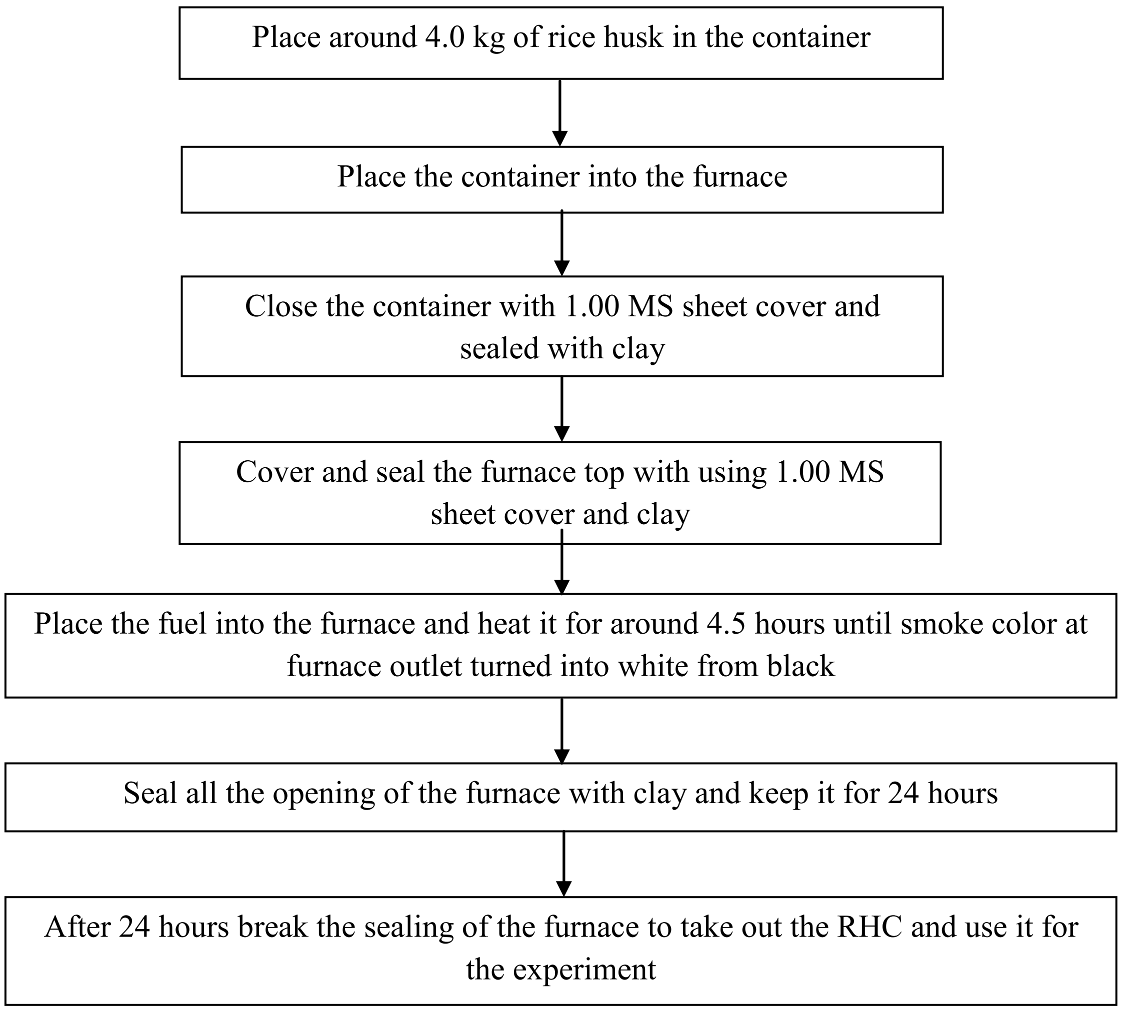


Figure 4.2 – Flow chart of the RHC manufacturing procedure

(a) Weight the container

(b) Placing the container in furnace

(c) Seal the container

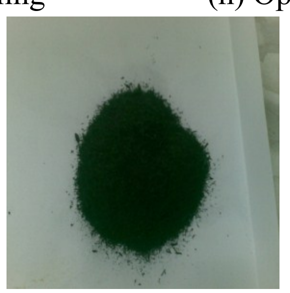
(d) Seal the furnace

(e) Heat the furnace

(f) Seal the furnace openings

(g) Keep for cooling

(h) Open furnace after cooling



(i) Bring out RHC

Figure 4.3 – Different stages of RHC manufacturing procedure (Mahmudur, 2010)

IRHC manufacturing procedure using laboratory design furnace was the same as RHC manufacturing describing in section 4.2.3. About 4 kg of oven dry IRHC cube (10x10 mm) was laid in three layers in a 25.4 cm diameter container. There was 2.54 cm gap between consequent two layers. The container was than sealed with clay and fueled for about 4.5 hours. After that all the openings of furnace was sealed with clay. Then the sealed furnace was kept for 24 hours and opened to bring out the produced IRHC. Continuous monitoring of sealing was provided because clay layer was cracked after some hours by propagation of heat. Figure 4.7 showed the resulting IRHC produced in laboratory design furnace.

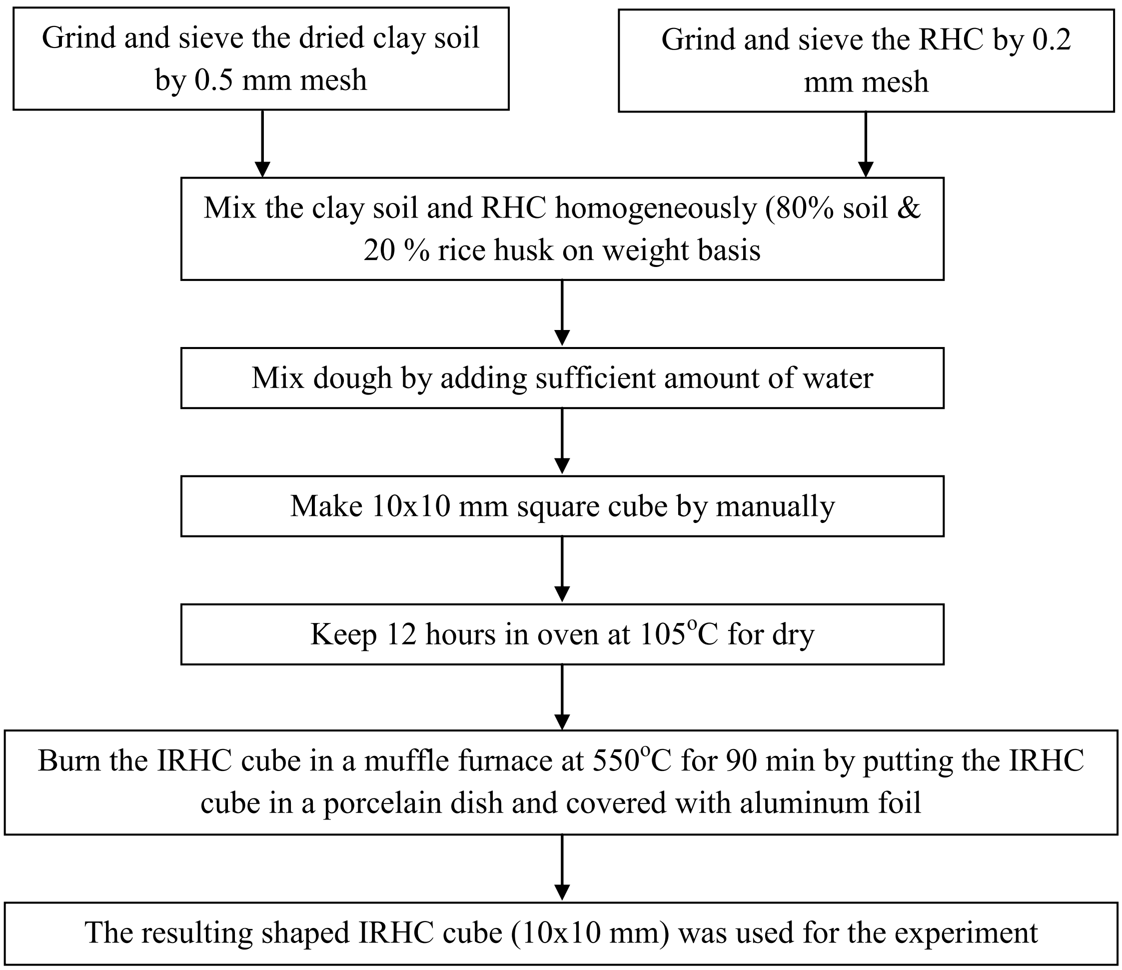


Figure 4.4 – Flow chart of the IRHC manufacturing procedure

(a) Mixing of RHC and clay soil

(b) Making dough with mixing water

(c) Shape the dough and check the measurement

(d) Cut the IRHC cube and dry in an oven at 105^{o}C

Figure 4.5 – Different steps of manufacturing oven dry IRHC cube (10x10 mm)

Figure 4.6 – IRHC cube after burning at 550^{o}C for 90 min

Figure 4.7 – IRHC cube manufactured in laboratory designed furnace

4.2.5 Comparison of RHC and IRHC

4.2.5.1 Batch study for release of turbidity, suspended solids (SS) and dissolved solids (DS)

Batch experiment carried out for checking the release of turbidity, SS and DS. For turbidity batch experiment, 1L pure water was taken in 1L beaker. Then 0.749 g of RHC or 3.016 g of IRHC (3 cubes) was added to the beaker. Next the water was stirred using glass rod slowly by hand for around 2 minutes only at the beginning of the experiment. Then 20 mL of duplicate sample was taken out from the beaker at 10, 20 and 30 minutes interval for measuring turbidity. In case of SS and DS experiment, 2 L pure water was taken in 2 L beaker. Then 1.254 g of RHC or 5 IRHC cube (5.214 g) was added to the beaker. Then water was mixed using glass rod slowly by hand for around 2 minutes. The duplicate samples of 100 mL were taken from the beaker at 10, 20 and 30 minutes interval for measuring SS and DS using the JIS K0102 (Japanese standard association, 2013). The RHC weight was taken 20% of IRHC weight.

4.2.5.2 Batch study for release of carbohydrate, T-P and PO_4-P

For carbohydrate, T-P and PO_4-P, batch experiment was carried out by taking 1 L pure water into a 1L beaker. Next 0.748 g RHC or 3 IRHC (3.062 g) was added to the beaker and stirred with glass rod slowly by hand for around 2 minutes. Around 20% of IRHC weight was used as RHC. Than 50 mL sample (triplicate) was taken every day same time for 7 days. The sample was than filtered through 5C filter paper (Advantec) and carbohydrate, T-P and PO_4-P of the samples were measured using the JIS K0102 (Japanese standard association, 2013), Shimadzu UV-1600 and Hitachi U-2900.

4.2.5.2.1 Carbohydrate measurement procedure

For carbohydrate measurement, standard was prepared by dissolving 80 mg glucose ($C_6H_{12}O_6$) in 1 L pure water using measuring flask. Then 10, 20 and 40 mg/L glucose solutions were prepared by diluting 80 mg/L glucose solution. Next 1 mL of 10, 20, 40, and 80 mg/L glucose solution and sample was taken in 50 mL test-tube. Then 1 mL of 5% phenol (C_6H_6O) solution and sulfuric acid (H_2SO_4) was added to the test tube and kept for 30 min. Next absorbance of the test tube solution was measured using UV-visible spectrophotometer at 490 nm wavelength.

4.3 Results and discussion

4.3.1 Ingredients ratio selection for IRHC manufacturing

The weight of one IRHC cube was found 0.82±0.21 g. Table 4.1showed the effect of different rice husk ratios in the ingredient. From table 4.1, it was seen that IRHC with higher percentage (40%) of RHC was broke after agitation while IRHC with lower percentage (10%) of RHC was little broke indicate that higher percentage of RHC was weak in strength and easy broke the as same as Hasan (2014), reported that higher percentage of rice bran combined with clay soil in burnt ceramic bar had weak flexural strength. Therefore, 80% soil and 20 % rice rusk was selected for manufacturing IRHC.

Table 4.1 – Effect of rice husk ratio

Rice husk ratio	Condition after agitation
40	Broke
20	Little broke
10	Little broke

4.3.2 Comparison of RHC and IRHC

4.3.2.1 Turbidity, SS and DS release in pure water

Figure 4.8 showed the concentration of turbidity, SS and DS in pure water after adding RHC/IRHC into it. It was clear that by addition of RHC in pure water turbidity and SS concentration of the solution was increased. This seemed to be caused by the powder form of RHC, which were in floating condition in the solution. It also showed that turbidity and SS could be decreased on time as floating RHC settle down at the bottom of the beaker. The concentration of DS was also increased by addition of RHC in pure water, indicating that RHC releasing dissolve particle in the solution.

On the contrary, it was seen that by addition of IRHC in pure water turbidity and SS were not increased, indicating that floating material was decreased in IRHC. Also DS showed no change indicating IRHC decreased releasing dissolved material.

4.3.2.2 Carbohydrate, T-P and PO_4-P release in pure water

Figure 4.9 showed carbohydrate, T-P and PO_4-P were released in pure water by RHC and IRHC. It was seen that carbohydrate was released by the RHC and IRHC. But carbohydrate released by IRHC was significantly ($p < 0.0001$) smaller than RHC indicating that by immobilization IRHC decrease carbohydrate release.

Also T-P and PO_4-P concentration was increased by RHC addition, which caused by phosphate release from the RHC because it contained high phosphate contents. On the other hand by IRHC addition phosphate release was decreased more than 97% suggesting that by immobilization of RHC could stop phosphate release.

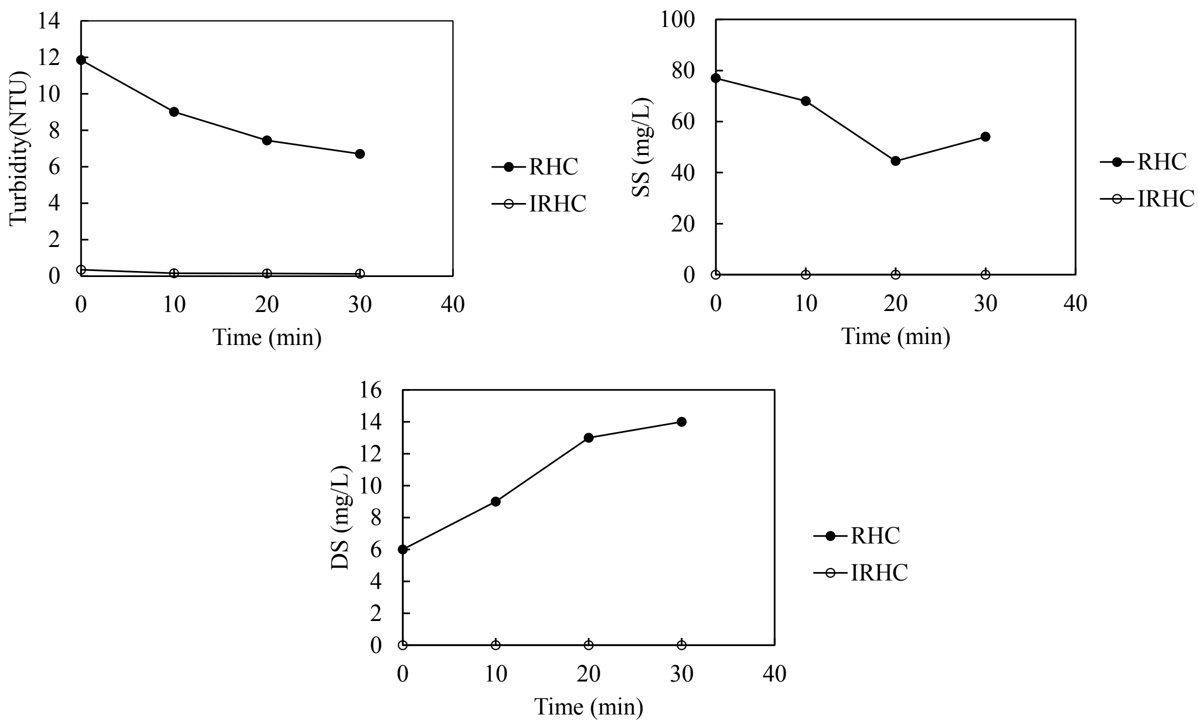


Figure 4.8 –Turbidity, SS and DS concentration in pure water by addition of RHC and IRHC. (Pure water turbidity, SS and DS concentration was <0.01 NTU, <5 mg/L and <5 mg/L, respectively)

4.4 Possible applications of the developed IRHC

The IRHC could be applied to reduce chromium (VI) into chromium (III) using as a reductant. Also it could be used as an adsorbent to remove bacteria by adsorption process. It was reported that fresh charcoal and activated carbon are widely used for bacteria removal by adsorption process (Sobsey, 2000). Activated carbon particle attached microorganisms such as negatively charged E.coli through Lifshitz-van der Waals forces by overcoming electrostatic repulsion between negatively charged cells and carbon particles (Bussscher, 2006). Among these possible applications, the applicability of IRHC for bacteria removal has been investigated.

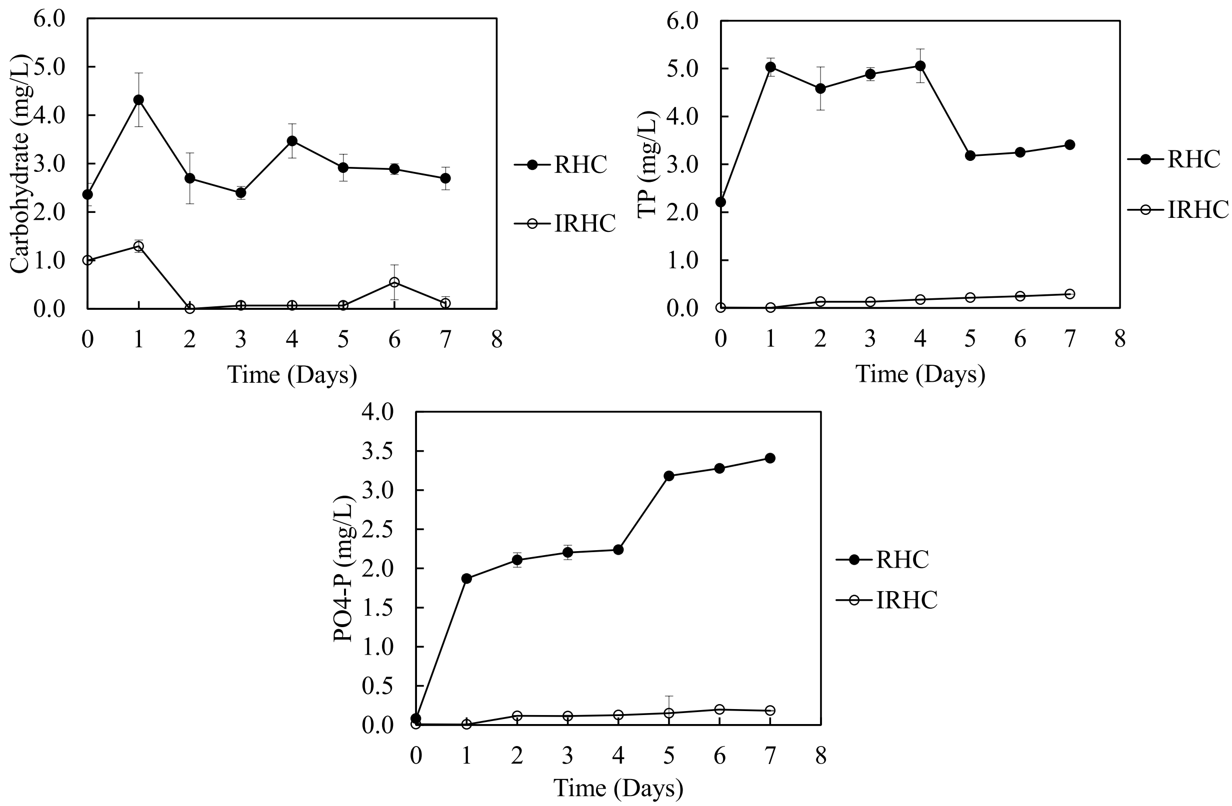


Figure 4.9 – Comparison of carbohydrate, T-P and PO_4-P release in pure water by RHC and IRHC

4.5 Summary

Immobilization of rice husk charcoal (IRHC) was successfully developed and manufactured with Bangladesh clay soil and RHC manufactured in laboratory design furnace. The ratio of 80% clay soil and 20% RHC on weight basis was found appropriate. IRHC was successfully manufactured using both muffle furnace and laboratory design furnace. Use of locally available materials and skills could reduce IRHC manufacturing cost. The release test showed that IRHC could decrease release of DS, carbohydrate, T-P and PO_4-P compare than RHC, which made them more efficient for water treatment.

IRHC could be used as adsorbent or reductant same the as RHC. The advantage of using IRHC will be easy handling, less release of DS, 100% involvement on treatment and disadvantage will be reduced active surface area which could be concentrated treatment efficiency. The possibility of IRHC application as an adsorbent for pond water treatment and Cr (VI) reduction has been investigated and described in the chapter 5 and Appendix A, respectively.

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Chapter 5

Development of a pond water purification assembly using a simple ceramic filter

5.1 Introduction

Water is an important element for human life. Most of the area of the Earth is covered by water, but the availability of safe water is limited. Approximately 1.6 million people die every year due to water and sanitation-related diseases caused by using unsafe water (Curry, 2010). Water-related problems are especially severe in Asia.

Bangladesh has abundant water, but there is a lack of safe water. People use both surface water and ground water. However, ground water is the most reliable source of safe water. The availability of safe ground water is decreasing due to a decreasing water table, arsenic contamination, and salinity (Abedin and Habiba, 2014; DPHE and UNICEF, 1994). Therefore, people have become more dependent on surface water, reconsidering surface water as an alternative source of water. In Bangladesh, ponds are used for productive purposes, such as irrigation and fish cultivation, and for non-productive purposes, such as cooking, bathing, ablution and washing clothes and kitchen utensils (Karäzlin, 2000; Milton *et al.,* 2006).

The Khulna and Bagerhat Districts, part of the coastal zone, located in southwestern part of Bangladesh, are seriously affected by salinity intrusion, arsenic and drought (Abedin and Habiba, 2014; Ministry of Water Resources, Bangladesh, 2005; Shafiquzzaman *et al.*, 2009 and Khan *et al.*, 2011). Rural people use pond water for domestic purposes as well as aquaculture in these districts. However, pond water that is contaminated with various impurities, such as algae and pathogenic bacteria, causes serious public health effects (Knappett *et al.*, 2011). In Khulna District, it was reported that 77.14 % of the people who used pond water were affected by waterborne diseases, such as skin disease, diarrhoea, dysentery, cholera and fever (Haque *et al.*, 2010). To reduce the health risks of pond water, treatment of the pond water must be applied. Filtration is one of the easy options for pond water treatment.

The pond sand filter (PSF) is a community-based pond water treatment system used in these areas. However, this system has high installation and maintenance costs (Jakariaya *et al.*, 2005), and most of the PSF users are not satisfied with the PSF performance (Harun and Kabir, 2013). Point of use (POU) systems for households using filters are an alternative pond water treatment system. There are many filters available in the local markets, such as the Safi filter and Bishuddhya filter, which are used for removing As and bacteria. However, they are too expensive for much of the rural population. The three pitcher filter is comparatively cheap, but the performance is questionable (Jakariya, 2005). Porous ceramic filters are reported to be one of the best treatment options for reducing turbidity and bacteria (Sobsey, 2002). In our laboratory, a low-cost, simple ceramic filter (SCF) was developed

(Shafiquzzaman *et al.*, 2011a) and successfully applied for arsenic removal from ground water (Shafiquzzaman *et al.*, 2011b and Hasan *et al.*, 2012). This filter was expected to be also useful for pond water treatment to obtain drinkable water quality.

Colloidal particles (turbidity) and bacteria can be removed by coagulation and flocculation with various salts of aluminum (e.g., alum), iron, lime and other inorganic or organic chemicals. Using iron as coagulant can reduce more than 90-99% of water borne pathogens (Sobsey, 2000). Fresh charcoal and activated carbon are widely used for bacteria removal by adsorption process (Sobsey, 2000). Iron as a coagulant or charcoal combining with SCF may increase the SCF performance for pond water treatment.

The aim of this chapter is to evaluate the effectiveness of suspended solid (SS) and bacteria (total coliforms (TC) and Escherichia coli (E. coli)) removal by filtration units using SCF, iron and charcoal (in different combination) from pond waters in a rural area of Bangladesh.

The main part of this chapter has been published in Japanese Journal of Water Treatment Biology (Rahman and Nakajima, 2015).

5.2 Materials and methods

5.2.1 Filter unit types and their structure

The SCF was manufactured in hollow cylindrical shape (one side closed) using Bangladesh clay soil and rice bran according to the previous report (Shafiquzzaman et al., 2011a). Figure 5.1 described the schematic diagram and the photograph of SCF. The surface area of the SCF was 0.039 m^2, calculated by the outer diameter and height of the SCF. One SCF was attached on the bottom of a 15 L clay flower pot (Shafiquzzaman et al., 2011b). This simple system was called the basic type unit and was denoted as B-filter (as shown in Figure 5.2 (a)). Secondly, a commercial iron net of 0.60 kg in weight was added to the B-filter to cover the SCF, as in the previous report, (Shafiquzzaman et al., 2011b) and was called an iron-covered type unit and denoted by I-filter (as shown in Figure 5.2 (b)). Next, rice husk charcoal (RHC; granular type with some powder) was prepared from rice husk from Bangladesh according to Rahman et al., (2013) and 0.10 kg of RHC was added to the bottom of the B-filter. The RHC contained 18.1 mg/g of phosphorus. It was called a charcoal-added type unit and denoted as R-filter (as shown in Figure 5.2 (c)). A filter unit containing both iron net and RHC was also prepared and was called a combination type unit and denoted as C-filter (as shown in Figure 5.2 (d)). Because phosphorus was released from RHC in the R-filter and C-filter, the RHC was immobilized by burning with clay as follows.

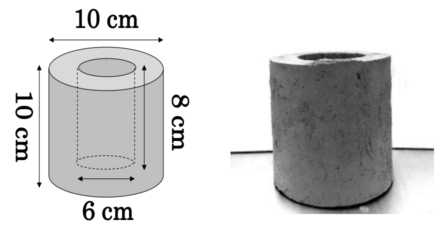


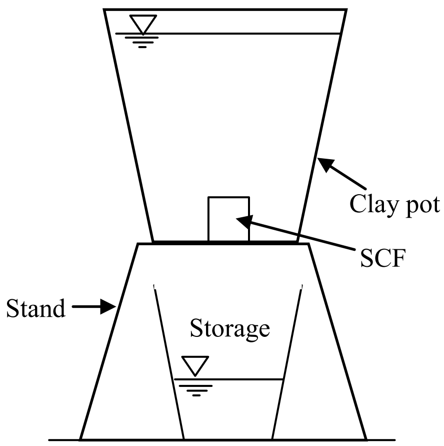
Figure 5.1 – Schematic diagram and photograph of SCF (Rahman and Nakajima, 2015)

RHC and clay soil was mixed in a 1:4 ratio on a dry weight basis, and a mud was made by adding water into it. The resulting material was cut into cubic shape pills with 10 mm sides. These pills were then burned in a muffle furnace at 500°C for 90 min. The burned pills were called immobilized rice husk charcoal (IRHC). On the bottom of the B-filter, 1.0 kg of IRHC was added, and this filter was called a hard-charcoal-added type unit that was denoted as H-filter (as shown in Figure 5.2 (e)). Figure 5.2 (f) showed actual filter set-up. The H-filter was used only during the dry season field test in place of the R-filter and C-filter that were used during the wet season test.

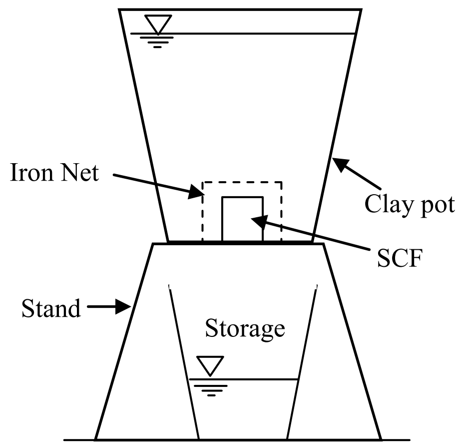
5.2.2 Laboratory test

A preliminary laboratory test of the B-filter for TC and E. coli removal was performed using bacteria-spiked test pond water in Japan. A phytoplankton community was cultivated in a 300 L, open-surface tank under the sun with phosphorus and nitrogen salts (2.0 mgN/L and 0.2 mgP/L, respectively) (Figure 5.3). Two types of bacteria-containing sample were used for spiking the test pond water in the tank: actual grey water taken from Ritsumeikan University campus and a portion of liquid media containing coliforms cultured from domestic wastewater, including toilet wastewater.

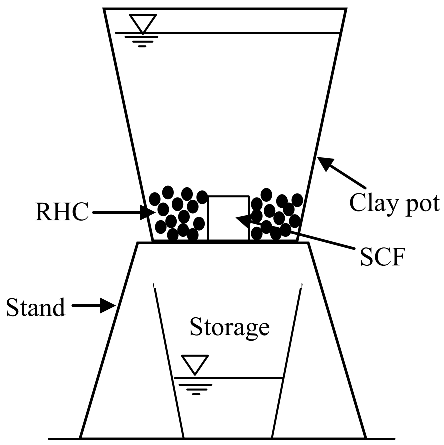
Fifteen litres of the bacteria-spiked test pond water was poured into a B-filter, and the filtrate during 60 min was taken for coliform measurement. Three B-filter units were used separately in the test. Figure 5.3 showed the laboratory experiment set-up. After one day filtration, the filtered water in the bucket was sampled for water quality measurement. Then the filter was washed and next the same bacteria-spiked test pond water was poured into the filter and the filtered water was taken again. The same filtration was performed on the third and fourth days. TC and E. coli in the test pond water and the filtered water of the four days were measured by the membrane filtering method using enzymatic substrate media XM-G (Nissui Pharmaceutical). The pH, DO, transparency, turbidity and SS of filtered sample was measured using HORIBA D-54SE, HACH HQ 30D and JIS K0102 (Japanese standard association, 2013).



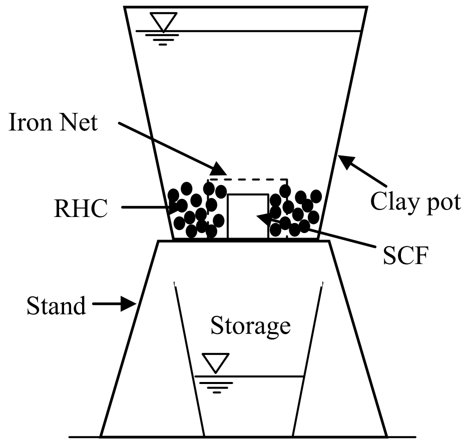
(a) B-filter



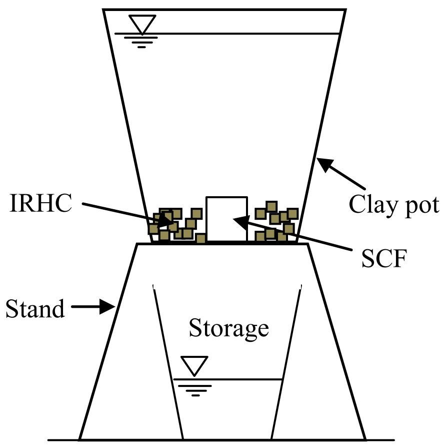
(b) I-filter



(c) R-filter



(d) C-filter



(e) H-filter

(f) Actual arrangement

Figure 5.2 – Schematic diagram of different filter units (Rahman and Nakajima, 2015)

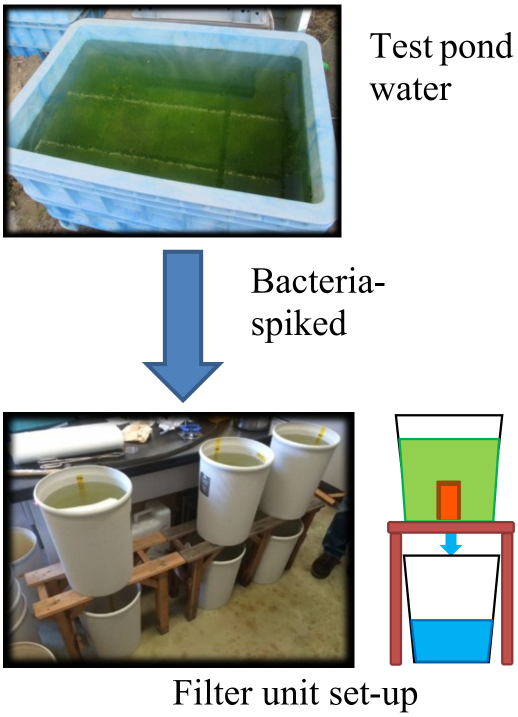


Figure 5.3 – Laboratory experiment set-up

5.2.3 Field test

5.2.3.1 Wet season test

The test was conducted using pond water in urban and rural areas near Khulna City, Bangladesh (Rupsha, Fakirhat and Mollahat Villages and Khulna City), where there are many wetlands and artificial ponds. The characteristics and water qualities of the ponds were described in a previous paper (Rahman et al. accepted). The village people use these ponds for many purposes, and the ponds were categorized into rural domestic use (denoted as D), wastewater storage use (denoted as W), agriculture use (denoted as A), fisheries use (denoted as F) and urban domestic use (denoted as U) in the previous paper21). Four filter units (B-filter, I-filter, R-filter and C-filter) were prepared for each category of pond (D, W, A, F and U; total 20 filters). The filters were placed in a laboratory at Khulna University of Engineering and Technology, Bangladesh.

Two ponds were selected from each pond category, and they were denoted as D-1, D-2, W-1, W-2, A-1, A-2, F-1, F-2, U-1 and U-2 in September, 2013 (wet season). The pond water (60 L) of D-1, W-1, A-1, F-1 and U-1 was collected in the morning for three consecutive days (from the first day to the fourth day). On the first afternoon, a portion of the collected pond water (15 L) was poured into the 4 types of filter units (B-filter, I-filter, R-filter and C-filter), and the filtered water was collected in 4 buckets. On the afternoon of the second day, the filtered water in the buckets was discarded and new pond water was poured into the filter units without discarding the remaining water until filling the units. A portion of the pond water was sampled for measurement. On the morning of the third day, the filtered water in the bucket was sampled for measurement, and in the afternoon, the bucket water was discarded, and the new pond water was poured into the filter following the same procedure as the second day. The pond water was also sampled for measurement. On the fourth morning, the filtered water was sampled for measurement.

The remaining water in the filter units and the buckets was then discarded, and they were washed with tap water. The same tests using the pond water of D-2, W-2, A-2, F-2 and U-2 were conducted on that day (the fourth day) and the following three days (the fifth day – the seventh day), and water samples were collected. Therefore, duplicate samples (taken on the third and fourth days for D-1, W-1, A-1, F-1 and U-1 and on the sixth and seventh days for D-2, W-2, A-2, F-2 and U-2) were obtained for the 10 pond waters and the filtered water using 4 types of filter units.

5.2.3.2 Dry season test

In February and March, 2014 (dry season) the similar test was carried out using the four ponds (D-1, D-2, U-1 and U-2) water and 3 types of filter units (B-filter, I-filter and H-filter). Duplicate samples were also obtained for the 4 ponds water and the filtered water using 3 type filter units. As a leakage of the filter was found during the test of U-1 using B-filter, the sample water of the test was not obtained. Then a new B-filter was prepared for the test of U-2 using B-filter.

5.2.4 Flux rate and water quality measurement

The pond water amount poured into the filter unit, the pond water remaining in the filter unit and the filtrate water amount were measured. The filtered water amount during 19 hours from the previous afternoon to the second, third and fourth mornings was converted to 24 hours and divided by the surface area of the ceramic filter to estimate the flux rate (m/day). Temperature, pH, DO, turbidity and SS were measured in Bangladesh using HORIBA D-54SE, HACH HQ 30D and JIS K0102 (Japanese standard association, 2013). TC and E.coli were also measured in Bangladesh using the same method as the laboratory test. Two hundred fifty milliliters of pond water and filtrate water samples was transferred into plastic bottles and stored in a refrigerator. The water samples were brought to Japan without acid addition during one day transportation. In Japan, the sample water was filtered through No. 5C filter paper (Advantec) and the dissolved organic carbon (DOC), dissolved nitrogen (D-N), dissolved phosphorus (D-P) and alkalinity were determined by SHIMADZU TOC-V WP and JIS K0102 (Japanese standard association, 2013).

5.2.5 Data analysis

Statistical data analysis for significant differences between the sample groups was conducted by non-parametric method (Wilcoxon Rank-Sum test) for the field test results because the data were not normally distributed. However, for the laboratory test results, the t-test was used because the data had a normal distribution.

5.3 Results and discussion

5.3.1 Laboratory test

Table 5.1 shows the average and standard deviations of the test pond water and the filtered water quality of a B-filter for the laboratory test pond (average and standard deviation of the four days measurements). The water filtered using three different B-filter units was denoted as LB-1, LB-2 and LB-3 in the table. The pH and DO of the B-filter was almost similar to test pond water indicating B-filter had no effect on pH and DO concentration. The transparency was greater than 50 cm, turbidity was less than 0.1 NTU, and SS was less than 5 mg/L after filtration, which suggested a good filtration performance to separate particulate matter from the test pond water. No significance ($p > 0.05$) difference was found between LB-1, LB-2 and LB-3 filter units for transparency, turbidity and SS suggesting that filter performance to separate particulate matter was similar for each filter unit.

Table 5.1 – Averages and standard deviation of the test pond water and the filtered water quality of a B-filter for the laboratory test pond.

Type	Unit	Grey water spiked				Cultured coliform spiked			
		Test pond water	LB-1	LB-2	LB-3	Test pond water	LB-1	LB-2	LB-3
		Avg±SD	Avg±SD	Avg±SD	Avg±SD	Avg±SD	Avg±SD	Avg±SD	Avg±SD
pH		8.4±0.3	8.4±0.1	8.4±0.1	8.4±0.1	8.7±0.4	8.4±0.3	8.5±0.2	8.5±0.2
DO	mg/L	8.7±1.4	8.2±1.4	8.1±1.2	8.1±1.1	8.3±1.2	7.7±1.0	7.6±0.9	7.6±0.9
Transperancy	cm	22±2.8	>50	>50	>50	12±2.5	>50	>50	>50
Turbidity	NTU	8.2±1.1	<0.1	<0.1	<0.1	14±3.9	0.1	0.2	0.1±0.1
SS	mg/L	33±9.2	<5	<5	<5	36±8.9	<5	<5	<5

Table 5.2 shows the TC and E. coli concentrations of the test ponds and the water filtered by a B-filter in the laboratory test (average and standard deviation of the four days measurements). The average removal is also shown in Table 5.2 using a log scale. The deviations of the log removal among the three B-filter units were small in all tests, indicating good repeatability and reliability of the obtained log removal values. In the grey water-spiked test pond water, the average log removals were approximately 2 log or more for TC and E.coli. In the cultured coliforms-spiked test pond water, the average log removals were approximately 3 log of more for TC and E.coli.

Table 5.2 – TC and E. coli concentrations of the test pond and the filtered water by a B-filter in the laboratory test (Rahman and Nakajima, 2015)

	Grey water spiked		Cultured coliforms spiked	
	TC (CFU/100 mL) (Avg±SD)	Average removal (log scale)	TC (CFU/100 mL) (Avg±SD)	Average removal (log scale)
Test pond water	29,000±22,000		280,000±120,000	
LB-1	540±310	1.7	5,400±8,400	2.2
LB-2	190±160	2.3	4,100±6,560	2.8
LB-3	440±510	2.0	1,300±1,300	2.7

	Grey water spiked		Cultured coliforms spiked	
	E.coli (CFU/100 mL) (Avg±SD)	Average removal (log scale)	*E.coli* (CFU/100 mL) (Avg±SD)	Average removal (log scale)
Test pond water	68±3.0		67,000±32,000	
LB-1	<1.0	>2.0	66±51	3.1
LB-2	<1.0	>2.0	34±37	3.4
LB-3	0.3±0.3	>2.0	150±280	3.4

The average log removal of E.coli was significantly larger than the average log removal of TC in the cultured coliforms spiked test pond water ($p<0.05$). The average log removal of TC was larger in the cultured coliforms-spiked test pond water than the grey water-spiked test pond water ($p<0.05$). The log removals of TC and E.coli were 2 – 3 log or more, although small variation was found in the different samples.

TC is removed by filtration, and the TC removal by depth filtration and microfiltration is 0 – 1.0 log and 2 - >4 log, respectively (Asano *et al.*, 2007; American Water Works Association, 2005; LeChevallier and Au, 2004 and Viessman and Hammer, 1993). Therefore, the TC and E. coli removal using a B-filter unit suggested higher removal performance than the depth filters and almost the same as the microfiltration. The pore size of SCF used in the B-filter unit was 1 – 5 µm (Shafiquzzaman *et al.*, 2011a), which was a little larger than the pore size of a microfilter. The removal performance of the B-filter unit in the laboratory test (2 – 3 log) showed feasibility of its application to pond water purification in Bangladesh.

5.3.2 Field test during wet season

5.3.2.1 Pond water quality

The temperature of the pond water was approximately 30^{o}C. The average of the duplicate measurements for pH, DO, turbidity, SS, DOC, D-N, D-P and alkalinity of the surveyed ponds are shown in Table 5.3. The pH had a positive correlation with DO (r = 0.956) and was high in the F and U ponds. The turbidity and SS were high in the ponds with high pH, which indicated that the pond water quality was strongly influenced by photosynthesis and primary production, causing the water colour to be greenish. The main SS in the D and U ponds was algae, whereas some solid waste contaminated the W ponds. The water quality of the A ponds was the clearest. The DOC was between 5 and 15 mg/L in all ponds. D-N was larger in categories D, W and U (except W-2), and D-P was also larger in categories D, W and U and lower in category F ponds. These results were the same as the previous survey (Rahman *et al.*, 2015).

The average pond water TC and E.coli concentrations during the wet season are shown in Table 5.4, which shows that the TC values were similar (n x 10^{3} CFU /100 mL), except A-1 (smaller) and W-1 (larger). The E.coli concentrations were mostly less than 300 CFU/100 mL, except in A-2, W-1 and U-2 (larger than 900 CFU/100 mL). No E.coli was detected in pond A-1. The TC concentration was higher than E. coli, and E. coli/TC was less than 0.1, except in A-2 (0.17) and U-2 (0.29). D ponds and U ponds were used for domestic activities. According to the Standard for Inland Surface Water in Bangladesh, TC should be less than 5000 CFU/100 mL for use as a source of drinking water after conventional treatment (Department of Environment, Ministry of Environment and Forests, Bangladesh, 1997). Although the TC concentrations of D-1 and D-2 almost satisfied the standard value, the TC concentrations of U-1 and U-2 were above the standard.

Table 5.3 – Averages of pond water and filtrate water quality of different filter units for different ponds during wet season (Rahman and Nakajima, 2015)

	Unit	D-1					D-2				
		Pond water	B	I	R	C	Pond water	B	I	R	C
pH		7.8	8.3	8.1	8.3	8.3	7.9	8.4	8.2	8.3	8.3
DO	mg/L	6.0	7.2	6.9	7.2	6.8	6.6	7.4	7.3	7.6	7.3
Turbidity	NTU	3.3	<0.1	7.9	<0.1	<0.1	36	<0.1	13.5	<0.1	0.2
SS	mg/L	7.3	< 5	<5	<5	<5	21	< 5	< 5	< 5	< 5
DOC	mg/L	15	15	12	14	12	9.1	8.9	7.2	8.7	7.6
D-N	mg/L	3.7	2.9	2.5	2.7	2.5	1.5	1.8	1.4	1.5	2.2
D-P	mg/L	0.96	1.06	0.04	1.40	0.32	0.13	0.47	0.04	0.94	0.18
Alkalinity	mg/L	170	180	140	180	160	160	180	130	170	190

	Unit	A-1					A-2				
		Pond water	B	I	R	C	Pond water	B	I	R	C
pH		8.1	8.4	8.6	8.7	8.2	7.6	8.2	8.3	8.6	8.3
DO	mg/L	7.0	7.3	7.1	7.3	5.8	5.8	7.5	7.4	7.7	6.5
Turbidity	NTU	9.3	<0.1	<0.1	<0.1	<0.1	17	<0.1	0.4	<0.1	<0.1
SS	mg/L	5.9	< 5	< 5	< 5	< 5	11	< 5	<5	<5	<5
DOC	mg/L	6.6	6.3	5.1	5.9	6.9	7.8	7.7	5.6	7.7	7.1
D-N	mg/L	0.8	0.8	0.6	0.7	1.1	1.1	1.0	0.8	0.9	1.7
D-P	mg/L	0.02	0.26	0.17	0.20	1.12	0.13	0.68	0.16	0.68	0.98
Alkalinity	mg/L	140	140	130	150	120	120	140	140	170	140

	Unit	F-1					F-2				
		Pond water	B	I	R	C	Pond water	B	I	R	C
pH		8.7	8.8	8.5	8.7	8.6	7.9	8.8	8.7	8.2	8.4
DO	mg/L	9.1	7.3	7.2	7.3	7.1	7.1	7.5	7.6	7.5	7.4
Turbidity	NTU	36	0.4	<0.1	0.1	<0.1	25	<0.1	<0.1	<0.1	<0.1
SS	mg/L	39	< 5	< 5	< 5	< 5	20	< 5	< 5	< 5	< 5
DOC	mg/L	12	11.4	9.2	11.7	10.0	5.5	5.8	4.7	8.0	5.9
D-N	mg/L	1.2	1.1	1.0	1.2	1.1	0.9	1.0	0.6	1.4	0.9
D-P	mg/L	0.02	0.14	0.10	0.53	0.59	0.05	0.54	0.08	-	1.49
Alkalinity	mg/L	110	130	80	130	110	80	100	80	120	90

	Unit	W-1					W-2				
		Pond water	B	I	R	C	Pond water	B	I	R	C
pH		7.9	8.8	8.5	8.7	8.5	8.3	8.7	8.6	8.6	8.5
DO	mg/L	6.5	7.2	6.8	7.2	6.5	7.8	7.5	7.5	7.6	7.0
Turbidity	NTU	48	<0.1	<0.1	<0.1	<0.1	9	<0.1	<0.1	<0.1	<0.1
SS	mg/L	78	< 5	<5	<5	<5	13	< 5	< 5	< 5	< 5
DOC	mg/L	9.0	8.3	6.4	8.1	7.9	5.7	5.3	4.8	5.9	5.4
D-N	mg/L	2.2	1.3	1.1	1.2	1.3	0.7	1.0	0.7	1.2	1.0
D-P	mg/L	1.23	0.86	0.15	1.47	1.68	0.07	0.37	0.12	0.82	1.49
Alkalinity	mg/L	160	200	160	200	170	170	200	150	200	170

	Unit	U-1					U-2				
		Pond water	B	I	R	C	Pond water	B	I	R	C
pH		8.5	8.7	8.8	8.8	8.3	8.4	8.7	8.8	8.8	8.5
DO	mg/L	9.8	7.2	7.0	7.2	6.3	8.7	7.5	7.6	7.6	7.1
Turbidity	NTU	17	0.1	<0.1	<0.1	<0.1	20	<0.1	<0.1	<0.1	<0.1
SS	mg/L	31	< 5	< 5	< 5	< 5	22	< 5	<5	<5	<5
DOC	mg/L	9.8	8.7	6.7	8.1	8.7	7.8	7.4	5.9	7.2	7.2
D-N	mg/L	2.2	1.6	1.1	1.4	1.6	1.7	1.6	1.2	1.4	1.5
D-P	mg/L	0.35	0.38	0.31	1.13	3.24	0.30	0.48	0.46	1.70	4.86
Alkalinity	mg/L	190	210	170	320	170	200	230	190	230	210

Table 5.4 – Average pond water TC and *E.coli* concentration and average log removal of TC and *E.coli* from filtrate water of different filter unit during wet season (Rahman and Nakajima, 2015)

	Pond water TC (CFU/100mL)	TC removal (log scale)				Pond water *E.coli* (CFU/100mL)	*E.coli* removal (log scale)			
		B	I	R	C		B	I	R	C
D-1	4300	3.1	2.1	2.1	3.1	290	1.7	2.7	>3.0	>3.0
D-2	4100	1.1	3.8	1.5	2.0	230	2.3	>3.0	1.9	>3.0
A-1	790	0.8	1.0	1.2	0.4	< 75	-	-	-	-
A-2	5600	2.3	3.5	2.2	2.5	950	3.2	>3.0	1.6	>3.0
F-1	4500	2.1	0.1	1.9	0.6	53	1.6	>3.0	1.0	1.2
F-2	3700	2.4	2.7	1.2	1.8	310	1.3	>3.0	1.8	>3.0
W-1	16000	1.7	0.4	1.4	0.4	1000	1.0	0.9	1.4	0.9
W-2	1900	1.3	<0.1	0.9	<0.1	150	0.8	0.6	0.9	0.5
U-1	6000	1.3	1.0	0.4	<0.1	210	1.0	2.2	0.9	0.5
U-2	5800	1.9	1.2	1.2	<0.1	1700	2.3	1.7	1.8	1.3

5.3.2.2 Filtration performance

The average of the duplicate measurements for pH, DO, turbidity, SS, DOC, D-N, D-P and alkalinity of the filtrated water of the B-filter unit are also shown in Table 5.3. The turbidity was less than 0.5 NTU (< 0.1 NTU except two cases), and SS was less than 5 mg/L after filtration, which suggested a good filtration performance to separate particulate matter (mainly algae) from the raw pond water. The removal of turbidity and SS were more than 99 % and 96 %, respectively and they were satisfied with Bangladesh standards for drinking water (10 NTU and 10 mg/L respectively (Department of Environment, Ministry of Environment and Forests, Bangladesh, 1997). These removals were higher than another household type filter named Biosand filters (BSF) using sand in Haiti (Sisson *et al.*, 2013) and in Nepal (Lee, 201) and similar to a household ceramic filter named Terafil Water Filter (www.scribd.com).

In contrast, DOC and D-N were not altered by filtration using a B-filter unit ($p > 0.05$). The filter cannot remove dissolved matter. However, D-P was slightly increased by filtration, although the difference was not significant ($p > 0.05$) between pond water and the B-filter water. The detected increase of D-P was caused by release of phosphate from the filter. Because rice bran containing phosphorus was used in the process of the filter manufacturing, phosphate salts remained in the filter. These phosphate salts will be removed by complete dissolution by water.

The averages of the duplicate measurements of the log removal for TC and E.coli by the B-filter unit during the wet season are shown in Table 5.4. The log removal was not described for E. coli in A-1 because no E .coli was detected in the pond water. The removals were 1 log to 2 log or higher (except A-1 for TC and W-2 for E.coli), which was lower than the log removal obtained in the laboratory test. However, the difference was not large. It was therefore confirmed that 1 log to >2 log removal of TC and E. coli could be obtained by the B-filter applied actual pond water. The removal was obtained by physical separation.

Table 5.5 shows a summary of the bacteria removal performance of different filters reported by other researchers. By comparing Table 5.4 and Table 5.5, it was seen that the log removal of TC by the B-filter unit from all ponds was higher than the PSF used in Bangladesh, a white clay filter without silver colloidal (candle filter), the Hong Phuc® candle filter, and a red clay disk filter without colloidal silver and showed similar result to the Bio Sand Filter (BSF), Ceradyn filter (candle filter), Gravidyn filter (candle filter), white clay disk filter without colloidal silver, red clay disk filter with colloidal silver, black clay disk filter without colloidal silver, and black clay disk filter with colloidal silver used in Nepal (except A-1 ponds). The TC removal performance of the B-filter was little lower compared to the white clay disk filter with colloidal silver used in Nepal and the TERAFIL water filter used in India. The E.coli log removal by the B-filter also had higher removal performance than the Ag^{+} wet chemical impregnated ceramic filter, Ag_2O wet chemical impregnated ceramic filter, the BSF filter used in Haiti, the PSF used in Bangladesh, the white clay filter without silver colloidal (candle filter), and the red clay disk filter without colloidal silver used in Nepal and showed similar results with other filters (except W-2 ponds), as shown in Table 5.5. These comparisons indicated that the B-filter unit could remove TC and E.coli to the same or higher level and could be used as an alternative filter to reduce the bacteria contamination risk from pond water in Bangladesh.

5.3.2.3 Effect of the additional devices

The average of the duplicate measurements for pH, DO, turbidity, SS, DOC, D-N, D-P and alkalinity of the filtrated water of different filters (I, R and C) are also shown in Table 5.3. The turbidity was less than 0.1 NTU, and SS was less than 5 mg/L after filtration by R-filter units, similar to the B-filter. However, for the I-filter units, the turbidity was 7.9, 13.5 and 0.4 for D-1, D-2 and A-2, respectively. Moreover, the turbidity of the D-2 filtrate using the C-filter was 0.2. These results indicate that the iron net sometimes increased the values due to the iron hydroxide colloid produced by oxidation after filtration as the ferrous ion dissolved from the added iron net.

The filtrate DOC using the R-filter was similar to that using the B-filter, although the DOC using I- and C-filters showed slightly lower concentrations. However, the average DOC in the filtrate using the I-, R- and C-filters was not significantly different from the B-filter filtrate ($p > 0.05$), which suggested that the iron net and/or RHC addition to the filter did not affect the DOC removal.

A clear difference of the filtrate D-N was not found between B-filter and the filters with added devices. The average filtrate D-N using the I-, R- and C-filters was not significantly different from the B-filter ($p > 0.05$). Therefore, the D-N removal in the filtration was not affected by the iron net or RHC addition.

Table 5.5 – Summary of bacteria removal test by different filters (Rahman and Nakajima, 2015)

Filter name	Bacterial type	Removal (log scale)	Study Location	References
Ceramic filter (burned in reductive atmosphere and 5 cm ceramic gravel layer)	*E. coli*	1.5-4	Haiti	Guerrero-Latorre, *et al.* (2015)
Ag^+ sputter coated ceramic filter	*E. coli*	1.5*	-	Simonis *et al.* (2014)
Ag^+ wet chemical impregnated ceramic filter	*E. coli*	0.3*		
Ag_2O wet chemical impregnated ceramic filter	*E. coli*	0.1*		
BSF Filters	*E. coli*	1.1*	Haiti	Sisson *et al.,* (2013)
Existing PSF (6 nos)	TC	< 0.6*	Bangladesh	Hasan *et al.* (2013)
	E. coli	0.1-0.7*		
Ceramic water filter	*E. coli*	2.2-3.8		Bielefedt *et al.* (2009)
Ceramic filter without colloidal-silver-impregnated	*E. coli*	1.7-3.5*	Mexico	Oyanedel-Carver and Smith (2008)
Ceramic water purifier (3nos)	*E. coli*	Around 2.0	Cambodia	Brown (2007)
Multi-stage filtration unit	*E. coli*	1.6 - >2.7	Bangladesh	APSU (2006)
BSF (5 nos)	TC	2.3*	Nepal	Lee (2001)
Ceradyn filter (candle filter)	TC	2.0	Nepal	Dies (2003)
	E. coli	1.8		
Gravidyn filter (candle filter)	TC	2.0		
	E. coli	1.8		
0White clay filter without silver colloidal (candle filter)	TC	0.8		
	E. coli	0.9		
White clay filter with silver colloidal (candle filter)	TC	2.0		
	E. coli	1.8		
Hong Phuc® candle filter	TC	0.8		
	E. coli	1.7		
White clay disk filter without colloidal silver	TC	2.3		
	E. coli	2.5		
White clay disk filter with colloidal silver	TC	3.5		
	E. coli	3.1		
Red clay disk filter without colloidal silver	TC	0.7		
	E. coli	0.7		
Red clay disk filter with colloidal silver	TC	2.6		
	E. coli	2.9		
Black clay disk filter with colloidal silver	TC	2.5		
	E. coli	2.8		
Black clay disk filter without colloidal silver	TC	2.3		
	E. coli	2.5		
TERAFIL water filter	TC	3.2-3.7*	India	www.scribd.com

*Calculated from the percentage removal data

D-P was increased in some cases after B-filter filtration because of phosphate dissolution from the rice bran in the filter. Because some contamination was found in the filtrate D-P of the F-2 pond using the R-filter, the D-P data of the sample were deleted, as shown in Table 5.3. The average and standard deviation of D-P in the pond water and the filtrates using different types of filter are shown in Figure 5.4. The average D-P after I-, R- and C-filter filtration was significantly lower, higher and higher than the B-filtrate, respectively ($p < 0.01$, $p < 0.05$ and $p < 0.05$, respectively). These results suggested a decrease of D-P by iron net addition and an increase of D-P by rice husk charcoal addition. In the application of SCF with an iron net for arsenic removal in previous research, ferric iron was produced from ferrous iron that was dissolved from the iron net, which worked to coagulate arsenic and phosphorus (Shafiquzzaman *et al.*, 2011). Therefore, the D-P decrease by the iron net found in this study was likely caused by the same mechanism, coagulation of phosphate by ferric iron (iron hydroxide) floc that was separated by the filter. The D-P increase by RHC addition was caused by phosphate release from the RHC because it contained high phosphate contents (1.8 % in the used RHC). The average D-P in the R-filtrate and C-filtrate was not significantly different ($p > 0.05$). Therefore, the iron net addition had less of an effect than RHC. Moreover, some RHC floated on the water surface, and powdered RHC made the mixed liquor colour blackish in the R-filter and C-filter, which caused difficulties with the operation of the filter. To mitigate these difficulties, RHC was immobilized by burning with clay (IRHC), as described in the methodology section.

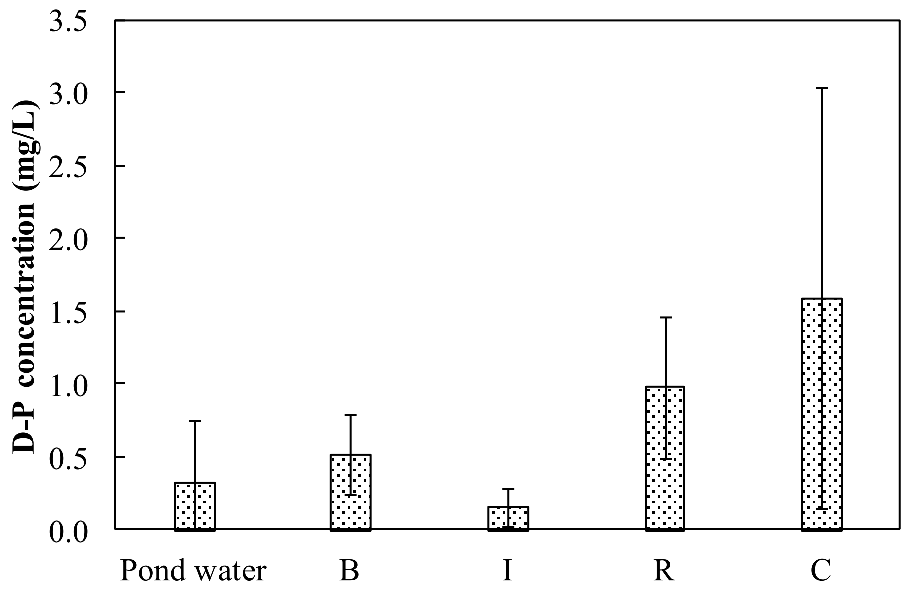


Figure 5.4 – The average and standard deviation of D-P in the ponds water and the filtrates using different types of the filter in the wet season test (Rahman and Nakajima, 2015)

The log removal of TC and E.coli by the I-filter, R-filter and C-filter was mostly approximately 2 log or greater, although the values varied from <0.1 to >3.0 (Table 5.4). No significant difference ($p > 0.05$) was found among the log removals of TC and E.coli by the B-filter, I-filter, R-filter and C-filter units, which indicated that the iron net and RHC additions had no effect on TC and E.coli removal by the filters. Bacterial coagulation by iron floc and adsorption onto the charcoal was not significant, and their removal was achieved mainly by physical separation.

5.3.2.4 Flux rate

The average and standard deviation of the flux rate of the B-filter on the second day (the first measurement of the flux rate) are shown in Table 5.6. The flux rate varied with pond category: higher in D and A ponds and lower in F, W and U ponds. The turbidity and SS of the pond water that was resistant to filtration were low in the former ponds and high in the latter ponds. Figure 5.5 shows the relationship between 1/SS of the pond water and the flux rate in the ponds, except W. The SS in the W ponds contained SS caused by excreta and was different from the SS in the other ponds that mainly consisted of algae. The good linear correlation suggested that the increase of SS deteriorated the filtration performance. The flux rate measured on the third and the fourth days was almost the same as the second day, although a slight decrease was found in some cases in the fourth day measurement. The filtration performance of the filter could be improved by scrubbing the SCF with a soft brush or cloth when the flow rate reduced to unsatisfactory level as mentioned in the previous paper (Shafiquzzaman *et al.*, 2011). The filter could be also replaced by new one in the strong clogging cases because its cost is inexpensive (Shafiquzzaman *et al.*, 2011).

The flux rates of the I-, R- and C-filters were also measured. The flux rate of the I-, R- and C-filters was not significantly different from the B-filter by ($p > 0.05$). Therefore, the iron net and RHC additions had no effect on the filtration flux.

5.3.3 Field test during dry season

The average pH, DO, turbidity, SS, DOC, D-N, D-P and alkalinity of the surveyed ponds (D and U ponds) during the dry season and the filtrates are shown in Table 5.7, and the average pond water TC and E.coli concentrations and log removal by filtration are shown in Table 5.8. Few differences in the pond water quality from the wet season were found. An increase in turbidity, SS, TC and E.coli was observed in the U ponds. The increase of turbidity and SS were caused by algal growth, which made the water colour greenish. The water temperature of all the ponds was decreased in the dry season to approximately 25^{o}C which was 5^{o}C lower than in the wet season.

The water quality of the B-filter filtrate was similar to that of the wet season, as shown in Table 5.7. The log removal of TC and E.coli was maintained approximately 1.5 log and showed no significant difference ($p > 0.05$) from the wet season results, although it showed a slight decrease. It was therefore confirmed that the B-filter could be applied to actual pond water during both seasons.

Table 5.6 – The average and standard deviation of flux rate of B-filter at the second day (the first measurement of the flux rate) for different pond categories (Rahman and Nakajima, 2015)

Pond Category	Flux rate (m/d) (Avg±SD)
D	0.35±0.09
A	0.36±0.09
F	0.10±0.04
W	0.17±0.05
U	0.12±0.07

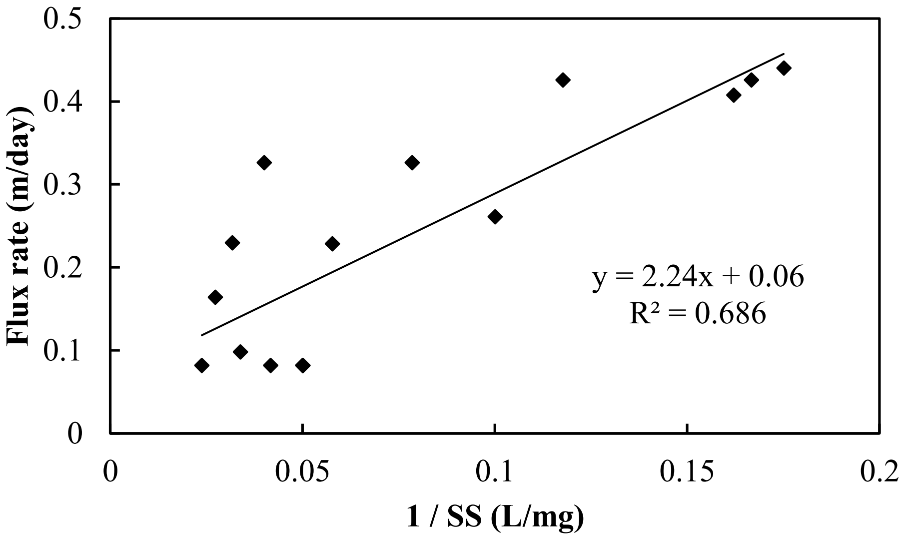


Figure 5.5 – Relationship between 1/SS of the pond water and the flux rate (except W ponds) (Rahman and Nakajima, 2015)

Iron net addition also caused a D-P decrease in the D ponds, similar to the wet season result, as shown in Table 5.7, whereas in the U ponds, D-P slightly increased in the filtrate of the I-filter compared to the B-filter due to an unknown reason. IRHC was added to the H-filter during the dry season instead of the RHC added to the R-filter during the wet season. The RHC floating and powdering observed in the R-filter were prevented by using IRHC in the H-filter. D-P in the filtrates of the B-filter and R-filter in the wet season and that of the B-filter and H-filter are shown in Figure 5.6. Although D-P increased in the H-filtrate compared to the B-filtrate, the increase was drastically mitigated from the D-P increase by the R-filter in the wet season. Therefore, immobilization of RHC successfully decreased floating, powdering and D-P release of RHC. The flux rate was similar to that in the wet season in the D and U ponds.

From Table 5.9 it was seen that the manufacturing cost of B-filter is low compared to the other filters and replacement element cost was also lower than other suggested that B-filter will be more affordable by the rural people of Bangladesh.

Table 5.7 – Averages of pond water and filtrate water quality of different filter units for different ponds during dry season (Rahman and Nakajima, 2015)

	Unit	D-1				D-2			
		Pond water	B	I	H	Pond water	B	I	H
pH		7.8	8.4	8.1	8.1	8.0	8.5	8.3	8.3
DO	mg/L	6.5	8.1	8.0	7.9	7.1	8.3	8.2	8.2
Turbidity	NTU	9.5	0.9	0.7	1	35	0.8	0.5	0.8
SS	mg/L	8	< 5	< 5	< 5	48	< 5	7.9	< 5
DOC	mg/L	18	18	17	17	12	11	11	11
D-N	mg/L	3.6	2.8	2.4	2.9	2.7	1.6	1.4	1.6
D-P	mg/L	1.44	1.29	0.12	1.49	0.58	0.58	0.16	0.66
Alkalinity	mg/L	260	290	260	280	290	310	270	290

	Unit	U-1				U-2			
		Pond water	B	I	H	Pond water	B	I	H
pH		8.1	-	8.6	8.5	8.5	8.6	8.5	8.6
DO	mg/L	6.3	-	7.9	8.7	9.8	8.2	8.2	8.1
Turbidity	NTU	33	-	1.3	0.9	93	2.6	0.7	0.8
SS	mg/L	63	-	< 5	< 5	93	< 5	< 5	< 5
DOC	mg/L	13	-	11	11	15	11	10	11
D-N	mg/L	2.9	-	1.5	1.9	2.4	2.0	1.5	1.8
D-P	mg/L	0.26	-	0.29	0.28	0.30	0.22	0.47	0.44
Alkalinity	mg/L	260	-	230	230	310	280	290	310

Table 5.8 – Average pond water TC and *E.coli* concentration and average log removal of TC and *E.coli* from filtrate water of different filter unit during dry season

	Pond water TC (CFU/100mL)	TC Removal (log scale)			Pond water *E.coli* (CFU/100mL)	*E.coli* removal (log scale)		
		B	I	H		B	I	H
D-1	12000	0.2	0.9	<0.1	300	1.5	1.7	1.6
D-2	4800	1.2	1.4	1.2	306	1.5	2.1	1.4
U-1	360000	-	1.2	2.5	4420	-	2.5	2.5
U-2	12100	1.5	1.7	2.0	3250	1.3	2.0	2.6

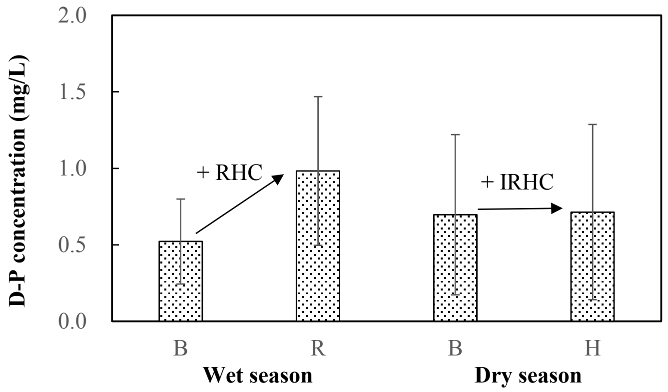


Figure 5.6 – Comparison of D-P concentration (average and standard deviation) increase by addition of RHC (wet season) and IRHC (dry season) (Rahman and Nakajima, 2015)

Table 5.9 – Comparison of different water filter manufacture cost and replacement element cost

Filter name	Manufacturing cost ($)	Replacement element cost ($)	Study location	Reference
B-filter	3.0-4.0*	0.2-0.3	Bangladesh	Shafiquzzaman *et al.*, (2011a)
Ceramic water purifier	<10	2.5-5.0	Cambodia	Brown (2007)
Silver impregnated ceramic	4.0	-	-	http://potterswithoutborders.com
Sediment filter with iron	100-1000	-		
Activated carbon filter	15-50	-		
TERAFIL filter	3.5	0.49	Nepal	Dies (2003)
Potters for peace (PFP)	9.0	-		
Indian candle filter	8.0-21	-		
Nepal candle filter	2.3-4.1	0.46		
Swiss Katadyn® Drip Filters	160-190	-		
Hong Phuc® Candle Filters	7.5	-		
PSF filter	600	-	Bangladesh	Jakariya *et al.* (2005)
Safi filter	15	4.0		
Bishuddhya filter	45	-		
Three-pitcher filter	<5.0	-		
Activated alumina filter (ALCAN filter)	52	12		

*Calculated in this study

5.4 Summary

To develop a sustainable purification system for pond water in a rural area of Bangladesh, TC and E.coli removal performance using several filtration units with simple ceramic filters was investigated. The results are as follows:

(1) The laboratory test showed that the log removal of TC and E.coli by B-filter was 2 – 3 log or more, suggesting higher removal performance than depth filters and similar to microfiltration.
(2) Good filtration performance of the B-filter to separate particulate matter (mainly algae) from raw pond water was shown in the wet season survey.
(3) TC and E. coli removal by the B-filter was also confirmed to be 1 log to >2 log for actual pond water in Bangladesh.
(4) The effect of the additional devices was not apparent, but a D-P decrease by iron net addition was found and was caused by coagulation of phosphate by iron hydroxide floc that was separated by the filter.
(5) The flux rate of the filter varied with ponds category and had a correlation with 1/SS, suggesting that increased turbidity and SS by algae deteriorated the filtration performance.
(6) The log removal of TC and E.coli was maintained at approximately 1.5 log and showed no significant difference from the wet season results.

It was therefore confirmed that the B-filter could be applied to actual pond water during both seasons. The B-filter unit is an alternative treatment system for the existing PSF system that is currently in use in the southern region of Bangladesh for treating pond water.

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Chapter 5: Development of a pond water purification assembly using a simple ceramic filter

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Chapter 6

6.1 Conclusions

A water purification unit was developed from natural material and its applicability in pond water treatment was investigated. As a result of developing water purification unit from natural material the performance of using it was evaluated in details and this study describes a promising approach.

In **Chapter 1** the purposes of this research were stated with background. As stated in the background pond water would be alternative safe water source in Bangladesh, although there is not so much information on pond categorization and water quality. However pond water needs to be treated before uses and some of the treatment system already used which had many limitations. Keeping in mind these limitations, especially high cost and bad water quality, using simple ceramic filter (SCF), prepared from natural material, for pond water treatment could be alternative option in Bangladesh. Therefore, pond water categorization, investigation of pond water quality and application of SCF for pond water treatment was proposed. In addition immobilization of rice husk charcoal was also proposed to investigate. The structure of this thesis was also described in this chapter.

Current literature review on natural material used in water treatment in terms of material composition, modification and application in water and wastewater treatment was reviewed in **Chapter 2**. Based on the current knowledge about different natural material and on studies dealing with the modification of the natural material such as immobilization of RHC, was developed and described in **Chapter 4**. Also, the removal of different impurities such as suspended solids and bacteria from pond water by pond water purification unit using natural material was investigated that described in **Chapter 5**.

Chapter 3 described pond water categorization of Bangladesh and its water quality. The 112 surveyed ponds were categorized according to their use into domestic (D), wastewater storage (W), agricultural (A), fisheries (F) and urban domestic (U; divided into two groups named urban domestic large (Ul) and urban domestic small (Us)) ponds.

During the wet season the area of ponds was U l> F > A > (D = Us) > W, volume of the ponds (Ul = F) > (A = D = Us) > W and the depth was approximately 1m for all five categories ponds. The average temperature was found 30^{o}C and the pH values ranged from 7.4 - 7.7 for all five categories ponds. The DO and ORP results showed aerobic condition in all the ponds. The EC in category W ponds was found high due to contamination of toilet waste containing high salinity. The average EC values of category F ponds divide into two groups named fresh water (0.30 – 1.4 mS/cm) and blackish water (2.0 – 5.7 mS/cm). The average transparencies was found low in category F, W and U ponds which seemed to be caused by high algal growth for category F and U ponds and polluted wastewater containing human excreta for category W ponds. The SS and TOC were found higher in category F and U ponds than the others. The T-N and T-P were found higher in category W and U ponds than other ponds, which seemed to be influenced by toilet wastewater that contains high amounts of nitrogen and phosphorus. The T-N and T-P of category F and D ponds were also higher than eutrophication level.

During the dry season the size of all ponds was decreased to less than half of the volume during the wet season. The temperature was found 4^{o}C lower than wet season. The transparency was also decreased to almost half that during the wet season. The pond water became more turbid during the dry season resulting from an increase of primary production. The first component (Z1) of principal component analysis had large contribution compared to others that described eutrophication progress (trophic state). And increase of Z1 during suggested that water quality improvement is necessary especially D and U ponds.

A detail description of developing and manufacturing immobilized rice husk charcoal (IRHC) using Bangladesh clay soil and rice husk charcoal (RHC) manufactured in laboratory design furnace was reported in **Chapter 4**. Bangladesh clay soil and RHC with a ratio of 80:20 (by weight) were used for its manufacturing. The firing temperature of IRHC was 550^{o}C for 90 min using muffle furnace. The size of on IRCH cube was 10x10 cm. The IRHC also successfully manufactured in laboratory design furnace. The IRHC showed decrease in release of dissolved solids (DS), carbohydrate, T-P and PO_4-P compared to RHC that could be more useful for water treatment.

Chapter 5 described application of pond water purification unit using SCF for pond water. The applicability of the pond water purification units to remove suspended solids (SS) and bacteria (total coliforms (TC) and *Escherichia coli* (*E.coli*) had been investigated through laboratory experiment as well as field level experiments in rural area of Bangladesh. The pond water purification units were assembled into five types denoted as B-filter, I-filter, R-filter, C-filter and H-filter according to their structure.

The laboratory test results showed that B-filter which contain only SCF into it, could remove turbidity and SS of <0.1 NTU and <5 mg/L, respectively and showed similar removal performance for different B-filter units. It was also showed that the log removal of TC and *E.coli* by B-filter was 2-3 log or more with good repeatability and reliability. This suggested that by B-filter higher removal performance of bacteria than depth filtration and similar to microfiltration could be possible.

The pond water purification units were also tested using actual pond water in a rural area of Bangladesh for both wet and dry season. After filtration by B-filter, the turbidity and SS was found <0.5 NTU and <5 mg/L, respectively which suggested good filtration to separate particulate matter mainly algae from pond water during wet season. The D-P was increased in some cases after B-filter filtration because of phosphate dissolution from rice bran in the filter. The log removal of TC and *E.coli* by B-filter was confirmed to be 1 log to > 2 log for actual pond water Bangladesh; suggested B-filter could be alternative filter to reduce the bacteria contamination risk from pond water in Bangladesh. The effect of the additional devices was not apparent, but a D-P decrease by iron net addition was found and was caused by coagulation of phosphate by iron hydroxide floc that was separated by the filter and increase by RHC addition was found that caused by high phosphate contents (1.8% in the used RHC) in RHC. The flux rate of the filter varied with ponds category and had a correlation with 1/SS, suggesting that increased turbidity and SS by algae deteriorated the filtration performance. The water quality of the B-filter filtrate was found similar to the wet season and log removal of TC and *E.coli* was maintained at approximately 1.5 log and showed no significant difference from the wet season results. By addition of IRHC instead of RHC during the dry season was drastically mitigated the D-P increase by the R-filter during the wet season.

Based on the above findings the conclusions of this thesis were made as follows:

1. Pond water was successfully categorized and water quality was investigated which could be important information for studying pond water condition in rural area of Bangladesh, suggested pond water (mainly categories D and U ponds) need to be treated especially during the dry season for safe water usage.

2. The pond water purification unit using SCF could efficiently separate SS and bacteria (TC and *E.coli*) from actual pond water in rural area of Bangladesh. This confirmed that, using pond water purification unit could be applied for pond water alternative treatment in rural area of Bangladesh.

3. The IRHC could be successfully developed using Bangladesh clay soil and RHC using both muffle furnace and laboratory designed furnace. Although the IRHC could not show additive performance in the pond water purification unit, it may be used for alternative usage of water and environmental applications.

Adding to such new findings the effective application of the purification unit, the development of facilities that can be widely used by the people and suitable maintenance system must be established in the future.

6.2 Limitations of this study and Scope of further studies

Although the pond purification unit could remove bacteria but the removal performance didn't meet the drinking water standard of Bangladesh. Moreover some important parameter such as filtration capacity, life span was not investigated in this study.

Considering the above limitations several scopes for further studies are described below:

The measures should be taken to increase removal performance of bacteria from filtered water, which could meet the standard. Although silver coat application on the SCF surface would be one option regarding this issue it offers high cost. Therefore alternative options especially use of natural material rather uses of silver are needed to clarify through further studies. Further research should be also carried out regarding the filtration capacity, long-term sustainability and maintenance system of pond water purification unit.

It is also essential to provide the developed pond purification unit to the local and vulnerable people at earliest time to protect their lives. And therefore all necessary steps should be taken to manufacture the pond purification unit commercially. These include finalizing the product design, policy making and collaboration with any marketing company or local NGO, taking permission from Bangladesh government and others.

Studies need to be carried out of the application of IRHC in different water sector. Although the application of IRHC as reluctant for Cr(VI) reduction was also carried out in this study more depth study was needed to find out the performance difference of IRHC with other charcoal. Moreover, heavy metals removal from water and wastewater using the mechanism of adsorption process using IRHC would be it’s another scope of application.

Appendix

Application of IRHC as a reductant to reduce Cr (VI)

A.1 Introduction

Rice husk charcoal (RHC) generally prepared from rice husk that a natural material is found from agricultural waste by-product. It was well known that RHC used as an adsorbent prepared by acid treatment, alkali treatment, carbonization (Daffalla *et al.*, 2010, Ye *et al.*, 2012 and Zhang *et al.*, 2011) for the removal of heavy metal like cadmium, copper, zinc, lead, chromium, mercury (Kumar and Bandyopadhyay, 2006; Ye *et al.*, 2012; Khan *et al.*, 2014; Oda *et al.*, 2013; Bansal *et al.*, 2000 and El-Shafey, 2010) and many other impurities from wastewater (Mohan *et al.*, 2008). RHC also used as a reluctant to reduce Cr (VI) to Cr (III) by ion exchange (El-Shafey, 2005). RHC normally used either as granular type or as powder type. Using RHC in water and wastewater treatment facilities faced some difficulties. The main problem is floating RHC (especially powder type) on the water or wastewater that could not participate fully in the treatment process that leads decrease in treatment efficiency. Another problem is to handling RHC in the maintenance process like removing used RHC for filter unit and installs new RHC. A certain amount of RHC was lost in the handling process. Moreover granular type RHC is become partially powdered in the treatment process. It would be more effective if RHC has suitable handling shape without changing their properties. By immobilizing RHC mixing with clay could be a good option to overcome the floating, broken and handling problem. To solve these problems, immobilized rice husk charcoal was prepared in previous study that described in Chapter 4.

This chapter aims to investigate application of IRHC on Cr (VI) reduction as an example of the usage of the IRHC.

A.2 Materials and methods

A.2.1 Ingredients

Cheap available materials – rice husk and clay soil were used as primary ingredients for manufacturing of IRHC. Rice husk was brought from local market and clay was collected from the paddy field of Khulna city in Bangladesh.

A.2.2 RHC and IRHC preparation

RHC was prepared by carbonizing rice husk in laboratory design furnace to Rahman et al. (2010) in Bangladesh. IRHC was prepared in the same procedure described in Chapter 4 at section 4.2.4.

A.2.3 Ingredients ratio and manufacturing condition

To select the ingredient ratio and manufacturing condition, IRHC cube (10x10 mm) were made manually with different ratios of clay soil and rice bran (90:10, 80:20 and 60:40) on dry weight basis and with different time (30, 60, 90 min) and with different burning temperature (450, 500, 550 and 600 ^{o}C).

A.2.4 Cr (VI) reduction test

A.2.4.1 Preparation and testing of synthetic chromium solution

Aqueous solution of chromium was prepared by dissolving 0.5 g of $K_2Cr_2O_7$ in 1 liter pure water using conical flask to obtained 500 mg/L synthetic solution. The aqueous solution was diluted with pure water to obtain the synthetic chromium solution of desired concentrations. The pH of the solutions was adjusted using different molar concentration of NaOH/HCl using pH meter. The standard solution was prepared from Cr (VI) solution (1001 mg/L) purchased from Wako Pure Chemical Industries, Ltd. The Cr (VI) concentration was determined by spectrophotometer (JIS K0102).

A.2.4.2 Selection test of ingredient ratio, burning time and burning temperature for IRHC

The selection test of ingredient ratio, burning time and burning temperature was done by taking 100 mL Cr (VI) solution in a 200 mL conical beaker and adding 1 IRCH square cube into it. The solution pH was adjusted to pH 1 by pH meter (Horiba 9615, calibrated with buffers of pH 4.01 and 6.82) using different molar concentration of NaOH/HCl solution and ORP was measured by ORP meter (HORIBA D-54SE). The conical beaker was then covered with warping sheet and placed in shaker machine to shake at 100 rpm for 1 hour. After that the solution was filtered through 5 μm filter paper (Advantec 5C). The initial and final Cr (VI) concentration was measured.

A.2.4.3 Reduction experiments

Batch experiments were carried out at various pH (1-5), IRHC dose (1-3 cube), different concentrations (10, 20, 40 and 80 mg/L synthetic solution) and stirring speed (100 rpm) for different contact time. For each batch experiment, 100 mL of synthetic solution was used using 200 mL conical beaker. After adding IRHC cube, pH was adjusted to desired value and ORP was measured using ORP meter (HORIBA). The mixture was than agitated on orbital shaker for desired time at predetermined temperature. After that the mixer was filtrated to separate IRHC from supernatant using 5μm filter paper (Advantec 5C). The residual concentration of chromium in supernatant was determined as stated in Section 4.2.4.1. All experiments were replicated thrice and results were averaged.

The reduction of (R) of chromium was calculated for each run by using Eq. (1):

$$R = \left(\frac{Ci - Ce}{M}\right) x\, 1 \qquad (1)$$

where C_i and C_e were the initial and final concentration of chromium (mg/L) in the test solution, respectively and M is the mass of adsorbent (g) used. The unit of R was mg/g.

A.3 Results and discussion

A.3.1 Ingredients ratio, burning time and burning temperature selection

The weight of 1 IRHC cube was found 0.82±0.21 g. Table 4.1, 4.2 and 4.3 showed the reduction capacity with different rice husk ratios in the ingredient, with different burning temperature and with different burning time. From table 4.1 it was seen that Cr (VI) reduction was high for IRHC cube made with 40 % RHC and low for IRHC cube made with 10% RHC indicate that IRHC with higher percentage (40%) of RHC reduced higher Cr (VI) concentration, while lower percentages (10%) presented a relatively low Cr (VI) reduction. On the other hand it was observed that IRHC with higher percentage (40%) of RHC was broke after agitation while IRHC with lower percentage (10%) of RHC was little broke indicate that higher percentage of RHC was weak in strength and easy broke as same as Hasan (2014) reported that higher percentage of rice bran combined with clay soil in burnt ceramic bar had weak flexural strength. Therefore, 80% soil and 20 % rice rusk was selected for manufacturing IRHC. From Table 4.2 it was seen that at 500 ^{o}C burning temperature of IRHC had higher chromium reduction than 450 and 550 ^{o}C burning temperature and from Table 4.3 it was seen that at 90 min burning time of IRHC had higher chromium reduction than 30 and 60 min burning time. Therefore, 500 ^{o}C burning temperature and 90 min burning time was selected for manufacturing IRHC.

Table A.1 – Relationship of reduction capacity and rice husk ratio

Burning time (min)	Cr (VI) reduction (mg/g) (Avg±SD)
30	0.68±0.07
60	0.67±0.11
90	0.70±0.08

Table A.2 – Relationship of reduction capacity and burning time

Rice husk ratio	Cr (VI) reduction (mg/g) (Avg±SD)	Condition after agitation
40	1.68±0.17	Broke
20	0.70±0.08	Little broke
10	0.57±0.06	Little broke

Table A.3 – Relationship of reduction capacity and burning temperature

Burning temperature (^oC)	Cr (VI) reduction (mg/g) (Avg±SD)
450	0.61±0.13
500	0.70±0.08
550	0.64±0.13

A.3.3 Cr (VI) reduction by IRHC

A.3.3.1 Effect of pH

Figure A.1 showed the Cr (VI) concentration at different pH for different time interval. It was seen that at low pH IRHC was active. The Cr (VI) concentration was decreased with decreasing solution pH. In particular, when the pH was lowered below 3 the reduction of Cr (VI) concentration suddenly increases. Similar results for RHC were gained by several researchers (Shindhu *et al.*, 2012; Guo *et al.*, 2002 and Bishnoi *et al.*, 2004). The improved reduction of Cr (VI) concentration at low pH may be caused by the reduction of Cr (VI) to Cr (III) according to Eqs. (2) and (3) (Sharma and Foster, 1994).

At low pH,

$$Cr_2O_7^{2-} + 14H^+ + 6e^- \longrightarrow 2Cr^{3+} + 7H_2O \qquad (2)$$

At moderate pH,

$$HCr_2O_4^- + 7H^+ + 3e^- \longrightarrow Cr^{3+} + 4H_2O \qquad (3)$$

The presence of large number of H^+ ions at low pH 1-3 may be the reason of high reduction of Cr (VI) concentration. At low pH, the positively charged carbon present in IRHC bind with negatively charged species ($Cr_2O_7^{2-}$/ $HCr_2O_4^-$) due to electrostatic force of attraction (Sindhu *et al.,* 2012).

Figure A.2 shows the pH vs Eh diagram for different pH experiment. It was seen that the final Eh was decreased at pH 1-3. The final Eh was not so changed at pH 4-5. According to pH-Eh diagrams for chromium, the Eh value for pH 1-3 were in the zone of Cr [3+] indicating the favorable condition for reducing Cr (III) from Cr (VI) [www.eosremediation.com]. Thus, by immobilizing RHC to IRHC, the properties of the IRHC in contrast effect of pH were remaining same and reduction of Cr (VI) to Cr (III) at low pH was done. The optimum pH for reduction of Cr (VI) was selected pH 1.

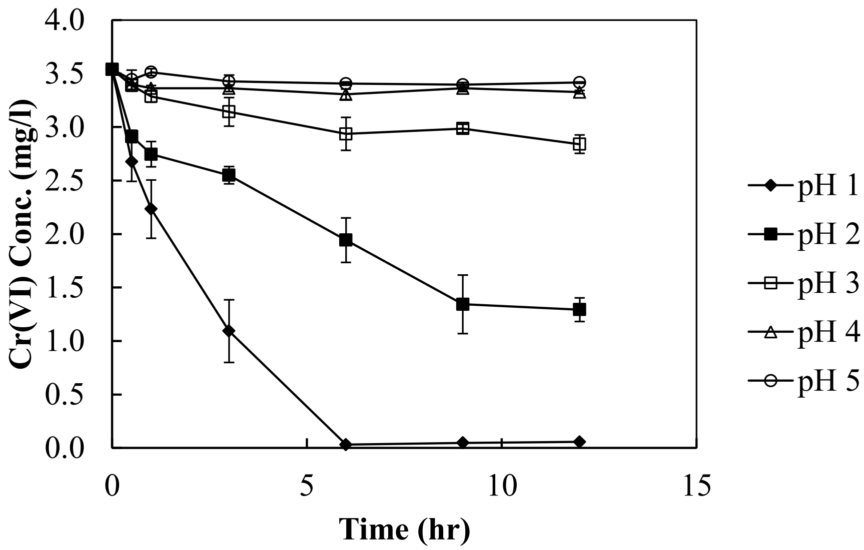

Figure A.1 – Cr (VI) conc. at different pH. Initial concentration of Cr(VI): 3.54 mg/L, 1 IRHC cube

A.3.3.2 Effect of IRHC dose

For IRHC dose test 1, 2 and 3 IRHC cube was used. The average weights of 1, 2 and 3 IRHC cube were found 1.06±0.13, 2.06±0.25 and 3.19±0.24 g, respectively. Figure A.3 showed the reduced chromium concentration for different IRHC dose in different time interval. It was seen that by increasing IRHC dose reduction of Cr (VI) concentration also increased due to the availability of more number of adsorbent sites for fixed number of Cr (VI) ions (Shindhu *et al.*, 2012). Also the contact time for Cr (VI) reduction was decreased.

A.3.3.3 Effect of initial Cr (VI) ion concentration

To check the effect of initial Cr (VI) ion concentration synthetic chromium solution of 10, 20, 40 and 80 mg/L was used where Cr (VI) concentration were 3.5, 7.0, 14 and 28 mg/L, respectively. From Figure A.6 it was seen that the reduced Cr (VI) was increased according to the increase of initial Cr (VI) conc. This may be caused by availability of high Cr (VI) ion for fixed amount of adsorbent (Shindhu *et al.*, 2012).

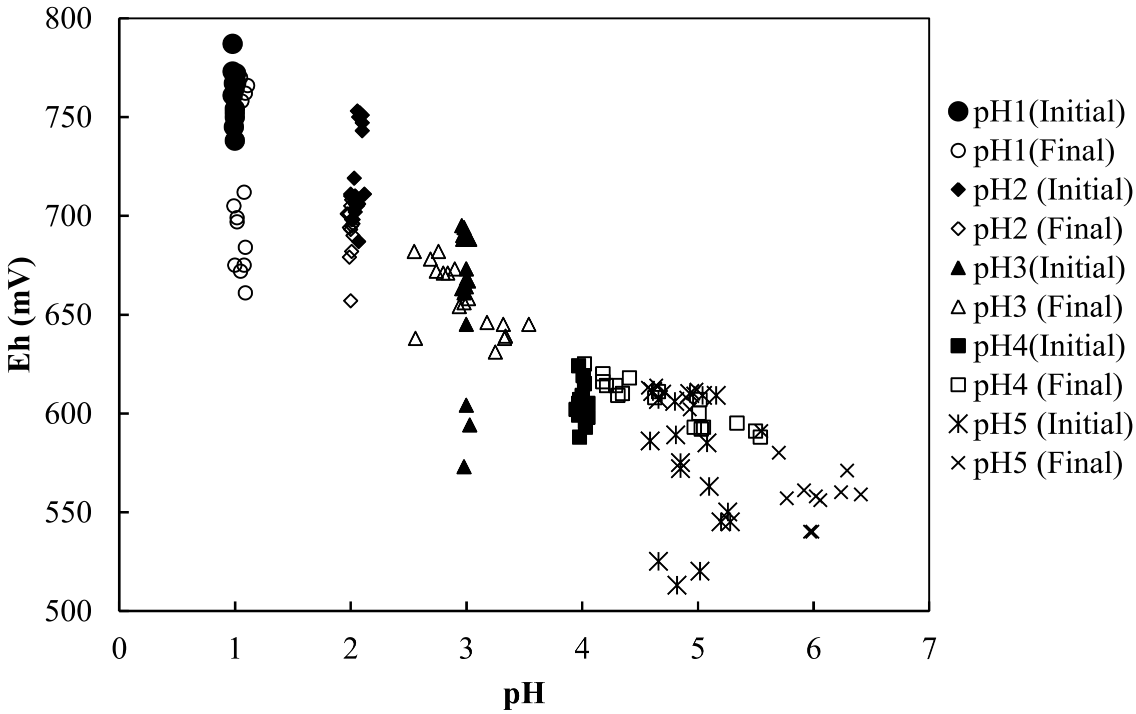

Figure A.2 – pH vs Eh diagram for the Figure A.1

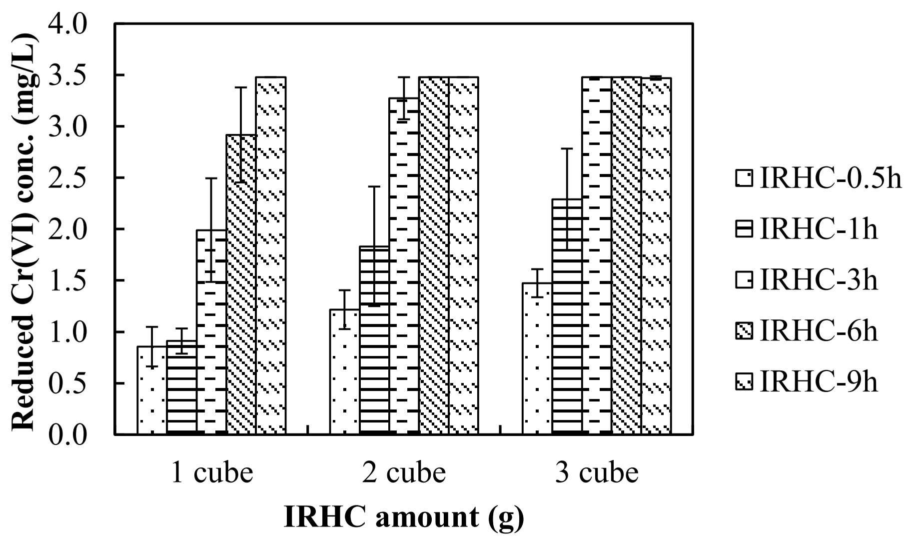

Figure A.3 – Relationship between reduced Cr (VI) concentrations and IRHC amount. Initial concentration of Cr (VI): 3.54 mg/L

It was observed that by immobilizing RHC to IRHC decreased floating, powdering of RHC which leads more efficient treatment. Also the reaction properties of IRHC showed similar properties as RHC suggesting that by immobilization of RHC could not affect the properties of RHC present in IRHC.

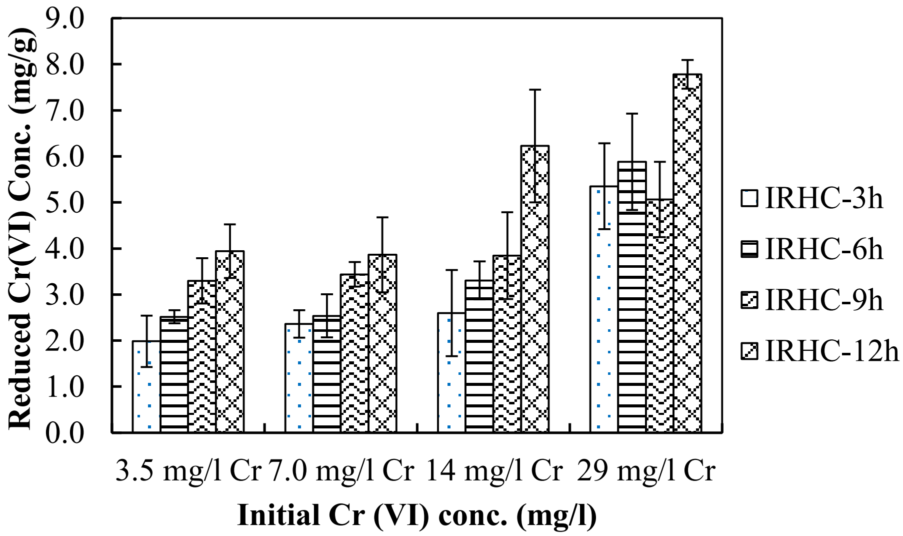

Figure A.6 – Relationship between initial Cr (VI) conc. and reduced Cr (VI) conc.

4.4 Summary

Based on the present investigation, it can be concluded that IRHC was successfully reduced Cr (VI) concentration. The removal was dependent on pH, initial concentration and IRHC dosage. It was observed that at low pH and higher initial concentration and IRHC dosage had higher reduction capacity. These properties were similar to RHC reduction properties', suggesting that the properties of RHC present in IRHC was not changed after immobilization.

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