

UNIVERSITI PUTRA MALAYSIA

CHARACTERIZATION OF GROUNDWATER QUALITY IN THE COASTAL AQUIFER OF TERENGGANU, MALAYSIA

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FPAS 2019 11



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By

ZAHIDI BIN HAMZAH

Thesis Submitted to the School of Graduates Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

October 2017

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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Rapid developments in agricultural, residential and industrial activities have increased pressure for the public water supply in Terengganu. Groundwater was touted to fulfil the demand thus commanded research on its quality through characterization study. The integration of geochemical methods such as multiple ion ratios and multivariate statistical analysis methods such as FA/PCA were carried out on 169 samples from 29 monitoring wells to characterize and to elucidate the factors and processes that influencing the geochemical composition of groundwater in coastal shallow aquifer of Terengganu. Spatial distribution maps of factors were obtained using geostatistical methods such as Ordinary Kriging. The samplings were carried out twice annually during the wetter Northeast Monsoon and drier Southwest Monsoon, respectively from 2006-2014. Generally, the groundwater is mildly acidic to alkaline in nature. Ca2+, Na+ and K+ ions were the dominant cations while HCO_3^- and Cl^- were the leading anions. The abundance of Ca^{2+} were contributed by carbonate and silicate weathering processes while HCO3⁻ and Cl⁻ were resulted from reverse ion exchange reaction as revealed by geochemical modelling using multiple ionic ratios. Saline intrusion probably cause the hydrogeochemical facies dominated by Ca-Mg-HCO3-Cl type based on Piper diagram plot. CI/HCO3 ratio further reinforced the influence of salinization was caused by seawater intrusion to the aquifer which slighty or moderately affected half (50.9%) of the groundwater samples. Therefore, groundwater in the study area constantly under threat of salinization mostly by seawater intrusion prominently in the major river confluences. Mapping of salinization affected area utilizing geostatistical analyses especially around the wells of Terengganu and Marang River Basin in the northeast, Dungun and Kemaman River confluence in southeast of study area were reliable with acceptable accuracy based on limited and sparsely located monitoring wells. Salinization, anthropogenic activities, reverse ion exchange, weathering processes, agricultural impact and seasonal were the regulating factors that influencing 63% of the major ion chemistry in study area as determined by factor analysis. Conclusively, the finding showed that for effective utilization, aquifer protection and prediction of changes, understanding the groundwater hydrogeochemical characteristics is crucial in order to minimize the effects of salinization and natural weathering processes. Reducing the anthropogenic pollution such as agriculture and urbanization impact also help to preserve and maintain groundwater resources for future usage. Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Sarjana Sains

PENCIRIAN KUALITI AIR BAWAH TANAH AKUIFER PANTAI DI TERENGGANU, MALAYSIA

Oleh

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Kemajuan pesat dalam aktiviti pertanian, kediaman dan industri telah meningkatkan beban kepada sumber air awam di Terengganu. Air tanah dicadangkan dapat memenuhi keperluan bekalan awam. Oleh itu, satu kajian ke atas ciri-ciri air tanah telah dilakukan untuk memastikan kualitinya. Integrasi kaedah analisis seperti kaedah nisbah ion pelbagai dan analisis statistik multi-variate seperti FA/PCA telah dilakukan ke atas 169 sampel daripada 29 telaga pemantauan untuk mencirikan dan mengenalpasti faktorfaktor dan proses yang mempengaruhi komposisi geokimia air tanah di akuifer cetek pantai Terengganu. Kaedah geostatistik seperti Ordinary Kriging digunakan untuk membuat peta taburan spatial faktor-faktor. Pensampelan dilakukan dua kali setahun bermula dari tahun 2006-2014 semasa musim Monsun Timur Laut (tengkujuh) dan Monsun Barat Daya (kering) masingmasing. Secara amnya, air tanah adalah sedikit berasid hingga beralkali secara semulajadi. Ion-ion Ca²⁺, Na⁺ dan K⁺ adalah kation dominan manakala HCO₃⁻ dan Cl⁻ adalah anion dominan. Kehadiran ion Ca²⁺ yang banyak adalah disumbangkan oleh proses luluhawa karbonat dan silikat sementara ion HCO3 dan Cl adalah berpunca daripada tindak balas pertukaran ion berbalik sebagaimana yang digambarkan oleh pemodelan hidrokimia menggunakan kaedah nisbah ion pelbagai. Plot rajah Piper menggambarkan berlakunya intrusi air masin yang menyebabkan fasis air tanah didominasi oleh jenis Ca-Mg-HCO₃-Cl. Nisbah Cl/HCO₃ menyokong akan pengaruh kemasinan disebabkan oleh pencampuran air laut dengan air tanah di dalam akuifer yang memberi sedikit kesan kepada separuh (50.9%) daripada sampel air tanah. Oleh itu, air tanah di kawasan kajian terdedah kepada ancaman kemasinan yang diakibatkan oleh kemasukan air laut terutamanya di kawasan muara sungai-sungai utama. Ketepatan dan kebolehpercayaan pemetaan kawasan terjejas akibat kemasinan menggunakan analisis geostatistik terutamanya di muara Sungai Terengganu dan Marang di utara kawasan kajian, Sungai Dungun dan Sungai Kemaman di selatan kawasan kajian berdasarkan telaga

pemantauan yang terhad dan tersebar luas boleh diterima pakai. Analisis faktor mengenalpasti kemasinan, aktiviti manusia, tindakbalas ion berbalik, proses luluhawa, kesan pertanian dan musim adalah faktor-faktor yang mempengaruhi 63% kesan geokimia ke aras air tanah di kawasan kajian. Kesimpulannya, kajian ini mendapati untuk pemahaman ke atas ciri-ciri hidrogeokimia air tanah adalah penting bagi memastikan ia dapat digunakan dengan mampan, akuifer dilindungi daripada ancaman kemasinan dan kesan luluhawa semulajadi. Pengurangan pencemaran yang diakibatkan oleh kegiatan manusia seperti pertanian dan perbandaran juga dapat memastikan pemeliharaan dan perlindungan akuifer air tanah untuk kegunaan pada masa akan datang.

ACKNOWLEDGEMENTS

Alhamdulillah...

Praise to Allah the Almighty for His blessings for giving me strength and willpower in order to complete the research as heart breaking events occurred during the study period. Firstly, the joy that arises by the birth of my 6th children, then the sorrow followed afterward caused by the death of my beloved mother two weeks later, is hard to bear. Fortunately with the steadfast support and unselfish encouragement of my beloved wife Norizan Yaso', coupled with adoration and affection shown by my magnificent six children namely Muhammad Hafidz Najmi, Nur Irdina Husna, Muhammad Afiq Aiman, Nur Hanin Najwa, Nur Inarah Insyirah and Muhammad Afif Zafran, I managed to to complete this MSc thesis despite many obstacles and challenges that arise.

Sincere million thanks to my supervisor, Prof. Dr. Ahmad Zaharin Aris for his coaching, fruitful discussion and valuable suggestions as well as continuous inspiration during thesis writing process. Million thanks to my co-supervisor committee Associate Prof. Dr. Mohammad Firuz Ramli, who have given valuable advice, clear instruction, alternative opinions and apparent vision. Gratefulness for my external co-supervisor, Associate Prof. Dr. Hafizan Juahir for his sparkling ideas on data presentations and motivation.

Thousands of thanks to my former boss Mohd Pauzi Abdullah, Director of DMG Terengganu Mohd Zukeri Ab Ghani, Abdul Hadi Abdul Rahman and Suhaimizi Yusoff for their invaluable encouragements, endless motivations, technical support and productive suggestions.

Besides, I would like to express my gratitude to my current employer the Department of Minerals and Geoscience (DMG), Meteorological Department (MET Malaysia) and Department of Rural and Town Planning (PLAN Malaysia) for providing the necessary data and information that essential for the study. I also would like to express my gratitude to the Government of Malaysia through the Public Services Department (PSD) for their sponsorship and financial assistances.

Last but not least, my warmest thanks to my colleagues and friends particularly in the DMG Terengganu, DMG headquarters in Kuala Lumpur, DMG state offices, NAHRIM, PHK Cycling Team (Kelantan), my brothers of Krazy Monkeys and Universiti Putra Malaysia (UPM) especially my classmates; Dr. Tahoora, Dr. Adamu, Dr. Noorain, Nordiani Sidi, Farhanna Mokhtar, Dr. Looi Ley Juen, Azrul Normi, Anuar Shafei, Dr. Shah Cristiani, Tuan Fauzan, Hanisah Nasir, Nasyitah, Afifah and others; for all your non-stop supports, magnanimous assistances and inspirations in completing the manuscript.

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LIST OF ABBREVIATIONS

Al	aluminium
ASE	average standard error
Ва	barium
Ca ²⁺	calcium
Cl	chloride
CO ₃ ²⁻	carbonate
DMG	Department of Minerals and Geoscience, Malaysia
EC	electrical conductivity
F	fluoride
FA	factor analysis
Fe	ferum
GIS	geographic information system
HCO ₃	bicarbonate
HDPE	high density polyethylene
K ⁺	potassium
ME	mean error
Mg ²⁺	magnesium
MINGEOSIS	Minerals and Geoscience Information System
MLD	million litre per day
Mn	manganese
Na⁺	sodium
NABC	needs, approach, benefits and challenges analyses
NH4 ⁺	ammonium
NO ₃ ⁻	nitrate
ОК	ordinary kriging
Ρ	phosphorus
PCA	principal component analysis
RMS	root mean square
RMSS	root mean square standardize
SI	saturation indices
SiO ₂	silica
SO4 ²⁻	sulphate
Sr	strontium
SWOT	strength, weakness, opportunity and threat analyses
TDS	total dissolved solid

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CHAPTER 1

INTRODUCTION

Groundwater is becoming the main water source for domestic, agricultural and other human activities in many places. It is also the primary resource for drinking water in the world amounting about 20% of the global water withdrawals (Yu et al., 2013). Groundwater is used as main water supply for drinking purposes and domestic consumption by 130 million U.S.A residents, 8 million Canadians (Kidd, 2002) and 80% of India's rural population (Reza et al., 2009) where rapid population growth and intensive economic activities are pushing the demand for fresh water to its limits (Kura et al., 2013). Groundwater also been utilised by residents of Republic of Korea, the Netherlands (Isa et al., 2014) and Northen China particularly in most arid and semi-arid areas (Yu et al., 2013).

Groundwater also fulfilled as many as 80% of New Zealand's (Raiber et al., 2012) and Iran's (Sheikhy Narany et al., 2014) agricultures' water need due to its reliability and availability to point of need at a lower cost.

Meanwhile, Malaysia's water demand for industries, domestic and other uses are predicted to increase to about 63% from the year of 2000 to 2050 (Department of Statistics, 2012). Malaysia's water consumption for domestic and non-domestic usage in year 2012 stands at 9,532 MLD and increased about 9.58% to 10,445 MLD in 2015. Meanwhile, Terengganu's water usage was at 389 MLD in year 2012 and rise to 417 MLD in 2014, an increment of 7.20%. In 2015, domestic water usage stood at 428 MLD (SPAN, 2016).

Groundwater has the potential to fulfill the increasing need as a sustainable water source as it found in abundant, easily accessible, and drought resistant. The volume of groundwater in the aquifers throughout Malaysia is estimated at 5,000 billion m³ (Mohamed, 1999). Groundwater only accounted about 10% from present water uses in Malaysia and largely limited to rural and remote area (Abdul Razak & Mohammed Hatta, 2009). However, in neighboring state of Kelantan, 70% of the public water supply of the state is provided by groundwater (Abdul Razak & Mohammed Hatta, 2009) while in Terengganu, its usage still limited to small islands (Isa et al., 2014), agriculture farms and some educational campuses (Hamzah & Ismail, 2012a).

Since the groundwater become more important as water resources for future generations, understanding of aquifer hydraulic properties (Masoud, 2014) and hydro-chemical characteristics (Islam et al., 2016; Pradhan & Pirasteh, 2011;

Ratan et al., 2016) of groundwater is crucial for effective utilization, aquifer protection and to predict changes in groundwater environments.

1.1 Problem Statement

Water quality analysis is an important issue in groundwater studies due to dynamic nature of its resources (Devic et al., 2014; Mustapha et al., 2013; Tlili-Zrelli et al., 2012). Variation in groundwater quality in area is a function of physical and chemical parameters (Lim et al., 2013; Muqtada et al., 2012) that are greatly influenced by geological formations (Giridharan et al., 2009; Jabal & Abustan, 2014; Yu et al., 2013) and anthropogenic activities (Belkhiri & Mouni, 2012; Devic et al., 2014; Jiang et al., 2009) such as expansion of irrigation activities (Ketata & Gueddari, 2012; Rahman et al., 2012), industrialization (Vasanthavigar et al., 2012; Wu & Sun, 2015) and urbanization (Onodera et al., 2008, 2009; Rao et al., 2013). Facing the combination of many factors, large uncertainties exist in determining the key processes controlling the ongoing hydrochemical transformations in the groundwater (Ratan et al., 2016; Wanda et al., 2013a; Yu et al., 2013).

One of the fundamental threat that encountered by groundwater aquifers is salt water intrusion or salinization particularly in coastal areas. The affected groundwater would contain high levels of sodium (Na), chloride (Cl) and TDS, which indicated salt water intrusion (Rao et al., 2013). Research on effect caused by salt water intrusion in Malaysia were concentrated mainly on oceanic aquifer system (Abdullah et al., 2010; Praveena et al., 2010) that supplying water needed by tourism industry such as in Manukan (Aris et al., 2008; Aris et al., 2009; Aris et al., 2010, 2012; Praveena et al., 2011; Praveena et al., 2010; Praveena et al., 2012; Praveena et al., 2010), Sipadan (Abdullah et al., 2008; Aris et al., 2009; Aris et al., 2011) and Kapas Islands (Isa & Aris, 2012; Isa et al., 2014; Isa et al., 2012a, 2012b; Kura et al., 2014; Kura et al., 2013). Surface water such as rivers (Lim et al., 2012; Sultan & Shazili, 2010) and dams (Shamsuddin et al., 2013) were crucial as major water source for public consumption as the nation which located in tropical zone received heavy rainfall averaging to 3,000 mm annually (DOSM, 2010a). However, rapid development in urbanization, industrialization and agriculture due to economic growth will require a sustainable supply of water (Chan, 2012; Saimy & Yusof, 2013) which could be provided by groundwater as conjunctive water source (Shamsuddin et al., 2013; Stek, 2008).

Therefore, a systematized study of groundwater properties and ionic concentrations had to be carried out in order to provide crucial information about groundwater hydrochemistry, water-rock interaction and hydrogeochemical processes that contribute to its current state in the aquifer system (Jabal & Abustan, 2014; Thivya et al., 2014) in order to preserve the water quality for future uses. The SWOT (Strength, Weakness, Opportunity

and Threat) analysis further highlighted the necessity of the current study for sustainable development in coastal aquifers (Table 1.1).

Table 1.1:	The	Strength,	Weakness,	Opportunity	and	Threat
	(SW	OT) analysis	for groundw	vater characte	rizatio	n

	Outputs
Strength	 Abundant groundwater in the study area due to heavy rainfall of more than 3,000 mm annually. Groundwater is easily assessable as it could be found in shallow wells less than 50m depth. Groundwater is drought resistant as it is available throughout the year.
	 Water consumption increases 6% annually and groundwater is capable to fulfil the demand. Groundwater utilization is still in infancy stage in the study area.
Weakness	 Groundwater monitoring wells are limited and sparsely located. Groundwater aquifers are exposed to various land use changes and contamination. Groundwater aquifer risk pollution at different degrees. There is an information gap on groundwater's aquifer particularly in the study area. There are inadequate information on groundwater quality especially in Malaysia and Terengganu. The findings are unique to Terengganu area and are confined to the context in which the study was carried out. Limited knowledge acquired especially on aquifer properties such as hydraulic conductivity and well drawdown.
Opportunity	 The study will aid to address the lack of information on groundwater geochemistry. Information gathered from the study will help the authorities in decision making on groundwater management Prevention measures could be taken to mitigate pollution from infiltrated into groundwater aquifers. Devastating environmental effect on groundwater could be reduced to minimum. Water security for future usage would be ensured.
Threat	 Groundwater aquifers especially in coastal areas are under constant threat of salt water intrusion or salinization. Depleted or contaminated groundwater resources would halt the rapid development in urbanization, industrialization and agriculture activities.

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1.2 Research Objectives

The main objective of the study was to characterize the groundwater and,

- i. To identify the hydrogeochemical facies of groundwater
- ii. To elucidate the factors dominating the groundwater quality and,
- iii. To assess the spatial variation of groundwater salinity

1.3 Research Questions

The study aimed to answer the research questions as below:

- i. What is the current groundwater quality in the study area?
- ii. What are the factors controlling the groundwater hydrochemistry conditions?
- iii. How vulnerable is the aquifer to salinization and other contaminations?

1.4 Scope and limitations of research

Principally this study encompasses the following scopes:

- i. This study focuses on the distribution of the elements such as of major ions: Ca^{2+} , Mg^{2+} , K^+ , CO_3^{2-} , HCO_3^- , Cl^- , SO_4^{2-} , NO_3^- , F, P, NH_4^+ and trace elements: Al, Fe, Mn, Zn, Sr, Ba in the groundwater wells along the coastal aquifers in the state of Terengganu, Malaysia.
- ii. The groundwater samples for this study were selected from 29 tube wells from coastal area that were collected during Southwestmonsoon and Northeast-monsoon season (refer Appendices A1). The wells were drilled at different depth between 5.0 m and 54.0 m from mean sea level (MSL). Therefore, the findings are unique to Terengganu area and are confined to the context in which the study was carried out.

1.5 Significance of research

Earlier reports on groundwater monitoring and assessment in Terengganu area (Hamzah & Ismail, 2012a; Hamzah & Ismail, 2012b; Hamzah & Ismail, 2013a) only relied on graphical representations such as Stiff and Piper diagrams for major ionic components. The graphs which are useful in visually describing and classifying water composition into identifiable groups (Subyani, 2010) only consider selected major water constituents in determining the groundwater type (Triki et al., 2013) and were limited to two dimensions only (Liu et al., 2003).

However, the use of geochemical modelling which utilizing combined approach of paired ionic ratios such as Ca^{2+}/Mg^{2+} , $Ca^{2+}+Mg^{2+}/HCO_3^-+SO_4^{2+}$, $Ca^{2+}+Mg^{2+}/Cl^-$, Na^+/Cl^- versus EC, Cl^-/HCO_3^- versus Cl^- , Na^+/Cl^- , Gibbs plot; $(Na^++K^+/Na^++K^++Ca^{2+}$ versus TDS), $(Cl^-/Cl^-+HCO_3^-$ versus TDS) and graphical methods in the elucidation of water quality indices (Rao et al., 2013; Ravikumar et al., 2013; Singaraja et al., 2014) will give better insight of the evolution of processes in groundwater chemistry (Glynn & Plummer, 2005; Kuldip-singh et al., 2011; Singh et al., 2012; Gimenez-Forcada & Vega-Alegre, 2015). The output from geochemical modelling software PHREEQC (Parkhurst & Appelo 1999), which asses the equilibrium state of groundwater and minerals particularly calcite, aragonite and dolomite will be employed to validate the geochemical reactions that took place in the aquifer (Aris et al., 2011; Salifu et al., 2012).

Massive groundwater dataset would require the use of multivariate statistical analysis such as factor analysis (FA) and principal component analysis (PCA) to identify ions or factors that controlling hydrochemistry in the groundwater aquifer along coastal areas (Gangadharan et al., 2016; Rekha et al., 2013; Triki et al., 2013), without neglecting valuable information such as spatial-temporal patterns (Arslan, 2014; Mustapha et al., 2012; Schot and Pieber, 2012), important parameters and hidden pollution sources (Adhikary et al., 2010; Jayalakshmi et al., 2014; Lim et al., 2012). Furthermore, the clarification can be simplified by using rotational procedures (Lim et al., 2012; Semar et al., 2013) to identify similarities between variables (Gangadharan et al., 2016; Lin et al., 2012; Mustapha & Aris, 2012b) in order to segregate the effect of natural activities (Jiang et al., 2009; Qiao et al., 2011) and anthropogenic pollution (Devic et al., 2014; van Geldern et al., 2014; Vissers & Van Der Perk, 2008) on the groundwater hydrogeochemistry.

The spatial distribution of groundwater hydrogeochemistry could be represented using geostatistical analysis (Cay & Uyan, 2009; Deutsch & Journel, 1998; Marko et al., 2013) such as ordinary kriging interpolation method. The accuracy of the interpolated value could be estimate using cross-validation technique (Triki et al., 2013; Uyan & Cay, 2013; Yimit et al., 2011a).

Recently, there is growing trend of using integrated approach which combining the graphical interface, geochemical modelling, ionic ratios, multivariate statistical analysis and geostatistical analysis in order to obtain important hydrogeochemical information in complex system

Contemporary researchers have integrated their approach in deciphering principal information from huge data set of groundwater samples from a complex system (Islami et al., 2012; Nazri et al., 2012; Sheikhy Narany et al., 2014). The combined application of ionic ratios, geochemical modelling using PHREEQC software, multivariate statistical analysis and geographical information system (GIS) have increased due to its advantages of characterizing and elucidating the various physiochemical processes that occurred in aquifer complex (Abdalla and Scheytt, 2012; Agoubi et al., 2013; Batayneh and Al-Taani, 2015; Rao et al., 2013; Hassan et al., 2014; Islami et al., 2012; Khashogji and Maghraby, 2012; Kura et al., 2014; Magesh et al., 2012; Masoud, 2014; Sheikhy Narany et al., 2014a, 2014b, 2014d; Swarna Latha and Nageswara Rao, 2012; Kura et al., 2013; Wanda et al., 2013b).

The NABC (Needs, Approach, Benefits and Challenges) analysis further verified the necessity of hydrogeochemical evolution research for sustainable development in coastal aquifers (Table 1.2). This analysis simplifies assessment for conditions as transpires in previous literatures and facilitate this study to fill the information gap or constraint as faced by other researches.

These integrated methods would offer a better understanding of groundwater quality in the state of Terengganu, allow identification of the possible factors/sources that influence the groundwater systems and present a valuable tool for reliable management of water resources as well as rapid solutions to improve groundwater quality.

	Outputs
Needs	 Water consumption increases 6% annually and groundwater is essential to fulfil the demand. Information on current condition of groundwater quality is needed to assess its suitability. Identifying factors or hidden pollution sources that influencing the groundwater hydrochemistry is essential for sustainable management of water resources. Variation in groundwater quality either in spatial or temporal is necessary in order to distinguish effect of natural activities and anthropogenic contribution.
Approach	 Analyse the groundwater samples using combined chemistry for major ion concentration. Ionic ratio assists to identify episodes of seawater intrusion. Statistical approach using multivariate analysis to streamlined influencing factors. GIS application to predict the attribute of physiochemical variables in the study well and its spatial distribution.
Benefits	 The study will aid to address the lack of information or groundwater geochemistry. Information gathered from the study will help the authorities in decision making on groundwater management. Prevention measures could be taken to mitigate pollution from infiltrated into groundwater aquifers. Devastating environmental effect on groundwater could be reduced to minimum.
Challenges	 There are inadequate information on groundwate quality especially in Malaysia and Terengganu. There is an information gap on groundwater's aquife particularly in the study area. Limited knowledge acquired especially on aquife properties. Monitoring groundwater wells are sparsely located.

Table 1.2:The Needs, Approach, Benefits and Challenges (NABC)outputsforrecentliteraturesongroundwatercharacterization

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1.6 The structure of the thesis

This thesis is divided into 5 chapters. The summary of each chapter is:

i. CHAPTER 1: INTRODUCTION.

This chapter briefly discussed the problem statement of the study, research gap, SWOT analysis, study objectives, its scope and limitations, and NABC analysis of the study. Also included in this chapter are significance of the study and the overall structure of the thesis.

ii. CHAPTER 2: LITERATURE REVIEW.

This chapter critically analysed the related publications relevant to the groundwater geochemistry. It means to illustrate the importance of water quality, limitation on characterization using graphical diagram, advantages of using multivariate statistical analyses such as Factor Analysis (FA)/ Principal Component Analysis (PCA), evaluating the relationship between groundwater and its surrounding aquifer materials using Saturation Indices (SI) and projecting or mapping the spatial variation of unsampling location using geostatistical analyst.

iii. CHAPTER 3: GROUNDWATER QUALITY ASSESSMENT USING INTEGRATED GEOCHEMICAL METHODS, MULTIVARIATE STATISTICAL ANALYSIS, AND GEOSTATISTICAL TECHNIQUE IN SHALLOW COASTAL AQUIFER OF TERENGGANU, MALAYSIA AND METHODS.

This chapter discussed on several methods that were utilized in order to elucidate processes that governing groundwater chemistry. This study employed combined approach using geochemical, multivariate statistical analysis and geostatistical analysis. Geochemical plots identified several processes such as carbonate and silicate dissolution while multivariate statistical analyses categorized several factors that influence the groundwater geochemistry. The spatial variations of those processes and factors influencing the groundwater geochemistry were plotted using geostatistical analyses.

iv. CHAPTER 4: GEOSTATISTICAL ANALYSIS OF GROUNDWATER SALINITY USING ORDINARY KRIGING IN TERENGGANU AND MARANG RIVER BASIN, MALAYSIA.

This chapter assessed on 11 different semivariogram models in order to obtain best fitted models that will predict the spatial distribution and uncertainty of groundwater salinity with acceptable accuracy based on limited and sparsely located monitoring wells in the study area.

v. CHAPTER 5: SUMMARY, CONCLUSION AND RECOMMENDATIONS FOR FUTURE RESEARCH.

vi. This chapter presented the conclusion from the research, recommendations and further research for the study area. Findings from chapter 3 and chapter 4 that addressed all the research objectives were summarized in this chapter. Recommendations that will advance future research were also stated in this chapter. Parts of the research that have been published in journals were provided.

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