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GEOSTATISTICAL MAPPING OF HEAVY METALS (Cd, Cr & Zn) DISTRIBUTION IN THE SEDIMENTS OF SALUT LAGOON, SABAH

WONG VUI CHUNG @ WEBSTER WONG

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**SCHOOL OF SCIENCE AND TECHNOLOGY
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DECLARATION

The materials in this dissertation are original except for quotations, excerpts, summaries and references, which have been duly acknowledged.

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ABSTRAK

PEMETAAN GEOSTATISTIK TABURAN LOGAM-LOGAM BERAT (Cd, Cr & Zn) DALAM SEDIMEN DI LAGUN SALUT, SABAH

Lima puluh (50) sample sedimen permukaan yang digeokod telah dikutip dari Lagun Salut yang terletak 15 km Timur Laut Kota Kinabalu, Sabah. Jumlah sedimen tersebut telah dianalisis dengan menggunakan Spektrofotometer Serapan Atom (SSA). Objektif kajian tersebut adalah untuk menilai kepekatan serta taburan spatial logam-logam berat (Cd, Cr dan Zn) serta membandingkan peta-peta yang dihasilkan dengan menggunakan teknik geostatistik iaitu *ordinary kriging* dan *indicator kriging*. Penilaian atas taburan logam berat dalam jumlah sedimen lagun Salut menunjukkan bahawa kepekatan min bagi Cd, Cr and Zn adalah masing-masing 6.1 mg kg^{-1}, 15.5 mg kg^{-1} dan 262 mg kg^{-1}. Taburan spatial logam berat dalam sedimen lagun Salut adalah terutamanya dikawal oleh factor-faktor fiziko-kimia dan corak penggunaan tanah. Dengan mempertimbangkan geologi sedimen dan perbandingan dengan kawasan lain, sedimen permukaan di Lagun Salut mencatatkan kepekatan Cd dan Zn yang tinggi. Kedua-dua peta *ordinary kriging* dan *indicator kriging* bagi taburan kepekatan Cd, Cr and Zn telah dihasilkan. Peta *Indicator kriging* telah terbukti lebih sesuai digunakan dalam membuat keputusan dan tujuan pengurusan logam berat.

ABSTRACT

GEOSTATISTICAL MAPPING OF HEAVY METALS (Cd, Cr & Zn) DISTRIBUTION IN THE SEDIMENTS OF SALUT LAGOON, SABAH

Fifty (50) geocoded surface sediment samples were taken from Salut Lagoon, which is situated about 15 km northeast of Kota Kinabalu, Sabah. These total sediments were analyzed by using Atomic Absorption Spectrometry method (AAS). The objective of this study was to assess the concentration and the spatial distribution of the heavy metal (Cd, Cr and Zn), and compare maps generated using geostatistical techniques i.e. ordinary and indicator kriging. Assessment on the concentration and spatial distribution of heavy metals in the surface sediments of Salut Lagoon showed that the mean concentration for Cd, Cr and Zn are 6.1 mg kg^{-1}, 15.5 mg kg^{-1} and 262 mg kg^{-1} respectively. The spatial distributions of the heavy metals in the surface sediments of Salut Lagoon are mainly controlled by physico-chemical factors and landuse patterns. Considering the sedimentary geology of the area, and comparison with other areas indicates that the Salut Lagoon shows high levels of Cd and Zn in the surface sediments. Both ordinary and indicator kriging maps for Cd, Cr and Zn concentrations were generated. Indicator kriging maps have proven to be more robust and suitable for decision-making and heavy metals management purposes.

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LIST OF ABBREVIATIONS AND SYMBOLS

$mg\ kg^{-1}$	milligram per kilogram
ppm	parts per million
km	kilometer
%	percentage
>	more than
<	less than
OK	Ordinary kriging
IK	Indicator kriging
USEPA	United States Environmental Protection Agency
KKIP	Kota Kinabalu Industrial Park
DOE	Department of Environmental
ECD	Environmental Conservation Department
DBKK	Dewan Bandaraya Kota Kinabalu

CHAPTER 1

INTRODUCTION

1.1 Introduction

Coastal zones have been the centre of intense housing, agricultural, fishery, industrial, and tourism development and estuaries perhaps are the most polluted of all marine environments (Garrison, 2005) mainly due to its unique characteristics for industrial location. Estuaries provide extensive flat land for setting up industrial bases and allow port developments which in turn ease the import and export of raw materials. They normally have high potential of water supply for industrial uses and historically they have been perceived as natural "waste disposal systems" (French, 1997). Thus the anthropogenic impact on the coastal and estuarine environmental is far-reaching and the uncontrolled development and increasing human activities have created a threat to these unique and economically valuable ecosystems. Among the pollutants being released into estuaries environments are the heavy metals which are particularly toxic in their chemically combined forms and some, notably mercury, are toxic in the elemental form (McBride, 1994).

Heavy metals are naturally occurring metals from earth materials and volcanic emanations. However these metals can also be introduced to the environment from anthropogenic sources. The anthropogenic sources of heavy metal can be point source or non-point source. Point sources may include discharges of waste effluents from factories, contamination from a landfill site or mining and non-point source may be due to agricultural practices surrounding the area. These pollutants can be transported to the estuaries by a variety of pathways and mediums. Sediments are by far the most significant medium in heavy metals distributions.

The behaviour of heavy metals in sediments depends on chemical and physico-chemical as well as biological factors associated with the microbial activities. It has been recognized that textural characteristics, organic matter content, mineralogy composition as well as depositional environment of the sediments play an important role in the concentration of heavy metals (Singh *et al.*, 1999; Rubio *et al.*, 2000).

Although some heavy metals are essential (e.g. haemocyanin – blood pigment of crustaceans, contains Cu) but increase concentration to toxic level will impair biological processes such as enzyme function (Boaden & Seed, 1985). Excessive concentrations of heavy metals in the sediments can affect marine organism and pose risk to human through the food web. These heavy metals can be absorbed by organisms from the surrounding water or ingested with their food. Thus heavy metals have a high tendency to accumulate in the organism (bioaccumulation). They often accumulate in the nervous system and brain, causing behavioural disorder and diverse neurological problems, such as the Minamata Diseases. They may also impair growth development and reproduction system, damage organs or disrupt the immune system. The adverse biological effects are complex and potentially fatal (Botkin & Keller, 2003).

The detrimental effects caused by the introduction of heavy metal pollutant into the coastal zone such as estuary is well known. Assessment and monitoring of these pollutants in environment like estuary is required to better manage and minimize if not totally eliminate these pollutants. Good management decision based on accurate risk assessment involved good understanding of the spatial variation and distribution of the variable of interest. However, long term assessment and monitoring is often synonymous with huge budget. Thus, an unbiased and good estimation method is required to quantify and model the variation and performs interpolation at un-sampled location and to delimit zones that need remedial treatment.

In this regard, geostatistics has proved to be a good estimation method in studying the environmental pollutants (Amstrong, 1998) especially when there is a budget constraint or limited resources. Geostatistics has been widely used in the mining sector for the past 40 years since its development in the 1960s due to the need for a methodology in evaluating recoverable reserves on ore deposits (Goovaerts, 1997). In recent years, geostatistics has also been successfully applied in other disciplines such as soil science, petroleum, hydrology, oceanography, as well as environmental science because geostatistical method incorporate the spatial and temporal aspect of the variables of interest.

Kriging method is one of the geostatistical methods that are widely used. It is an estimation method which gives the best unbiased linear estimates of point values or block averages. Kriging also proved to be an exact interpolator because it takes into account the number of samples, quality of the variables, spatial relationship of the samples (position and distance), and also their spatial continuity (Amstrong, 1998). There are two kinds of spatial analysis in geostatistics, i.e. spatial interpolation and uncertainty assessment. Commonly used spatial interpolator is Ordinary Kriging (OK) while Indicator Kriging (IK) has been applied in uncertainty assessments. The advantages of both methods are elaborated in Chapter 2.

Risk map generated from an appropriate kriging technique is useful in good decision making and management especially in managing high risk areas of contamination that needs immediate remedial action. Taking into account the advantages of both the OK and IK techniques, it is the objective of this study to assess the suitability of these two techniques in mapping the distribution of heavy metals in an estuary setting such as Salut Lagoon.

1.2 Background of Research Location

Salut lagoon is located about 15km northeast of Kota Kinabalu (Figure 1.1), on the west coast of Sabah. Towards the west, the Mengkabong-Salut Lagoon systems developed into a five-kilometre beach before open up to the South China Sea, whereas to the east of the system is bordered by mangrove buffer, which changes into a secondary vegetation further inland, commonly found in the west coast of Sabah. The lagoon systems comprise the best mangrove forest remaining near Kota Kinabalu, Sabah and have been identified as important nursery and feeding grounds for fish and shellfish and of high eco-tourism potential (Gassner *et al.*, 2004).

With the expansion of KKIP just next to Salut Lagoon as well as population increase and extensive resource exploitation such as quarrying and industry developments nearby, more pollutants will be brought into the estuary. These toxic pollutants will subsequently be accumulated in the estuary. Hence, an appropriate estimation technique is required to evaluate the level and distribution of the contamination in the estuary in order to draw up a mitigation/management measures as well as for monitoring purposes.

The Mengkabong-Salut Lagoon Area has also been identified as a project area by Global Environmental Centre with the objective to improve the quality and status of biodiversity through community participated river management. Universiti Malaysia Sabah (UMS) was given the responsibility as the lead agency for Working Group 1-Monitoring, with the objective to monitor water quality, habitat and hydrology in the Salut-Mengkabong estuary. The group focuses on the caring capacity of the lagoon systems, which is related to the anthropogenic utilisation on the available resources and services in the lagoon or their catchments, and has since started the survey in 2004 (Gassner *et al.*, 2004).

1.3 Research Statement

For environmental management of pollutants, the concentration and distribution of the pollutants need to be quantified. As most environmental studies have the constraint of funding and manpower, maximum information with minimum sample density is needed. In many applications, the basic tool in geostatistics, the variogram

(Wackernagel, 1995 ; Goovaerts, 1997; Amstrong, 1998), is used to quantify spatial correlation between observations and can be used to estimate values at un-sampled points. However it is vital to choose the best geostatistical estimation technique for a particular environmental pollutant at a study site. In this research, the heavy metal concentrations in the surface sediments of Salut Lagoon will be studied by using geostatistical mapping techniques.

1.4 Objectives

The objective of the study is to assess the concentration and distribution of the heavy metals (Cd, Cr & Zn) and create spatial distribution maps using two different kriging methods for the management of heavy metal contamination.

To accomplish the above objective the following tasks will be carried out:

a. To determine the concentration of Cd, Cr, and Zn in surface sediments of the Salut Lagoon.

b. To model the spatial distribution of the heavy metals (Cd, Cr, & Zn) using variography.

c. To create surface map of the heavy metals (Cd, Cr, & Zn) using Ordinary Kriging and Indicator Kriging.

1.5 Scope of Work

To accomplish the objective of this research, a total of 50 sediment samples were collected in Salut Lagoon. The samples were analyzed for heavy metals (Cd, Cr, & Zn) by using Atomic Absorption Spectrometer (AAS) at Minerals and Geoscience Department.

For the purpose of geostatistics, 40 samples were used for variography and kriging (ordinary kriging and indicator kriging) while 10 samples were used as validation set. Estimated values at the locations of the 10 samples were compared with the laboratory results to test the appropriateness of the two kriging techniques used. Software such as *Variowin 2.21* and *Surfer 7* were used to do the variography, kriging and construct the heavy metals spatial distributions maps.

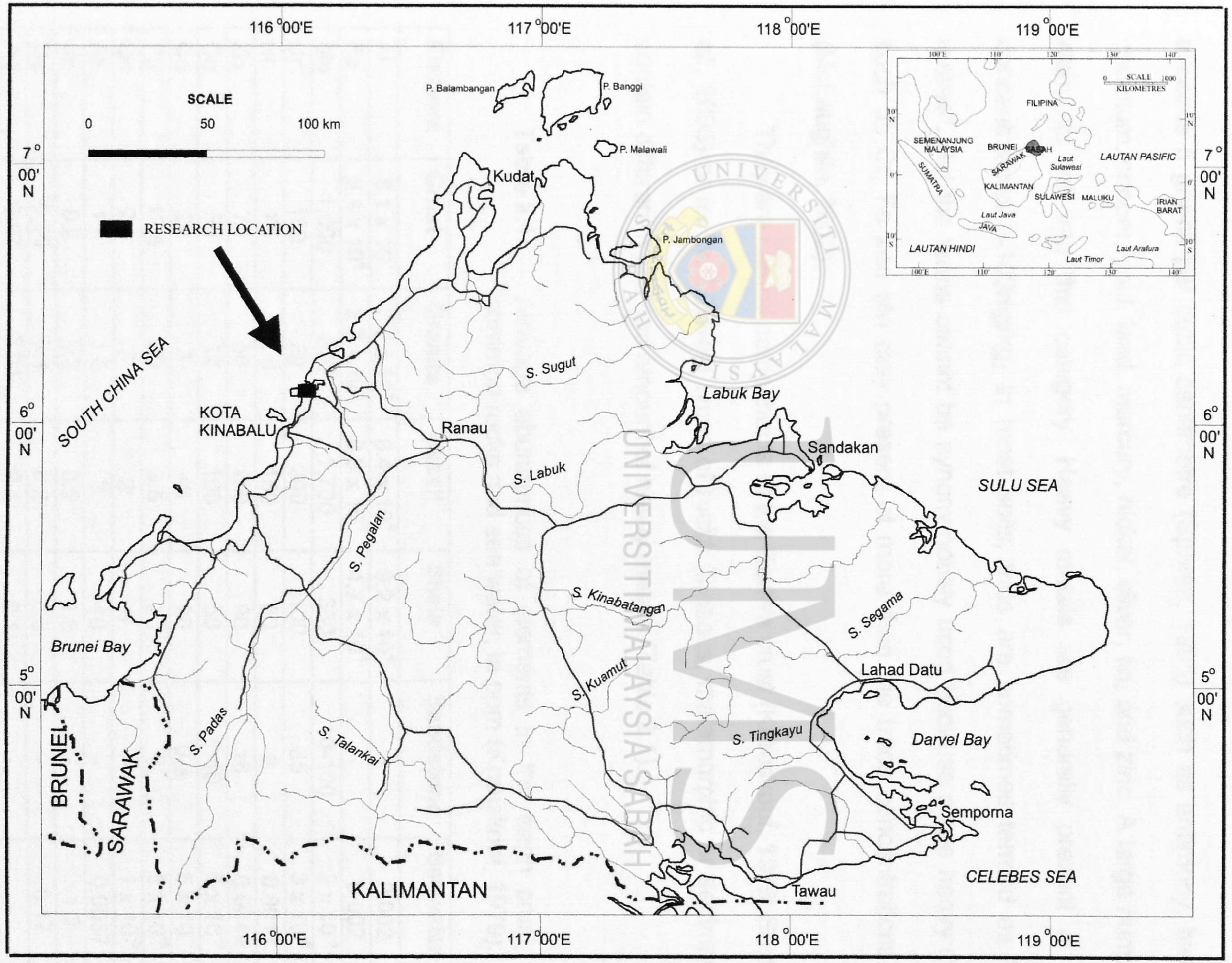

Figure 1.1 Location map of the research area.

CHAPTER 2

LITERATURE REVIEW

2.1 Heavy Metals

Heavy metals are metallic element with relatively high atomic mass which density exceeds 5 grams per cubic centimetre (Sparks, 1995) such as antimony, bismuth, cadmium, copper, gold, lead, mercury, nickel, silver, tin, and zinc. A large number of elements fall into this category. Heavy metals are generally present at trace concentrations (<100mg/kg) in most soils, thus are sometimes termed as "trace metals". But the terms cannot be synonymously used because some heavy metals such as Cr, Fe and Mn may present at more than the trace concentrations level (McLaughlin, 2002).

They are natural components of the Earth's crust (Krauskopf, 1979; Santos *et al*., 2005) or rocks. Different types of rocks (igneous, metamorphic or sedimentary) contain different types and amount of heavy metals (Table 2.1).

Table 2.1: Average abundances of elements in the earth crust, three common rocks and sea water, in ppm (Krauskopf, 1979).

Element	Crust	Granite	Basalt	Shale	Sandstone	Seawater	Sediments (Sparks, 1995)
Al	8.1×10^4	7.7×10^4	8.4×10^4	9.2×10^4	-	0.002	7.2×10^4
Fe	5.4×10^4	2.7×10^4	8.6×10^4	4.7×10^4	-	0.002	4.1×10^4
Mn	1,000	500	1,700	850	10-100	2×10^{-4}	770
Cr	100	20	200	100	35	3×10^{-4}	72
Ni	75	0.8	150	80	2	0.0017	52
Zn	70	50	100	90	16	0.0049	95
Cu	50	12	100	50	1-10	5×10^{-4}	33
Co	22	3	48	20	0.3	5×10^{-5}	14
Pb	12.5	20	3.5	20	7	3×10^{-5}	19
Sn	2.5	3	2	6	-	1×10^{-5}	4.6
As	1.8	1.5	2	10	-	0.0037	7.7
Sb	0.2	0.2	0.2	1.5	-	1.2	2.4×10^{-4}
Cd	0.15	0.1	0.2	0.3	-	0.17	1×10^{-4}
Ag	0.07	0.04	0.1	0.1	-	-	4×10^{-5}
Se	0.05	0.05	0.05	0.6	0.05	0.42	2×10^{-4}
Hg	0.02	0.03	0.01	0.3	-	0.19	3×10^{-5}

Heavy metals weathered from these rock formations spread widely in the environment, occurring in particulate or dissolved form in soils, rivers, lakes, seawater and sea floor sediments. Other sources of heavy metals are volcanoes where these heavy metals are being released into the atmosphere.

Background levels of heavy metals usually reflect the composition of the parent rock materials. However the background levels are sometimes difficult to determine because of anthropogenic inputs (McLaughlin, 2002).

Industries such as mining and ore processing, metallurgical procedures (e.g. electroplating), paper manufacturing, petroleum refining etc. may cause heavy metals such as cadmium, lead, tin, plutonium and mercury be released to the environment (Murck *et al.*, 1996). Some of the primary and secondary sources of heavy metals contaminations are shown in Table 2.2.

Table 2.2: Sources of heavy metal contamination in soils (McLaughlin, 2002).

Source	**Main heavy metals**
Primary Sources	
Fertilizers	Cd, Cu, Mo, Pb, Zn
Irrigation water	Cd, Fe
Manures and composts	Cd, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb, Zn
Pesticides	Cu, Hg, Pb, Zn
Sewage biosolids (sludges)	Cd, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb, Zn
Soil amendments (lime, gypsum, etc)	Cu, Mn, Pb, Zn
Secondary sources	
Automobile aerosols	Pb
Coal combustion	Pb
Mine waste and effluents	Cd, Cu, Fe, Hg, Mn, Ni, Pb, Zn
Nonferrous smelter waste	Cd, Cu, Hg, Mn, Ni, Pb, Zn
Paint dispersal	Cd, Pb
Tire wear	Cd, Zn
Waste combustion	Cd, Pb