HYDROCHEMISTRY EVOLUTION IN THE SHALLOW AQUIFER OF PULAU KAPAS, TERENGGANU, MALAYSIA

NOORAIN BINTI MOHD ISA

FPAS 2016 1

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By

NOORAIN BINTI MOHD ISA

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Doctor of Philosophy

February 2016

Abstract of thesis presented to the Senate of the Universiti Putra Malaysia in fulfillment of the requirements for the degree of Doctor of Philosophy

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Chairman : Assoc. Prof. Ahmad Zaharin Aris, PhD
Faculty : Environmental Studies

Classified as a small island, Pulau Kapas, Terengganu, Malaysia has become renowned locations among tourists as it served clean beaches with attractive scenery, ocean activities and turtle hatching. In Malaysia, islands are one of the biggest contributors to ecotourism activities, which support the national economic growth and help to sustain Malaysia's development. Apart from playing an important role in tourism, most of the small tropical islands faces similar problems regarding supplying freshwater. Due to the absence of surface water, small islands experiences major problems especially in supplying freshwater where groundwater abstraction is the only way to meet the demand of drinking water and domestic use. Higher demand of groundwater usage in the small island would possess a threat to the groundwater quality for example seawater intrusion and climate variation. The excess groundwater withdrawal may lead to vertical and lateral seawater intrusion. Meanwhile, climate variability is referring to low groundwater quantity due to dry season may eventually decreased its quality. Without proper planning and management, tourism could lead to degradation of an island environment. This study aimed to provide data on the temporal distribution of groundwater hydrochemistry in the small tropical islands and the important factors controlling the groundwater evolution as well as the spatial variation based on the study area characteristic.

A total of 216 groundwater samples were collected during different monsoon of pre-, (dry season; August – October 2010) and post-monsoon, (wet season; February – April 2011) in six constructed monitoring boreholes (KW 1, KW 2, KW 3, KW 4, KW 5 and KW 6). Triplicate samples in each monitoring boreholes were analyzed including physicochemical parameter of *in-situ* and major ions as well as cation exchanges capacity analysis and heavy metals measurement. Based on ANOVA test, all of the parameters show significant effects ($p < 0.01$) with the changes of monsoon seasons. The groundwater hydrochemistry facies were displays as Na-rich with Na-HCO_3 type during pre-monsoon while Ca-rich with Ca-HCO_3 type during post-monsoon, respectively. Based on the environmental investigation via analytical analyses, the ionic ratios were used to distinguish the origin and chemical behavior of the groundwater where the groundwater was influenced by the cation exchanges processes, simple mixing and water-rock interaction. The saturation indices (SI) with respect to calcite and aragonite described that 76% of the groundwater samples were in under-saturation state (during pre-monsoon) while post-monsoon shows contrary results where most of the groundwater samples were under super-saturated condition.

Additional of multivariate analyses of Principal Component Analysis (PCA) and Discriminant Analysis (DA) in present study can summarized the responsible factors controlling the groundwater evolution, which are the natural processes (ions exchanges process and saturation state of mineral) and human activities (over pumping which caused the up-coning of transition zone and simple mixing process). Meanwhile, the spatial variation was examined through the condition of groundwater in each monitoring boreholes using Hierarchy Cluster Analysis (HCA). There are two major groups which were classified as fresh groundwater (KW 3 and KW 4) and slightly affected/moderate condition of groundwater (KW 1, KW 2, KW 5 and KW 6). This classification was justified by the ionic strength calculation which reveals the same condition of groundwater in Pulau Kapas.

The investigation of heavy metals concentration reveals the domination order of Sr > Fe > Mn > Al > Cr > Zn > Ni > As > Pb > Cu > Cd. None of these heavy metals exceeded the guidelines regulated by World Health Organization (2011) and Ministry Of Health (2012). Only Sr concentration was reported higher than permissible limits as this metal might abundance in the aquifer bedrock of study area.

The cation exchange capacity (CEC) values shows a strong relationship with Ca concentrations, indicates the contribution of $CaCO_3$ (aquifer bedrock) either by dissolution or precipitation state in determining the groundwater evolution. The structure and morphology of the sediment were explained by X-Ray Diffraction (XRD) test. Aragonite which has the highest percentage with 68% was the dominant mineral due to the deposition of corals. The existence of this mineral was confirmed by the Scanning Electron Microscopy - Energy Dispersive X-ray spectroscopy (SEM-EDX) data where the major elements were carbonate minerals ($CaCO_3$).

This study recommends an illustration on the complex system of groundwater aquifer as to reveal the groundwater hydrochemistry status and chemical mechanisms as well as the fingerprint of groundwater evolution in small tropical islands and specifically, Pulau Kapas. This study also offers a better understanding on groundwater hydrochemistry as it's provide continuous data for future guidelines especially in the developing pristine environment.

Keywords: Kapas Island, ions exchanges, hydrogeochemistry mechanisms, heavy metals, groundwater

Abstrak tesis yang dikemukan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsafah

EVOLUSI HIDROKIMIA DALAM AKUIFER CETEK DI PULAU KAPAS, TERENGGANU, MALAYSIA

Oleh

NOORAIN MOHD ISA

Februari 2016

Pengerusi : Prof. Madya Ahmad Zaharin Aris, PhD
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Diklasifikasikan sebagai sebuah pulau kecil, Pulau Kapas, Terengganu, Malaysia telah menjadi lokasi terkenal di kalangan pelancong kerana ia menyediakan pantai yang bersih dengan pemandangan yang menarik, aktiviti di lautan dan penetasan penyu. Di Malaysia, pulau-pulau merupakan antara penyumbang terbesar kepada aktiviti eko-pelancongan, yang menyokong pertumbuhan ekonomi negara dan membantu mengekalkan pembangunan Malaysia. Selain memainkan peranan penting dalam bidang pelancongan, kebanyakan pulau-pulau tropika kecil menghadapi masalah yang sama berhubung membekalkan air tawar. Oleh kerana tidak mempunyai air permukaan, pulau-pulau kecil mengalami masalah utama terutamanya dalam membekalkan air tawar di mana pengekstrakan air bawah tanah adalah satu-satunya cara untuk memenuhi permintaan air minuman dan penggunaan domestik. Permintaan penggunaan air bawah tanah yang lebih tinggi di pulau kecil akan membawa satu ancaman kepada kualiti air bawah tanah sebagai contoh pencerobohan air laut dan perubahan iklim. Pengeluaran air bawah tanah yang berlebihan boleh membawa kepada kemasukan air laut secara menegak dan mendatar. Sementara itu, kepelbagaian iklim merujuk kepada kuantiti air bawah tanah yang rendah disebabkan musim kering akhirnya mengurangkan kualiti. Tanpa perancangan dan pengurusan yang betul, pelancongan boleh membawa kepada kemerosotan persekitaran pulau. Kajian ini bertujuan untuk menyediakan data mengenai pengagihan temporal hydrokimia air bawah tanah di pulau-pulau tropika kecil dan faktor-faktor penting yang mengawal evolusi air bawah tanah serta variasi spatial berdasarkan ciri-ciri kawasan kajian.

Sebanyak 216 sampel air bawah tanah telah dikumpulkan pada waktu monsoon yang berlainan iaitu pra- (musim kering; Ogos – Oktober 2010) dan pasca monsun, (musim basah; Februari – April 2011) dalam enam lubang gerudi pemantauan (KW 1, KW 2, KW 3, KW 4, KW 5 and KW 6). Tiga replika sampel dalam setiap lubang gerudi pemantauan telah dianalisis merangkumi fizikokimia parameter iaitu *in-situ* dan ions utama serta analisis kapasiti pertukaran kation dan pengukuran logam berat.

Berdasarkan ujian ANOVA, semua parameter menunjukkan kesan kolerasi ($p < 0.01$) dengan pertukaran musim monsun. Hidrokimia fasies bagi air bawah tanah menunjukkan masing-masing sebagai kaya-Na dengan jenis Na-HCO_3 sewaktu *pre*-monsun manakala kaya-Ca dengan jenis Ca-HCO_3 sewaktu *post*- monsun.

Berdasarkan kepada penyiasatan alam sekitar melalui penggunaan analisis analitikal, nisbah ion digunakan untuk mengenalpasti asal dan perlakuan kimia air bawah tanah dimana air bawah tanah telah dipengaruhi oleh pertukaran kation proses, percampuran mudah dan interaksi air dan batu. Indeks ketepuan (SI) berkaitan dengan kalcit dan aragonit menggambarkan bahawa 76% sampel air bawah tanah kebanyakkanya adalah dalam keadaan pembubaran (pra monsun) manakala pasca monsun menunjukkan hasil yang bertentangan di mana kebanyakkan sampel air bawah tanah berada di bawah keadaan super tepu.

Tambahan kepada analisis multivariat analisis komponen utama (PCA) dan analisis diskriminasi (DA) dalam kajian ini boleh merumuskan faktor yang bertanggungjawab mengawal evolusi/perubahan air bawah tanah, adalah dari aktiviti semulajadi (proses pertukaran ion dan keadaan ketepuan mineral) dan kegiatan manusia (proses pengepaman yang berlebihan yang mana menyebabkan kenaikan dari zon peralihan dan proses percampuran mudah). Tambahan pula, kepekatan Ca di dalam air bawah tanah telah berkolerasi dengan data CEC yang menunjukkan penglibatan $CaCO_3$ sama ada dalam keadaan pembubaran atau pemendakan dalam menentukan perbahan air bawah tanah. Sementara itu, taburan ruang telah diteliti melalui keadaan air bawah tanah dalam setiap lubang telaga pemantauan menggunakan analisis hirarki kelompok (HCA). Terdapat dua kumpulan utama yang diklasifikasikan sebagai air bawah tanah segar (KW 3 dan KW 4) dan keadaan sedikit terjejas/sederhana bagi air bawah tanah (KW 1, KW 2 KW 5 dan KW 6). Klasifikasi ini telah dijustifikasikan dengan pengiraan kekuatan ion yang mana mendedahkan keadaan yang sama bagi air bawah tanah di Pulau Kapas.

Penyiasatan tentang kepekatan logam berat mendedahkan rangka dominasi Sr > Fe > Mn > Al > Cr > Zn > Ni > As > Pb > Cu > Cd. Tiada satu pun dari logam berat ini melebihi garis panduan yang ditetapkan oleh Pertubuhan Kesihatan Sedunia (2011) dan Kementerian Kesihatan Malaysia (2012). Hanya kepekatan Sr dilaporkan lebih tinggi daripada had yang dibenarkan kerana logam ini berkemungkinan wujud dengan banyaknya di batu hampar akuifer di kawasan kajian.

Nilai kapasiti penukaran cation (CEC) menunjukkan hubungan yang kuat dengan kepekatan Ca, menandakan sumbangan $CaCO_3$ (akuifer batu hampar) samada melalui pembubaran atau pemendakan dalam menentukan evolusi/perubahan air bawah tanah. Struktur dan morfologi sedimen telah dijelaskan melalui ujian Pembelauan sinar-X (XRD). Aragonit, yang mempunyai peratus paling tinggi dengan 68% adalah mineral yang dominan kerana pemendapan batu karang. Kewujudan mineral ini telah disahkan dengan data Pengimbas Mikroskopi Elektron – Tenaga serakan spektroskopi sinar-X (SEM-EDX) dimana elemen utama adalah mineral karbonat ($CaCO_3$).

Kajian ini mencadangkan gambaran pada sistem akuifer air bawah tanah yang kompleks untuk mendedahkan kedudukan hidrokimia air bawah tanah dan mekanisma kimia serta identiti bagi perubahan air bawah tanah di pulau tropika kecil dan khususnya, Pulau Kapas. Kajian ini juga menawarkan pemahaman yang lebih baik mengenai hidrokimia air bawah tanah kerana ia menyediakan data berterusan sebagai garis panduan pada masa hadapan terutamanya dalam persekitaran yang membangun.

Kata kunci: Pulau Kapas, pertukaran ion, hidrogeokimia mekanisma, logam berat, air bawah tanah

ACKNOWLEDGEMENTS

In the name of Allah, the most beneficent, the most eternally merciful. First and foremost I would like to express my sincerest gratitude to my supervisor, Assoc. Prof. Dr. Ahmad Zaharin Aris for his patience, motivation, advices and spending of time and energy throughout my research period and thesis writing. Besides my advisor, I would like to thank the other members of my committee, Prof. Dr. Wan Nor Azmin Wan Sulaiman for the assistance he provided throughout my PhD program. This study was funded by the Ministry of Higher Education, Vot no. 5523724 (07/11/09/696FR). The provision of an allowance, Graduate Research Funding (GRF) by Universiti Putra Malaysia and MOHE Budget Mini Scholarship and MyPhD, are gratefully acknowledged. The valuable help from the Faculty of Environmental Studies and Faculty of Engineering, Universiti Putra Malaysia, in preparing boreholes for this research was much appreciated. High appreciation is also extended to the Department of Minerals and Geoscience, Terengganu, and the Malaysian Nuclear Agency for providing helpful information. Parts of this thesis have been published, thus, I would to thank the anonymous reviewers who gave valuable comments prior to publication. My appreciation is extended to my beloved parents, Mohd Isa Yusof and Faridah Jaafar, my family members, my sister, my fiancé, my colleagues and staffs upon their assistance, friendship and support.

I certify that a Thesis Examination Committee has met on 17 February 2016 to conduct the final examination of Noorain binti Mohd Isa of her thesis entitle “Hydrochemistry Evolution in the Shallow Aquifer of Pulau Kapas, Terengganu, Malaysia” in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The committee recommends that the student be awarded the Doctor of Philosophy (PhD).
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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillments of the requirements for the degreed of Doctor of Philosophy. The members of the Supervisory Committee were as follow:

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Signature: ______________________________

Name of Member of Supervisory Committee: Prof. Wan Nor Azmin Wan Sulaiman

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LIST OF ABBREVIATIONS

Al	Aluminium
ANOVA	Analysis of Variance
APHA	American Public Health Association
As	Arsenic
Ba	Barium
Ca	Calcium
Cd	Cadmium
CEC	Cation Exchange Capacity
Cl	Chloride
Cu	Copper
Cr	Chromium
DO	Dissolved Oxygen
EC	Electrical Conductivity
Eh	Redox Potential
EPA	Environmental Protection Agency
FAAS	Flame Atomic Absorption Spectrometry
Fe	Iron
H	Hydrogen
HCA	Hierarchical Cluster Analysis
HCO_3	Bicarbonate
IAP	Ion Activity Product
ICP-MS	Inductively Coupled Plasma Spectrometry
K	Potassium
Ksp	Equilibrium Solubility Product
Mg	Magnesium
Mn	Manganese
MOH	Ministry of Health
Na	Sodium
Ni	Nickel
NO_3	Nitrate
PCA	Principal Component Analysis
SD	Standard Deviation
SI	Saturation Indices
SO_4	Sulfate
PASW	Statistical Package for Social Science

TDS	Total Dissolved Solids
WHO	World Health Organization
XRD	X-Ray Diffraction
SEM	Scanning Electron Microscopy – Energy Dispersive X-ray
Zn	Zinc

LIST OF UNITS

%	Percent
<	less than
>	more than
°C	degree Celsius
µS/cm	micro Siemens per centimeter
µm	Micrometer
cm	Centimeter
g	Gram
km	Kilometer
km^2	kilometer square
m	Meter
m^2	meter square
meq/L	miliequivalent per liter
mg/L	miligrams per liter
mL	Millimeter
mV	Millivolt
ppb	parts per billion
ppt	parts per trillion

CHAPTER 1

INTRODUCTION

1.1 Background

Islands is an area that surrounded by ocean, with local resident or migration from the mainland (Whittaker and Fernandez 2007). Islands come in many shapes and sizes, their arrangement in space, geology, environments and variable characteristic of biotic which make them special and known to be isolated, remoteness, real or simulated and natural or artificial (Thronton 2007).

Islands are similar to the mainland that has similar ecosystem and hydrological theory cycle, yet, in a small group. Up to date, islands have developed into urbanized area as it's served a potential economic growth in the countries through the ecotourism sector. Industrial and agriculture also developed along with the population growth. It has become a challenges to islands in term of sustainable an ecosystem especially in small islands. On small islands, there a limited resources, economic and social activities however the interconnectivity between economic, environment, social and cultural is strong and pervasive (Twining-Ward and Butler 2002).

Despite being one of the major economic contributions, however, small islands sustainability is in jeopardies because of tourism development. In other point, the negative impact of ecotourism can give more pressure on this vulnerable, limited resource of island. As has been well documented, small islands depend on groundwater as its main water supply. Generally, there's no such surface water exist in most small islands that can support the needs of freshwater for entire island. Due to increasing number of population including tourists, high water demands are required.

Groundwater is water that filled up in an open space of rocks below the earth's surface (Nonner 2010) under a pressure larger than atmospheric pressure. Groundwater has been widely used in developed countries for industrial and agriculture sector while has extensively used for drinking water supplier and domestic uses in developing countries (Hiscock 2009). Small tropical islands are unexceptional area to exploit groundwater for daily consumption from drilled boreholes that usually comes with negative impact of uncontrolled basis, untreated, unmonitored and etc. Such behaviors were further threated the availability of fresh groundwater for human consumption and other activities such as water planting.

Like surface water, groundwater can become polluted or contaminated. Groundwater pollution is associated with natural and anthropogenic activities. Pollution of groundwater is a legacy of past and present land-use practiced, poor waste disposal (Hiscock 2009), inundation, overexploitation, seawater intrusion and climate variability. Low quality of groundwater due to untreated pollutions were influenced the diurnal activities in the small islands which later on, in a long-term will affect the economic contribution.

Regardless, groundwater is poorly understood and often undervalued. A complex and hidden resources of groundwater is difficult to conceptualize and it is the reason of working hard to understand more about groundwater. It remains one of the greatest challenges for the future to provide the amenity of a safe reliable groundwater supply to the entire islands annually without degrades its quality.

Since the groundwater control management in small islands is still lacking, this study attempt to determine the spatial and temporal distribution of groundwater quality by integrating monitoring, analytical and modeling approaches. The groundwater composition will be examined and the inter-relationship between seasonal changes will be investigated. Application of modeling will be used to identify the connection of the complex geochemical reactions such as ion exchanges, oxidation/reduction and precipitation/dissolution (Mao et al. 2006). Further discussion on water-rock interaction and simple mixing processes of groundwater are also will be determined.

1.2 Problem statements

Small tropical islands have urbanized in these decades as it served the important role in promoting economic development of the country. The profits gained by the small tropical islands are from ecotourism, water-sports activities, recreation and hereditary center. Rapid development have possessed a threat to the natural ecosystem especially groundwater quality (Chen and Feng 2013; Ravikumar et al. 2011). Groundwater is a paramount importance in small tropical islands as it represents the main water resources to satisfy the different needs of the various applications (Lu et al. 2012). For examples, the groundwater is use for drinking water purposes, water consumption by the chalet maintenance, domestic activities, water planting and etc. Since the tourist and local resident numbers have increased remarkably, high groundwater demands are required to fulfill their needs. Hence, it is necessary to understand the influence of groundwater deterioration which could happen via natural processes or anthropogenic influxes (Barragán-Alarcón 2012: Isa et al. 2012a). However, inadequate information and knowledge of water preservation are the challenges that need to overcome by the community (Nosrati, and Van Den Eeckhaut 2012), therefore, to protect the ecological balance of island's ecosystem.

In worldwide, coastal aquifer are vulnerable to seawater intrusion (Aris et al., 2012a; Singh et al. 2012; Russak and Sivan, 2010; Werner et al., 2009; Lee and Song 2007; Fleeger et al., 2009) due to overexploitation of groundwater. Since the small islands have developed tremendously, the numbers of population as well as the production of groundwater have increased. As results from over-abstraction, the groundwater lens might shrink and enable the vertical (i) and lateral (ii) intrusions (Figure 1.1) of seawater which later on mixing with fresh groundwater and later will degrades its quality.

Recently, the groundwater deterioration due to the climate variability has caused the groundwater issues. Previous studies have discussed the alteration of groundwater quality during monsoon periods (Negrel et al. 2011; Mondal et al. 2010; Kazama et al. 2007; Panda et al. 2007; Kumar and Ahmed 2003). Small islands do not have any surface water reservoir except for ephemeral rivers that only exist in an exploitable form during heavy rainfall. That will make the groundwater are the only freshwater resources in the small islands. Pre-monsoon which defined as dry season with less rainfall usually received tons of tourists that led to extract more groundwater. With less groundwater recharge versus high extraction of groundwater, it is definitely degrade the groundwater quality.

It is necessary to understand the nature of seasonal changes. The seasonal variation affects the groundwater including the groundwater storage and its quality (Green et al., 2011; Dragoni and Sukhija, 2008). Since present study is referring to tropical regions, two different tropical seasons are discussed – pre- and post-monsoon seasons (Wong et al. 2009; Desa and Niemczynowicz 1996). The seasonal variation in tropical regions is associated with the dry and wet seasons, which occur during the South-West Monsoon (SWM; pre-monsoon) and North-East Monsoon (NEM; post-monsoon), respectively. Most of the tourists are free to pay a visit to these related islands usually throughout the year, except during monsoon interchanges due to safety issues. The association of seasonal changes in the complex nature of the groundwater system results in the alteration of the groundwater composition. During rainfall, the groundwater aquifer widens, eliminating the contaminants by dilution and simultaneously freshening the groundwater quality. Meanwhile, during drought, the groundwater storage declines, pollutants are concentrated and they lead to the brackish water as the groundwater quality decreases. In order to sustain the groundwater availability and to supply good quality groundwater, the impact of the climate variability is required to be understood.

Despite those problems, large of scientific works discussed on the hydrochemistry of groundwater aquifer in various sector (Cavalcanti de Albuquerque and Kirste 2012; Christodoulidou et al. 2012; Ekwere and Edet 2012; Haloi and Sarma 2012; Kargar et al. 2012; Krishna Kumar et al. 2012; Norrström and Knutsson 2012; Papaioannou et al. 2010; Recep and Tuncan 2011; Shamsudduha et al. 2009; Lee and Song 2007; Lee et al. 2007; Leung and Jiao 2006; Nouri et al. 2006; Lakshmanan et al. 2003). There are several factors happened that affect the groundwater quality through its chemical mechanisms including the composition of the precipitation (Farmaki et al. 2012), geological structure (Kim et al. 2012: Bouchaou et al. 2008), mineralogy (Harrington et al. 2008), geological processes (Sena and Teresa Condesso de Melo 2012) along with external pollution such as effluents (Khelfaoui et al. 2012; Currell et al. 2010), seawater intrusion and domestic activities (Aris et al. 2012a; Srinivasamoorthy et al. 2012).

A good knowledge of the groundwater hydrogeological model in small islands is a prerequisite to investigate the mechanisms that control the groundwater quality. The developed model aims to explain the interaction between the complex hydrogeochemical environments of small islands and their surroundings (surrounded by seawater), which makes them more susceptible to seawater disturbance. Figure 1.1 shows the conceptual model of a small tropical island, which was developed to understand the groundwater hydrogeochemistry process. This conceptual model developed consists of static and dynamic components. The static component symbolizes the aquifer matrix or soil and the dynamic component represents the groundwater as the only water reservoir in the island since no other surface water exists in exploitable form, including evapotranspiration and precipitation. The main input of the conceptual model starts from precipitation, which infiltrates into the groundwater as groundwater recharge, discharge of groundwater from the constructed boreholes, evapotranspiration process from the surface level and to complete the model the water returns as precipitation. These elements/components are shown to be related by exact conceptual, depending on the situation in present day (Toth, 1970) that controlled the groundwater environment. Another factor that contributes to the process of the conceptual model is climate, comprising the temperature, precipitation and winds while the geology factor is made up of the distribution of different parent rocks and the groundwater. Therefore, the physical and chemical conditions resulting from the combination of hydrogeology and climate may be said to contribute to the hydrogeological groundwater composition. It is possible to describe and analyze the groundwater conditions in the context of the hydrogeological observations of the conceptual model. The hydrogeological conceptual model developed is very important to understand the interaction between the static and dynamic component to identify the mechanisms involved that control the groundwater composition. The use of such a conceptual model for the precise hydrogeological of the sampling area in this study appears to be a prerequisite for groundwater development and management, especially in small tropical islands, as it answers several key issues, which are salinization, mineralization and the chemical processes.

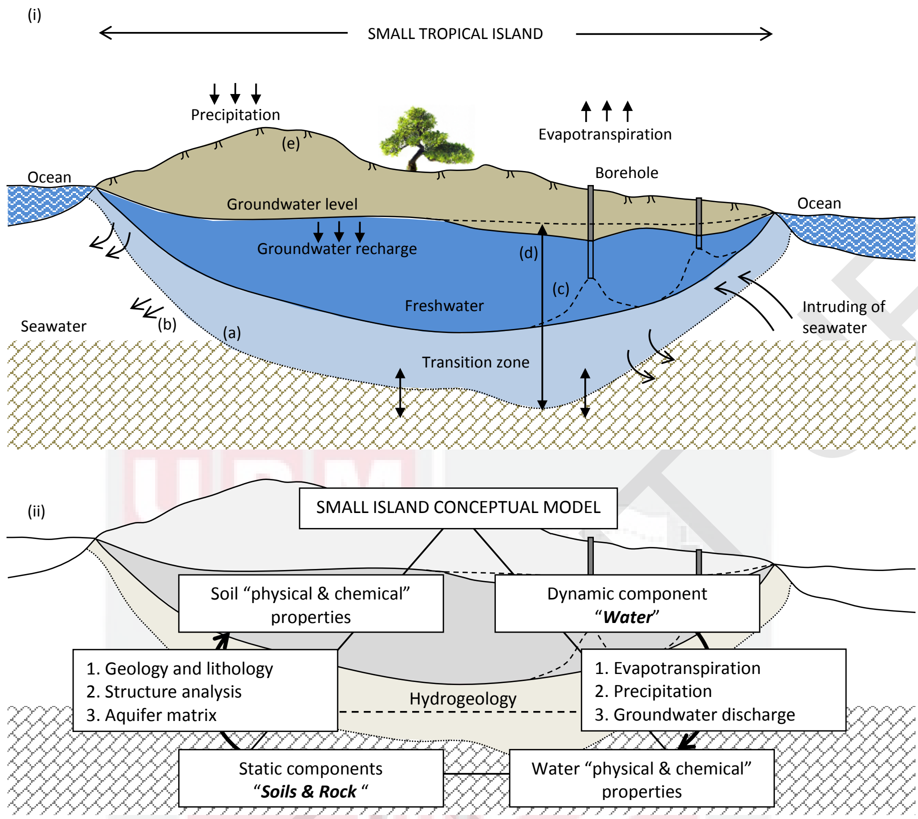

Figure 1.1 (i) A simple hydrological conceptual model for Pulau Kapas aquifer and (ii) the hydrological conceptual model approach used in this study, modified from Winkler et al. (2003). Showing (a) seawater wedge, (b) density-driven circulation-force seawater zone underneath groundwater, (c) seawater upconing due to borehole pumping, (d) average aquifer thickness and (e) ground surface

Depletion of groundwater level and deterioration of its quality is the major concerned that needs immediate attention. Therefore, an understanding of the hydrogeochemical processes of groundwater is a must for a sustainable development in small tropical islands. Many studies have been conducted to ascertain the groundwater quality by demarcating the character of groundwater to solve different hydrogeochemical problems. Groundwater monitoring and hydrochemical data are the methods that usually practiced (Chowdhury et al. 2012; Isa et al. 2012a; Papaioannou et al. 2010). Among these methods, additional techniques of multivariate analyses (Kargar et al. 2012: Khelfaoui et al. 2012: Subyani and Ahmadi 2010; Petalas and Anagnostopoulos 2006), calculation of saturation index (Krishna Kumar et al. 2012: Gupta et al. 2011; Pulido-Leboeuf 2004), ionic ratios along with ionic strength evaluation (Cavalcanti de Albuquerque and Kirste 2012; Tay 2012; Edet and Okereke 2001), and inverse geochemical modeling (Recep and Tuncan 2011; Belkiri et al. 2010; Von der Heyden 2004) have been applied to the study, evaluate and characterize the possible sources and minerals of groundwater hydrochemistry variation. In literature, there is a dearth of information on the hydrochemistry approaches particularly in small developing tropical islands where the groundwater sustainability is almost impossible. Thus, this kind of investigation is required to carry out extensively, to conserve the existing natural environment for the future generation.

1.3 Scopes of research

Essentially, the study involves the following scopes;

1. Seven boreholes were drilled in Pulau Kapas to identify the distribution of major ions and heavy metals in groundwater aquifer and the exchangeable capacity of sediments. The in-situ parameters of the groundwater were also recorded. The temporal and spatial variations of major ions were investigated in relations to different seasons. Measurements of physicochemical parameters were used to ascertain the interconnectivity within the hydrochemical mechanisms in the groundwater system.
2. This study involved of environmental study approaches that combined analytical analyses, different geological indices, and multivariate analyses, hydrochemical analyses including types of groundwater and capability of ions to exchanges as well as modeling approaches to illustrate the movement of freshwater-seawater interface and predicting the impact of groundwater chemistry evolution related to seasonal changes.
3. In general, the scopes of this study were to understand the chemical mechanism of groundwater system through the variation of physicochemical parameters as to interpret the important of hydrogeochemical processes in relation to seasonal changes.

1.4 Choices of the research study area

Pulau Kapas was chosen for this study because of the following reasons;

1. Pulau Kapas was chosen in this study due to the limited information available concerning the hydrogeochemistry, either pertaining to protection or preservation, and the fact that this small island has become a high water demand area due to the development of tourism activities.
2. Anticipate that present study will provide basis in partially polluted area for future studies on groundwater hydrochemistry especially in small tropical islands
3. Exposed to the monsoon interchanges as Pulau Kapas located at the high seas of South China Sea.
4. Researches overview the facts that small island at the open sea especially in tropical region facing enormous problems arising from population growth, urbanization, shortage of water resources quantitatively and qualitatively.
5. Groundwater wedge is a fragile environment as the water balance is exposed to the fresh and seawater interaction.

It is important to study on this particular area in order to conserve and protect the natural resources as well as initial steps for responsible organization to manage and control the small tropical island’s developments.

1.5 Objectives

In general, this study intends to assess the hydrochemistry of groundwater in Pulau Kapas, Terengganu, Malaysia. It provides a hydrochemical dataset including the changes of concentration and groundwater composition in different seasons as well as sediments facies. The research objectives are stated as follows:

1. To identify the hydro-chemical facies of the groundwater as a chemo-indicator to the water chemistry equilibrium disturbance.
2. To describe the chemical reactions involve in the islands aquifer based on different monsoon seasons.
3. To determine the impact of monsoon changes to groundwater hydrochemistry due to the climate variability.
4. To define the controlling factor of groundwater evolution in Pulau Kapas using the hydrochemistry data based on the holistic environmental forensics approaches.

1.6 Significance of study

As expected, the issues of groundwater pollution and protecting the groundwater resources provide an important reason to study the groundwater. Scientists and engineers in developed countries have aware of the health threat posed by the contamination involving in cleaning up contaminated groundwater (Schwartz and Zhang 2003). Developing area such as Pulau Kapas is far away in managing the sustainable of groundwater, yet, lack of expertise and information which can be applied from other tropical islands. This study has offers a better understanding on groundwater hydrochemistry of a pristine environment by providing an excellent interpretation of the results and enhanced the used of selected variables in hydrochemistry studies, as a source tracer for groundwater contaminations. Such hypothesis on more and more people attends becoming less quality of water supply needs to be eliminated in order to gain a sustainable development as well as concrete profitable.

Due to scarcity of information and workloads on groundwater in tropical region, present study was conducted to fill the gaps and provides an understanding of groundwater hydrochemistry characteristics. The methods approaches discussed in section 1.2 were carried out in order to better assess of the chemical mechanism as well as to concern the status of groundwater degradation. Hence, with right and suitable facts, it could provide explanation and solution to the sector responsible for the variation of groundwater constituents.

There are important attributes from present study which are the quantitative and qualitative assessment of the natural groundwater hydrochemistry and contaminated systems. Quantitative outputs exemplify the identification of chemical concerns, signify the spatial and temporal distribution and elucidate the groundwater status. These datasets of groundwater were used to extinguish the hydrochemical mechanism along with the data analyses which provide valuable information on the characteristic performed during two different seasons. The evaluation between natural and anthropogenic influences based on the degree of saturation and modeling application are also described.

Whereas, the qualitative outputs includes the evidences of affected area, comparison with previous researches on groundwater monitoring and either to create a new databases or to improve an existing guidelines. Finding in this research will be used as a platform to agencies and the governments for detail investigation on specific area to spot available risks to the ecosystems in the future. In addition, the outputs could be used to generate new regulations incorporate with sustainable, manageable and conservable developments specifically in small tropical islands.

1.7 Thesis Outlines

The main body of this thesis consists of five chapters. Each of the chapter was purposely explains on the groundwater hydrochemistry and the chemical mechanism controlling the groundwater evolution as well as their potential contaminant risks. The chapters in this thesis have been organized as follows. In Chapter 1, a brief introduction on the groundwater aquifer system is provided including the objectives, problem statements and scope of study.

A comprehensive literature review is given in Chapter 2. The information on water security in small islands was highlighted based on previous scientific research. Chapter 3 covers on the detail description of the study area and the material and methods used in present study. The groundwater samples were the main subject utilized throughout this study. The laboratory analysis applied in this study was divided into groundwater and sediment samples. The data collected were analyzed by the related mathematical calculation and hydrochemistry analysis.

The results and discussion of this research were explained in Chapter 4. The arrangement of data presentation was based on the finding of groundwater hydrochemistry based on the contribution of monsoon seasons and followed by the explanation on the hydrochemical mechanism in the groundwater system. Part of the discussion was based on the published manuscripts which are; Finding of Cation Exchange Capacity (CEC) values (Isa and Aris 2012), the heavy metals distribution in the study area (Isa, et al. 2014), the contribution of monsoon seasons against the groundwater constituents (Isa, et al. 2012; Isa et al. 2014) and identification of factor controlling the groundwater composition using statistical approach (Isa et al. Revised).

Chapter 5 was detailed out the main conclusions that draw from the results and discussion session while SWOT analysis was performed to identify the advantages and the limitation of this study. The recommendations were presented in this chapter as to provide appropriate recommendations to improve the status of groundwater quality especially in small tropical islands.

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APPENDICES A

An example of output for PHREEQC calculation

```
Input file: D:\pendrive\project\data\phreeqc\metal\KW
1.7.1.pqi
Output file: D:\pendrive\project\data\phreeqc\metal\KW
1.7.1.pqo
Database file: C:\Program Files\USGS\Phreeqc Interactive
2.17.4468\database\phreeqc.dat

------------------
Reading data base.
------------------

      SOLUTION_MASTER_SPECIES
      SOLUTION_SPECIES
      PHASES
      EXCHANGE_MASTER_SPECIES
      EXCHANGE_SPECIES
      SURFACE_MASTER_SPECIES
      SURFACE_SPECIES
      RATES
      END
------------------------------------
Reading input data for simulation 1.
------------------------------------

DATABASE C:\Program Files\USGS\Phreeqc Interactive
2.17.4468\database\phreeqc.dat
      SOLUTION 1
          temp      30.6
          pH        7.1
          pe        4
          redox     pe
          units     mg/l
          density   1
          Ca        107.51
          Mg        14.02
          Na        18.06
          K         0.63
          Alkalinity 336.72
          Cl        27.99
          S(6)      14
          Zn        2.717 ug/L
          Pb        0.00005 ug/L
          Mn        32.952 ug/L
          Cu        0.566 ug/L
          water    1 # kg
```

```
-------------------------------------------
Beginning of initial solution calculations.
-------------------------------------------

Initial solution 1.



-----------------------------Solution composition---------
-------------------

      Elements             Molality       Moles

      Alkalinity         6.732e-003  6.732e-003
      Ca                 2.684e-003  2.684e-003
      Cl                 7.899e-004  7.899e-004
      Cu                 8.912e-009  8.912e-009
      K                  1.612e-005  1.612e-005
      Mg                 5.770e-004  5.770e-004
      Mn                 6.001e-007  6.001e-007
      Na                 7.860e-004  7.860e-004
      Pb                 2.414e-013  2.414e-013
      S(6)               1.458e-004  1.458e-004
      Zn                 4.158e-008  4.158e-008

----------------------------Description of solution-------
-------------------

                                          pH  =   7.100
                                          pe  =   4.000
       Specific Conductance (uS/cm, 30 oC) = 774
                              Density (g/cm3)  =   0.99594
                            Activity of water  =   1.000
                               Ionic strength  =  1.045e-002
                           Mass of water (kg)  =  1.000e+000
                        Total carbon (mol/kg)  =  7.698e-003
                           Total CO2 (mol/kg)  =  7.698e-003
                          Temperature (deg C)  =  30.600
                      Electrical balance (eq)  = -4.884e-004
 Percent error, 100*(Cat-|An|)/(Cat+|An|)  =  -3.35
                                   Iterations  =  10
                                      Total H  = 1.110191e+002
                                      Total O  = 5.552891e+001

----------------------------Distribution of species-------
-------------------
```

Species	Molality	Activity	Log Molality	Log Activity	Log Gamma
OH-	2.119e-007	1.901e-007	-6.674	-6.721	-0.047
H+	8.717e-008	7.943e-008	-7.060	-7.100	-0.040
H2O	5.551e+001	9.998e-001	1.744	-0.000	0.000
C(4)	7.698e-003				
HCO3-	6.513e-003	5.879e-003	-2.186	-2.231	-0.044
CO2	9.864e-004	9.888e-004	-3.006	-3.005	0.001
CaHCO3+	1.485e-004	1.340e-004	-3.828	-3.873	-0.044
MgHCO3+	2.836e-005	2.551e-005	-4.547	-4.593	-0.046
CaCO3	1.210e-005	1.213e-005	-4.917	-4.916	0.001
CO3-2	5.807e-006	3.855e-006	-5.236	-5.414	-0.178
NaHCO3	2.326e-006	2.332e-006	-5.633	-5.632	0.001
MgCO3	1.442e-006	1.445e-006	-5.841	-5.840	0.001
MnHCO3+	1.455e-007	1.309e-007	-6.837	-6.883	-0.046
MnCO3	7.629e-008	7.647e-008	-7.118	-7.116	0.001
NaCO3-	7.427e-008	6.679e-008	-7.129	-7.175	-0.046
ZnHCO3+	1.054e-008	9.479e-009	-7.977	-8.023	-0.046
ZnCO3	9.826e-009	9.850e-009	-8.008	-8.007	0.001
Zn(CO3)2-2	1.241e-009	8.117e-010	-8.906	-9.091	-0.184
PbCO3	2.154e-013	2.160e-013	-12.667	-12.666	0.001
PbHCO3+	1.674e-014	1.506e-014	-13.776	-13.822	-0.046
Pb(CO3)2-2	3.197e-015	2.091e-015	-14.495	-14.680	-0.184
Ca	2.684e-003				
Ca+2	2.497e-003	1.656e-003	-2.603	-2.781	-0.178
CaHCO3+	1.485e-004	1.340e-004	-3.828	-3.873	-0.044

```
   CaSO4                   2.577e-005  2.584e-005    -4.589
-4.588     0.001
   CaCO3                   1.210e-005  1.213e-005    -4.917
-4.916     0.001
   CaOH+                   3.848e-009  3.460e-009    -8.415
-8.461    -0.046
   CaHSO4+                 1.439e-011  1.294e-011   -10.842
-10.888    -0.046
Cl              7.899e-004
   Cl-                     7.899e-004  7.091e-004    -3.102
-3.149    -0.047
   MnCl+                   8.023e-010  7.215e-010    -9.096
-9.142    -0.046
   ZnCl+                   3.464e-011  3.115e-011   -10.460
-10.507    -0.046
   MnCl2                   2.228e-013  2.233e-013   -12.652
-12.651     0.001
   ZnCl2                   2.359e-014  2.364e-014   -13.627
-13.626     0.001
   PbCl+                   1.160e-016  1.043e-016   -15.936
-15.982    -0.046
   MnCl3-                  4.850e-017  4.361e-017   -16.314
-16.360    -0.046
   ZnCl3-                  2.162e-017  1.944e-017   -16.665
-16.711    -0.046
   PbCl2                   1.055e-019  1.058e-019   -18.977
-18.976     0.001
   ZnCl4-2                 1.103e-020  7.217e-021   -19.957
-20.142    -0.184
   PbCl3-                  6.853e-023  6.163e-023   -22.164
-22.210    -0.046
   PbCl4-2                 3.336e-026  2.182e-026   -25.477
-25.661    -0.184
Cu(1)           1.098e-010
   Cu+                     1.098e-010  9.816e-011    -9.960
-10.008    -0.049
Cu(2)           8.802e-009
   Cu(OH)2                 5.867e-009  5.881e-009    -8.232
-8.231     0.001
   Cu+2                    2.658e-009  1.777e-009    -8.575
-8.750    -0.175
   CuOH+                   2.488e-010  2.236e-010    -9.604
-9.650    -0.046
   CuSO4                   2.791e-011  2.798e-011   -10.554
-10.553     0.001
   Cu(OH)3-                4.960e-015  4.460e-015   -14.305
-14.351    -0.046
   Cu(OH)4-2               1.712e-020  1.120e-020   -19.766
-19.951    -0.184
H(0)            8.438e-026
```

```
   H2                        4.219e-026  4.229e-026   -25.375
-25.374     0.001
K                1.612e-005
   K+                        1.611e-005  1.446e-005    -4.793
-4.840    -0.047
   KSO4-                     9.235e-009  8.305e-009    -8.035
-8.081    -0.046
   KOH                       6.296e-013  6.312e-013   -12.201
-12.200     0.001
Mg               5.770e-004
   Mg+2                      5.399e-004  3.605e-004    -3.268
-3.443    -0.175
   MgHCO3+                   2.836e-005  2.551e-005    -4.547
-4.593    -0.046
   MgSO4                     7.213e-006  7.231e-006    -5.142
-5.141     0.001
   MgCO3                     1.442e-006  1.445e-006    -5.841
-5.840     0.001
   MgOH+                     3.010e-008  2.707e-008    -7.521
-7.568    -0.046
Mn(2)            6.001e-007
   Mn+2                      3.737e-007  2.498e-007    -6.427
-6.602    -0.175
   MnHCO3+                   1.455e-007  1.309e-007    -6.837
-6.883    -0.046
   MnCO3                     7.629e-008  7.647e-008    -7.118
-7.116     0.001
   MnSO4                     3.654e-009  3.663e-009    -8.437
-8.436     0.001
   MnCl+                     8.023e-010  7.215e-010    -9.096
-9.142    -0.046
   MnOH+                     1.406e-010  1.265e-010    -9.852
-9.898    -0.046
   MnCl2                     2.228e-013  2.233e-013   -12.652
-12.651     0.001
   MnCl3-                    4.850e-017  4.361e-017   -16.314
-16.360    -0.046
Mn(3)            4.477e-028
   Mn+3                      4.477e-028  1.723e-028   -27.349
-27.764    -0.415
Na               7.860e-004
   Na+                       7.833e-004  7.053e-004    -3.106
-3.152    -0.046
   NaHCO3                    2.326e-006  2.332e-006    -5.633
-5.632     0.001
   NaSO4-                    3.022e-007  2.718e-007    -6.520
-6.566    -0.046
   NaCO3-                    7.427e-008  6.679e-008    -7.129
-7.175    -0.046
   NaOH                      5.851e-011  5.865e-011   -10.233
-10.232     0.001
```

```
O(0)              2.762e-040
   O2                        1.381e-040  1.385e-040   -39.860
-39.859     0.001
Pb                2.414e-013
   PbCO3                     2.154e-013  2.160e-013   -12.667
-12.666     0.001
   PbHCO3+                   1.674e-014  1.506e-014   -13.776
-13.822    -0.046
   Pb+2                      4.929e-015  3.224e-015   -14.307
-14.492    -0.184
   Pb(CO3)2-2                3.197e-015  2.091e-015   -14.495
-14.680    -0.184
   PbOH+                     8.799e-016  7.913e-016   -15.056
-15.102    -0.046
   PbSO4                     1.343e-016  1.346e-016   -15.872
-15.871     0.001
   PbCl+                     1.160e-016  1.043e-016   -15.936
-15.982    -0.046
   Pb(OH)2                   3.865e-018  3.875e-018   -17.413
-17.412     0.001
   PbCl2                     1.055e-019  1.058e-019   -18.977
-18.976     0.001
   Pb(SO4)2-2                8.022e-020  5.247e-020   -19.096
-19.280    -0.184
   Pb(OH)3-                  6.226e-022  5.599e-022   -21.206
-21.252    -0.046
   PbCl3-                    6.853e-023  6.163e-023   -22.164
-22.210    -0.046
   PbCl4-2                   3.336e-026  2.182e-026   -25.477
-25.661    -0.184
   Pb(OH)4-2                 2.468e-026  1.615e-026   -25.608
-25.792    -0.184
   Pb2OH+3                   1.484e-028  5.711e-029   -27.828
-28.243    -0.415
S(6)              1.458e-004
   SO4-2                     1.125e-004  7.426e-005    -3.949
-4.129    -0.180
   CaSO4                     2.577e-005  2.584e-005    -4.589
-4.588     0.001
   MgSO4                     7.213e-006  7.231e-006    -5.142
-5.141     0.001
   NaSO4-                    3.022e-007  2.718e-007    -6.520
-6.566    -0.046
   KSO4-                     9.235e-009  8.305e-009    -8.035
-8.081    -0.046
   MnSO4                     3.654e-009  3.663e-009    -8.437
-8.436     0.001
   HSO4-                     7.225e-010  6.498e-010    -9.141
-9.187    -0.046
   ZnSO4                     2.320e-010  2.326e-010    -9.634
-9.633     0.001
```

```
   CuSO4                      2.791e-011  2.798e-011   -10.554
-10.553     0.001
   CaHSO4+                    1.439e-011  1.294e-011   -10.842
-10.888    -0.046
   Zn(SO4)2-2                 2.057e-013  1.346e-013   -12.687
-12.871    -0.184
   PbSO4                      1.343e-016  1.346e-016   -15.872
-15.871     0.001
   Pb(SO4)2-2                 8.022e-020  5.247e-020   -19.096
-19.280    -0.184
Zn                   4.158e-008
   Zn+2                       1.939e-008  1.281e-008    -7.713
-7.893    -0.180
   ZnHCO3+                    1.054e-008  9.479e-009    -7.977
-8.023    -0.046
   ZnCO3                      9.826e-009  9.850e-009    -8.008
-8.007     0.001
   Zn(CO3)2-2                 1.241e-009  8.117e-010    -8.906
-9.091    -0.184
   ZnOH+                      2.982e-010  2.682e-010    -9.525
-9.572    -0.046
   ZnSO4                      2.320e-010  2.326e-010    -9.634
-9.633     0.001
   ZnCl+                      3.464e-011  3.115e-011   -10.460
-10.507    -0.046
   Zn(OH)2                    2.548e-011  2.554e-011   -10.594
-10.593     0.001
   Zn(SO4)2-2                 2.057e-013  1.346e-013   -12.687
-12.871    -0.184
   ZnCl2                      2.359e-014  2.364e-014   -13.627
-13.626     0.001
   Zn(OH)3-                   1.131e-015  1.017e-015   -14.947
-14.993    -0.046
   ZnCl3-                     2.162e-017  1.944e-017   -16.665
-16.711    -0.046
   ZnCl4-2                    1.103e-020  7.217e-021   -19.957
-20.142    -0.184
   Zn(OH)4-2                  3.101e-021  2.028e-021   -20.509
-20.693    -0.184


------------------------------Saturation indices----------
-------------------

      Phase               SI log IAP  log KT

      Anglesite       -10.86  -18.62   -7.76  PbSO4
      Anhydrite        -2.52   -6.91   -4.39  CaSO4
      Aragonite         0.18   -8.19   -8.37  CaCO3
      Calcite           0.32   -8.19   -8.51  CaCO3
      Cerrusite        -6.84  -19.91  -13.06  PbCO3
```

```
        CO2(g)             -1.47   -3.00   -1.53  CO2
        Dolomite            0.17  -17.05  -17.22  CaMg(CO3)2
        Gypsum             -2.33   -6.91   -4.58  CaSO4:2H2O
        H2(g)             -22.20  -25.37   -3.17  H2
        H2O(g)             -1.37   -0.00    1.37  H2O
        Halite             -7.90   -6.30    1.59  NaCl
        Hausmannite       -14.68   44.99   59.67  Mn3O4
        Manganite          -6.64   18.70   25.34  MnOOH
        O2(g)             -36.93  -39.86   -2.93  O2
        Pb(OH)2            -8.25   -0.29    7.96  Pb(OH)2
        Pyrochroite        -7.60    7.60   15.20  Mn(OH)2
        Pyrolusite        -10.70   29.80   40.50  MnO2
        Rhodochrosite      -0.87  -12.02  -11.15  MnCO3
        Smithsonite        -3.25  -13.31  -10.06  ZnCO3
        Zn(OH)2(e)         -5.19    6.31   11.50  Zn(OH)2

------------------
End of simulation.
------------------

------------------------------------
Reading input data for simulation 2.
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End of run.
-----------
```
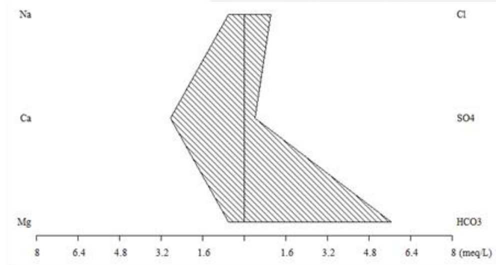
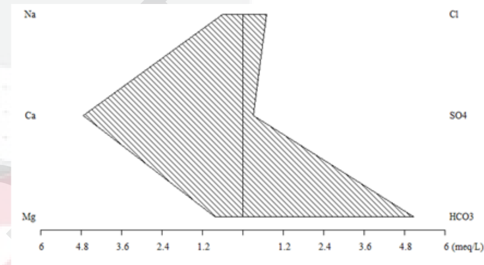
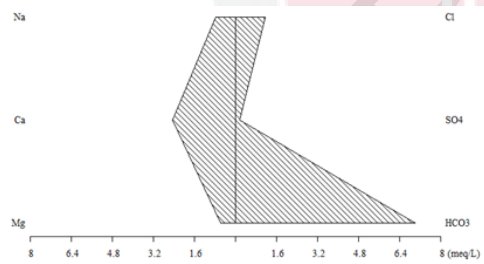
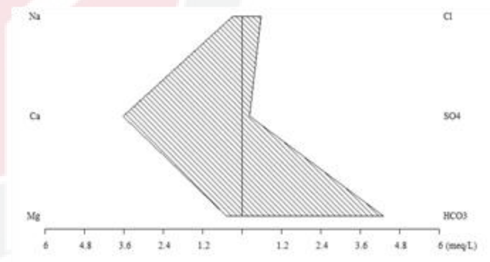
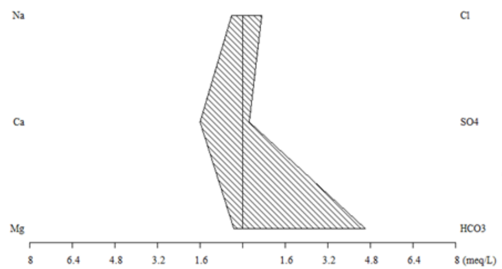
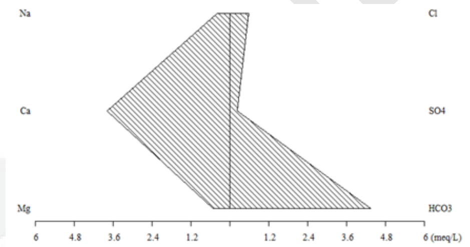
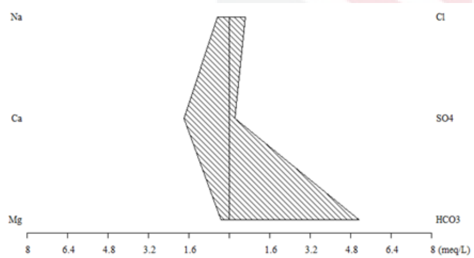
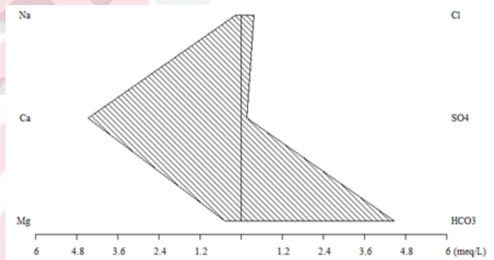

APPENDICES B

Station	Pre- monsoon	Post-monsoon
KW 1		
	Water type: Ca-HCO_3	Water type: Ca-HCO_3
KW 2		
	Water type: Ca-HCO_3	Water type: Ca-HCO_3

<table>
<tr><td>KW 3</td><td>
</td><td>
</td></tr>
<tr><td></td><td>Water type: Ca-HCO_3</td><td>Water type: Ca-HCO_3</td></tr>
<tr><td>KW 4</td><td>
</td><td>
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<tr><td></td><td>Water type: Ca-HCO_3</td><td>Water type: Ca-HCO_3</td></tr>
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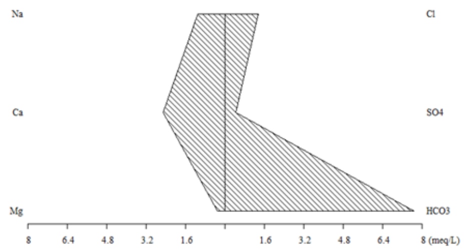
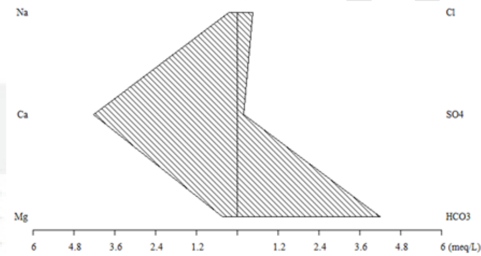
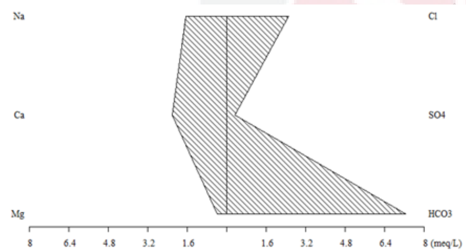
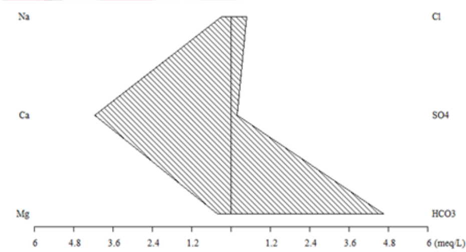
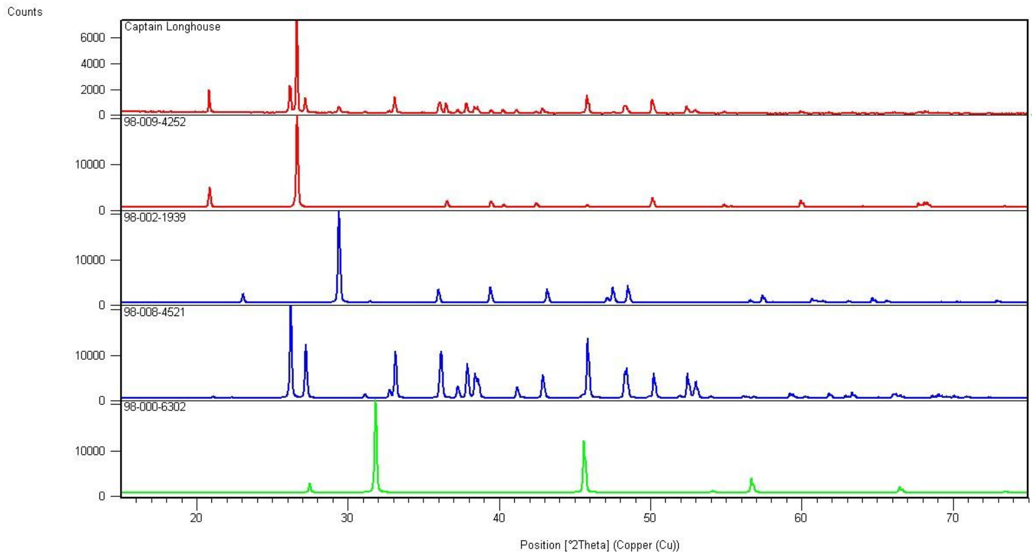
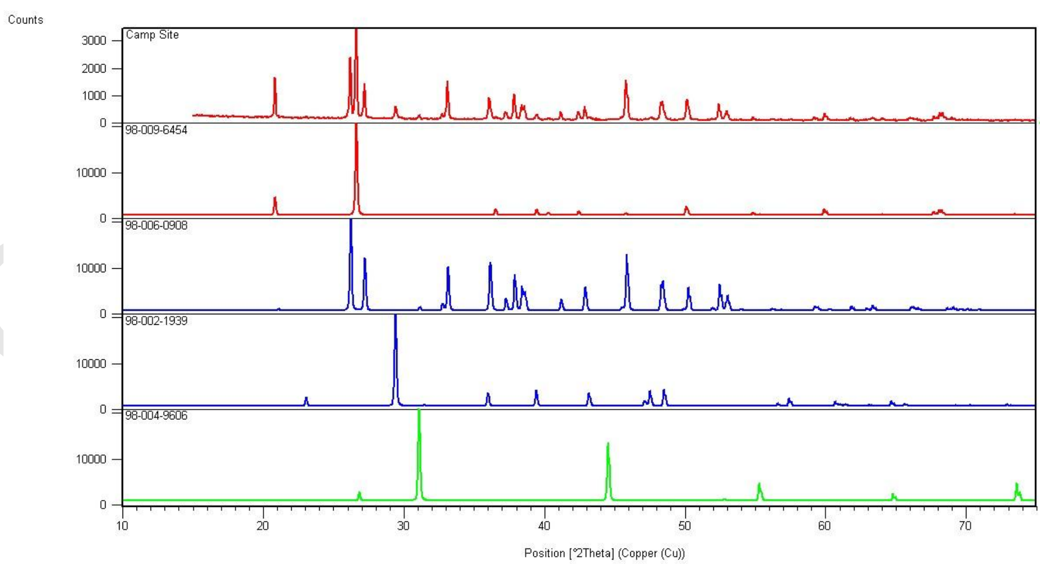
KW 5		
	Water type: Ca-HCO_3	Water type: Ca-HCO_3
KW 6		
	Water type: Na-HCO_3	Water type: Ca-HCO_3

APPENDICES C

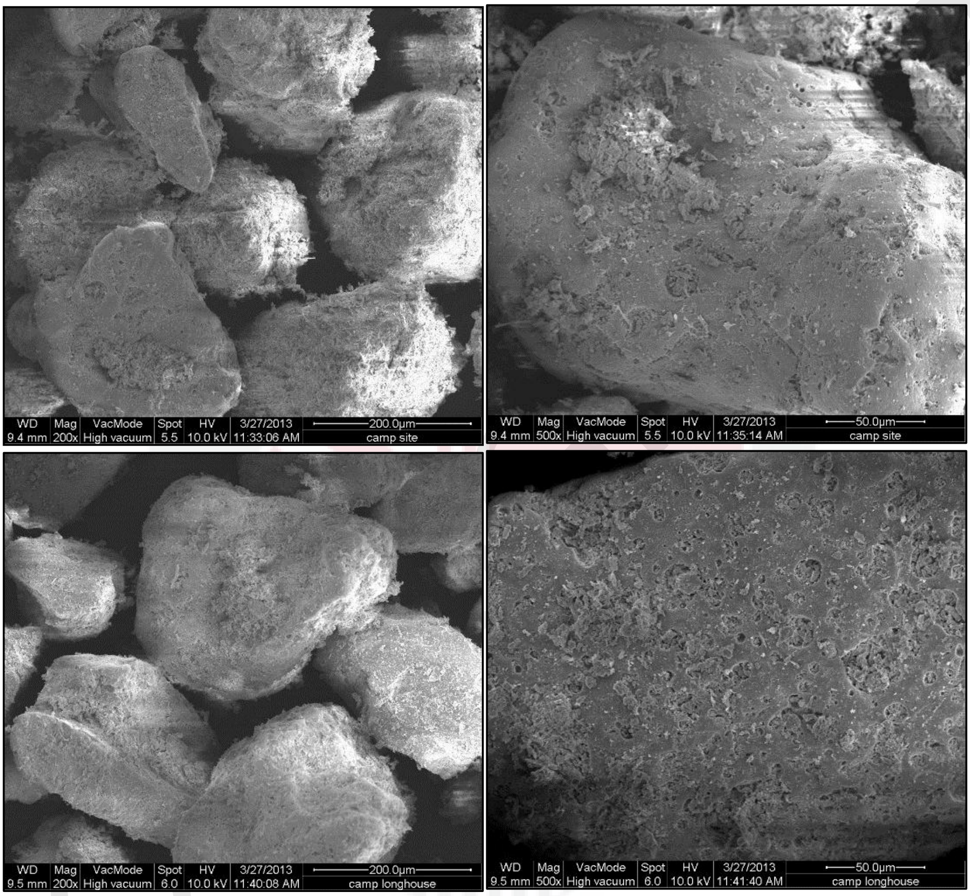
Example of XRD outputs





APPENDIX D

Example of SEM outputs



BIODATA OF STUDENT

Noorain Mohd Isa was born in 1987, in Alor Setar, Kedah. She grew up in Johor Bahru, eventually studying at the Sekolah Menengah Kebangsaan Agama Johor Bahru, Johor. She attended Johor Matriculation College in 2006 and continued with Bachelor degree at Universiti Putra Malaysia (UPM). She graduated with Bachelor degree of Science (Environment) in 2009. During her degree study, she attended an Internship Program and worked as a trainee in Palm Oil Factory in Maran, Pahang for three month. She finished her final year project regarding on river water quality.

After undergraduate study, she entered the PhD program at the UPM under the Environmental Hydrology and Hydrogeology course. She was offered a research grant by Universiti Putra Malaysia in year 2010 with research project entitled “Simulation of the Evolution of the Groundwater Hydrochemistry in Small Tropical Island”, with no project of 5523724 (07/11/09/696FR) under Fundamental Research Grant Scheme (FRGS). She also had been trained as a demonstrator and research assistance throughout the study. Currently, her PhD project involves the contribution of natural and anthropogenic influences toward the groundwater hydrochemistry. She managed to publish several journals in national and international levels. She also took part in numerous seminars, symposiums and conferences as a speaker as well as participant both in internally and abroad.

LIST OF PUBLICATIONS

Journals

Isa, N.M., Aris, A.Z., Lim, W.Y., Wan Sulaiman, W.N.A. and Praveena, S.M. (2014) Evaluation of heavy metal contamination in groundwater samples from Kapas Island, Terengganu, Malaysia. Arabian Journal of Geosciences 7(3), 1087-1100.

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Noorain Mohd Isa and Ahmad Zaharin Aris (2011) Controlling Factors of Hydrogeochemistry Mechanism in Sandy Coastal Aquifer at Kapas Island, Terengganu, Malaysia. The Second Asia-Pacific Coastal Aquifer Management Meeting : 18-21 October 2011, Jeju, Korea, Research Institute of Construction Technology, Dong-A University

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Chapter in book

Isa, N.M. and Aris, A.Z. (2014) Application of Geochemical and Geostatistical Analyses in Observing the Controlling Factors of Groundwater Compositions. Aris, A.Z., Tengku Ismail, T.H., Harun, R., Abdullah, A.M. and Ishak, M.Y. (eds), pp. 171-174, Springer Singapore.